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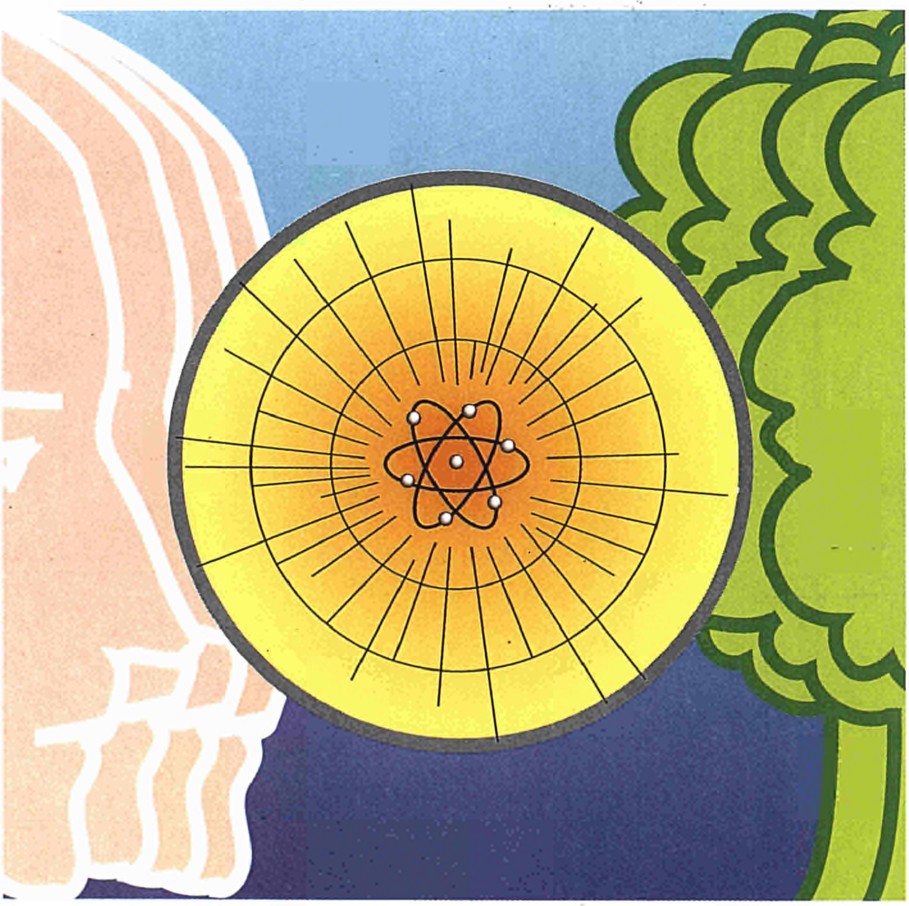
European Commission

nuclear science and technology

Annual progress report 1996 on exploring innovative approaches, reactor safety, radioactive waste management and disposal and decommissioning research areas of the 'Nuclear fission safety' programme 1994-98



Report

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Annual progress report 1996 on exploring innovative approaches, reactor safety, radioactive waste management and disposal and decommissioning research areas of the 'Nuclear fission safety' programme 1994-98

Directorate-General
Science, Research and Development

1997

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FOREWORD

The Council of the European Union adopted with its decision 94/920/Euratom of 15 December 1994 (O.J. N° L 361 dated 31 Dec. 1994, pp. 143-145) the specific programme "Nuclear Fission Safety 1994-1998" under a framework programme of Community activities in the field of research and training for the European Atomic Energy Community (Euratom) 1994-1998[1].

The specific programme "Nuclear Fission Safety" which has a total budget of 170.5 million ECU consists of five research areas: A-Exploring Innovative Approaches; B-Reactor Safety; C-Radioactive Waste Management and Disposal and Decommissioning; D-Radiological Impact on Man and the Environment; E-Mastering Events of the Past.

This report covers the progress of the 62 multi-partner research projects running in 1996, for the area A, B and C of the programme, under the responsibility of European Commission Unit XII/F/5 "Nuclear Fuel Cycle, Radioactive Waste".

The running contracts are those resulting from the selection procedure of the proposals received at the first deadline of the call for proposals on shared cost projects and concerted actions.

Cooperation among teams within the Member States is assured through the presence of at least two partners from different countries.

The programme also includes awards of training and mobility grants, as well as international cooperation agreements and concerted actions with states outside of the European Union.

The European Commission is assisted in the implementation and the management of the programme by the Consultative Committee for the Programme (see the annexed list Committee members).

A total of 20 "clusters" have been set up for the areas A, B and C to assure the best follow up of groups of projects covering special items of the programme.

An introduction to each area of research gives a general overview of the objectives and items being developed. The objectives, the working programme and a synopsis of progress and results achieved for each contract in 1996 are presented as prepared by the contractors, under the responsibility of the project coordinator.

The European Commission wishes to express its gratitude to all scientists who have contributed to this report.

R. Simon
Programme Manager for the Areas A, B and C

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[1] Council decision 94/268/Euratom of 26 April 1994 - O.J. N° L 115 of 6 May 1994, pp. 31-37. This Euratom framework programme is linked to activity one of the Fourth Framework Programme of the EC. RTD activities 1994-1998 (Co-decision N° 1100/94/EC of the European Parliament and of the Council of 26 April 1994-O.J. N° L 126 of 18 May 1994).

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[1]This Committee was established by the Council Decision 94/920/Euratom of 15 December 1994, Art. 5, Par. 2 (O.J. N° L361 pp. 143-145)

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PROGRESS OF THE R&D WORK

on:

A - Exploring Innovative Approaches

B - Reactor Safety

C - Radioactive Waste Management and Disposal and Decommissioning

AREA A

EXPLORING INNOVATIVE APPROACHES

A. EXPLORING INNOVATIVE APPROACHES

INTRODUCTION

The activities in this area are aimed at exploring and developing new concepts for further improving the safety of nuclear reactors and the fuel cycle, including innovative strategies for the reduction of the radiotoxicity of nuclear waste. They should also provide a basis for industry to select those features which best reply to the development trends of future nuclear installations.

The following research tasks are being undertaken:

A.1 Conceptual Reactor Safety features

Assessment of new, especially passive and inherent conceptual safety features with regard to their feasibility and their contribution to the overall safety of a nuclear plant.

A.1.1 Passive decay heat removal : problems of natural convection, flow stability, stratification and mixing due to small driving forces, specific heat transfer problems, influence of incondensable gases. Special emphasis on structural integrity aspects. Code development and validation.
A.1.2 Passive safety measures : initiation, depressurisation and injection measures e.g. self-powered or self-acting valves, steam injection pumps.

A.2 Fuel Cycle Concepts - Partitioning and Transmutation (P&T)

Investigation of alternative fuel cycle concepts with regard to waste minimisation, actinide burning and safeguards. Evaluation of the impact of P&T on waste management, contribution to the development of new separation techniques, assessment of different transmutation techniques from the point of view efficiency and safety.

A.2.1 Strategy studies : potentialities of different fuel and P&T strategies on the use of resources, the reduction of the inventory of long-lived radionuclides and waste production. Studies of partitioning and transmutation scenarios, waste conditioning, storage and disposal. Assessment of different devices for transmutation. Studies to investigate fission product retention capabilities of the fuel, especially with regard to direct storage.
A.2.2 Partitioning techniques : flow sheets to separate actinides including the stability of extractants to radiation, of various aspects of P&T application e.g. secondary waste, additional shielding requirements for fuel and waste handling, proliferation.
A.2.3 Transmutation techniques : Investigation of fuel/target fabrication problems, of the physics and safety aspects of different transmutation devices.

For each of the two sub-areas a "CLUSTER" has been created, where projects on the same topic are grouped:

- A.1- Conceptual reactor safety features - INNO;
- A.2- P&T strategy studies and transmutation experiments - TRANSMUT.

A.1.1-1 Thermalhydraulics of large pools with immersed heat exchangers and natural convection heat transfer - POOLTHY

Contract No: FI4I-CT95-0003 **Duration :** 1 Jan. 1996 - 31 Dec. 1998
Coordinator : D. Tenchine, CEA, Grenoble/FR
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Partners : FZK Karlsruhe/DE, Univ. "Victoria" Manchester/GB, Univ. "La Sapienza" Roma/IT

A. OBJECTIVES AND SCOPE

New safety requirements for the next generation of nuclear power plants are formulated. Among them, the use of passive decay heat removal systems has to be investigated, as for:

- the secondary condensing systems (SCS) with condensers immersed in pools
- the sump cooling through immersed heat exchangers and condensers after core meltdown, in case of a severe accident
- the emergency core cooling systems coupling immersed heat exchangers and condensers in a passive circuit

Based on separate effect experimental studies, the POOLTHY project will provide a data bank for code validation. The following aspects will be addressed : efficiency of immersed heat exchangers operating at low pressure in a two-phase flow regime, natural convection in pools with local boiling conditions, efficiency of condensers for steam/gas mixtures, and coupling of the previous processes in a passive circuit.

B. WORK PROGRAMME

B.1 Immersed Heat Exchangers at low pressure

The EPICE test facility will be used to investigate the thermalhydraulic behaviour of immersed heat exchangers operating at low pressure in two-phase natural circulation. The expected information from the tests is : (i) heat transfer correlations along a vertical tube immersed in a pool, for the various two phase flow regimes, (ii) critical heat flux as a function of the steam quality, and (iii) interaction between two phase heat transfer process along the vertical tube and natural circulation in the pool.

B.2 Pool natural convection with local boiling conditions

The SUCOT test facility will be used to investigate the short term two-phase phenomenology of the natural circulation within a flooded spreading compartment. Several separate effect tests under steady state and transient conditions will be performed to investigate : subcooled and saturated pool boiling, flow aspects (pattern, flashing, instabilities), formation and collapse of bubbles and influence of non-condensable gases on boiling behaviour. A data base for development and validation of advanced multi-dimensional computer codes will be elaborated.

B.3 Condensation heat transfer for steam/gas mixtures

Several separate effect tests will be performed at the MUCON facility to investigate the effects of geometry and arrangement of heat transfer surfaces on the efficiency of vapour removal by condensation from flowing steam mixed with known quantities of air. Methods of enhancing the condensation process by means of extended surfaces or mixing devices will also be examined.

B.4 Coupling of previous processes

In this task the global thermal hydraulic behaviour of the passive Emergency Core Cooling System (ECCS) of the innovative MARS reactor will be investigated. A test facility will be designed and constructed to this end (QUSCOCS), which will be used to execute a tests matrix previously defined. The experimental data will be available for the validation of computer codes applied to passive heat removal through heat exchangers immersed in a pool and condensation of steam-gas mixtures.

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The four work-packages of the POOLTHY project are concerned with new test facilities to provide a data bank for code validation.

The first phase of the project has consisted in building the four test facilities. At the end of 1996, the situation is the following :

- the construction of the EPICE, SUCOT, MUCON and QUSCOCS test facilities is completed or nearly completed
- the final design reports of the four test facilities are produced
- the commissioning tests are performed from December 1996 to March 1997 for the four test facilities
- the testing phase is planned to start from January 1997 for SUCOT (first one) to April 1997 for EPICE (last one).

The delay of few months during the construction phase is not critical as the 3 years project is sufficient to perform the test program.

C.1 Immersed heat exchangers at low pressure

The EPICE test facility is described in the EPICE design report (Ref. [1]). The EPICE 1 tests will be performed with an electrical heated pin and the EPICE 2 tests will be performed with a heat exchanger tube (Figure 1).

The detailed design was achieved by mid 1996 and the construction of the EPICE 1 test facility will be completed by March 1997.

The EPICE circuit is including a production of demineralized and deaerated water to fill the main circuit. The fabrication of the various components of the circuit was achieved by October 1996.

The main difficulty was the realization of the EPICE 1 test facility with the fabrication and the instrumentation of the electrically heated pin, and the instrumentation of the glass made housing tube. These elements were successfully installed in December 1996. A specific instrumentation is mounted along the heated pin to detect rapidly the dry out situation to prevent the destruction of the pin.

At the end of 1996, the construction of the EPICE 1 test facility is nearly completed. During February and March 1997, the electrical connections, the insulation and the installation of the measurement lines will be realized.

The commissioning tests are scheduled for March 1997 and the testing phase on EPICE 1 test section should start in April 1997.

C.2 Pool natural convection with local boiling conditions

The SUCOT test facility is described in the SUCOT design report (Ref. [2]). The SUCOT test section is a volumetrically similar representation of the core melt spreading compartment at a power scale of 1:356 (figure 2).

The building up of the SUCOT test section was achieved by July 1996, as for the cooling circuits the installation was finished by September 1996.
An original aspect of the SUCOT experiment is that the front wall and the back wall of the test facility are made of large glass windows allowing for flow visualization. Various flanges are provided to introduce instrumentation.
An important part of the work consisted in the preparation of the instrumentation : thermocouples, Laser Doppler Anemometer, impedance probes, optical probes. An associated data acquisition system was developed and installed by November 1996. This instrumentation will allow the local measurement of the velocity and temperature fields, the void fraction and the bubble sizes.
At the end of 1996, the commissioning tests are in progress on the SUCOT experiment. The testing phase will start early in 1997, by performing steady state tests.

C.3 Condensation heat transfer for steam/gas mixtures

The MUCON test facility is described in the MUCON design report (Ref. [3]). The MUCON test section is installed on a previous facility to measure the efficiency of condensation on staggered horizontal, inclined or vertical tubes or plates (Figure 3).
The design, construction and commissioning of the modified loop were carried out during the period January 1996 to September 1996. A new circulation pump and a new condenser were chosen to replace the existing one. In parallel with this some refurbishment work was carried out : cleaning the degassing column, the boiler and associated pipework, checking the electrical systems.
The new test section and the associated air supply system were designed during the same period. The test section is made from a pyrex vessel which allows visualisation of the condensation behavior on the cooled plates. The instrumentation is mainly composed of flowmeters to measure the different flowrates (steam, gas, condensate, cooling water), and thermocouples mounted on the condensing plates and at the inlet and outlet of the test section to evaluate the efficiency of the condensation process.
The construction of the test section is in progress at the end of 1996, as for the air supply system the work will start early in 1997.
The commissioning tests will commence during February 1997 and the testing phase is scheduled to begin in March of April 1997.

C.4 Coupling of previous processes

The QUSCOCS test facility is described in the QUSCOCS design report (Ref. [4]). The QUSCOCS test section consists of a transparent tank with immersed bended tubes of different materials and with various angles (Figure 4).
The detailed design of the test facility and the purchasing phase are completed at the end of 1996. Most of the components and materials are available.
Some problems have been encountered during the manufacturing of the polycarbonate transparent tank, and this component has been re-manufactured. This tank has a maximum content of 100 liters of water, open at the upper side to maintain the atmospheric pressure and with all the connections located at the bottom side. Different types of tubes, 1 m long, may be assembled in the tank. The tubes to be tested will be bended with different angles ;

different materials will be used and tests to evaluate the effect of surface finishing are also scheduled.
The instrumentation has been purchased, mainly thermocouples and flowmeters to establish the heat transfer efficiency.
The construction of the test section has started in November 1996 and the assembly of the experimental facility will be completed in February 1997.
After the commissioning tests planned for February 1997, the testing phase will start in March 1997.

References

[1] BERTHOUX, M, INNO-POOLTHY Report P005 (1996)
[2] KNEBEL, JU, INNO-POOLTHY Report P003 (1996)
[3] JACKSON, JD, INNO-POOLTHY Report P002 (1996)
[4] CARUSO, G and NAVIGLIO, A, INNO-POOLTHY Report P004 (1996)

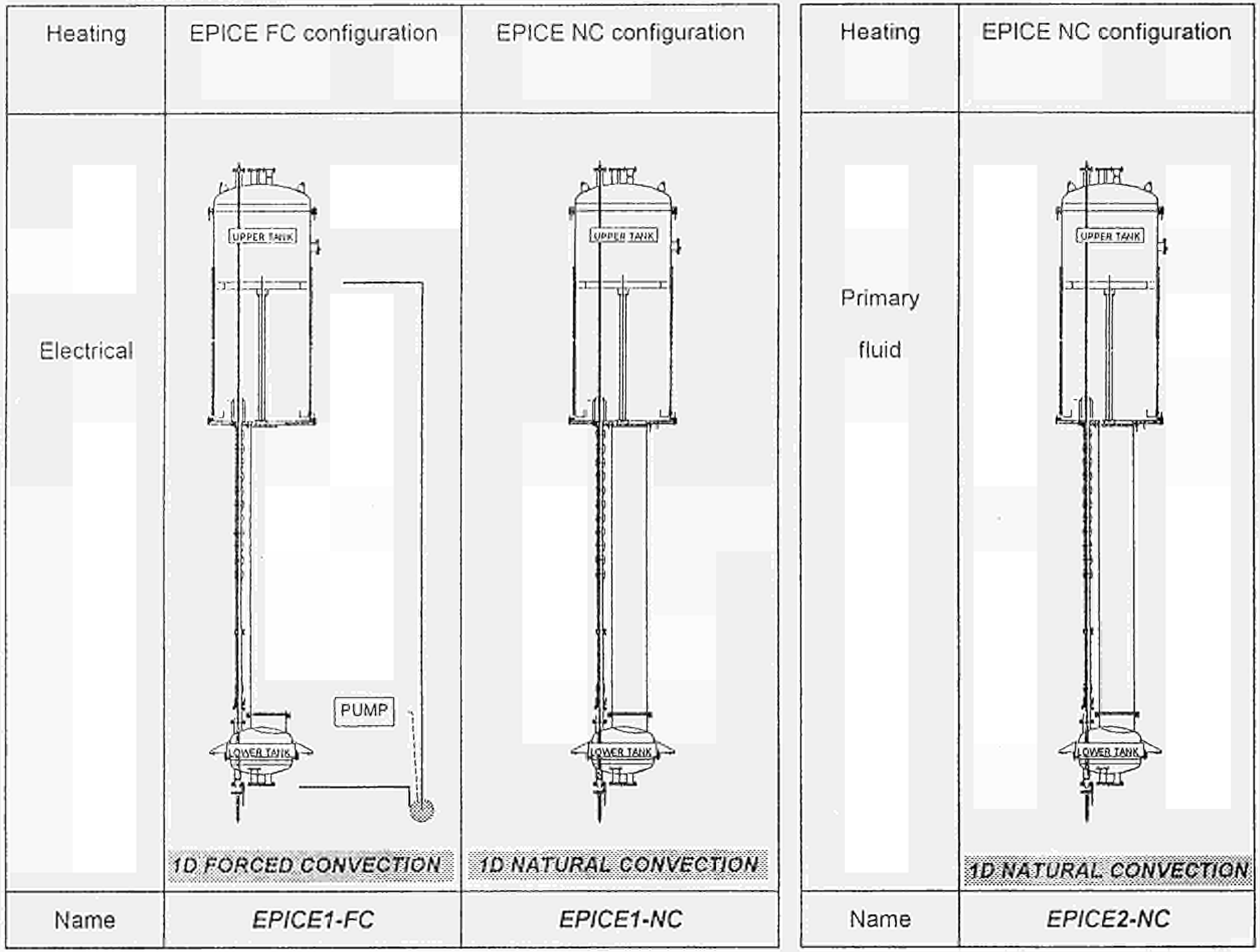

Figure 1 : EPICE Experiments

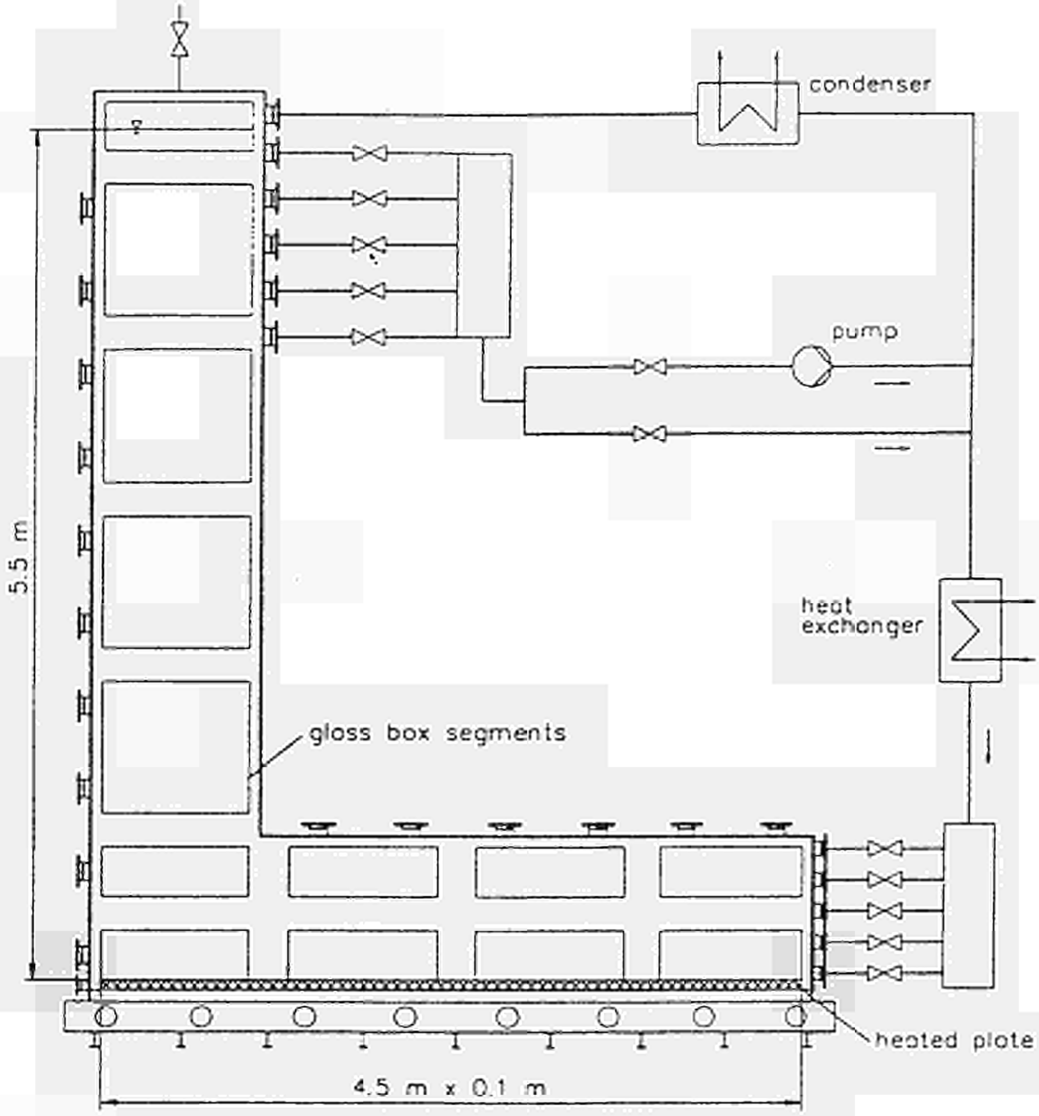

Figure 2 : Sketch of the SUCOT test facility at the Forschungszentrum Karlsruhe

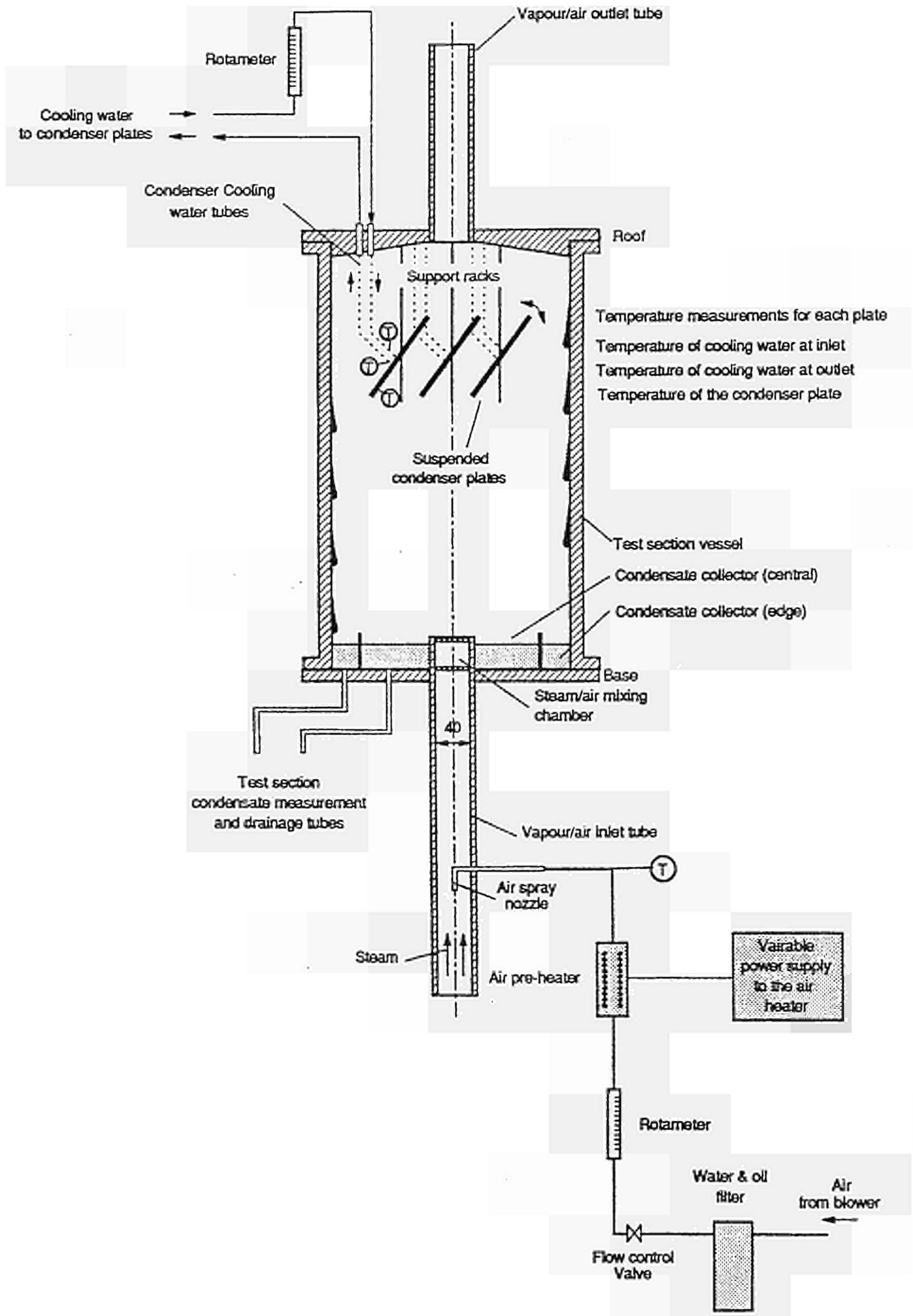

Figure 3 : Test section and air supply and mixing arrangement

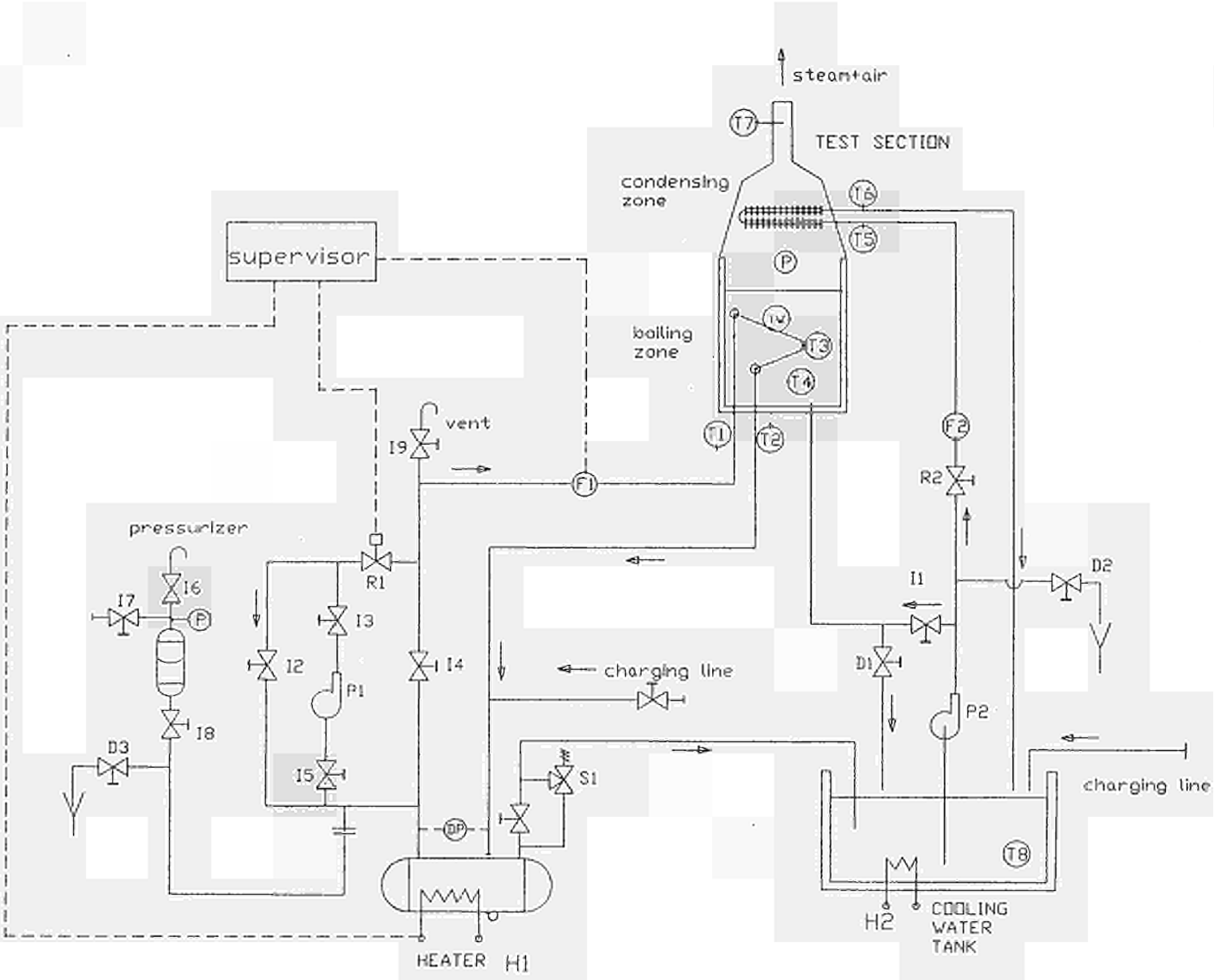

Figure 4 : QUSCOCS test facility

A.1.1-2 Assessment of passive safety injection systems of advanced light water reactors - APSI

Contract No: FI4I-CT95-0004 **Duration :** 1 Jan. 1996 - 31 Oct. 1998	
Coordinator :	**J. Tuunanen, VTT Energy, Espoo/FI**
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A. OBJECTIVES AND SCOPE

In many Advanced Light Water Reactors (ALWRs), pump driven Emergency Core Cooling Systems (ECCS) are replaced by gravity driven passive safety injection systems. The performance of these passive systems in accidental conditions, particularly Loss of Coolant Accident (LOCA), must therefore be investigated in order to assure that it will achieve the desired functions.

This project is based on experiments carried out in the PACTEL integral test facility on the performance of a passive core make-up tank (CMT). Of particular interest are the phenomena occurring in the CMT, such as possible rapid condensation and temperature stratification of water. The work to be done consists of :

- a review of ALWR thermal-hydraulic phenomena of interest and the evaluation of current system code capabilities in modelling them;
- the execution of experiments with gravity driven core cooling (CMT) and the analysis of the experimental data;
- the validation of computer codes (APROS, CATHARE, RELAP5)

B. WORK PROGRAMME

B.1 Review of thermal-hydraulic phenomena and evaluation of system code capabilities

Based on the existing experience, the applicability of available computer codes for modelling passive safety injection systems, will be reviewed, and the results will serve as a basis for the specification of the test parameters.

B.2 Experiments on passive safety injection systems performance

The experimental part of the project will be performed at the PACTEL facility. Since PACTEL does not model any of the proposed passive ALWR designs, the investigation will focus on phenomenology, and the CMT will be used to simulate the gravity driven flow to the primary system. Three series of tests are planned to investigate the effects of break sizes and location, the CMT size, and the CMT elevation. The experimental data produced will be provided to the code analysts.

B.3 Computer code analyses, including feedback from code users to experimenters

After each series of experiments, selected tests will be analyzed with computer codes like APROS, CATHARE and RELAP5. The main phenomena to be analyzed are the condensation and temperature stratification occurring in the CMT.

B.4 Preparation of final report

This report will collect the findings from the experiments and code analysis, and will also propose a methodology for consistent code assessment.

C. Progress of work and results obtained

Summary of main issues

The project has followed the schedule as planned. See Table I for the progress made in the project within the first year. The project group wrote a summary report of experiments and analyses of passive safety injection systems (work package 1). The report has been submitted to the commission and to the partners of the project. The experiment team has run two series of experiments on PACTEL. The results of the first series have been published and submitted to the commission and to the partners of the project. The analysts of Lappeenranta University of Technology (as a subcontractor of VTT Energy), University of Pisa and AEA Technology have calculated experiment GDE-24 with APROS, CATHARE and RELAP codes. The results of analyses are ready for and will be submitted to the commission early 1997. The project group had three meetings. VTT Energy organised a kick-off meeting in Lappeenranta, Finland, February 7th, 1996. The University of Pisa organised the second meeting in Pisa, Italy on June, 18th, 1996. The University of Pisa hosted also the third meeting on November 22nd, 1996. The project group visited SPES test loop in Piasenza the day before the meeting. The project co-ordinator has submitted the minutes of the meetings, and a short overview of the visit to SPES, to the partners of the project and to the commission. Table II presents the planned activities for the next reporting period. The computer code simulations of the second test series will start after delivery of the quick look report of the second series. The experiments will continue in August, 1997.

Progress and results

1. Review of thermal-hydraulic phenomena and evaluation of current system code capabilities

The objective of the first work package of the project was to carry out a **review of thermal-hydraulic phenomena and evaluation of current system code capabilities important in the modelling of passive safety injection systems**. In the kick-off meeting in Lappeenranta, the project group decided to expand the contents of the report to include also a review of experiment work done. The report of the first work package is ready and the co-ordinator has distributed it to the partners and to the commission.

2. Experiments on passive safety injection system performance

The objective of the second work package of the project is to **perform a relevant set of experiments with gravity driven core cooling, to analyse the experimental data, and to prepare qualified experiment reports**. The experiment work consists of three series of five experiments each. VTT Energy and Lappeenranta University of Technology completed the first and second series within the first year. The experiments simulated cold leg small break LOCA's. The series included SBLOCAs with four different break sizes and three different break positions. In addition, experiments studied the reproducibility of the phenomena and the influences of the flow distributor (called sparger) on the condensation phenomena in the CMT. VTT Energy has analysed the first experiment series and delivered the experiment data report to the partners of the project and to the commission. The analyses of the experiment data of the second experiment series will start January, 1997. The experiment group has also measured flow resistance of the passive safety injection system lines. This data forms an important boundary condition in the computer simulations of the experiments.

The Passive Safety Injection System (PSIS) studied consisted of a Core Make-up Tank (CMT) and two pipelines. The Pressure Balancing Line (PBL) connected the CMT to one cold leg. The Injection Line (IL) connected the CMT to the downcomer. The break size affected on the CMT behaviour as following:

- the recirculation phase was longer; and the resulting hot liquid layer, thicker in the experiments with smaller break size;
- the oscillation phase between the injection and recirculation phases was longer in the experiments with small break size; and
- local wall heat flux to the CMT wall was higher in the experiments with larger break size.

In all experiments the CMT ran as planned. There were no problems with rapid condensation in the CMT, such as was seen in the earlier passive safety injection experiments in PACTEL. The main reason was the new CMT arrangement, with a flow distributor (sparger) installed to the CMT. The sparger spread incoming flow to the CMT horizontally, and the breakdown of the saturated water layer due to incoming water was not possible. The hot liquid layer between the steam and cold water in the CMT remained stable, even in the experiments where the hot liquid layer was less than 5 cm thick. The experiments examined the CMT behaviour up to 5 millimetre break diameter. This corresponds to about 9 cm break diameter in the reference VVER-440 plant. Investigations of condensation phenomena in the CMT with larger break sizes would be of great value. When the break size increases, decreases the thickness of the hot liquid layer in the CMT. In large break LOCAs, the hot liquid layer may not exist. The water in the CMT has a large condensation potential. Condensation and resulting water hammer may occur in the PBL or in the top of the CMT. This may delay or disturb injection from the CMT.

The density difference between the IL and the PBL drives the CMT flow. If this density difference disappears, the CMT flow stops. The density difference may disappear in different ways. First, it may happen during SBLOCAs if the recirculation phase is so long that the CMT becomes full of hot water before the injection phase starts. Second, cold water may flow to the PBL during injection in LOCAs, if the flow in the loop having PBL connection reverses. The density difference may also disappear during the normal operation, if the IL check valve leaks or if the PBL heating fails. The first case above requires a very small break size, but all the CMTs will be affected. In the other cases, the driving force disappears only in one CMT. In all cases, the fact the PBL becomes full of hot water may disturb safety injection from the CMT.

The experiments of the second series focused on the break location influences on the CMT behaviour and possible flow reversal in the broken cold leg. If the flow reverses, cold water may flow from the downcomer to the CMT through the PBL. This may lead to condensation in the CMT. Experiment GDE-34 started with CMT and PBL full of hot water. The CMT may become full of hot water during normal operation of the plant if IL valve leaks. In GDE-35 experiment, the objective was to study influences of sparger on the CMT behaviour. All experiments used smaller CMT than in the first series. This made possible to study CMT size (scaling) effects on the primary system behaviour. The use of smaller CMT influenced only on the length of the injection phase of the CMT operation. The experiments did not include significant reversed flow in the broken cold leg. No condensation due to flow of cold water from downcomer to the CMT occurred. The CMT operated without problems also in the experiment GDE-34, where the CMT was initially full of hot water. There were no recirculation phase of CMT operation in the GDE-34 experiment, but the CMT started to inject when the water-level in the cold leg dropped below the PBL connection. The sparger did have a significant influence on CMT behaviour. The operators had to terminate the experiment GDE-35 when strong condensation in the CMT collapsed the pressure in the tank.

3. Computer code analyses, including feedback from code users to experimenters

The objective of the third work package of the project is to **analyse a relevant set of experiments with APROS, CATHARE and RELAP5 codes**. The purpose of the analyses is to **evaluate the capability of existing computer codes on the simulation of passive safety injection systems, and to provide feedback to the experimenters**. The main phenomena of interest in the simulations of experiments were

a) thermal stratification in the CMT (temperature profile; formation of hot liquid layer),
b) condensation in the CMT (wall condensation; condensation to water),
c) heat transfer from hot liquid to the CMT wall,
d) CMT injection flow (possible oscillations), and
e) influences of CMT in the overall system behaviour (core heat-up; primary pressure).

In addition, the project manager asked the analysts to make suggestions for

f) possible model improvements to the codes,
g) modelling the CMT (nodalization of the CMT; description of the walls of the CMT), and
h) modified or additional instrumentation for PACTEL.

3.1 Activity 1: Validation of the APROS code

Lappeenranta University of Technology, as a subcontractor of VTT Energy, has completed the input preparation and the simulation of the GDE-24 experiment with the APROS code. The documentation of the analyses results is ready and will be distributed early 1997. APROS code calculated successfully the first part of the simulated transient i.e. the CMT recirculation phase. Major problems occurred when the draining mode began. The injection flow started to oscillate continuously never reaching the full magnitude. The injection with full power seemed to be possible only when saturated water was present in the boundary node. The explanation for these flow oscillations was the lack of stable saturated liquid layer which would prevent direct contact of vapour and cold water. The reason for this was numerical diffusion, which transferred energy from top to bottom nodes in the CMT. A test run with reduced condensation in the CMT showed that it is possible to reduce flow oscillations by reducing condensation rate in the CMT. The condensation heat transfer coefficient was reduced by using a hydraulic diameter of 10 metres in the upper CMT nodes. Hydraulic diameter is an important parameter in the Shah correlation used in APROS to calculate condensation heat transfer. In the experiments, the thickness of the thermally stratified layer depended on the break size. When the break was smaller, the recirculation mode became longer and the layer of hot water thicker. The calculations showed that it was necessary to use dense nodalization in the CMT. None of the used nodings could, however, solve the oscillation problems. If there would be a solution with a certain node size for a particular break size, a common model for any break size can not be introduced. So, the CMT would need special treatment to ensure reliable calculation models. One solution could be a special module with dense moving mesh around the water surface. Nevertheless, this method relies on the basis of using nodes and can still lead to numerical diffusion. The ideal solution to avoid numeric diffusion and flow oscillations would probably be a separate continuous model to solve the temperature distribution in the CMT. However, even without a special CMT model the APROS code was able to estimate the overall behaviour and the main phenomena of the PSIS in the simulated SBLOCA.

3.2 Activity 2: Validation of the CATHARE code

University of Pisa has completed the input preparation and the simulation of the GDE-24 experiment with the CATHARE code. The documentation of the analyses results is ready and will be distributed early 1997. The CATHARE code succeeded to simulate the overall behaviour of the passive safety injection system, although the code predicted too early core heat-up and too high CMT flow rates. Differences between the measured and calculated values were to great extend due to well known reasons. Due to large computing time (typically 4 to 5 days for one simulation), some of the discrepancies were not yet corrected. For more accurate simulation, adjustment of the pressure losses of the CMT lines, heat losses to the environment and modelling of secondary side of steam generators would be necessary.

3.3 Activity 3: Validation of the RELAP5 code

The AEA Technology has completed the input preparation and the simulation of the GDE-24 experiment for the RELAP5 code. The documentation of the analyses results is ready and will be distributed early 1997. A RELAP5 mod 3.2 deck for PACTEL has been modified for the calculation of test GDE-24. The deck has been run to provide an initial steady state and perform the pre-transient part of the GDE-24 test, successfully providing initial conditions for the transient. Two full calculations have been made. Both represented the full transient. In the first calculation there was a significant fall of pressure which occurred as the core make-up tank started to empty. In the second calculation the rate of condensation in the core make-up tank was artificially reduced and it was found that the results followed the experimental data almost precisely. This highlights some inadequacy in the wall condensation modelling in the RELAP5 code. A study of how the code calculated the heat transfer coefficients revealed the deficiency in the model, the principle feature being that the thickness of the film of condensation on the wall is not modelled realistically.

Stand-alone modelling of the CMT indicated:

(a) that replacing the mesh in the CMT wall, over a practical range of sizes, produced little benefit. The implication is that it may be difficult to get the resolution necessary using a system code like RELAP5.

(b) flow oscillations could be reduced by refining the volumetric mesh, but it was not possible to eliminate them. Again this is a limitation of the approach inherent in system codes.

(c) the results were not sensitive to the noding refinement in the PBL. This is comforting because of practical limitations of running a complete rig model with fine noding in the PBL.

The implication of this work is that, apart from the wall condensation modelling issue, the modelling in RELAP5 is broadly adequate for this application. As part of this analysis some observations on the rig instrumentation have been made:

(a) some improvement in the mounting of the thermocouples employed for the heat flux determination in the CMT wall is desirable to ensure that the results are truly representative of the body of the CMT

(b) a means of determining, or estimating, the water film thickness would be very beneficial. Possibilities might involve ultrasonic techniques or laser interferometry noting that in practice that film thickness could be significantly less than 1mm.

Table I: Project management tasks, documentation (D1-D10) and milestones (M0-M6). Completed activities are presented in *italic*.

Tasks & milestones	Deliverables	Responsible organisation	Actual completion date or deadline (day or week/year)
Start of the project (M0)			*1.1.1996*
Work Package 1			
Review of thermal-hydraulic phenomena of interest in passive safety injection systems & evaluation of current system codes capabilities in their modelling (M1)	*Summary report (D1)*	*VTT, AEA and Univ. of Pisa*	*28.6.1996*
Work Package 2			
Test specification of the first series (M2)	*Test specification report (D2)*	*VTT*	*16.2.1996*
Prepare PACTEL facility for the first test series (M3) *-design and constr. of passive safety injection system (CMT)* *-modification of the instrumentation* *-calibration and testing of equipment*	*Facility ready for the 1st test series*	*VTT & LUT*	*14.3.1996* *21.3.1996* *28.3.1996* *28.3.1996*
First test series (M4)	*Experimental data*	*VTT & LUT*	*3.5.1996*
Deliver test data to code users (M5)	*Test data (D3)/first set (full data)* *Quick-Look report (D4)*	*VTT*	*18.6 (15.7) 1996* *18.6.1996*
Document and analyse test results (first series) (M7)	*Experimental data report (D11)*	*VTT*	*7.10.1996*
Test specification of the second series (M8)	*Test specification report (D12)*	*VTT*	*31.10.1996*
Prepare PACTEL facility for the 2nd test series (M9) *-design and constr. of passive safety injection system (CMT)* *-modification of the instrumentation* *-calibration and testing of equipment*	*Facility ready for the 2nd test series*	*VTT & LUT*	*13.11.1996* *6.11.1996* *13.11.1996* *13.11.1996*
Second test series (M10)	*Experimental data*	*VTT & LUT*	*11.12.1996*
Deliver test data to code users (M11)	Test data (D13) Quick-Look report (D14)	VTT	7/97
Document and analyse test results (2nd series) (M12)	Experimental data report (D15)	VTT	22/97
Test specification of the third series (M14)	Test specification report (D19)	VTT	31/97
Prepare PACTEL facility for the 3rd test series (M15) -design and constr. of passive safety injection system (CMT) -modification of the instrumentation -calibration and testing of equipment	Facility ready for the 3rd test series	VTT & LUT	31/97 32/97 33/97 33/97
Third test series (M16)	Experimental data	VTT & LUT	39/97
Reconstruct the test facility (M17)		VTT & LUT	41/97
Deliver test data to code users (M18)	Test data (D20) Quick-Look report (D21)	VTT	45/97
Document and analyse test results (3rd series) (M19)	Experimental data report (D22)	VTT	13/98
Work Package 3			
Prepare PACTEL input for APROS *code (M5)*	*Input data file (D5)*	*VTT*	*1.8.1996*
Prepare PACTEL input for CATHARE code (M5)	*Input data file (D6)*	*Univ. of Pisa*	*15.6.1996*
Prepare PACTEL input for RELAP5 code (M5)	*Input data file (D7)*	*AEA*	*1.8.1996*
Document results of analyses of the first test series with APROS code (M6)	*Code validation report (D8)* *Feedback to the experimenters*	*VTT*	*31.1.1997*
Document results of analyses of the first test series with CATHARE code (M6)	*Code validation report (D9)* *Feedback to the experimenters*	*Univ. of Pisa*	*3.1.1997*
Document results of analyses of the first test series with RELAP5 code (M6)	*Code validation report (D10)* *Feedback to the experimenters*	*AEA*	*20.1.1997*
Document results of analyses of the second test series with APROS code (M13)	Code validation report (D16) Feedback to the experimenters	VTT	26/97
Document results of analyses of the second test series with CATHARE code (M13)	Code validation report (D17) Feedback to the experimenters	University of Pisa	26/97
Document results of analyses of the second test series with RELAP5 code (M13)	Code validation report (D18) Feedback to the experimenters	AEA	26/97
Document results of analyses of the third test series with APROS code (M20)	Code validation report (D23)	VTT	18/98
Document results of analyses of the third test series with CATHARE code (M20)	Code validation report (D24)	University of Pisa	18/98
Document results of analyses of the third test series with RELAP5 code (M20)	Code validation report (D25)	AEA	18/98
Work Package 4			
Write the final report of the project (M21)	Final Report (D26)	VTT, AEA, Univ. of Pisa	30.9.1998

Table II: Planned activities for the third reporting period

Activity	Work Package	Partner	Deadline	Status & Comments
Quick look report	2	VTT	February, 15	
Data delivered to code users	2	VTT	February, 15	
Experiment data report	2	VTT	May, 30	
Input data file for APROS code (2nd series analyses)	3	VTT	February, 28	modif. to CMT only
Input data file for RELAP code (2nd series analyses)	3	AEA	February, 28	modif. to CMT only
Input data file for CATHARE code (2nd series analyses)	3	Univ. of Pisa	February, 28	modif. to CMT only
Code comp. report, APROS code (2nd series analyses)	3	VTT	June, 31	
Code comp. report, CATHARE code (2nd series analyses)	3	Univ. of Pisa	June, 31	
Code comp. report, RELAP5 code (2nd series analyses)	3	AEA	June, 31	
Deliver progress report to the Commission		VTT	January, 31	

Contract No: FI4I-CT95-0005 **Duration :** 1 Jan. 1996 - 31 Dec. 1998
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A. OBJECTIVES AND SCOPE

R&D on innovative Boiling Water Reactors (BWR) is to a large extent being focused on a broader implementation of self-acting passive safety systems for the prevention and mitigation of severe accidents. Within this project the generic thermal-hydraulic phenomena of these systems will be investigated using existing BWR-relevant large scale test facilities (NOKO and PANDA) as well as the Dodewaard Reactor (NL).

The experimental results will be used to improve models, validate different computer codes (APROS, ATHLET, RELAP5, TRAC, RALOC, etc.), and assess uncertainties with regard to passive decay heat removal (both from the core and the containment).

B. WORK PROGRAMME

B.1 Natural Convection Cooling in the RCS
Compile relevant steady state and transient data from the Dodewaard reactor operation, and compare them with (i) existing calculations (TRAC-BF, RELAP5) and (ii) calculations with the ATHLET code to be performed as part of the project.

B.2 Passive Decay Heat Removal from the Core Region
A number of tests will be performed at the NOKO (Germany) and PANDA (Switzerland) facilities in order to investigate the thermal-hydraulic phenomena in "Isolation Condensers" (IC) and "Emergency Condensers" (EC). These results, plus Dodewaard operational data will be compared with several code calculations (APROS, ATHLET, PHOENICS, RELAP5, TRAC).

B.3 Steam Jet Pumps (SJP) for Decay Heat Removal (DHR)
A conceptual study on the use of SJPs or steam injectors for DHR will be elaborated.

B.4 Passive Initiators
A number of tests will be performed at the NOKO facility in order to study the behaviour of different types of passive Initiators with different fluids and at low temperatures. The results will lead to recommendations of possible improvements and applications of passive initiators.

B.5 Passive DHR from the BWR - Containment
Integral and separate-effect tests will be conducted at the PANDA and NOKO facilities respectively, in order to assess different principles for passive DHR from the containment and the associated thermal-hydraulic phenomena. The experimental results will be compared with the calculations from different thermal-hydraulic (ATHLET, RALOC, FLOW3D) and coupled codes (RELAP/MELCOR).

C. Progress of work and results obtained

Summary of main issues

Schematic views on the main test facilities PANDA and NOKO as well as on the DODEWAARD Isolation Condenser are shown in figures 1-3. Results from the operation of these facilities offer the input data for the validation of different code systems. Several papers have been published on the European BWR R&D Cluster; a poster session together with other related European R&D activities on passive safety systems has been organized at the Annual Meeting on Nuclear Technology '96 at Mannheim (see List of References).

Although the Dodeward reactor will be shut down early in 1997 this will not influence the tasks within this project related to natural convection cooling. The exchange of geometrical and operational data has been mainly done allowing new calculations for steady state and transient operation with the ATHLET code in comparison to the codes TRAC-BF1, RELAP5.

Based on already performed test series with the emergency condenser in NOKO calculations with ATHLET have been performed with a good agreement between measured and calculated data. The experimental data base will be broadened by using experimental data from the PANDA and Dodeward ICs. Related data input decks are under preparation for ATHLET, TRAC, APROS and RELAP5. The pool side of the NOKO-tests have been calculated with PHOENICS, those for the PANDA IC-tests are underway.

The application of steam jet pumps for NPPs is investigated by 4 organisations world-wide. An assessment and proposals for beneficial use of steam jet pumps will be issued in 1997.

3 different types of Passive Initiators have been tested by FZJ with promising results. Further tests will be performed in 1997. Recommendations for improvements and applications will be derived from these experiments.

The assessment of the effectiveness of decay heat removal systems from the containment is up to now mainly based on PCC-tests in PANDA. Tests with the building condenser in PANDA and NOKO have been prepared and partially installed in 1996; the experiments will start in the 2nd quarter of 1997.

In 1996 the Concerted Action on 'BWR Physics and Thermalhydraulics - Complementary Actions to the BWR R&D Cluster (BWRCA)' could be established to further assist and broaden the objectives of this project. The partners of the BWRCA are: CEA-France, CIEMAT-Spain, ENEA-Italy, FZR-Germany, Siemens-Germany and TU-Delft-Netherlands.

C.1 Natural convection cooling in the reactor cooling system (RCS)

The ownership of the Dodewaard reactor has been taken over by the SEP company (Samenwerkende Electriciteits Productie Bedrijven); it is assessed that the scheduled shut down of the plant early in 1997 will not influence the tasks of this work package. The relevant data from the Dodewaard reactor system and related drawings have been delivered to the BWR R+D CLUSTER to allow the preparation of an input deck for the thermal hydraulic code ATHLET. In addition, data from steady operations and a comparison with TRAC-BF1 and RELAP5 calculations are given.. A report by KEMA with data from reactor transients and related calculations with the TRAC-BF1 code is being drafted. These informations form the basis for the matrix of cases to be calculated. A report on a shutdown experiment in the Dodewaard reactor with decreasing pressure causing core-wide oscillations which resulted later in a scram is under preparation. ECN has produced a report with RELAP5 calculations (performed in the late 80's) for steady state conditions and postulated transients; due to major upgrades of the Dodewaard reactor system these results have to be used carefully. The preparation of the input deck for steady state and transient calculations have been delayed; however, the final date will be met.

C.2 Passive Decay Heat Removal from the core region

Part of the experimental data base has been established. After the completion of 40 tests series with the emergency condenser bundle in the NOKO facility for the project funded by German organisations. 8 test series have been performed for the EU cluster with a 4-tube bundle arrangement. Most of the data of these tests are already available via Internet; some Quick Look Reports have been drafted. In addition, 4 tests with the so-called single tube which is placed parallel to the bundle and has a much higher instrumentation density (thermocouples and needle probes) have been performed to check the sensitivity of the needle probes and the flow pattern inside the condenser tubes. The available data from the Dodewaard Isolation Condenser have been transferred to the BWR R&D Cluster. The preparation of tests with the Isolation Condenser in the PANDA facility - planned for April 1997 - has continued with the installation of a Helium injection system (for simulating severe accidents with Hydrogen generation) and a recalibration of the instrumentation.

The experimental data of 6 tests in NOKO are shown in fig. 4. The results show that the emergency condenser works very effective. One tube is capable to transfer up to 1 MW. The heat transfer characteristics are influenced by higher concentrations of non-condensable gases. It can also be seen that the pressure losses in the bundle and in the orifices lead to large differences in the geodesic heights of the water niveaus in the bundle and in the RPV. The results have to be considered for the design of an optimized new NOKO bundle.

The NOKO facility has been modelled with ATHLET; a comparison of measured and calculated data for six tests showed a good agreement; however, the heat transfer package in ATHLET has been improved (funded by another project).

The data transfer to VTT via Internet could also be established in a way that the input deck for the APROS calculations on NOKO tests can be prepared.

Input data from the Dodewaard IC for the ATHLET nodalisation scheme has been transfered; calculations will be performed in late 1997.

The PHOENIX calculations for the pool side of the NOKO bundle have been performed and are consistant with the observed behaviour of the flow and temperature distribution in the pool.

RELAP5 and GOTHIC input decks for NOKO and PANDA-IC tests have been set up. First calculations are in good agreement with the test data.

C.3 Conceptual study on steam jet pumps application in NPP

NUKON carried out a literature study and started an assessment for different applications. SIET/ENEL (Italy), Toshiba (Japan), VNIIAES (Russia) and Ohio State University (USA) are performing R&D on steam jet pumps issues. The Russian design, e.g., is planned to be implemented in VVER 440/213 reactors as an additional decay heat removal system. Other identified applications of steam jet pumps are internal coolant recirculation in BWRs, High Pressure Core Injection Systems and feedwater injection with steam jet pumps. SIET will issue a report in the first half of 1997 on their relevant experience.

C.4 Passive Initiators (PIs)

Three different types of PIs have been tested by FZJ as shown in fig. 5. The evaluation reveiled that the reaction time was strongly dependent on the ratio of heat transfer area to water content. Further analyses will allow recommendations for improvements (e.g. internal structures with a low heat capacity to reduce the water content), for the choice of alternate working fluids for covering start-up and shut-down conditions at lower temperatures and for different applications in NPPs.

SIET elaborated first function principles how to start a steam jet pump with a PI. The simulation shows the potential of PIs also for this purpose.

C.5 Passive decay heat removal from the BWR-Containment

Three different condenser designs for the passive decay heat removal from the containment are being investigated in this work package. PCC and IC tests are combined as these components are more or less identical.

In 1996 the installation of the building condenser in PANDA as well as in NOKO (see fig.6.) has been planned and started; the first tests are expected in the 2nd quarter of 1997 for both facilities. The test matrices have been discussed and agreed upon. Conditions under severe accidents with Hydrogen generation are also included in the tests by simulating the influence of light non-condensable Hydrogen by Helium.

Three PANDA text facility configurations (M3, M10A, M10B) have been selected out of the PCC-test series performed in 1995 for further evaluation. The PANDA BC tests will also be used to perform 3D-calculations on the flow pattern in the pool above the BC arrangement.

The BC tests will be succeeded by tests end of 1997 in NOKO and beginning of 1998 in PANDA with the cast iron plate condenser that shows strong resistance against mechanical impacts under hostile accident conditions. The first out of two plate condensers has already been delivered by Siempelkamp foundry which is planning to test the other one under extreme heat radiation loads.

Pre-test calculations with RALOC for the NOKO building condenser tests as well as with RELAP5 and GOTHIC for the PANDA PCC tests have been performed to optimize the experimental procedures as well as the instrumentation schemes.

Publications in 1996

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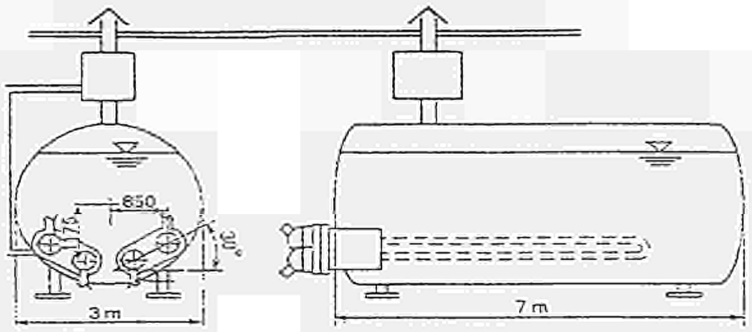
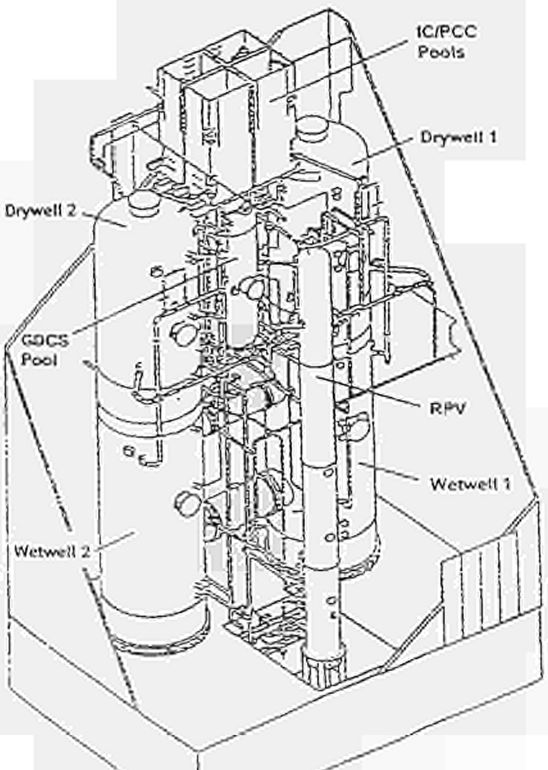
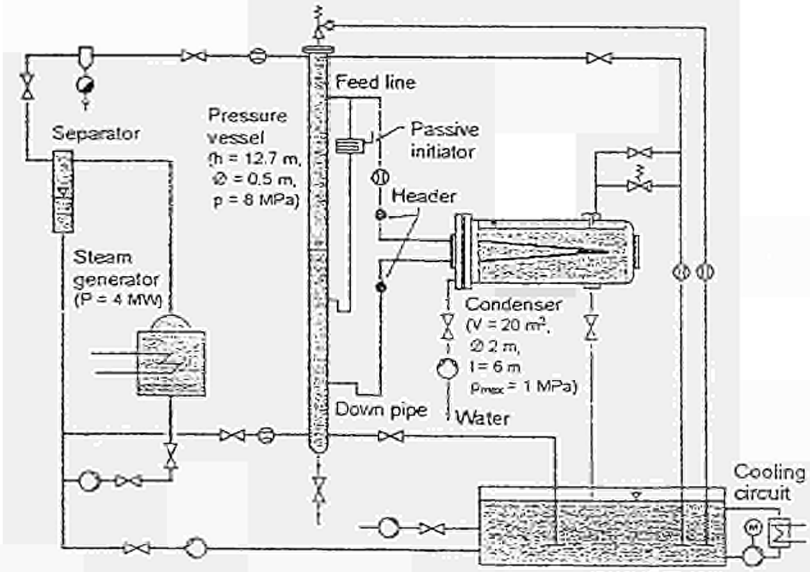
Schematic view of the Dodewaard IC



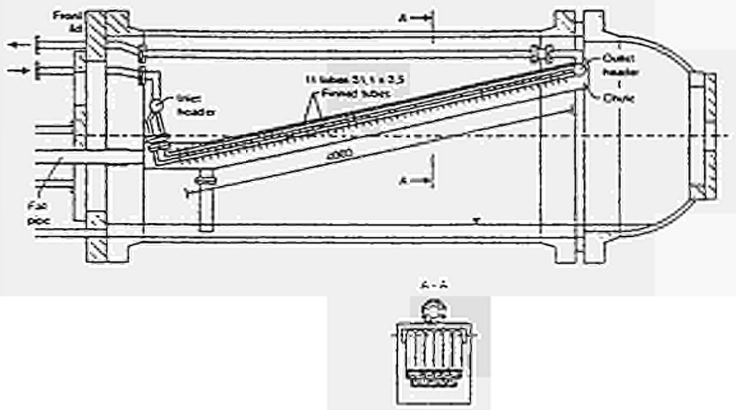
PANDA Experimental Facility: Vessels, Pools and System Lines



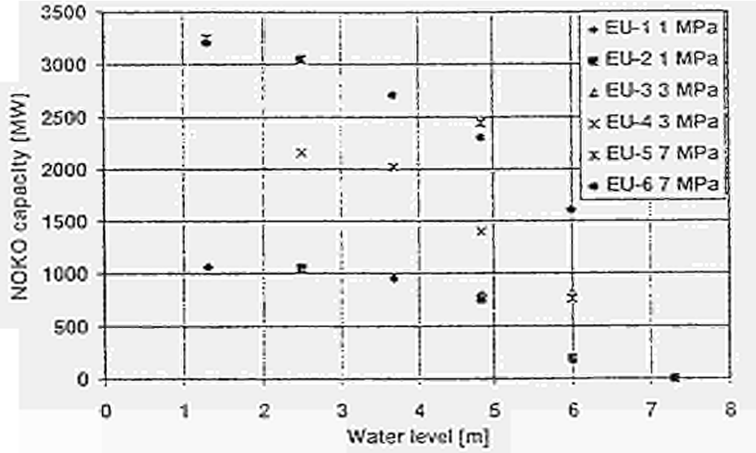
Schematic view of the NOKO test facility



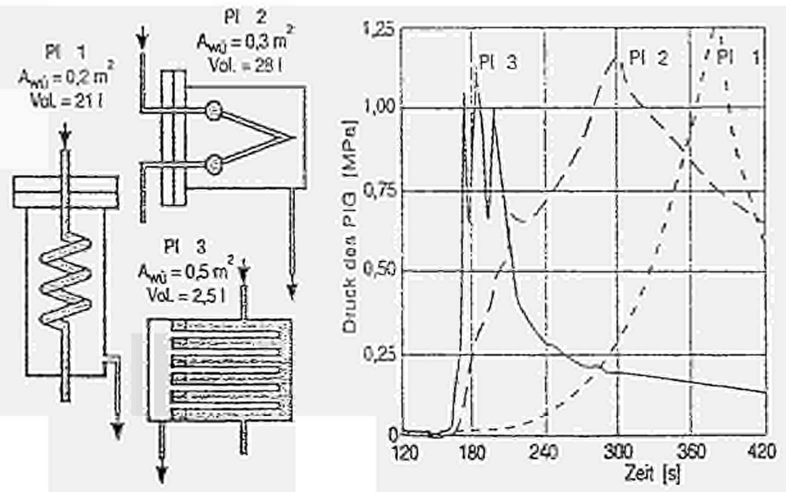
The Building Condenser in the NOKO test facility



Results of the NOKO 4 tube bundle experiments



Design and first test results of the Passive Initiators



Complementary interaction of BWR R&D-Cluster and BWR-CA activities

WP 1

Experiments or plants	European BWR-R&D-Cluster	BWRCA
Dodewaard Reactor	Transfer of operational Data and TRAC-BF-calculations (KEMA) Results from RELAP-Calcul. (ECN) 20 ATHLET-calculations (FZJ)	3D-power distribution and neutronic characteristics (CEA/DRN, ENEA) DYN3D-ATHLET-calculations (FZR)
		Experimental and theoretical research on natural circulation for BWR (TU Delft)

WP 2

Experiments or plants	European BWR-R&D-Cluster	BWRCA
1. NOKO-Bundle - 40 experiments Germany-NOKO - 10 experiments EU 2. NOKO-Bundle - 40 experiments EU	4 APROS-Calculations (VTT) 10 ATHLET-Calculations (FZJ) 4 RELAP 5-Calculations (ECN) 4 RELAP 5-Calculations (PSI) 3 TRAC-Calculations (KEMA) 5 PHOENICS-Calculations (NUKON)	Special Instrumentation (FZR) Condensation Phenomena (SIEMAT) CATHARE-Calculations (CEA/DRN) Additional NOKO-Data (SIEMENS) ATHLET-Calculations (FZR)
PANDA IC	APROS-Calculations (VTT) 10 ATHLET-Calculations (FZJ) 5 PHOENICS-Calculations (NUKON) 4 RELAP 5-Calculations (ECN) 4 RELAP 5-Calculations (PSI)	
Dodewaard IC	4 APROS-Calculations (VTT) 5 ATHLET-Calculations (FZJ)	
PANTHERS IC	Inform about generic experience (SIET)	
Thermal Valve		Simulation and assessment with: CATHARE-calcul. (CEA/DRN/DER) RELAP-calcul. (ENEA/ERG/FISS)

WP 3

Experiments or plants	European BWR-R&D-Cluster	BWRCA
Steam Jet Pumps (SJP)	Conceptional study (NUKON) Information on ongoing R&D and plan for future activities (SIET)	

WP 4

Experiments or plants	European BWR-R&D-Cluster	BWRCA
NOKO-Passive Initiators (PI)	30 tests with water and alternative fluids (FZJ) Assess the use of PI for SJP (SIET) Recommendation of possible improvements and applications (FZJ)	

WP 5

Experiments or plants	European BWR-R&D-Cluster	BWRCA
PANDA-Experiments (PSI)		
- Passive Containment Cooler (PCC)	FLOW 3D-calcul. (PSI) 5 RALOC-calcul. (FZJ) RELAP/MELCOR-calcul. (ECN) MELCOR-calcul. (VTT) ATHLET 3D-calcul on large pool. (GRS)	
- Building Condenser (BC)	FLOW 3D-calcul. (PSI)	TUBCO and TUBEX-calcul. (CEA/DRN)
- Plate Condenser (CPC)	FLOW 3D-calcul. (PSI) RALOC-calcul. (GRS)	TUBCO and TUBEX-calcul. (CEA/DRN)
NOKO-Experiments (FZJ)		
- 27 Building Condenser (BC)	5 RALOC-calcul. (FZJ) RELAP/MELCOR-calcul. (ECN) APROS-calcul. (VTT)	
- 13 Plate Condenser (CPC)	5 RALOC-calcul. (FZJ)	
		Pressure and Temp.-calcul. for a containment (CEA/DRN)
PANTHERS-PCC	Document tests and give information (SIET)	

A.1.1-4 Technology enhancement for passive safety systems - TEPSS

Contract No: FI4I-CT95-0008 **Duration :** 1 Jan. 1996 - 31 Dec. 1998
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A. OBJECTIVES AND SCOPE

The objective of this project is to undertake research needed to support further development of the technology base related to Advanced Boiling Water Reactors (BWR) of passive-type design. The research will focus mainly on mixing and stratification phenomena in large water pools, passive decay heat removal, and effects of aerosol deposition inside heat exchangers tubes.

The experimental work will be performed in three existing European facilities : LINX-2, PANDA, and AIDA. It will be supported by analytical work to identify and understand the governing phenomena, and to produce usable correlations and reliable physical models. Different computer codes (RELAP5/MOD3, GOTHIC, TRAC/BF1, MELCOR) will be used.

B. WORK PROGRAMME

B.1 Suppression Pool Mixing and Stratification

Two series of tests will be performed at the LINX-2 experimental facility in order to study and improve the design of spargers to be used in suppression pools. The effects of spargers in promoting deep-layer mixing and preventing hot stratified pool regions will be investigated. A third series of tests will be used to identify simple mixing models that can be applied in system analysis codes.

B.2 Passive Decay Heat Removal

Eight experiments will be performed at the PANDA facility in order to test the performance of a containment configuration in which the gravity-driven cooling system is made topologically part of the wetwell airspace in a pressure suppression containment. Scaling analyses will be used to assure that the experimental observations will be in known relationship to the phenomena in the reference design (European SBWR). For each test run, pre-test analyses will be performed with RELAP5/MOD3, in order to better define the test configuration and parameter range extensions. In addition, post-test analyses for each test run will be performed with RELAP5/MOD3, GOTHIC, TRAC/BF1, and MELCOR to benchmark the codes.

B.3 Passive Aerosol Removal

The AIDA-PCCS experimental facility will be used to investigate the degradation of decay heat removal due to the fission product aerosols which might deposit on the inside surfaces of the heat exchanger tubes. One key benchmarking experiment using SnO aerosol as simulant will be performed. Pre- and post- test analyses will be performed using the MELCOR code.

C. Progress of work and results obtained
Summary of main issues

The main achievements for the TEPSS project over the year 1996 are the completion of workpackage 3, the completetion of the modifications to the PANDA facility, and the scaling study which is about to be completed.

Concerning the deliverables for 1996, as mentioned in the Technical Annex of the TEPSS project, all but one deliverable have been submitted to the European Commission. The one missing is the scaling report which will be completed february 1997.

The conclusion with respect to progress is that the project is doing well. The upcoming year will be more challenging with all the experimental and analytical work going on. A possible problem may occur with the schedule of the PANDA experiments if the experiments for the European IPSS project which are performed in the same facility are delayed too much.

C.1 Suppression Pool Mixing and Stratification

No activities were performed for the reporting period.

C.2 Passive Decay Heat Removal

The work to be performed under this work package consists of a scaling study, the experimental work, and the analytical work.

The scaling analysis presents a comparison between the PANDA containment cooling test facility at the Paul Scherrer Institute in Switzerland and a reference design of a BWR with passive safety systems. The scaling analysis has been performed in 1996 according the "Hierarchical Two-Tiered Approach to Scaling" methodology, which has been developed by the U.S. Nuclear Regulatory Commission. This methodology involves two types of analyses: a top-down analysis on system level and a bottom-up analysis of relevant phenomena. The results have been described in a document which will become available february 1997.

The main conclusions of the scaling analysis are:

- All major processes are scaled properly. Corrections can be made for other processes, following guidelines contained in the report.
- The PANDA tests will form a good benchmark for pressure response of the reference containment design, and will yield valuable insight into the thermo-hydraulic phenomena of the reference containment design, and as such form an indispensable tool for code qualification.
- The proposed modifications of the PANDA installation and test matrix will have limited consequences for pre-test and post-test numerical analysis. The main qualitative change is, that the gas volume above the suppression pool is a function of time, as in the reference design this volume is coupled with the gravity driven core cooling pool volume.

The PANDA test facility has been modified to reflect the containment configuration and the passive decay heat removal systems of the reference design. These modifications include rerouting of the Passive Containment Cooler (PCC) drain lines to the Reactor

Pressure Vessel and connection of the GDCS vessel air space with the Wetwell air space.

A test series involving eight test runs will be performed. The PANDA test matrix as defined at this moment consists of:

- P1 Simulating a Main Steam Line Break (MSLB) with the new containment configuration. Experiment starts at 1 hour into the LOCA transient.
- P2 Simulating also a MSLB but starting earlier to include the GDCS injection phase.
- P3 Investigating start-up behaviour of PCC units when the drywell is initially filled with air.
- P4 Similar to P1, with a supply of air later in the test to simulate hidden air somewhere in the containment.
- P5 Similar to P1, with an additional supply of air and turning off one of the PCCs to simulate a severe accident.
- P6 Investigating the interaction between Isolation Condenser functioning in parallel with the PCC units. In addition, the effect of bypass leakage from drywell to wetwell will be investigated.
- P7 Similar to P5, but with both active PCC units connected to the same drywell.
- P8 Similar to P1, but with lower PCC pool levels.

The main analytical activity in 1996 concerns the implementation of a PCC model in the TRAC-BF code by UPV. This model accounts for both the primary side heat transfer within the tubes as the secondary side heat transfer in the pool. The default version of the TRAC-BF code does not properly model the steam condensation phenomena in the presence of non-condensable gases. UPV has compared three different condensation heat transfer coefficients for steam condensation in presence of non-condensable gases against experimental data from the University of California at Berkeley (UCB) experiments. These models are: (1) the correlation which was originally derived from the UCB data and used by GE for the TRACG code, (2) a UPV-FIT model in which the degradation effects of non-condensables have been substituted by a correlation fit, and (3) a diffusion layer model in which the heat transfer coefficients are expressed in terms of the condensate Reynolds number. These models have been implemented in the TRAC-BF code with an option to choose a specific correlation. The calculational results obtained using the first two models were compared to the UCB experimental data. The results show that the UPV-FIT model gives a better prediction than the GE model, in particular at the entrance of the condenser tube. The secondary side heat transfer model developed by UPV concerns the development of natural convection correlations for vertical cylinders immersed in a pool. In order to capture the boundary layer phenomena in the TRAC-BF code, correlations have to be developed. UPV has developed correlations for the Nusselt number for both laminar and turbulent natural convection. These correlations have been implemented in the TRAC-BF code.

C.3 Passive Aerosol Removal

AIDA test A-TEPSS04 has been executed for the TEPSS program by PSI, following a series of thermal-hydraulic tests which were devoted to a) investigate the thermal-hydraulic characteristics of the model condenser and b) demonstrate the functions of various measurement and data acquisition systems. The objective of Test A-TEPSS04 is to demonstrate the general behaviour of the model condenser in removing the aerosol particles and condensing the steam under specially selected thermal–hydraulic and aerosol conditions. Experiment A–TEPSS04 consists of two phases. The first phase, Phase A, determines the condensation behaviour of the condenser at the specified thermal–hydraulic boundary conditions (without aerosol particles). The second phase, Phase B, determines the condensation behaviour of the condenser at the specified thermal–hydraulic conditions of the first phase and in addition, at the specified aerosol boundary conditions. Phase B of the experiment starts with the initiation of the tin powder flow into the plasma torch at a desired rate. This will cause the generation of the SnO_2 aerosol particles. Table I shows the measured thermal–hydraulic and aerosol deposits.

The experiment indicated that under overloaded condenser conditions and using SnO_2 particles, the efficiency of the AIDA model condenser was degraded by about 20 % at the end of the experiment. Significant aerosol deposition occurred in the upper plenum lower surface. As a result, a thick layer of deposits was formed. Deposition also took place inside the tubes, but it is believed that this did not lead to heat transfer degradation in the tubes. The overall degradation in heat removal could only be explained by a possible interaction between aerosol distribution in the upper plenum and the flow distribution to the individual tubes. The deposition pattern presumably caused a non–homogenous flow distribution between the tubes. As a result, the total available heat transfer area is not fully used and the condenser efficiency decreases. It is not clear whether the decreasing trend in the condenser efficiency would last if the experiment duration would have been longer or if the aerosol deposits would be purged at some point.

Pre– and post–test analysis were performed using the MELCOR computer code for which an AIDA input deck was developed. With the MELCOR model developed two calculations have been run. The base case calculation uses the default MELCOR steam condensation model in which a condensate layer is build–up on a heat structure until the maximum thickness of 0.5 mm is reached. The second calculation uses the film tracking model in MELCOR. This model tracks in detail the build–up of the condensate layer on the wall. The MELCOR results are compared with the experimental data in Table I.

MELCOR underpredicts the cooling efficiency of the condenser unit by about 30 % during the non–aerosol phase. The best thermal hydraulic results have been obtained by using the film–tracking model. The measured degradation in condenser efficiency is not calculated by MELCOR during the aerosol feed phase.

All MELCOR calculations strongly underpredicted the aerosol deposition in the upper plenum lower surface in contact with the water. Using the film–tracking option causes an overprediction of aerosol mass in the tubes and scrubber tank and an underprediction of aerosol mass in the drain tank.

The overall conclusion is that the modelling present in MELCOR at this moment is not adequate for simulating the AIDA experiment which was performed under this TEPSS project.

Table I: Measured and calculated results of AIDA test A-TEPSS04.

	Post-test results		Experimental data
	Base case	Film tracking	
Heat removed by the cooling flow in the water jacket [kW]	61	75	90
Temperature difference between the inlet and out of the cooling jacket [°C]	3.1	3.8	4.5
Water temperature in the drain line [°C]	105	101	90
Steam temperature in the lower plenum [°C]	150	156	120
Steam flow to the scrubber tank [kg/s]			
	0.033	0.027	0.018
Water flow to the GDCS tank [kg/s]			
Aerosol mass distribution:	0.023	0.028	0.038
Upper plenum [%]			
Tubes [%]	0.4	2.1	31.1
Lower plenum [%]	2.4	36.4	15.9
Drain tank [%]	0.6	1.2	--
Scrubber tank [%]	30.2	1.5	9.9
	66.4	58.7	37.3

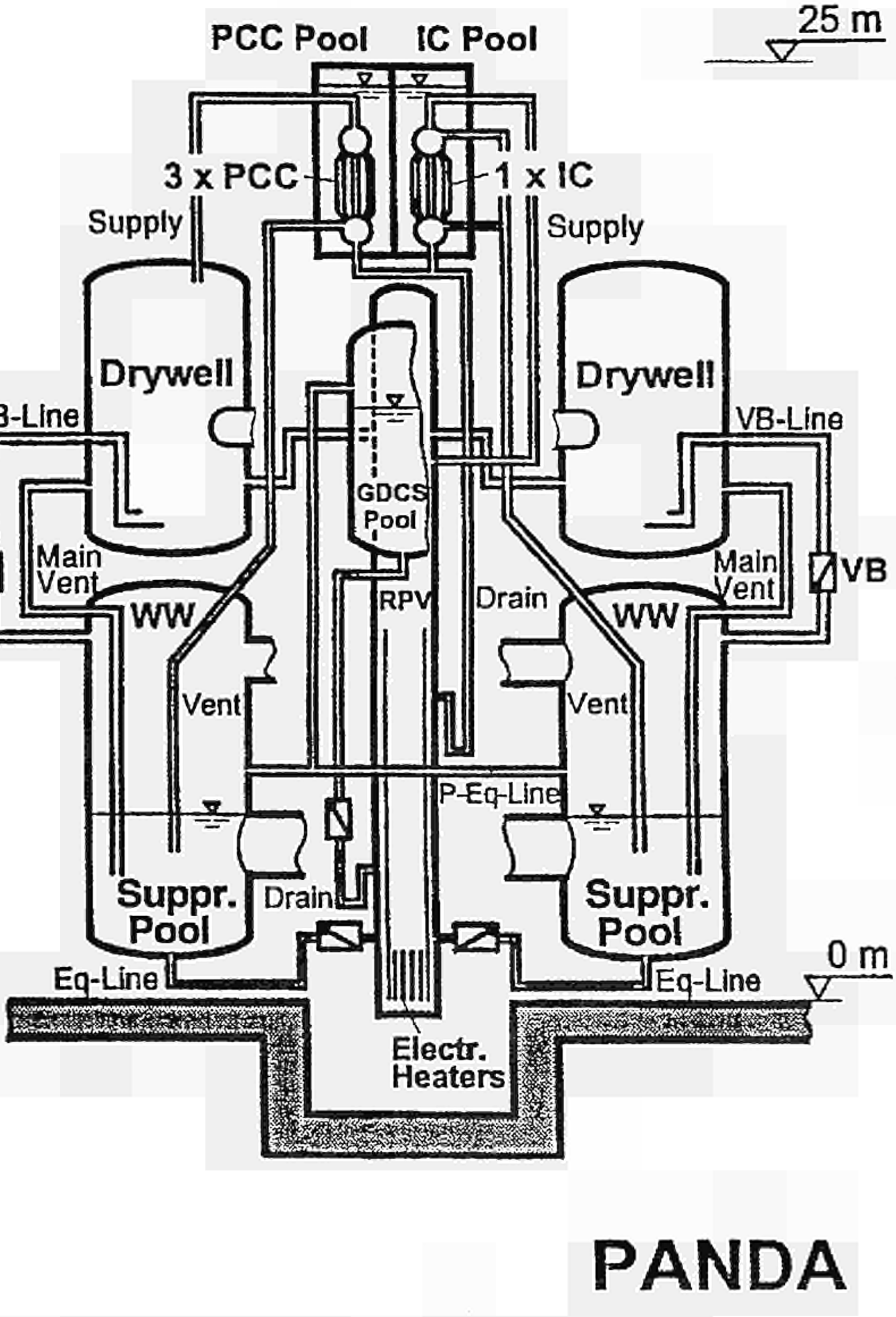

Figure 1: Schematic drawing of the modified PANDA Facility

A.1.1-5 System for emergency core cooling through high performance steam injector - SYNTHESIS

Contract No:	FI4I-CT95-0001 **Duration :** 1 Jan. 1996 - 30 March 1998
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Partners :	**Siemens KWU Münich/DE, CISE Milano-Segrate/IT**

A. OBJECTIVES AND SCOPE

To demonstrate the feasibility of an injection system (SYNTHESIS), based on a passive device (High Performance Steam Injector, HPSI). This system should be able to pump water into a Reactor Cooling System (RCS) at high pressure (maximum pressure about 9 MPa), taking water from an atmospheric tank. SYNTHESIS will be equipped with automatic actuators (valves) for startup and for keeping the optimum operating conditions during transients. However, the feasibility of a completely passive version of SYNTHESIS will also be assessed. In this case the above mentioned automatic actuators will be replaced by self-acting valves or by valves powered by passive mechanisms.

B. WORK PROGRAMME

B.1 Definition of SYNTHESIS Requirements

To define the system functional requirements for a reference application (BWR 1000 design), as well as for an alternative application to an innovative PWR plant. Based on the results of these two activities, functional requirements enveloping as much as possible both applications will be defined.

B.2 Prototype testing with active valves

To design and build the HPSI prototype in accordance to the functional requirements defined in B.1. A test facility and a test matrix for the prototype with active valves will also be defined. Then the tests will be performed, and the results analyzed (including simplified numerical modelling) to check compliance with the SYNTHESIS functional requirements and to define the final functional requirements of the passive actuators.

B.3 Passive Actuators Development

According to the functional requirements defined in B.1, a set of possibly required types of passive actuators will be identified, and, if necessary, their design will be updated.

B.4 Benefits Evaluation

A simplified mathematical model of SYNTHESIS will be developed to be introduced into a thermalhydraulic computer model of target plants. For the BWR 1000 reference case the computer code to be used is RELAP5 (for which there is a SYNTHESIS model available), and for the alternative PWR application, a similar approach will be adopted. Different scenarios will be analyzed, and as a result the benefits of SYNTHESIS, in terms of plant simplification and cost reduction, will be pointed out.

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The activities related to the identification of the reference plant and system functional specification have been performed according to the schedule shown in the figure 2.2 of the Technical Annex to the contract.

On the other hand, there is delay in the activities related to the experimental part of the project in spite of the anticipated preparation of the rev.0 of the Technical Specification for the contract to SIET (ENEL major subcontractor).

In fact ENEL had some difficulties to finalize the contract with SIET. Essentially these difficulties have been caused by the need to finalize the scope of work and the economical part in order to get as much as possible useful data, limiting the extra cost in comparison to the preliminary SIET economical evaluation. Essentially the problem was to reproduce as much as possible the real plant condition, especially regarding to RPV pressure effect on the HPSI discharge. At the moment the contract is going to sign and the related activities are ready to start. Most of the delay will be absorbed from the particular attention given to the component choice in order to avoid long procurement time, so the project is able to maintain the foreseen overall duration.The partners agreed to distribute among them the extra cost of the contract to SIET.

C.1 Definition of SYNTHESIS functional requirements (WP1)

As results of this activity the reference application for the project will be an injection system for the German concept SWR 1000 which is an innovative Boiling Water Reactor (BWR). The expected benefit coming from these kind of system would a reduction of Reactor Pressure Vessel (RPV) volume and height, because the high pressure water provided by SYNTHESIS would reduce the inventory losses during Automatic Depressurization System (ADS) intervention. In this way plant cost would be decreased and plant layout would be simplified.

As far as alternative solution PWR applications are concerned, two main alternatives have been considered: a primary system emergency injection system, operating at high-medium pressure and a secondary side (steam generator) emergency feedwater system, operating at a near secondary side design pressure

At the end. preference was given to the second application.

This WP was concluded with the identification of system functions, interfaces, preliminary layout, operating conditions and required performances.

C.2 Prototype testing with active valves (WP2)

According to the functional requirements identified in WP1 the HPSI optimization and design has been completed by ENEL/CISE, developing a preliminary configuration of HPSI prototype and then checking it by appropriate computer models.

The preliminary HPSI configuration has been optimized by performing sensitivity analyses on different geometrical parameters; the main optimization criteria has been to minimize the average steam consumption and the overflows.

The HPSI detailed drawings has been completed.

The component construction has been completed too.

About the test facility, the modifications to the already existing test loop, needed to perform the experimental program as per the objectives of the project, have been discussed and finalized with the subcontractor SIET. The related Technical

Specification has been prepared, discussed among the partners and issued in order to send the official request for bid to the SIET itself.
Finally the contract to SIET for facility modification and test execution is ready to be signed.
Furthermore, based on the outputs coming from the functional requirements definition, from the HPSI design and from the finalization of the facility technical specification, the draft version of the test specification, matrix and active component specification have been issued for comments.

C.3 Passive actuators development (WP3)

There was only activity related to preliminary design and calculation of passive actuators. A report on this topic is going to be issued. The existing design contains 7 different armatures. Two are check valves which work passively, one is a small active valve which may be opened or closed independently from the others. The functional operation times of the remaining four armatures (with minimum diameters between 80 mm and 200 mm) are restricted and the sequence of their operation is fixed. It cannot be predicted theoretically which exact values should be realized. The exact requirements will be found experimentally in WP2.3.

A.2.1-1 Supporting nuclear data for advanced MOX fuels

Contract No:	FI4I-CT95-0002 **Duration:** 1 Jan. 1996 - 31 Dec. 1998
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Partners:	**ECN Petten/NL, SCK/CEN Mol/BE, CEA-Cadarache/FR, JRC-ITU Karlsruhe/EC, ENEA Bologna/IT**

A. OBJECTIVES AND SCOPE

The objective is to supplement the results of the previous European Strategy Study in the field of Partitioning and Transmutation (P&T). The use of advanced MOX fuels, either in thermal (PWR) or fast reactors will be investigated. To this end the necessary nuclear data working libraries will be updated with new information from basic data evaluations and available integral experiments. The accuracy of the strategy studies to minimize wastes will be assessed.

B. WORK PROGRAMME

B.1 Accuracy of strategy studies involving the use of thermal reactors, especially Pressurized Water Reactors **(BN Brussels, ECN Petten, SCK×CEN Mol, CEA Cadarache)**

The contributions from SCK•CEN, BN and ECN are closely coupled, and related to the use of advanced MOX fuels in LWR, with an emphasis on high burnup and high transmutation rates (effects of over-moderation).

The SCK•CEN contribution consists of reviewing post irradiation examinations made in the past on a number of fuel samples from LWRs, especially PWRs (like BR3), so as to establish an extended database. At a later stage, recent irradiation results should be used to that aim.

Mass balances from these high burnup irradiations are to be re-calculated at BN for MOX fuel pins irradiated up to very high burnup (e.g. 80,000 MWd/t), using adequate modelling and cross-sections from the JEF 2 database. ECN will correlate the measured and calculated masses with the cross-sections, and estimate the uncertainties of mass inventories and of reactor parameters.

At CEA, the French LWR programme has comprised analyses of spent fuel isotopics for a wide range of UOX and MOX fuel types and burnups. Their analysis will be used to reduce uncertainties in nuclear data for actinides in MOX fuel from the JEF 2 database.

B.2 Accuracy of strategy studies involving the use of fast neutron reactors **(SCK×CEN Mol, CEA Cadarache, JRC-ITU Karlsruhe)**

ITU will verify actinide and fission product nuclear data, based on irradiation experiments in the fast reactor KNK-II/2. This involves composition measurements using IDMS, alpha- and gamma-spectrometry and ICP-MS.

SCK×CEN will review post irradiation examinations made on fuel samples from fast reactors (like KNK-II), so as to establish an extended data base.

CEA will interprete the results of the analysis of the MOX fuels and samples irradiated in the fast neutron reactor PHENIX with JEF2 nuclear data.

B.3 Supporting work on the evaluation of basic isotopic data **(ENEA Bologna)**

ENEA will revise nuclear data files for the isotopes Pu240, Pu242 and Am241, especially in the range of resonances.

B.4 Assessment of the accuracy of P&T strategy studies **(BN Brussels, ECN Petten, CEA Cadarache)**

The activities described above will be structured, by comparing first the computational techniques, and by integrating all results in a common data base, so as to agree on the final trend analysis.

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

From the 4 Work Packages, shortly designated by :

WP1 : Analysis of Irradiation in Thermal Reactors

WP2 : Analysis of Irradiation in Fast Reactors

WP3 : Complementary Basic Data

WP4 : Assessment of the Accuracy of P&T Strategy Studies,

WP4 cannot be engaged before 1997, as it relies on the work made in the 3 first WPs. Work has begun in 1996 for WP1, WP2 and WP3, and is described below. Most of the activity has been spent on WP1 and WP2, to set up a comprehensive compilation of irradiation results for high burnup fuels.

On WP1, the work was mainly carried out by SCK and BN on the one hand, and CEA on the other hand. Two tasks : Compilation of MOX fuel irradiation results from BR3 and other LWR reactors (SCK), and Compilation of SLB1 irradiation results (CEA) have been completed in 1996. ECN has also started preparation work for the sensitivity analyses.

On WP2, work was mainly carried out by CEA ; their task, comparison of calculated and measured values for PHENIX, has substantially progressed. ITU has also started work.

WP3 was started by ENEA with a re-evaluation of Am241 data files.

C.1 Work Package 1 : Accuracy of strategy studies involving the use of thermal reactors, especially Pressurized Water Reactors

At SCK, a data bank has been established on the basis of post-irradiation examination results coming mainly from BR3, a 40 MWth, 10.5 MWe experimental reactor, characterized by very high burnup values reached on MOX pins.

The data bank contains the irradiation history, the actinide vectors and some major fission product concentrations (Cs137, Ce144, Ru106 and all neodymium isotopes) for 71 samples irradiated in BR3, including 23 MOX samples, covering a burnup range from 1 to nearly 10 atom percent. The irradiation conditions have been ranked according to a "plutonium transmutation quality factor".

The data bank also contains the results of re-irradiations in BR2, a material testing reactor, for 25 samples including 19 MOX samples. It furthermore contains the results of 101 samples obtained from PWR and BWR power stations (39 MOX samples) with a burnup ranging from 0.2 to 6 atom percent. The data bank has been made available at SCK on CD-ROM.

BN has selected with SCK the irradiation results which fit best to the contract objectives (high burnup MOX, and also check the effect of high moderation), see Table I.

The needed calculational tools have been checked to be operational. They are contained in the WIMS-7 code package [1]. The calculation has begun with the follow-up of BR3 high burnt MOX fuel rod (80 Gwd/t). Special care has been taken to represent exactly the complex geometry of BR3 in a 2D heterogeneous modelling which describes the rod environment including changes in the shuffling of the concerned assembly. The cross-section file is a 172-group one, derived in 1996 from the JEF 2.2 data file.

At CEA, the compilation of Saint-Laurent B1 (SLB1) MOX fuel irradiations is completed.

The SLB1 reactor, a 900-MWe PWR, is characterized by a 30 % MOX fuel loading. The U235 enrichment of the UO_2 driver fuel is 3.25 % and the Pu content of the MOX fuel is 5.3 % in average ; both types of fuels are normally staying 3 calendar years, aiming at an average burnup of 33,000 MWd/t. In this reactor, well characterized MOX assemblies have been examined so far after 1, 2 or 3 years of irradiation ; the fuel rods examined range from 9 to 41 GWd/t HM.

Analyses have been made to determine the concentrations in U, Pu, Am, Cm, Np isotopes and in Nd and Cs ; an experimental determination of burnup is given for each sample, either through the Nd/U ratio or through the Cs ratio measured by gamma spectroscopy. While these experimental results are proprietary, the C/E ratios (calculational over experimental values) for the isotopic compositions will be available.

The depletion calculation for fuel inventory assessments versus burnup is sensitive to the local neutronic spectrum ; therefore, an accurate 2D heterogeneous assembly pattern is required. The APOLLO 2 code, with its collision probability transport theory methods, offers several options for the physical modelling of resonance self-shielding, 2D spatial representation and burnup. The process used aims at optimizing the calculational scheme for MOX assemblies, starting from a standard reference calculation and comparing the results with those of the reference. In doing so, accuracies can be assessed [2].

ECN has started work directed to a sensitivity analysis for MOX recycling in PWRs by applying their OCTOPUS burnup and criticality code system [3] on the OECD benchmark "Multiple Recycling in Advanced Pressurized Water Reactors". The agreement with other participants (among them BN and CEA) is very good, which gives confidence in the code system.

C.2 Work Package 2 : Accuracy of strategy studies involving the use of fast neutron reactors

The work carried out at ITU in 1996 has mainly concerned an optimisation of the analytical tools, by a dissolution of a blank stainless-steel container to determine its effect on separations for mass spectroscopy ; a check has also been made of the sensitivity for the fission gas measurement.

The analysis is in progress on samples from the KNK-IIb campaign (remeasuring fresh material composition) and the KNK-IIa campaign (irradiated fission product targets). A delay has been caused by the hot cell operation schedule.

At CEA, the analysis of sample irradiation experiments carried out in the PHENIX reactor has been performed using the JEF 2.2 basic nuclear data. In a standard subassembly placed in the first row of the inner core of PHENIX, 46 pure actinide and fission product isotope capsules corresponded to the PROFIL 1 experiment, while 2X42 such capsules corresponded to PROFIL 2. Samples are placed inside two stainless steel containers, loaded in standard pin claddings, see Figure 1.

The pure actinide isotopes are, in addition to the U ones, 5 Pu, 3 Am, Np237 and Cm244. Nd148 was used as a burnup indicator. Mass spectrometry was used, with simple or double isotopic dilution and well-characterized tracers. All experimental results are presented as ratios of actinide or fission product concentrations and a global experimental accuracy has been estimated, accounting also for the reproducibility of the measurements.

The calculations make use of basic nuclear data from the JEF 2.2 file. First, it has been observed that the performance of the unadjusted JEF 2.2 data was fairly good already. Some important trends have been denoted for the major actinides.

An adjusted nuclear data library is being set up, ERALIB1, in view to reach a better safety and improved performances in future fast reactor applications. The adjustment covers 2 U isotopes and 4 Pu isotopes. An example of adjustment is the reduction by 15 % of the capture cross-section of Pu242 below 0.5 keV.

C3. **Work Package 3 : Supporting work on the evaluation of basic isotopic data**

At ENEA, a critical analysis has been performed of available data files for Am241 against experimental data. It was concluded that the recent work by Maslov et al. [4] in a collaboration between Belorussia and the Japanese JAERI was satisfactory. This has been taken as a starter file into which to add the ENEA high quality photon production data. Plots of the different file comparisons are available.

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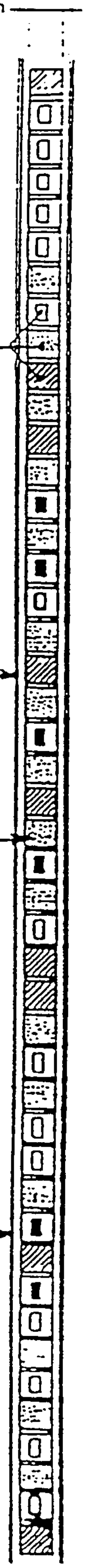
[3] KLOOSTERMAN, J L et al, International Conference on the Physics of Reactors, PHYSOR 96, Mito, Japan, 16-20 September 1996 :
The OCTOPUS Burnup and Criticality Code System

[4] MASLOV et al, International Conference on the Physics of Reactors, PHYSOR 96, Mito, Japan, 16-20 September 1996 :
New Evaluation of Minor Actinide Nuclides

TABLE I

MOX Fuels Irradiation Data Bank (SCK / BN)
Excerpts

Designation	% Pu	% burnup	'Pu transmutation quality factor'
<u>BR3 reactor</u>			
36.13 1	0	7.01	-
36.13 8	0	8.22	-
36.13 9	0	9.45	-
ZO-100 bu6	3.7	3.93	8.94
ZO-100 bu8	3.7	4.09	9.15
3G54/pf 07	8.28	2.13	42.97
B2000 BU1/1	11.3	7.43	7.35
B2000 BU 1/5	11.3	9.95	6.28
B2000 BU 3/3	11.3	3.23	9.87
<u>BR3/BR2 reactors</u>			
P3/363/BU2	3.7	6.23	10.68
P65-B1	7.77	2.66	10.04
<u>CHOOZ-A reactor</u>			
DO120 BU1	5.01	1.57	7.11
DO120 BU2	5.01	3.14	6.36
<u>DODEWAARD reactor</u>			
B201 DO11	2.71	2.11	8.72
B201 DO14	2.71	2.55	14.46
PB10 B66	5.44	6.03	7.68
PB12 U09	0	5.79	-
PG34 RBU1	3.28	3.56	14.57
PG34 RBU2-5	3.28	6.09	7.44



Position (mm)	Isotope	Chemical form	Mass (mg)
180	^{235}U	UO_2	22
170	^{145}Nd	Nd_2O_3	22
160	^{105}Pd	Palladium	21.7
150	^{133}Cs	CsF	23.4
140	^{101}Ru	Ruthenium	19.6
130	^{95}Mo	Molybdenum	21.5
120	^{238}Pu	PuO_2	14
110	^{97}Mo	Molybdenum	21.7
100	^{239}Pu	PuO_2	~10
90	^{235}U	UO_2	22.5
80	^{241}Am	AmO_2	10.0
70	^{238}U	UO_2	22.3
60	^{240}Pu	PuO_2	3.7
50	Natural lithium	LiF	3.8
40	^{242}Pu	PuO_2	29
30	Natural boron	Boron	1.1
20	^{149}Sm	Sm_2O_3	25.8
10	^{241}Pu	PuO_2	4.3
0	^{235}U	UO_2	24.1
−10	^{239}Pu	PuO_2	~10
−20	Natural boron	Boron	1.0
−30	^{240}Pu	PuO_2	6
−40	^{238}U	UO_2	23
−50	^{241}Pu	PuO_2	5
−60	Natural lithium	LiF	3.7
−70	^{239}Pu	PuO_2	~10
−80	^{101}Ru	Ruthenium	20
−90	^{235}U	UO_2	23.9
−100	^{238}U	UO_2	23.7
−110	^{240}Pu	PuO_2	6.3
−120	^{133}Cs	CsF	22.4
−130	^{242}Pu	PuO_2	27
−140	^{95}Mo	Molybdenum	21.9
−150	^{105}Pd	Palladium	20
−160	^{241}Pu	PuO_2	12.5
−170	Natural boron	Boron	1.3
−180	^{235}U	UO_2	24.5
−190	Natural lithium	LiF	4.2
−200	^{101}Ru	Ruthenium	19
−210	^{242}Pu	PuO_2	25
−220	^{145}Nd	Nd_2O_3	20.2
−230	^{238}Pu	PuO_2	12.6
−240	^{97}Mo	Molybdenum	19.7
−250	^{241}Am	AmO_2	13.7
−260	^{149}Sm	Sm_2O_3	22.9
−270	^{235}U	UO_2	25

Figure 1 : *PROFIL PIN IRRADIATION IN PHENIX*

A.2.1-2 Evaluation of possible P&T strategies and of associated means to perform them

Contract No:	FI4I-CT95-0006	**Duration:**	1 Feb. 1996 - 31 Jan. 1999
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A. OBJECTIVES AND SCOPE

The objective of this study is to give indications on what can be expected from P&T strategies. This will be achieved by the evaluation of these strategies according to several criteria, relative to their main advantages or drawbacks. To take into account their time dependence and the national position of each participating country, the global evaluation will be performed on scenarios defined in common by representatives of all nations involved in the study.

The criteria include of course the technical feasibility of the different operations needed to perform the strategies: fuel reprocessing to achieve the partitioning of the radionuclides to be transmuted, transmutation techniques and related fuel or target fabrication. They also include their costs, the amounts of waste they generate and their associated risks. Short term risks arise from possible reactors accident or from workers exposure to radioactivity in the fuel cycle operations. On the other hand, the existence of separated fissile material is also considered by some as a risk. Finally, a long term risk induced by the radionuclides remaining at earth's surface or by those returning to the biosphere from a deep underground repository continues to exist.

B. WORK PROGRAMME

B.1 Global evaluation (CEA Valrhô, FZK Karlsruhe, BNFL Risley, SCK-CEN Mol, ECN Petten, ENEA Roma)

The global evaluation of possible P&T strategies must consider the time dimension and it is proposed to develop a small number of scenarios, based on these possible strategies. These scenarios will have to take into account, not only conceptual aspects, characterised e.g. by the type of steady state expected, but also the operational aspects characterized, e.g., by political or public opinion constraints, urging some countries to take decisions on radioactive waste management.

B.2 Assessment of partitioning techniques (CEA Valrhô, ENEA Roma)

The consequences of the use of plutonium highly enriched fuel for PWRs and FRs on partitioning will be assessed. The possibility of using enhanced "PUREX based" and/or pyrometallurgical processes will be investigated.

B.3 Assessment of the feasibility of transmutation (CEA Valrhô, FZK Karlsruhe, BNFL Risley, BN Brussels, ECN Petten, GRS Köln, ENEA Roma, Univ. "Politecnico" Milano)

Transmutation techniques in PWRs and FRs will be assessed on the basis of core computations with assumed loads of "advanced" MOX fuels or specific targets containing americium and/or curium. "U-free" reactor concept will be investigated. The optimisation of plutonium consumption will be investigated, while keeping the core safety features at a reasonable level.

B.4 <u>Assessment of the feasibility of advanced fuel or target fabrication</u> **(BN Brussels, JRC-ITU Karlsruhe, ENEA Roma)**

Fuel and target requirements for these transmutation devices will be established, and related fabrication processes derived from them. The operation risks due to dose received by the workers when handling radioactive materials in the fuel cycle operations will be assessed.

B.5 <u>Assessment of geological barrier efficiency</u> **(AEA Technology Harwell, SCK·CEN Mol, GRS Köln)**

The efficiency of three geological barriers (clay, hard rock, salt) will be assessed for several waste types containing different radionuclide inventories (spent UO_2fuel, spent MOX fuel, vitrified HLW, actinide depleted vitrified HLW, separated actinides in advanced conditioning matrices, spent FR-MOX fuel after multiple recycling).

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

In the area of global evaluation, 5 scenarios were defined for further assessment. The first one is the open cycle, with only one evolution, 40 years after the beginning of the scenario, in the burnup and fuel management of the UOX-fuelled PWRs. The total installed electric power remains at the constant rate of 120 GWe all over the time. Three other scenarios are considering actual P&T, after a 40-year period during which the plutonium is only recycled once. These scenarios differ by the type of reactors that are implemented (only PWRs, both PWRs and FRs or only FRs). The power rate is the same as in the case of the open cycle scenario. The last scenario is supposed to start from the equilibrium state of one P&T scenario and to describe the shutdown of the nuclear system in a way that limits the amounts of remaining radioactive materials after its complete stop. This has been reported [1] in accordance with the contract dispositions. The first scenario has been simulated and evaluated with respect of the short term risk to the public [2]. To support the work of other Work Packages, the characteristics of two reference fuels (UOX and MOX for conventional PWRs) have been provided [3][4]. In the area of waste repositories, preliminary calculations were performed and models adapted to cover the situations resulting from the different scenarios in the 3 selected geological formations. As regards transmutation, highly moderated (HM) PWRs and CAPRA FRs have been investigated both on the standpoints of incineration performances and safety requirements. Inert matrix fuel suitability for P&T purposes is being evaluated, as well as its fabrication by sol-gel or impregnation techniques. Preliminary assessments of PUREX-based reprocessing applied to CAPRA fuels and pyrometallurgical processes were performed.

C.1 Global evaluation

In the period covered by this report, the planned activities in this work package were to define 5 scenarios that would be evaluated in the following of the work and to study the first of them. The scenario definition was supposed to take place within the first 3 months of the contract (i.e., by 04/96) and the study of the first one six months later (i.e., by 10/96). This was actually achieved roughly in time. Reports on these 2 tasks were planned as contractual deliverables. The first one has been published but the second is still in progress although a report on the radiological impact of the first scenario was also published.

Moreover, it appeared necessary to provide homogeneous data to all the participants in the different Work Packages in order to ensure a relevant comparison of solutions. This was particularly needed in the case of fuel characteristics that are essential in the definition of partitioning processes, fabrication facilities and simulation of radioactive material migration in the geological formations. Thus, the delivery of the characteristics of the fuels used in the selected scenarios was decided and two of them have been provided in 1996.

As regards the evaluation of doses to the workers, it was considered that any new facility will have to meet the present requirements in this respect and therefore, only upper regulatory limits could reasonably be given. This could be done, if required, but has no discriminatory value for the global evaluation.

In the area of proliferation resistance, there is no currently accepted criterion to evaluate a situation or a strategy. It is proposed to build a multiple criteria methodology to reach this goal, this methodology could be tested with the first available scenario results and then generalised.

Finally, as regards the cost evaluation, it appeared that the costs of new reactors, both on the investment and the operation standpoints, were not possible to assess with the present tools and with the allocated resources. As these costs represent the main part of the whole cost of nuclear electricity, the economical evaluation of the scenarios was abandoned.

1. Scenario studies

The first selected scenario illustrates an open cycle strategy. It is supposed to begin in 2000 with an existing reactor population composed of UOX-fuelled PWRs generating a total electric power of 120 GWe. In the first 40 years, the reactors are supposed to be managed of 1/5 core with a fuel burnup of 47.5 GWd/t. After 2040, the fuel management is enhanced to 1/6 corresponding to a burnup of 55 GWd/t.

The three P&T scenarios begin with a 40 year common period during which the reactor population is composed of fully UOX-fuelled PWRs in the one hand and of 30% MOX-fuelled PWRs in the other hand. The portion of each type of reactors is adapted so as to ensure that all the plutonium produced by the UOX fuel is reused in the MOX fuel and that this MOX fuel is not reprocessed. It is then kept in interim storage to be processed and recycled in the continuation of the scenarios. The reactors have the same characteristics as those used in the same period in the open cycle scenario. After 2040, MOX-fuelled reactors are gradually replaced by HM-PWRs with a burnup of 56 GWd/t or by CAPRA FRs with a burnup of 210 GWd/t, depending on the scenario. The simulation is performed until an equilibrium state is reached. In the case of the FR implementation, the evolution of the reactor population is continued with the replacement of UOX-fuelled PWRs by CAPRA FRs in a breeding configuration. In these scenarios the total electric power remains constant at 120 GWe.

Finally, a scenario with a gradual decrease of the electric power has been selected. In this case the intention is to obtain a minimal radioactive material remainder after the complete stop of nuclear power generation.

In all the cases where reprocessing is operated, the losses at the plants are supposed to be equal to 0.12% for the actinides. The minimum cooling time before reprocessing is 5 years and the plutonium ageing time before MOX fuel fabrication is 2 years.

The open cycle scenario has been simulated. Of course, its equilibrium is reached very soon : in 2050 the annual natural uranium requirements are stabilised at 15,300 tons, the enrichment at 11.4 MSWU (SWU=Separation Work Unit), the depleted uranium production at 13,600 tons, the fuel fabrication at 1,660 tons and the equivalent for irradiated fuel production.

The radiological impact on the human population during the open cycle scenario has also been assessed. It appears that mining and milling and reactor operation are the two main sources of risk, amounting to 120 man.Sv each at equilibrium.

2. Data for reference fuels

The fuel contents in terms of mass and activity of actinides, fission products, tritium (^3H) and ^{14}C at cooling times ranging from 5 to 500,000 years has been provided for 2 reference fuels used in the scenario studies : UOX an MOX for conventional PWRs.

C.2 Assessment of partitioning techniques

There are two main aspects concerned in this Work Package. The first one is to determine whether a PUREX-derived reprocessing process would be suitable for partitioning CAPRA fuels similar to those considered in some scenarios. The second one is to evaluate the alternative possibility provided by pyrochemical processes for all highly enriched in plutonium advanced fuels considered in the scenario studies or in other transmutation studies.

As regards the reprocessing of CAPRA fuels by liquid-liquid extraction, the reference CAPRA fuel characteristics were not yet available in 1996. Therefore, only a preliminary assessment have been performed on the standard CAPRA fuel (burnup of 140 GWd/t). It showed that the main issue is the possibility of dissolving the fuel in nitric acid. It also appeared that the current head-end of the process, in particular, with respect to the mechanical operations would have to be designed in a more appropriate way. Finally, the treatment of additional components to fuel assemblies, such as diluent or moderator pins, was also identified as a problem requiring a careful study.

As regards the pyrometallurgical partitioning process, the selected process is originally applicable to metal fuel. The first issue for its application to oxide fuels is to devise a reduction stage to obtain metal. The second one is to obtain a specific separation of actinides which otherwise would remain associated with the plutonium at the same content as that of the irradiated fuel. These two aspects are currently on progress.

C.3 Assessment of the feasibility of transmutation

This Work Package is mainly intended to confirm that the hypotheses taken in the scenario studies with respect to the possibility to massively recycle actinides, and in the first place plutonium, in advanced burner reactors are well grounded. Two solutions are specially emphasised, highly moderated (HM) PWRs and CAPRA type FRs, and this because, although plutonium recycling in PWRs is already an industial reality, its generalisation in the current conditions comes up against two redhibitory hurdles. The first is that present core configurations (30% MOX fuel, 5 to 8% of plutonium in the fuel content) are not sufficient to balance the plutonium build-up and consumption. The second is that calculations show that the successive plutonium recyclings make its isotopic content evolve towards a depletion in odd fissile isotopes. To keep the required reactivity would lead to an increase in the plutonium content of the loaded MOX fuels, which would not meet any longer the current safety criteria.

As regards the HM-PWRs, several parametric studies are in progress. The criteria are on the one hand the respective actinide build up and consumption of the various core configurations, on the other hand the usual safety related criteria. In this respect, the effect of varying the plutonium content of the fuel, the MOX content of the core, the moderating ratio, the fuel burnup and the nature of the fuel matrix is being investigated. Besides, specific core configurations allowing for minor actinide transmutation are also being addressed. There was no milestone associated with this task in 1996 and so no official report have been published.

As regards the transmutation in CAPRA FRs, three aspects are being considered. The first one is to devise a high burnup reactor core suitable for the objectives determined in the scenario studies, i.e., 210 GWd/t. This task is roughly finished and the corresponding report will be published in the beginning of 1997. The second one is to evaluate the effects of minor actinide additions in this kind of reactor. The first step has consisted in reviewing the possible options for incorporation. The third aspect is the study of a core disruptive accident and its effects on safety. The first part of this study was dedicated to the initiation phase and it appeared that, in this phase, CAPRA cores were very similar to that of the European Fast Reactor (EFR) core. The recriticality effects are currently being evaluated. In a preliminary approach they seem more detrimental than in the case of EFR.

C.4 Assessment of the feasibility of advanced fuel or target fabrication

Three kinds of fabrication processes are being evaluated for the fuel and targets needed in the various P&T strategies : sol-gel ones, impregnation ones and processes derived of the current powder blending ones.

In the latter case, the main issue is to evaluate the additional shielding that would be necessary to handle safely the new fuels and targets. This requires to know the characteristics of these materials. These characteristics were not available in 1996 and so this task has been postponed.

As regards the sol-gel methods, a gel supported precipitation (GSP) processed has been devised is being investigated. Some experiments are in progress with plutonium simulated by cerium in the form of sol-gel microspheres and with various inert matrix. Simulated fuel material has thus been fabricated and characterised and the results of that seem promising.

As regards the impregnation methods, an americium target has been fabricated using such a process, in the framework of the EFTTRA program. The obtained material has been characterised and the target is now being irradiated in the HFR reactor in Petten.

No specific milestone was planned in this area in 1996.

C.5 Assessment of geological barrier efficiency

The purpose of this work package is to evaluate the long term risk resulting from the disposal, in deep underground repositories, of the radioactive wastes arising from the nuclear systems considered in the scenario studies. The evaluation is carried out for three possible geological formations, namely clay, hard rock and salt, in which repositories could be built. The first step of this study was to determine how this will be performed. To avoid unnecessary modelling efforts it was decided to have a site specific approach where a site modelling was already available, i.e., in the case of clay and salt formation. In these cases, the selected sites are Mol, Belgium, and Gorleben, Germany, respectively. On the contrary, in the case of hard rock, a generic approach was selected. To take into account the actual waste streams arising in each scenario, the repositories are sized, in a first step, according to their design capacities, and then the long term effects are extrapolated from that after normalisation to 1 TWh.

In accordance with this, first calculations were performed on the case of spent fuel disposal. The maximum resulting dose rates to the most exposed people (3 μSv/a in clay for 5,000 tons of fuel, 8 10^{-3} μSv/a in hard rock for 15 tons of fuel, 7.6 μSv/a in salt for 10,000 tons of fuel), when evaluated in a deterministic way, appear roughly at the same time (100,000 years after disposal) and at the same level (between 0.5 and 1 μSv/a per 1,000 tons of fuel) for all geological formations. The main contribution to that comes from iodine.

Preliminary evaluations of the relative effects of spent fuel in comparison with vitrified high level waste resulting from reprocessing show that the latter have generally lower contribution except in the case of repositories in salt formations in which the effects are similar.

A sensitivity analysis on several chemical characteristics in the neighbourhood of the repositories showed that the effects could be drastically enhanced and modified in their times of occurrence if uncertainties were taken into account.

References

[1] CEA Note SPRC/LEDC 96/4126
[2] ECN Report 71038/DD/1996/004133
[3] CEA Note SPRC/LEDC 96/4111
[4] CEA Note SPRC/LEDC 96/4184

A.2.1-3 Thorium cycles as a nuclear waste management option

Contract No:	FI4I-CT96-0011	**Duration:**	1 May. 1996 - 30 Apr. 1999
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A. OBJECTIVES AND SCOPE

The objective is an assessment of thorium cycles in the context of limitation of nuclear waste production and of prospectives for waste burning. The major cycle steps have to be reviewed, focused on the European situation with thorium fuelled PWRs and FRs as candidate reactors.

For one thorium fuelled hybrid system, the fuel cycle aspects will be considered with emphasis to the radiotoxicity of fuel inventory and waste production.

B. WORK PROGRAMME

The work consists of work packages with respect to the following subjects:

B.1 **Radiological aspects of mining** (CNRS)
B.2 **(Re)fabrication aspects of thorium based fuels** (BN)
B.3 **Reactor assessments**
B.3.1 **Thorium fuelled PWR cores with minimum actinide production (ECN)**
B.3.2 **Thorium fuelled PWR cores for burning transuranium elements (KFA)**
B.3.3 **Thorium fuelled fast reactor cores (CEA)**
B.3.4 **Accelerator-driven systems with thorium (ENEA)**
B.4 **Reprocessing of thorium fuel by the THOREX process** (KFA)
B.5 **Residual risks of long-term disposal** (CNRS)
B.6 **Technical issues related to non-proliferation** (JRC-ITU)

For each work package the results will be reported, including a review of the state of the art and prospectives and problems to be envisaged.

C. Progress of work and results obtained
Summary of main issues

During a kick-off meeting held at Petten , end of June 1996 the action plans of the various working packages have been reviewed. Some adjustments have been made in the planning, agreement has been obtained on standards and methodology and it was decided to have the next meeting at CNRS, Orsay on April 7/8. Since there are already many reviews on the thorium cycle, it was stressed that the present investigation should address the topic of nuclear waste. New in this respect is that the entire fuel cycle, from mining to storage, will be evaluated on possible advantages (compared with U-loaded PWRs) with respect to both long-lived radiotoxicity and radiological effects to the present and future populations. Also the topic of Pu and possibly TRU burning is addressed. Overlap with the current IAEA programme is avoided. More detailed results will be given after the progress meeting in April 1997.

C.1 Radiological aspects of mining (CNRS)

A 4-step action plan was defined at the kick-off meeting consisting of (1) Data collection from literature, (2) Radiotoxicity calculation of tailings from mining, (3) Residual short-term and long-term risks and (4) Health impact to radiological workers. Action (1) has been completed and calculations have been performed on action (2) . The radiotoxicity of the academic case of pure thorium ore is dominated by Ra-228, disappearing after about 60 years. For U-tailings the radiotoxicity is much higher and disappears only after 10,000 to 100,000 years. However, in practice ore contains thorium as well as uranium and therefore the picture is more complicated. Results will be reported later in the contract period.

C.2. (Re)fabrication aspects of thorium based fuels (BN)

A 2-step action plan was formulated : (1) Review of radiation exposure to radiological workers , (2) Evaluation of additional protection measures. Some literature survey has been made and it was decided to limit this activity to fuels considered in thermal reactors only. The work has been postponed until the fuel composition resulting from work packages C.3.1 and C.3.2 is known.

C.3. Reactor assessments

In total four reactor types are being considered from the point of view of minimum
long-lived radiotoxic waste production and Pu- or THR-burning.

C.3.1 Thorium fuelled PWR cores with minimum actinide production (ECN)

The action plan contains the following elements: (1) Reference PWR calculation, (2) Assembly calculations, (3) Evaluation of once-through PWR with Th, (4) Evaluation of PWR core with Th and U-recycling. For the reference core the N4 reactor assembly was selected. Calculations have been performed for this core with the WIMS-7 code package and the results have been communicated to KFA (Work package C.3.2) for intercomparison. Also cell calculations have been performed for a Th/Pu core for intercomparison with KFA. Action (2) starts in 1996.

C.3.2 Thorium fuelled PWR cores for burning transuranium elements (KFA)

The action plan consists of (1) Literature study, (2) Assembly type of burnup calculations, (3) Core calculations, (4) Evaluation. Both once-through and U-recycling options will be considered for Pu and TRU burning. In addition Pa-231 recycling is considered. The same PWR reference core as used in Work Package C1 will be calculated. The literature study has been completed. An important point to take into account is the fact that thorium bearing fuels can withstand a very long burnup. At present assembly type of calculations are being performed.

C.3.3 Thorium fuelled fast reactor cores (CEA)

The CEA action plan consists of the following points: (1) Characteristics and performances, (2) Physical cycle variables, (3) Comparison with standard fast reactors, (4) U-recycling in a park of reactors. Some results have already been obtained by comparing a standard $(U,Pu)O_2$ core with a $(U,Pu,Th)O_2$ core without blankets. An important advantage of the thorium fuelled core is that the reactivity loss over burnup is strongly reduced. Further calculations with the cell code ECCO and the data file JEF-2.2 are in progress.

C.3.4 Accelerator-driven systems with thorium (ENE)

The action plan consists of three points: (1) Validation of codes and libraries, (2) Determination of evolution of k_{elf} and proton current during burnup and (3) Evaluation of fuel inventory and radiotoxicity flow. Action (1) has been completed by performing benchmark tests with the Monte Carlo code for materials occurring in the Fast Energy Ampli-fier, proposed by CERN. The results are satisfactory.

C.4 Reprocessing of thorium fuel by the THOREX proces (KFA)

The KFA action plan consists of three items: 1) Literature survey, (2) Full actinide recycling options, (3) New developments. The literature survey has been completed. The THOREX process for separation of thorium and uranium promises small losses. If Pu has to be recycled as well, THOREX and PUREX have to be combined. Pa recovery is possible in a pre-separation step before the THOREX process starts.

C.5 Residual risks of long-term disposal (CNRS)

A 4-stage action plan was presented by CNRS: (1) Production of ThO_2, (2) Solubility determination, (3) Dissolution determination, (4) Risk calculations using the determined constants. Action (1) has been completed with the production and characterization of ThO_2 in crystalline form. This experimental work is continued with the determination of parameters, necessary as input for the risk calculations of geologically stored spent fuel from the thorium cycle.

C.6 Technical issues related to non-proliferation (JRC-ITU)

The action plan of ITU contains the following points: (1) Identification of relevant nuclides and (2) Isotopic vectors, (3) Definition of parameters expessing non-proliferation resistance, (4) Evaluation. Action (1) has been completed. Action (2) has been completed for the case of FEA (C.3.4).

A.2.1-4 Impact of the accelerator-based technologies on nuclear fission safety (IABAT)

Contract No:	FI4I-CT96-0012	**Duration:**	1 May. 1996 - 30 Apr. 1999
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A. OBJECTIVES AND SCOPE

The overall objective of the IABAT project is to make a European assessment of the possibilities of accelerator-driven hybrid reactor systems from the point of view of safe energy production, minimum waste production and transmutation capabilities.

B. WORK PROGRAMME

The working programme includes the following items :

B.1 System studies on an accelerator driven hybrid

B.1.1 ADS safety and economical assessments, accident scenarios (KTH)

B.1.2 Physics of ADS (CEA)

B.1.3 Analysis of the ADS dynamics (Univ. Chalmers)

B.1.4 Neutronics of ADS (ENEA Casaccia)

B.1.5 Incineration of transuranium elements with the accelerator-driven liquid Lead system (KFA)

B.1.6 Spallation target optimisation (FZK)

B.1.7 Yields and radiotoxicity of spallation products and validation of spallation computer codes (ENEA Casaccia)

B.1.8 Objective for ADS transmutation based on HLW repository risk (AEA Techn.)

B.2 Assessment of the technology and cost of linear and circular accelerators (AEA Techn.)

B.3 Basic nuclear and material data

B.3.1 Evaluated Nuclear Data File (ENDF) for protons on Lead (ECN)

B.3.2 Validation and measurements of some cross-sections above 20 MeV and measurements of the fission yields for ^{233}U thermal fission and ^{232}Th fast fission (Univ. Uppsala)

B.3.3 ENDF for selected isotopes and projectiles up to 200MeV (ENEA Bologna)

B.3.4 Radiation damages at the spallation target enclosure walls (ENEA Casaccia)

B.4 Studies of the fuel cycle for ADS

B.4.1 Studies of ADS for LWR-waste transmutation (KTH)

B.4.2 Accelerator breeding based on Thorium cycle and liquid Lead coolant / carrier (KFA)

B.4.3 Strategy and radiotoxicity consideration for ADS fuel cycles (JRC-ITU)

B.4.4 Evaluation of ADS burning capabilities, fuel inventory and radiotoxicity flow for minor actinide and plutonium burning (ENEA Casaccia)

C. PROGRESS OF WORK AND RESULTS OBTAINED

Specific ADS systems have been chosen for future analysis. These systems cover a wide range of different ADS parameters like: neutron spectra varying from superthermal to fast neutrons, fuel form from solid through suspension in liquid lead to molten salt solutions. ADSs studied in this project involve different fuel cycles: Thorium based Plutonium burnurs, LWR waste incinerators and minor actinides incinerators. A number of code systems to simulate, validate and benchmark different ADS concepts were setup. For high energy transport simulations, codes: : HETC [1], LAHET [2], FLUKA [3] and NMTC/JAERI [4] were used. For neutron transport below 20 MeV, codes MCNP [5], MORSE [6] and TWODANT [7] were adapted. Burnup calculations were performed with ORIGEN [8] and KARBUS [9] codes.

For assessment of the accelerator technology the basic requirements for the accelerator power and performance have been preliminary formulated.

First experiments on ^{232}Th fission yields ^{233}U fast fission yields have been performed at OSIRIS mass-separator facility in Sweden.

The basic nuclear parameters and nuclear model calculations for neutron and proton transport data for intermediate energy range from 20 to 150 MeV. It has been agreed that the optimal way of performing ADS calculation in the future will be the creation of the intermediate energy range cross-section library for neutrons and protons and extending the existing reactor cross section libraries to energy up to 150 MeV.

First calculations of the radiotoxicity of the Th-based ADS fuel cycle were performed.

C.1. System studies on an accelerator driven hybrid

Preliminary physics studies of the different ADS systems were performed with special attention to neutron spectrum, transmutation rates, spallation processes in the target and spallation target optimization. It was decided to setup, use and develop a number of code systems to simulate, validate and benchmark different ADS concepts. For high energy transport simulations, codes: HETC, LAHET, FLUKA and NMTC/JAERI were used, for neutron transport below 20 MeV, codes MCNP, MORSE and TWODANT. Burnup calculations were performed with ORIGEN and KARBUS codes. First studies were performed for a wide range of ADS as thermal and fast neutron systems based on molten salt coolant/fuel and liquid lead based fast and thermal neutron systems with different fuel forms.

For spallation targets studies of the different materials, optimal dimensions and spallation product generation were performed.

Moreover, in this workpackage some analytical studies were started to proof the feasibility of computational models of critical reactor, like point reactor kinetics, for subcritical systems. Also the space kinetics in 2-D cylindrical geometry with fuel recirculation has been studied.

C.1.1. ADS safety and economical assessments, accident scenarios – KTH, Sweden

System studied:

- Molten salt and liquid lead systems

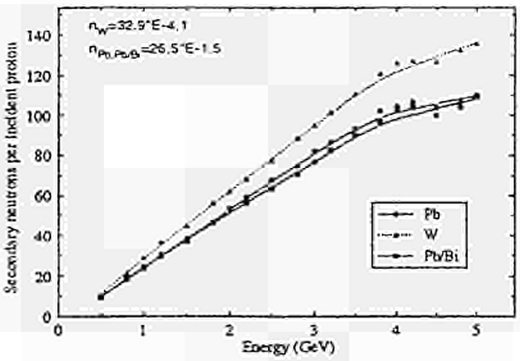
Tasks:

- Performance analysis of the ADS-system for different spectrum. MCNP + ORIGEN
- Coupling FLUKA -MCNP and FRITJOF/DUBNACE –MCNP [10] under way
- Optimization of the spallation target
- Collecting of the accelerator performance data

- MSc thesis completed on analysis of the spallation target with Fluka-code.

Two examples of the project results are presented on Figs 1 and 2.

Fig. 1 presents the results of FLUKA calculation of the neutron spallation yield for Lead/Bismuth target. Fig. 2 shows the dependence of the total neutron production from Pb/Bi spallation target as a function of target dimensions: radius and length.



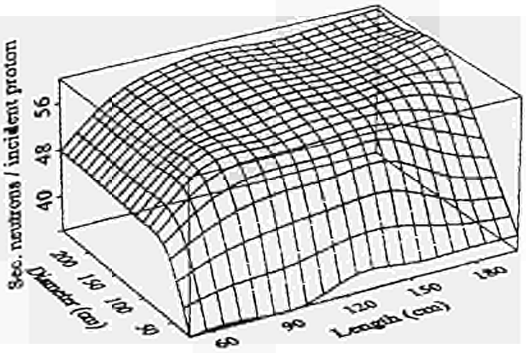


Fig. 1. Spallation neutron production in different target materials as a function of the incident proton energy. Target diameter 25 cm, length 100cm. The expression for the neutron production (upper left corner) is valid for 0.8 < E < 4 GeV.

Fig. 2. Spallation neutron production per incident proton as a function of spallation target size. Incident proton energy - 1.6 GeV, Pb/Bi target.

C.1.2. Physics of ADS – CEA, France

Systems studied:

- a superthermal neutron spectrum system (LANL – ATW system), 500 MWth and or a fast neutron spectrum (JAERI Molten Salt System - JMS), 800 MWth.

Studies on the physics of ADS's have been developed for two systems using molten salts. The composition permits to obtain a superthermal neutron spectrum (ATW) or a fast neutron spectrum (JMS).

The first part of these studies consisted of :

- Defining the molten salt composition to obtain a given type of spectrum.
- Determining the main characteristics of the system.
- Defining the way to obtain an equilibrium.

Calculations use :

- The HETC code to characterize the interactions between protons and target elements down to a cut-off energy of 15 MeV.
- Below 15 MeV the JEF2.2 data are processed through the MICROX2 cell code.
- Core calculations and burn-up are done with 2DTB.

Sample results for ATW system:

A parametric study on the fraction of TRU's in the salt gives the best k_∞ and the composition formula.

Salt Formula : 79.27 LiF - 20.67 BeF_2 - 0.06$(TRU)F_4$.

Table I. TRU in atomic %.

^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{237}Np	^{241}Am	^{243}Am
1.40	51.51	23.77	7.92	4.83	4.58	5.07	0.92

Sample results for JMS system:

Salt Formula : 64 NaCl + 36 $(MA)Cl_3$

Table II. MA's in atomic %.

^{237}Np	^{241}Am	^{243}Am	^{244}Am
56.87	26.27	11.85	5.01

C.1.3. Analysis of the ADS dynamics - CTH, Sweden

Survey of existing literature on ADS dynamics has been performed. A good overview of work by various groups that had been done in the area has been acquired, as well as the main points of interest in ADS dynamics were identified. The concrete way of executing the subsequent items of the work schedule has been specified further.

The acquiring of the basic material constants is underway.

A certain piece of work has been performed that has some relevance to the dynamics of ADS systems. The problem consists of the description of the dynamic response of a multiplying system with a non-constant volume (varying boundaries), and the necessary modification of reactor physics approximations that are used in dynamic calculations [11].

C.1.4. Neutronics of ADS - Politecnico di Torino in contract with ENEA-Casaccia

This workpackage is performed in a contract ENEA - Politecnico di Torino.

Assessment of point reactor kinetics model for source injected subcritical reactor dynamics

The effective delayed neutron fraction for a fully-mixed multiplying system where delayed precursors are instantly redistributed in the core has been evaluated. Typical results are presented in Table III . An overall reduction of 20% is experienced. Therefore, this effect is important and cannot be overlooked in safety assessments.

Solution of space kinetics problems in one-D with fuel recirculation

A one dimensional diffusion problem with an externally-imposed velocity field has been studied. Both analytical and numerical techniques have been employed. Results show a remarkable spatial deformation of the precursor distribution. In Fig. 3 the evolution following an instantaneous and homogeneous change in multiplicativity is reported in a one-group system. Although of course point kinetics would not be applicable, the possibility of a consistent definition of the kinetic parameters is currently being investigated.

Solution of space kinetics in two-D cylindrical geometry with fuel recirculation

The multigroup diffusion equation are being discretized, in the presence of fuel recirculation and neutron source. Preliminary results have been obtained for the reference stationary reactor.

Table III - Effective delayed neutron fraction in a fully mixed subcritical reactor, for different spatial configurations of the source; t_s/H is the fraction of the reactor volume occupied by the source.

t_s/H	$(\beta^{mix}/\beta)_{CPK}$	$(\beta^{mix}/\beta)_{MPK}$
0.005	0.81163	0.80692
0.01	0.81163	0.80692
0.05	0.81163	0.80697
0.1	0.81163	0.80714
0.3	0.81163	0.80885
0.5	0.81163	0.81202
0.8	0.81163	0.81795
1	0.81163	0.82010

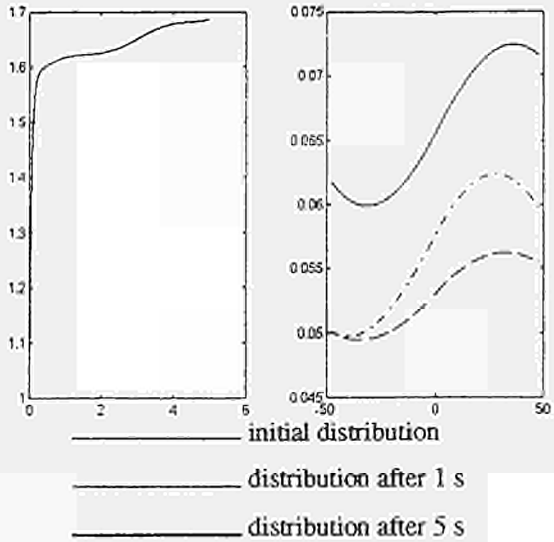


Fig. 3. Power (on the left) and precursor space distribution (on the rigth) transients in a subcritical (k=0.99) reactor following an istantaneous increase of multiplicativity of 200 pcm.

C.1.5. Incineration of transuranium elements with the accelerator-driven liquid Lead system - KFA, Germany

System studied:

- Liquid Lead cooled and fuelled (TRU suspensions) thermal neutron system

Preliminary results have been obtained in studies of the multiplication ratio by cell calculations depending on:

- TRU loading
- TRU composition
- Moderation ratio (pitch)
- Burn-up

C.1.6. Spallation target optimisation - KfK, Germany

System studied:

- (^{233} U, ^{232} Th - core with a hard neutron spectrum

Verification of HETC-TWODANT versus HETC-MCNP

From code comparisons we can conclude that the relative deviations of the one group cross sections of the individual isotopes are less than 20%, except for threshold reactions. The largest relative deviation has been found for the inelastic scattering reaction of ^{233}U (86%). When comparing the results of HETC-TWODANT and HETC-MCNP it has to be kept in mind that the cross section libraries used with MCNP or with TWODANT are based on different nuclear data.

The relative deviation for the total neutron flux density per source neutron is about -40% corresponding to a relative deviation in k_{eff} of about -4% (relative to MCNP).

Investigation of the influence of the location of the proton source within the target

The proton pencil beam enters the cylindrical core from the bottom along its center axis through a vacuum channel of radius 0.5 cm and of variable length. The beam hits the target at the end of the vacuum channel. The energy of the proton beam is 1.5 GeV per proton. The results shown in TableI have been obtained with the HETC-MCNP chain. The impact point of the neutron beam on the target is varied from 0.0 cm (bottom of the core) to 67.0 cm (coremidplane) with step widths of 10 cm to 17 cm.

Investigation of the influence of different target materials

In an further investigation the material composition of the otherwise homogeneous core has been modified in a cylindrical region of 0.5 cm of radius around the center axis of the core. In this region the homogeneous mixture has been replaced in different studies by ^{232}Th, ^{208}Pb ^{186}W , Zr^{nat} , ^{233}U, ^{239}Pu (with 100% and 90% of its theoretical density) and core: ^{232}Th, ^{233}U mixed oxide with 14 at% of ^{233}U. For the different material substitutions along the center of the core the total number of neutrons per proton (produced by intranuclear cascade reactions (INC), evaporation (EVAP), and high energy fission (HIFISS)) has been calculated using HETC. Moreover the number of neutrons produced in the center zone (r=0.5 cm), the number of neutrons with E 20 MeV and the number of neutrons with E 10 MeV have been determined. The largest number of spallation neutrons is obtained, if the central region is filled with ^{239}Pu with 100% theoretical density, namely 49.2 neutrons per proton. The smallest number of neutrons (36.6) is obtained, if in the center region the homogeneous mixture of the core is replaced by Zr^{nat}.

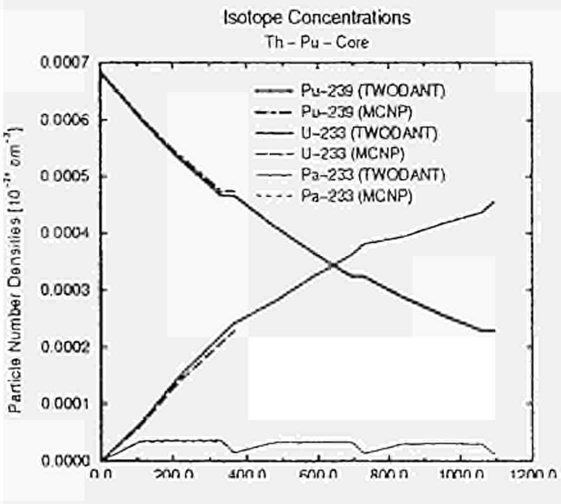


Fig. 4. The isotopic concentration of ^{239}Pu, ^{233}U, and ^{233}Pa during the burnup time

Incineration of LWR waste Pu in a 3000 MWth core with Th^{232} - Pu fuel

The code chain HETC-TWODANT-KARBUS has been applied to calculate the incineration of Pu in a 3000 MW(th) core with ^{232}Th - Pu fuel.

Figure 4 shows the development of the isotopic concentration of ^{239}Pu, ^{233}U, and ^{233}Pa during the burnup time specified above. For the results that are represented by dashed

lines neutron flux density spectra from MCNP calculations have been used for the determination of the one group cross sections used in the burnup calculations.
Table IV gives the burnup dependant concentrations of 20 actinides.

Table IV. Change of the Actinide composition of a ^{232} Th-Pu core during Burnup

Proton Energy 1600 MeV/ Power of the Core 3000MW(th)							
	weight in kg						
Isotop	Begin	365 d	Change	$2 \star 365$ d	"Change	$3 \star 365$ d	"Change
Pu^{238}	64.18	48.90	-15.27	39.82	-9.08	34.33	-5.49
Pu^{239}	2267.72	1545.80	-721.92	1076.45	-469.35	757.80	-318.65
Pu^{240}	966.70	967.38	6.81E-1	923.59	-43.80	859.96	-63.62
Pu^{241}	519.89	369.74	-150.15	283.19	-86.55	228.44	-54.75
Pu^{242}	220.24	227.57	7.33	225.60	-1.97	219.28	-6.32
$\sum Pu$	4038.72	3159.40	-879.32	2548.64	-610.58	2099.80	-448.84
Th^{232}	19121.13	18060.89	-1060.24	17105.83	-955.05	16223.61	-882.22
Pa^{233}		47.17	47.17	42.55	-4.61	39.31	-3.24
U^{232}		4.65E-1	4.65E-1	8.78E-1	4.13E-1	1.25	3.75E-1
U^{233}		782.98	782.98	1232.35	449.38	1469.36	237.01
U^{234}		31.02	31.02	75.96	44.93	122.13	46.17
U^{235}		1.77	1.77	7.75	5.98	17.14	9.38
U^{236}		1.45E-1	1.45E-1	5.97E-1	4.53E-1	1.62	1.02
Np^{237}		1.93E-2	1.93E-2	7.35E-2	5.42E-2	1.81E-1	1.07E-1
Np^{239}		1.29E-5	1.29E-5	2.18E-5	8.92E-6	2.80E-5	6.19E-6
Am^{241}		17.65	17.65	25.92	8.27	29.32	3.40
Am^{242}		2.85E-6	2.85E-6	7.64E-6	4.79E-6	1.17E-5	4.04E-6
Am^{242m}		2.38E-1	2.38E-1	6.38E-1	4.00E-1	9.76E-1	3.38E-1
Am^{243}		15.04	15.04	25.42	10.38	32.62	7.21
Cm^{242}		1.04	1.04	2.18	1.14	2.76	5.78E-1
Cm^{244}		2.00	2.00	6.34	4.35	11.63	5.29

Pu incinerated : 597 kg/GW(e)a, U^{233} accumulated in irradiated fuel: 451 kg/GW(e)a

total production of U^{233} in 3 years: 2.8t, U^{233} contained in irradiated fuel: 1.5t

reduction of U^{233} by in situ fission: 1.2t, reduction of U^{233} by transmutation into U^{234}: 0.1t

C.1.7. Yields and radiotoxicity pf spallation products and validation of spallation computer codes . - ENEA Casaccia, Italy

Calculational tools: LAHET, HETC and NMTC/JAERI codes.

Table V. Comparison between experimental and calculated neutron yields. E_B = 960 MeV. LAHET/MCNP calculations

Material	Target size (cm)	Experimental Data	Calculation Results
Uranium	10X60	40.5	39.5
Lead	20X60	20.5	21.5
Lead	10X60	17.2	18.3
Tin	10X60	12.5	12.2
Beryllium	10X10X91	2.7	1.79

Simulation of COSMOTRON Experiments.

The COSMOTRON experiments have been simulated for the validation of the code systems LAHET/MCNP and NMTC-JAERI/MCNP.

Preliminary LAHET/MCNP calculation results, corresponding to a proton beam having energy of 960 MeV, see Table V, show an excellent agreement with the experimental data except for Be.

C.1.8. Objective for ADS transmutation based on HLW repository - AEA Techn., United Kingdom

A survey of the literature on both the objectives of partition and transmutation and on the risk assessment of repositories for spent fuel and HLW has been started. The risk assessments are being used to identify which radionuclides it would be important to transmute from a safety point of view. The aim is to provide a provisional list of which radionuclides need to be transmuted.

C.2. Assessment of the accelerator technology – AEA Techn., UK.

Work in the first six months on this task has been restricted to planning and starting to survey the recent literature on accelerator technology for relevant data. The aims for the next reporting period are to continue the literature survey and then to start developing conceptual designs for prototype and commercial circular accelerators which will in future periods be assessed and costed. A decision will be made on what circular accelerator types will be assessed.

C.3. Basic nuclear and material data

This workpackage comprises some cross-section measurements and cross-section data formatting and processing and the validation of nuclear reaction model codes. First experiments on ^{232}Th fission yields ^{233}U fast fission yields have been performed at OSIRIS mass-separator facility in Sweden. The basic nuclear parameters and nuclear model calculations for neutron and proton transport data for intermediate energy range from 20 to 150 MeV. It has been agreed that the optimal way of performing ADS calculation in the future will be the creation of the intermediate energy range cross-section library for neutrons and protons and extending the existing reactor cross section libraries to energy up to 150 MeV. For the first sample case , ^{56}Fe data file has been extended up to 150 MeV and soon will be available for the community.

C.3.1 Evaluated Nuclear Data File (ENDF) for protons on Lead - ECN, Netherlands

The work proceeds in two parallel directions with focus iron and lead isotopes. In first instance neutron induced reactions are considered only since these have a direct application in applied (MCNP) calculations [12].

Assembling basic nuclear parameters and nuclear model calculations

- A new proton and neutron optical model between 0 and 200 MeV is under construction, using the interactive optical model visualisation program ECISVIEW.

- Initial nuclear model calculations have been performed with GNASH for neutron energies between 20 and 150 MeV with the Ignatyuk parametrization. The results have not yet been benchmarked against experimental data.

ENDF-6 data Format and processing

The data file for Fe-56, extension of the EFF-3.0 fusion file up to 150 MeV, has been accomplished. This library can be soon be used by the community.

C.3.2. Validation and measurements of some X-sections above 20 MeV and measurements of the fission yields for ^{233}U thermal fission and ^{232}Th fast fission - UU, Sweden

Fission yields

^{232}Th fission yields

The data set obtained early in 1996 is still being analyzed. These data are somewhat limited due to the rather low fission cross section of ^{232}Th . It is expected to obtain yield values only for the products on the peaks of the yield distribution.

^{233}U fast fission yields

Calculations and experiments performed early in 1996 showed that a filter of boron carbide would fulfil the requirements that more than 70% of the fission events are induced by fast neutrons. The experimental study is presently planned for the early part of 1997. The neutron filter is under construction. A special data-taking system is also under construction, to permit the use of a multi-detector counting system. This will improve the quality of the data obtained.

The validation of nuclear reaction model codes for some cross section data above 20 MeV

Several neutron induced cross section measurements are in progress or planned at the intermediate energy neutron facility at the The Svedberg Laboratory in Uppsala, Sweden. In a report recently accepted for publication in Nucl. Phys. A [13], the experimental data for the $^{208}Pb(n,p)$ reaction are published and compared with theoretical calculations based on the quantum-mechanical statistical direct reaction multistep model according to Tamura, Udagawa and Lenske (TUL) [14,15].

C.3.3. ENDF for selected isotopes and projectiles up to 200 MeV - ENEA, Bologna, Italy

This workpackage is scheduled to begin later.

C.3.4. Radiation damages at the spallation target enclosure walls –ENEA, Casaccia, Italy

Acquisition of the MCNP cross sections for Hastelloy-N to calculate the atomic displacements" is completed, Fusion Evaluated Nuclear Data Library was chosen to perform the evaluations of radiation damage at the spallation target enclosure walls.

C.4. WORKPACKAGE D : Studies of the fuel cycle for ADS

Studies in this workpackage have been concentrated on different fuel cycles for ADS:

- U-Pu fuel cycle for LWR-waste transmutation and Pu-burning
- Th-based fuel cycle and its radiotoxicity
- Mixed U-Pu-Th fuel cycle

Preliminary analysis were done on the impact of neutron spectrum on transmutation rates and radiotoxicity of residuals.

C.4.1. *Studies of ADS for LWR-waste transmutation and Pu incineration - KTH, Sweden*

Calculations of the transmutation rates at BoL have been performed for different neutron spectra. Inventory of the necessary nuclear data, which are missing in the standard libraries, has been accomplished. Fig. 5 presents a typical calculation of the effective microscopic cross-section for transmutation (capture+fission) as a function of average neutron energy in the system (indication of the type of neutron spectrum). It is clear that some of the isotopes can be transmuted most effectively in the thermal neutron spectrum, other in intermediate spectrum.

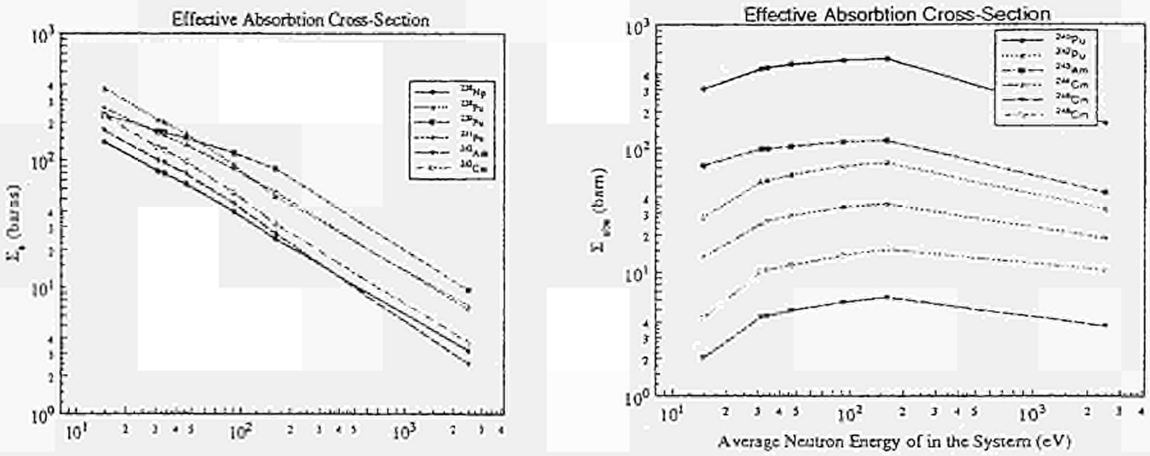


Fig. 5. Effective transmutation cross-sections for 2 different group of Actindes as a function of the neutron spectrum in the system (indicated by the average neutron energy in the system).

C.4.2. *Accelerator breeding based on Th-cycle and liquid lead coolant/carrier – KFA, Germany*

Investigations have been started, but have not yet been come to a final result. It can be stated already, that loadings of TRU-dioxides between 0.5 to 2 w/o-% homogeniously distributed as a suspension in liquid lead are sufficient to establish a criticality level of about 0.95. This is strongly dependant on the moderator, which is selected as graphite graphite, and the pitch. Besides the TRU-loading, the variation of the pitch seems to be an effective means for controlling the criticality.

C.4.3. *Strategy for the ADS fuel cycles, radiotoxicity consideration of these cycles - ITU, EC*

Calculational protocol for multiple irradiation and recycling using ORIGEN2 has been established. Cross section data used were for a fast lead cooled system [16].

Waste resulting from 10th cycle spent fuel (1 cycle is 5 years irradiation plus 1 year cooling time) from a fast Thorium fuelled ADS has been characterised. Actinide reprocessing losses were taken to be 1% of the core inventory at the end of each cycle. Ingestion radiotoxicity of this waste has been calculated for cooling times up to 10^6 years after discharge. In addition to the evolution of total toxicity, the results are also grouped by chemical element.

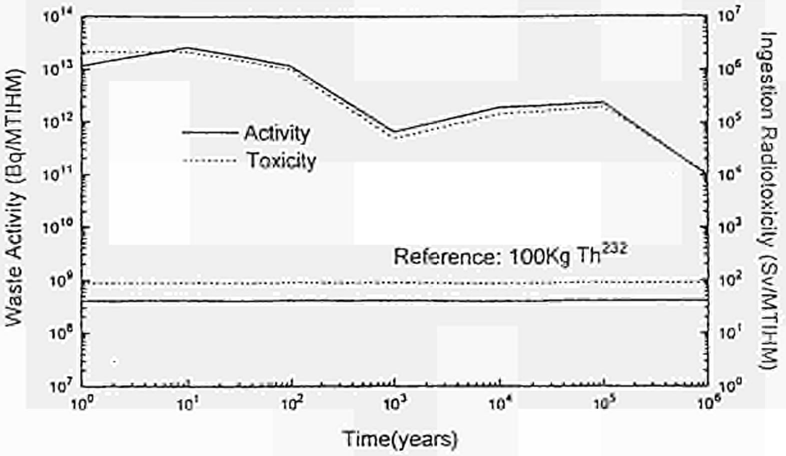


Fig. 6. Time evolution of the waste activity and ingestion radiotoxicity from 10^{th} cycle spent fuel from a fast ADS. Losses to waste are assumed 1% of core inventory. After 5 years irradiation, approx. 100 kg ^{232} Th must be added to replenish material losses through energy production.

From the waste activities at discharge and the effective dose coefficients, the radiotoxicity was calculated. The results are summarised in Fig. 6 which shows the time evolution of the waste activity and ingestion radiotoxicity for a period of 10^6 y after discharge. For reference purposes, the activity and toxicity of 113 kg of pure Th^{232} are shown. This is the amount of fresh material which must be added at each cycle to make up for losses after the 5 year irradiation period. It should also be noted the total mass of actinides is approximately 9 kg /MTIHM.

It can be seen from Fig.6, that the total activity and total ingestion radiotoxicity can be related by an "average dose coefficient".

For the first thousand years, the radiotoxicity is dominated by uranium (mostly U^{232} (half-life≅70y)) and plutonium (mostly Pu^{238} (half-life≅88y)). Thereafter, Thorium dominates (mostly Th^{229}) with contributions from Pb (Pb^{210}), Po (Po^{210}), and Ra (Ra^{226}) becoming more important.

C.4.4. *Evaluation of ADS burning capabilities fuel inventory and radiotoxicity flow for minor actinide and for Plutonium burning - ENEA, Casaccia, Italy*

Evaluation of burning capability of a liquid fuel Pu burner

A Monte Carlo calculation model has been developed to determine the criticality safety conditions and the burning capability of Pu and its associated MAs. The target was analyzed using NMTC-JAERI Monte Carlo code.

Both the neutron multiplication factor calculations and the flux and power ones, have been performed with MCNP Monte Carlo code. Pu burning calculations have been obtained with ORIGENS, using the Molten Salt Breeder Reactor (MSBR) Library. Liquid fuel has been considered and continuous reprocessing and fission product removal has been envisaged. A Pu burner, with a core loaded with Th to compensate reactivity due to the even Pu isotopes, can operate at a low proton current using perhaps a cyclotron, incinerating 70% of the charged Pu; its burning capability would be the production of about 1.5 PWR.

The influence of the even plutonium isotopes on the plutonium burning capability.

In the case of reactor-grade Pu-fulled core, after 11 irradiation days the percentage of even Pu isotopes (neutron poisons) is practically equal to the one of fissile isotopes, while at the beginning of irradiation this percentage was about 31.6. The increase in the even Pu isotope concentration explains the need for continuous enhancement of the proton current in order to keep the thermal power constant.

Poisoned plutonium burners

Pu-fuelled core with ^{99}Tc as a poison, that would be removed to compensate for the reactivity drop due to the even Pu isotopes, can operate at a low proton current using perhaps a cyclotron; but this system could burn only 27% of the charged Pu. A better system using Er instead of ^{99}Tc would burn 62% of the charged Pu; while the best solution based on the poisoning philosophy would use ^{232}Th instead of ^{99}Tc and burn 70% of the initial Pu load. The burning capability of these three burners would be the production of about 1.5 PWRs.

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A.2.2-1 New partitioning techniques (NEWPART)

Contract No:	FI4I-CT96-0010	**Duration:**	1 May. 1996 - 30 Apr. 1999
Coordinator:	**C. Madic, CEA-Valrhô/FR**		
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Partners:	**Univ. Reading/GB, Univ. Chalmers Göteborg/SE, JRC-ITU Karlsruhe/EC, ENEA Saluggia/IT, FZK Karlsruhe/DE, KFA Jülich/DE, Univ. "Politecnico" Milano/IT**		

A. OBJECTIVES AND SCOPE

The main objectives of the project are to develop processes for the separation of minor actinides:

1. without the generation of secondary solid wastes.
2. minimising the majority of the drawbacks encountered in the previous processes, such as the precipitation of radionuclides, difficulties in the stripping of the actinides from the solvent, etc.

The following principles for the design of the processes are:

1. to develop molecules (extractants, diluents, aqueous reagents) that, when they are at the end of their use in the processes, can be converted into gases which can be released into the atmosphere (CHON principle).
2. no reduction of the acidity of the HLLW to be processed.

Accordingly, the separation strategy includes two routes and broad sets of measurable objectives:

1. the first route includes several extraction cycles, the first to extract the actinides and the lanthanides from HLLW, and the second cycle to separate the An from the Ln. Some effort will be devoted to the Am/Cm separation according to the time available.
2. the second route corresponds to a single cycle process for which the fundamental criteria using molecular design, synthesis and evaluation for the simultaneous selective extraction of An (III) from Ln (III) in HNO_3 aqueous solutions (<2M) must be established.

B. WORK PROGRAMME

B.1 Cooextraction of actinides and lanthanides

B.1.1 Basic studies on diamides (CEA Valrhô, Univ. Reading, Univ. Chalmers Göteborg)

This task involves the study of extraction thermodynamics, mechanisms and kinetics. Structural determinations of metal nitrate solvates of diamide extractants and molecular modelling are performed. The diamide formula will be optimised and new molecules will be synthesized.

B.1.2 Development of the DIAMEX process (CEA Valrhô, Univ. Chalmers Göteborg, JRC-ITU Karlsruhe, ENEA Roma, KFA Jülich, Univ. "Politecnico" Milano)

The reference DIAMEX process using the dimethyldibutyltetradecylmalonamide (DMDBTDMA) extractant is being developed. Data banks concerning the distribution coefficients of the metallic species present in the reference HLLW and the loading capacities of the solvent versus solutes of interest will be completed. The stability of the solvent during α, β, γ irradiation will be studied and the hydrolytic and radiolytic degradation compounds of the solvent will be identified. Counter-current integrated tests (mixer-setters or/and centrifugal contactors) will be performed. Different DIAMEX processes will also be investigated. In addition, computer modelling of the DIAMEX processes will be carried out.

B.2 Separation of actinides (III) from lanthanides (III)

B.2.1 Basic studies of heterocyclic nitrogen compounds (CEA Valrhô, Univ. Reading, Univ. Chalmers Göteborg, FZK Karlsruhe)

B.2.1.1 Hydrophobic tripyridyltriazine (TPTZ derivatives)

Extraction thermodynamics, mechanisms and kinetics will be studied with various molecules. The structures of metal salt solvates of hydrophobic TPTZ derivatives will be measured and modelled. The TPTZ derivative formula will be optimised and new molecules will be synthesized.

B.2.1.2 New heterocyclic N donor atom extractants, e.g. oligomeric pyridines

The new extractant molecules will be designed with molecular modelling, synthesized and characterised and their properties will be studied.

B.2.2 CYANEX 301 (KFA, Jülich)

The use of CYANEX 301, a sulphur bearing acidic extractant, for the An(III)/Ln(III) separations will be studied.

B.2.3 Process development (CEA Valrhô, JRC-ITU Karlsruhe, ENEA Roma, KFA Jülich, Univ. "Politecnico" Milano)

A process based on the tritertiarybutylpyridyltriazine will be developed. This involves the definition of the solvent composition and the determination of distribution coefficients for An (III), Ln (III) and other metal ions. The loading capacities of the solvent for the major solutes of interest, its hydraulic behaviour in the extraction devices and its degradation/regeneration properties will be investigated. Counter-current integrated tests (mixer-settlers or/and centrifugal extractors) will be performed. Finally, the An (III)/Ln (III) separation processes will be modelled.

C. PROGRESS OF WORK AND RESULTS OBTAINED

. Summary of the main issues

Numerous results were obtained during the first semester of the research. They can be summarised as follows.

1. Coextraction of An(III) and Ln(III).

To achieve this goal without modifying the acidity of the high active effluent to be processed, diamide extractants were selected (DIAMEX process). Basic research and process development were the subjects of several studies carried out by partners 1, 2, 3, 4 and 5.

The main achievements obtained in the basic studies are the following : (i) synthesis of several new diamides, (ii) distribution studies of actinides (III), and lanthanides (III) nitrates, including the evidence of a synergistic phenomenon with mixture of diamide+carboxylic acid, (iii) crystal structure determination of some Ln(III) nitrates solvated by diamide molecules, (iv) initial experiments carried out at the synchrotron LURE facility at Orsay for SAX studies of diamide solvates, (v) molecular modelling of diamide extractants, (vi) proposition of a new optimised diamide : the dimethyldioctylethoxyhexylmalondiamide (DMDOEHMDA) with improved properties compared to that of the reference diamide (DMDBTDMA).

DIAMEX process development. This development is based on the use of DMDBTDMA. (i) Scrubbings sections for the process flowsheet, in order to prevent the coextraction of Zr(IV) and Mo(VI) fission products with the An+Ln(III) mixture, were studied. Two scrubs were tested in cold counter-current tests, one based on the use of oxalic acid and the second involving a double scrub using hydrogen peroxide and ketomalonic acid, successively. Both systems can be proposed for the design of a DIAMEX flowsheet but the use of only one scrub (oxalic acid) is recommended. (ii) A first version of a computer code of the DIAMEX process was established.

2. Separation of An(III) and Ln(III).

Basic studies carried out in this field by partners 1, 2, 3, 6 and 7 can be summarised as follows : (i) terpyridine (terpy) was found effective in synergistic combination with carboxylic acid(s) to selectively extract An(III) over Ln(III), (ii) lipophilic polypyridine (terpy, quinquepy) were synthesised successfully. Instead of improved extracting properties, it was found that alkylated terpy are less efficient than terpy itself. This point must be understood in the future, and we need to consider especially the electronic density on the pyridinic nitrogen which certainly must be kept low, (iii) other N donor extractants were studied by partner 6. Bidendate ligands, like 2-substituted benzimidazoles, and also tridendate ligands, were studied for the selective extraction of An(III) over Ln(III). Interesting separation factors between An(III) *vs* Ln(III) were often obtained, (iv) important results were obtained by partner 7 demonstrating the effectiveness of purified CYANEX 301, a dithiophosphinic acid, for the An(III)/Ln(III) separation. The results obtained confirm the necessity to purify the CYANEX 301 by the Chinese method (precipitation of the ammonium salt of CYANEX 301) and that the American purification method is inefficient. Tremendously high An(III)/Ln(III) separation factors were obtained (higher than 10^3) confirming the data published in 1995 by Y. ZHU *et al.* from Beijing (China).

C.1 Coextraction of actinides and lanthanides

C.1.1 Basic studies on diamides

Task A1 and Task A2. Several new diamide molecules were synthesised and their extracting properties studied. Particularly, dimethyldicyclohexanotetradecylmalonamide (DMDCHTDMA) and dimethyldiphenyltetradecylmalonamide (DMDPHTDMA) prepared at Reading were studied at Göteborg. The extraction behavior of nitric acid and of some lanthanides (III) and actinides (III) nitrates by these malonamides were studied according to the concentration of nitric acid or lithium nitrate in the aqueous phase. Whereas the affinity of the two diamides for nitric acid were similar, definite differences were observed between these two extractants regarding the metallic nitrates. The diphenyl malonamide (DMDPHTDMA) exhibits higher affinities than that of the DMDCHTDMA. The question of the dual extraction mechanisms, solvate formation for low aqueous nitric acid concentration and lithium nitrate (every concentration) and ion-pair formation for high aqueous nitric acid concentration suggested in the past by L. NIGOND (CEA) is still open and must be addressed in the future.

Task A4. To have a better description of the extracting properties of malonamides, structural determinations using X-ray diffraction or X-ray spectroscopic methods were undertaken either on single crystals or on liquids. For example, the structure of crystals made by Nd(III) and Yb(III) nitrates with DMDCHTDMA or DMDPHTDMA were determined. The stoichiometries of the compounds prepared are as follows : $M(NO_3)_3L$ (L = DMDPHTDMA) and $M(NO_3)_3L_2$ (L= DMDCHTDMA). Both extractants can certainly generate 1/1 and 1/2 complexes in organic solutions but only the 1/1 complex precipitates in the case of DMDCHTDMA. Attempts to prepare suitable crystals of the possible Ln(III)/malonamide ion-pairs have been unsuccessful up to now. Efforts will be done in the future to succeed in this topic. To determine the coordination polyhedra of metal ions extracted in organic phases, it was decided to use X-ray absorption (XAS) spectrometries (EXAFS and XANES). Such techniques can be operated at synchrotron facilities like LURE Orsay, France), ESRF (Grenoble, France) or Daresbury (U.K), *etc*... Experiments in that field were undertaken at the CEA. To test the method, a known system was selected. It consits in the study of the solvates formed when uranyl nitrate is extracted with neutral organo-phosphorus compounds, like the tri-n-butylphosphate (TBP), the extractant used to implement the PUREX process (nuclear fuel reprocessing). For this system, it was demonstrated that the coordination polyhedron of U(VI) in the organic liquid phase resembles that of U(VI) in the crystal of a related compound $UO_2(NO_3)_2(TiBP)_2$. It was thus demonstrated that SAX methods are useful to study solvent extraction systems. Co-operative work between partners 1 and 2 is continuing in that field.

Several experiments have been proposed to be undertaken at LURE and Daresbury to study malonamide/lanthanide nitrate solvates.

Task A5. Molecular modelling was undertaken in order to try to understand the differences in extraction affinities for metal ions of the two diamides mentioned above : DMDCHTDMA and DMDPTDMA. The lowest energy conformations of the two molecules were found to be different, in agreement with their crystal structures. This data is mirrored with the extraction affinities of the two diamides for An(III) and Ln(III) ions.

Task A6. Malonamide formula optimisation was the subject of intense research at the CEA. Several criteria were selected for this selection, such as the affinity of the diamides for An(III) and Ln(III) metal ions, the loading capacities of the solvent without third phase formation, the extractant degradation and regeneration, *etc*... To optimise all the required properties, one can adjust the nature of the groups attached to the amide functions : one can select the length of alkyl groups and/or introduce either function in these groups. Note that one group branched to each nitrogen must be a methyl group. The main conclusions of that study are the following : (i) the total number of carbons of the groups must be higher or equal to 26, (ii) the second alkyl group branched on each nitrogen must be alkyl, while the group branched on the central carbon bridge must be : $C_2H_4OC_nH_{2n+1}$, (iii) sharing the carbons between the central group and the two groups branched on nitrogens is recommended. Consequently, the new diamide : dimethyldioctylethoxyhexylmalonamide (DMDOEHMA) is the optimised molecule which will be synthesised and studied in the near future.

Task A7. As mentioned above, several new diamides were synthesised at Reading and in France (by PANCHIM).

C.1.2 Development of the DIAMEX process.

Task B1. Active tests carried out at CEA Fontenay-aux-Roses in 1993 have demonstrated the necessity to define efficient scrubbing sections in the DIAMEX flowsheet based on the use of DMDBTDMA to prevent the extraction of two important fission products : the zirconium (Zr(IV)) and the molybdenum (Mo(VI)). Two scrubbing sections were designed and tested successfully during counter-current tests carried out in mixer-settler batteries. The first scrub tested was based on the use of oxalic acid to selectively complex both Zr(IV) and Mo(VI) ions, while the second scrub (dual) uses two different complexing agents : hydrogen peroxide for Mo(VI) removal and ketomalonic acid for Zr(IV) complexation. Both scrubbing systems operate efficiently. Thus, it is recommended to use the simplest scrub based on the use of oxalic acid for the definition of the reference DIAMEX flowsheet. Active contacts between partners 1, 4 and 5 were established in order for partners 4 and 5 to be able to test the DIAMEX process in a near future.

Task B4. A first version of the DIAMEX process was defined by partner 1 and tested using the data collected during the active trials of the process (1993) and those obtained during the cold tests carried out for the definition of the Zr(IV) and Mo(VI) scrubbing sections. Good agreements between calculated and experimental concentration profiles were obtained for the following species : ° nitric acid, ° Ln(III) nitrates, ° Zr(IV) and ° Mo(VI). One problem is still pending : the behaviour of Fe(III) (a corrosion product) is not well calculated by the model. This difference between calculated and experimental data is surely due to the slow kinetics of extraction of this species by the solvent. This point must be addressed in the future.

C.2 Separation of actinides (III) from lanthanides (III)

C.2.1 Basic studies of heterocyclic nitrogen compounds

Task C2. New heterocyclic N donor atom extractants were studied. These extractants include oligopyridines, like terpyridines and quinquepyridines, studied by partners 1, 2 and 3, and other molecules, like 2-substituted benzimidazoles and 2,6-Bis(4,5-dihydro-(4S)-isopropyloxazol-2-yl)pyridine, studied by partner 6. The main results obtained can be summarised as follows.

(i) Terpyridine (terpy) is a suitable ligand, able, in synergistic combination with carboxylic acids, to selectively extract the trivalent actinides over the trivalent lanthanides with a good separation factor ($S_{Am/Eu}$ close to 10). This data is similar to that corresponding to the tripyridyltriazine (TPTZ) which is the reference compound in this field. Nevertheless, to obtain similar distribution coefficients for M(III) ions with terpy for a particular aqueous acidity it is necessary to use a more concentrated solution than in the case of the TPTZ (0.1 mol/L instead of 0.02 mol/L). Nevertheless, it was thus concluded that terpy is a suitable « platform » for the design of good extractants if more lipophilic molecules can be synthesised by branching lipophilic groups on the pyridinic rings of the terpy. Two alkylated terpy derivatives were made by partner 1 by branching octyl or dodecyl groups in position 4 on the central pyridinic ring. The solubilities in water of these compounds decreased drastically in comparison with the corresponding terpy data. Meanwhile, solubilities of these new terpy compounds in aliphatic dilutents increase a lot in comparison with terpy. Consequently, better affinities for M(III) ions for these new terpy in comparison with terpy were expected. The data obtained so far exhibit exactly the reverse trend : if $S_{Am/Eu}$ are similar (or slightly lower), the distribution coefficients of the trivalent metal ions are shifted to lower values. One possible explanation for this unfortunate result is that the branching of the alkyl group on the pyridinic ring enhances the basicity of the nitrogen. Molecular modelling will be used in the future to check this hypothesis.

(ii) Two new quinquepyridines have been synthesised at Reading with lipophilic groups branched in position 4 on the pyridinic rings 2 and 4. One of these new compounds has been characterised by X-ray cristallography. Samples of these ligands will be transfered to partners 1 and 3 for the determination of their extracting properties.

(iii) 2-substituted benzimidazoles. These ligands were studied by partner 6 for the selective extraction of An(III) over Ln(III) when the metal ions are extracted as MA_3L_n, where A^- is an anion (I^-, SCN^-, ClO_4^-, NO_3^-, thenoate) and L the 2-substituted benzimidazole. Important $S_{Am/Eu}$ (50 to 70) were observed under certain conditions, for example using 6-methyl-2(2-pyridyl)benzimidazole in the dilutents chlorobenzene and xylene/4-methyl-2-pentanone, with an aqueous phase consisting of 1.0 mol/L NH_4SCN in 0.04 mol/L formate buffer. It was demonstated that the higher the number of ligand L included within the metallic solvates, the higher the $S_{Am/Eu}$ is. Nevertheless, the good results obtained for pH values may be too high for practical uses.

C.2.2 Basic study with CYANEX 301

CYANEX 301 is a commercial extracting agent which contains about 75-80% of bis(2,4,4-trimethylpentyl)dithiophosphinic acid. Even if this compound does not fulfill the CHON principle, its istudy is considered important because Chinese and American scientists published papers in 1995 demontrating that, under certain conditions, highly efficient An(III)Ln(III) separations can be obtained using this extractant.

The crude extractant is unable to perform the An(III)Ln(III) separation unless it is saponified, *i.e.* neutralised with sodium hydroxide. Efficient separations were observed at KFA with unpurified CYANEX 301 for high Ln(III) concentrations, but not for low Ln(III) concentrations. Purification of CYANEX 301 was performed using the Chinese (Y. ZHU) and American (G. JARVINEN) methods. The purities of the purified compounds were checked by ^{31}P NMR. Both methods produced pure bis(2,4,4-trimethylpentyl)dithiophosphinic acid but the yield corresponding to the American method was rather poor (5%) in comparison with that corresponding to the Chinese method (40%).

Purified CYANEX 301 (both methods) were studied for its ability to separate An(III) over Ln(III). Very high Am/Eu separation factors (higher than 10^3) were obtained for micro or macro concentrations of Eu(III). This corresponds to the first confirmation of the Chinese results published in 1995. A problem was identified in regards to the instability of the purified CYANEX 301, generating impurities with detrimental properties for An(III)Ln(III) separations. This point needs to be solved in the future.

It can be forecast that the use of CYANEX 301 will be good for efficient An(III)Ln(III) separations from real active effluents.

C.2.3 Process development

Task D1. An An(III)Ln(III) separation process based on the use of tri-tertiobutyltripyridyltriazine (T*t*BTPTZ) will be developed. This requires the synthesis of kg amount of T*t*BTPTZ. Preliminary contacts with an industrial synthesis company were made for the preparation of this extractant.

A.2.3-1 Joint EFTTRA experiment on Am transmutation

Contract No:	FI4I-CT95-0007	**Duration:**	1 Feb. 1996 - 31 May 1999
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A. OBJECTIVES AND SCOPE

The objective of the project is to study the transmutation of Am-241 embedded in an inert matrix by preparing, performing and analysing an irradiation experiment in the High Flux Reactor (HFR) at Petten. It serves as a European pilot project to demonstrate the feasibility of Am transmutation and includes the fabrication of a target pin, the execution of irradiation and post-irradiation experiments and the analysis of the results.

The work will be performed jointly by the existing EFTTRA group of laboratories and institutes. EFTTRA is an acronym for "Experimental Feasibility of Targets for Transmutation".

B. WORK PROGRAMME

In order to meet the objectives described above the work is divided into 8 work packages, according to the following scheme:

B.1 Co-ordination (ECN)
B.2 Design and safety report by the irradiation in HFR (ECN, JRC-ITU, JRC-IAM)
B.3 Target preparation and transport (JRC-ITU, FZK)
B.4 Irradiation facility preparation (JRC-IAM)
B.5 Irradiation in HFR during one-and-half year (JRC-IAM, ECN)
B.6 Non-destructive analysis (ECN)
B.7 Transport and post-irradiation experiments (JRC-ITU, FZK)
B.8 Interpretation and final report (all participants)

C. Progress of work and results obtained

Summary of main issues

The irradiation of a target of ^{241}Am in a host matrix of spinel has been started in the HFR, according to the planning. The preparation of the experiment has been completed successfully. A novel fabrication technique (infiltration) has been developed and has been used for the fabrication of the target. The target has been transported from Karlsruhe, where the fabrication was done, to Petten, where the irradiation takes place. For the irradiation, a sample holder, which can be placed in a standard irradiation rig, has been designed and fabricated. Nuclear analysis of the experiment has been performed. A design and safety report has been written and was approved by the safety authorities.

Progress and results

C.1 Design and safety report

Nuclear analysis of the experiment has been performed at ECN using the code WIMS-97. The calculations were concentrated on nuclear constants, reactivity effects, relative and absolute fluence rates, fission power, burnup, gas production and activation of materials. The main conclusions are:

- the maximum fission power is about 280 W cm^{-3},
- the maximum reactivity effect is 267 pcm,
- no strong radial variations in power density and burnup are expected,
- the actinide contents of the sample will be reduced to about 60% of the original content after 400 full power days, wheras americium will be transmuted almost completely.

On the basis of these results, a design and safety report, which is required for irradiation in the HFR, has been prepared by JRC-IAM [1]. This report was approved by the safety authorities.

C.2. Target fabrication/transport

The selected target composition is americium oxide (^{241}Am) embedded in a support matrix of spinel ($MgAl_2O_4$). The fabrication of the target and the assembly of the target capsule was performed at JRC-ITU. To this purpose a novel fabrication technique has been developed, called in INRAM-process (Infiltration of Radioactive Materials).

The principle of this method is the infiltration by capillary forces of a porous host material with a solution containing the infiltrant, i.e. ^{241}Am. For the present study, the host material was a green pellet of spinel (diameter 6.55 mm, height 8.5-9.2 mm, 49% density) which was immersed in an americium nitrate solution (ca. 400 g/l). The green pellets were prepared from commercial product (Baikalox S33CR, Baikowski

Chemie) and were calcinated at 650 °C for 4 hours. After the impregnation, the pellets were thermally treated in a H_2/Ar mixture, for 4 hours at 700 °C and for 6 hours at 1600 °C.
The resulting pellets had the following characteristics:

Density	: 96–97 %
Dimensions	: ϕ = 5.38 mm, height = 7.06 ± 0.1 mm
Am content	: 11.1 ± 0.7 %
Homogeneity	: good

The stack of pellets was placed in tube of 15/15Ti stainless steel, and were enclosed on either side by a spinel pellet of 5 mm length, a HfO_2 pellet of 6 mm length and a stainless steel pellet of 17 mm length. The purpose of these pellets is to obtain a homogeneous flux profile and to separate the target material from the structural materials. The pellets are held in position by means of a spring in the plenum volume. The spring and the end plugs were also made of 15/15Ti stainless steel. The capsule has been filled with pure helium at atmospheric pressure during welding.

After the fabrication was completed, the sample was transported from Karlsruhe to Petten, as arranged by FZK.

C.3 Irradiation facility preparation

The irradiation facility was prepared jointly by JRC–IAM and ECN. The facility, an existing standard TRIO–131 irradiation rig, is suited for in–pile use in the High Flux Reactor at Petten. A sample holder was designed and fabricated that was placed in the rig. The sample holder consists of three sections:

- The lower section consists of a stainless steel containment containing the target capsule and the instrumentation (thermocouples, dosimeters and gamma scan wires). The target capsule is positioned in a central hole of an aluminium drum and the instrumentation is arranged around it. The drum with the target capsule is supported below by one and above by two aluminium drums. The stack of four drum (600 mm long) is positioned inside the containment.
- The middle section is formed by the shielding plug and a filter in the downstream tube of the first containment.
- The upper section consists of the penetration plug of the TRIO channel.

Defined gas gaps are designed between the containment wall and between the drums and the first containment wall for the purpose of temperature control, either by means of helium or by means of helium/neon gas mixture technique.

C.4 Irradiation in the HFR

The irradiation facility was placed in core position C5 of the HFR at Petten. The irradiation started in HFR cycle 96.08 and during 1996 four irradiation cycles were completed. The cumulative irradiation time is 101.55 full power days.

References

[1] CONRAD R, Joint EFTTRA Experiment on Am Transmutation at the HFR Petten. Design and Safety Report. Tech. Mem. HFR/96/4327, JRC–IAM, Petten, 1996.

A.2.3-2 Neutron driven nuclear transmutation by adiabatic resonance crossing (TARC)

Contract No: FI4I-CT96-0009 **Duration:** 1 Sep. 1996 - 31 Aug. 1998
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A. OBJECTIVES AND SCOPE

The main objective of this project is to develop both theoretically and experimentally a new method, the adiabatic resonance crossing (ARC), which enables to enhance strongly the capture rate of neutrons by the radionuclides to be incinerated. The peak cross sections for neutron energies corresponding to the resonance region are much larger than for the other neutron energies. For instance, the cross section of ^{99}Tc is 4000 barn at the peak of a resonance at 5eV, but it is only 20 barn at thermal energy. Access to the resonance region can be achieved by using a transparent medium like Lead where the neutrons lose their energy in very small decrements. An experimental test will be carried out on ^{99}Tc with a neutron spallation source driven by the CERN proton synchrotron.

B. WORK PROGRAMME

B.1 Lead assembly and beam line (CERN, Univ. Autonóma Madrid, Univ. Athens, Univ. Thessaloniki)

B.1.1 Setting-up of the Lead assembly

B.1.2 Preparation and instrumentation of the beam line

B.2 Experiments on ARC

B.2.1 Timing experiments (CeF_3 counters) (CNRS)

B.2.2 Activation experiments (delayed g counting) (all partners)

B.3 Study of advanced neutronics in Lead

B.3.1 Electronic experiments (He^3 ionization and scintillation counters, Li F counters, fission counters) (CERN, Univ. Autónoma Madrid, CNRS,Univ. Athens, Univ. Thessaloniki)

B.3.2 Activation measurements (Ge g counters, track etched detectors, thermoluminescence counters) (CERN, Univ. Autónoma Madrid, CNRS, Univ. Athens, Univ. Thessaloniki)

B.3.3 Temperature measurements (thermometers) (Univ. Autónoma Madrid)

B.4 Developing appropriate formalism and computational tools for ARC (CERN, Univ. Athens, Univ. Thessaloniki)

B.5 Conceptual design of an incinerating device based on ARC (CERN)

B.6 Other applications of ARC (CERN,CNRS, Univ. Athens, Univ. Thessaloniki, Sincrotrone Trieste)

C. Progress of work and results obtained

Summary of main issues

The project is proceeding on schedule. The different components of the equipment are in place and the first tests shows them to work according to expectations.

The experiment calls for high purity Lead devoid of such know impurities as Silver, Antimony and Cadmium. The 4N quality Lead offered by Britannia Refined Metals (UK) proved to be satisfactory after thorough chemical analyses were performed on samples sent to several independent laboratories world-wide. The Lead ingots had to be transformed into carefully machined blocks in order to construct the assembly with the required geometry. The machining process, which should avoid introducing impurities but also keep the price acceptable, was based on moulding followed by a machining pass to take away the surface crust. This was done by Calder Industrial Materials. The blocks (613 kg each) , have been assembled in the experimental hall at CERN. Since Lead is very soft, a special suction device has been built to handle the blocks without deformation and position them precisely.

The proton beam line has been set-up by the PS staff and is capable as required to supply beam in two modes:

-"slow extraction" with about 10^4 particles extracted over 350 ms every 14. 4 seconds. Secondary particles (pions, protons etc..) of a given momentum can be distinguished by Time -of Flight.

- "fast extraction" with about 10^9 protons extracted in bunches of 30ns recurring every 14.4 seconds. Of special concern is the accurate measurement of the intensity of the beam. A new type of beam transformer, developed for our purpose by industry has been shown to be adequate by an absolute calibration by bombarding Aluminium foils with protons and counting the ^{24}Na produced.

A CeF_3 scintillator has been mounted, using a special quartz photomultiplier and working in the current integrating mode. The purpose is to record the prompt gammas associated with the neutron capture on selected elements. The first tests of the device readily show a time spectrum with a series of peaks which, through the time- energy relationship are at the position of the ^{99}Tc neutron capture resonances.

A pneumatic fast transport has been developed which allows the irradiation of ^{99}Tc and the subsequent counting of gammas in the daughter ^{100}Tc nucleus. A systematic mapping of the capture quantitative yields according to the position in the Lead assembly has been undertaken.

To measure the neutron spectra over a wide energy range (including timing properties after a proton impact) a number of different devices have to be used. These include specially developed ^{3}He ionization and scintillation counters, Si diodes (with ^{6}Li and ^{233}U targets), as well as more conventional flux measurements methods such as gamma activation of foils, track-etched detectors and thermo-luminescence. Finally an original high sensitivity (liquid Helium temperature) thermometric device to measure the residual heat deposited by neutrons in Lead has been built and tested. All these methods have now been used and the first results show that they work according to expectations.

The planning of these experiments required the development of a Monte Carlo simulation (combining production of neutrons, transport and chemical evolution) which is also the cornerstone of the development of the Energy Amplifier. TARC is in fact the first direct validation of this new code. To reach the required statistical accuracy, the code had to be optimized and in addition is now running on a parallel (Convex SPP 1200) computer (6 processors simultaneously)

The combination of these measurements and of the simulation point to the possibility of incinerating e.g. ^{99}Tc in some parts of the Energy Amplifier at a high rate and without any loss in power production.

AREA B

<u>REACTOR SAFETY</u>

B. REACTOR SAFETY

INTRODUCTION

The main scope of research in the reactor safety is the understanding of severe accident phenomena and the measures to prevent possible radioactivity release under severe accident conditions. Also considered are measures to mitigate the consequences of a severe accident.

The research should develop a consensus on how to treat phenomena in accident analyses, define mitigation and accident management measures, provide experimental results for code and their validation.

The main research tasks of this area are:

B.1 In-vessel Core Degradation and Coolability

Better understanding of core degradation phenomena, of interaction with the coolant of the behaviour of melt in contact with the primary circuit and its cooling potential.

B.1.1 Corium formation and behaviour: investigation of various phenomena with a special view to accident management measures.
B.1.2 Molten corium coolant interactions: understanding of basic phenomena, experimental and theoretical investigations on accident scenarios, taking into account scaling effects, extrapolation from simulant to real material.
B.1.3 In-Vessel Corium coolability: heat transfer mechanisms from either the debris bed or the pool to its environment (atmosphere, RPV wall) by radiation and natural convection, cooling potential of the RPV from the outside.
B.1.4 RPV behaviour: thermalhydraulics of the melt, behaviour under thermal and mechanical load.

B.2 Ex-Vessel Corium Behaviour and Coolability

Better understanding of basic phenomena of melt release from the RPV, corium spreading, interactions with coolant and structures to investigate generic aspects of corium cooling.

B.2.1 Thermochemical modelling and data: improved modelling of thermochemistry of interactions with structures, fission product retention, improvement of database of various chemical data.
B.2.2 Corium release and spreading: investigation of the effect of different RPV failure and corium release modes, potential for direct containment heating.
B.2.3 Corium retention and cooling: experimental and theoretical investigations to describe corium behaviour on the core retention device, investigation of corium interactions with coolant and structures. Generic studies on retention devices (core catchers).

B.3 Source Term

Release and behaviour of the fission product from the fuel into the containment. Integral tests (Phébus) to validate source term computer codes. New model and code developments for specific phenomena.

B.3.1 In-vessel fission product behaviour: separate effects tests for modelling the fission products release from the fuel, their transport, resuspension, revaporisation, taking into account effects from circuit chemistry are complement to the Phébus experiments.
B.3.2 Ex-vessel fission product behaviour: modelling of fission product transport including resuspension and revaporisation, influence of sprays and other mitigation measures, multicomponent aerosol behaviour, iodine chemistry based on separate and integral tests (in particular Phébus).
B.3.3 Benchmark calculations: validation of new code against experimental results.

B.4 Containment Performance and Energetic Containment Threats

Better understanding of phenomena having the potential to threaten the containment integrity. Assessment of new (passive and inherent) conceptual features with regard to their feasibility. Prevention and assessment of containment leakages.

B.4.1 Hydrogen distribution and combustion: experimental and theoretical investigations on hydrogen combustion modelling and evaluation of various mitigation measures.
B.4.2 Containment thermalhydraulics and cooling: experimental and theoretical investigations on thermalhydraulics codes, with regard to hydrogen distribution and natural convection for decay heat removal. Generic studies on passive decay heat extraction.
B.4.3 Material data and structural response: provision of dynamic concrete behaviour data (strain rate effect) at high impact velocity (shock waves, missiles). Structural response with respect to the identification of load conditions and material data.
B.4.4 Containment Leakage: Leakage flows through cracks and penetrations and prevention measures.

B.5 Supporting Activities

Activities are mainly intended to exchange information, to coordinate projects executed in the member states and at the JRC and to develop a common approach.

B.5.1 Accident management measures
B.5.2 Ageing: because of the increasing life time of nuclear power plants ageing phenomena are getting more importance, e.g. irradiation embrittlement, changes in mechanical properties.

The following "CLUSTERS" have been set up to assure the best coordination in each sub-area:

- B.1- In-vessel core degradation and coolability - INV;
- B.2- Ex-vessel corium behaviour and coolability - EXV;
- B.3- Radiological source term - ST;
- B.4- Containment performance and energetic containment threats - CONT;
- B.5- Accident management measures - AMM;
- B.5- Ageing of structural components - AGE.

B.1.1-1 Experimental and computational modelling of corium formation and behaviour during a severe accident in a light water reactor - investigation of core degradation - COBE

Contract No.: FI4S-CT95-0013 **Duration :** 1 Feb. 1996 - 31 Jan. 1999
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A. OBJECTIVES AND SCOPE

The main objectives are :

1) to develop the capability of calculating the behaviour within a reactor vessel during a severe accident so that the efficacy of accident management strategies can be assessed.
2) to understand how accurate the results of such calculations are.

The capability is achieved by developing the existing European codes ICARE and KESS to the point that they are capable of calculating such events. The accuracy is demonstrated by checking the results of calculations of these codes against past experiments, other codes and experiments to be carried out during the lifetime of this project. Models to be developed include quenching models and models for material relocation; both to a debris bed/molten pool and afterwards towards the lower head.

The codes will be checked against TMI-2, Phebus-FP, the Sandia MP and XR tests and also against quenching tests to be performed on a new "Quench" facility consisting of an electrically heated bundle of rods. These tests will investigate the quenching of a bundle of simulated rods.

B. WORK PROGRAMME

The project is divided into four main work packages :

B.1. Execution of experiments that can be used for the calibration and validation of codes used for reactor sequences (FZK).

B.2. Scaling studies to make sure that the experiments carried out in this project represent as well as possible phenomena expected to be encountered in reactors (AEA, Univ. Ruhr, Univ. Pisa).

B.3. Development of codes so that they can better represent reactor sequences (ENEA, Univ. Ruhr, JRC, Univ. Provence, Univ. Politécnica Madrid, Univ. IKE, Univ. Dresden, CEA).

B.4. Validation that codes are able to calculate experiments. Some of the experiments used are performed as part of this project. Others are the heritage of past programmes (ENEA, JRC, Univ. IKE, Univ. Politécnica Madrid, AEA, FZK, Univ. Ruhr, Univ. Dresden, CEA).

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of Main Issues

The main experimental work will be the bundle tests to be performed in the Quench facility. Although the design and construction of the facility has taken longer than planned, FZK still feel that they will be able to perform the two tests stipulated in the contract and write a final report before the end of 1999. The latest timetable for this work is:

Design and construction of facility	April 1996 - March 1997
Scoping test	June 1997 - October 1997
First bundle experiment	October 1997 - March 1998
Second bundle experiment	January 1998 - July 1998
Final Report	December 1998

While waiting for the results of the bundle tests the single rod tests are providing valuable insights into the cracking of cladding and the consequent enhancement of oxidation rates during the quenching process.

The pre-test calculational work, the code development and code validation are proceeding. There have been three modifications with respect to the original plan:

1) FZK are assisting with the scaling calculations for the Quench facility. Their effort is not paid by the project but it will be summarised here because it is an integral part of the overall effort.
2) IKE will perform pre-test calculations instead of post-test calculations for the Quench experiments. This decision was taken because it was felt that an independent confirmation of the SCDAP/RELAP5 scaling calculations was required in order to increase the confidence in the boundary conditions chosen for the Quench tests.
3) UP are developing models for the movement of material to the lower head instead of models for debris bed/ molten pool transformation. Debris bed/molten pool models already exist and are already being calibrated as part of this project. The state of knowledge on relocation is much more scanty.

A report on the validation of the code ICARE-2 on the first Phebus test, FPT-0, has been submitted as a deliverable to the project.

Progress and Results

1. EXPERIMENTAL WORK (FZK)

The main experimental results obtained so far have been from a set of single rod experiments. These were conducted in the following way:

1) pre-oxidation in air to a pre-defined thickness of zirconium dioxide (0, 100 or 300μm).
2) heat-up to 1200, 1400 or 1600°C in an inert atmosphere.
3) quenching either by:

i) water at $90°C$ and 1.5cm/sec flooding rate
ii) a rapid steam flow at 140°C and 2 grams per second.

The heat losses is these experiments were such that no temperature escalation was seen. The main effects of quenching were:

- A release of hydrogen. Figure 1 shows the mass released for the water-quenched cases.
- A storage of hydrogen in the zirconium. Figure 2 shows the mass stored for each of the water-quenched cases.

These results are, at first sight, surprising. Most models assume that the oxide layer acts as a diffusional barrier so we would have expected an exponential increase of hydrogen release with increasing temperature of the specimens and a decrease of hydrogen release with increasing oxide layer thickness. Similarly we would have expected that the tests without pre-oxidation would have absorbed most hydrogen. That this did not happen is due to the cracking of the cladding that exposed bare metal.

For the 300μm pre-oxidized specimens large cracks, penetrating both the oxide layer and metal substrate, were observed in both the water-quenched and steam-cooled specimens. Metallographic examinations indicated that:

1) The formation of the cracks is more pronounced for the experiments where the quench is initiated from low temperatures of 1200°C than those where it was initiated at 1600°C.
2) Most of the metallic Zircaloy is converted into the brittle oxygen-stabilised α-Zr(O).
3) The surfaces of the cracks are partially oxidized.

During cooldown zirconia changes its crystal structure from tetragonal to monoclinic at about 1200°C. This phase transition is accompanied by a volume increase that generates mechanical stresses in the oxide. The crack formation at specimen temperature of about 1200°C was also directly observed during an experiment with reduced inductive heating and slow cooldown. We therefore believe that it is is this phase transition that leads to the cracking of the cladding and the enhanced hydrogen storage and release.

These experiments are leading to an increased understanding of phenomena governing quenching that can be used for model validation and calibration.

2. SCALING STUDIES

2.1 Bundle Quench Tests (AEA, FZK, RUB)

The main objective of this task is to make sure that the tests to be performed in the Quench bundle facility are representative of reactor accidents. The first year's work has concentrated on making sure that the tests are feasible.

Early decisions were taken firstly build the shroud liner from Zircaloy so that the heat liberated during oxidation could then compensate for heat lossses and, secondly, base the bundle layout on Phebus-FP.

The calculational work was divided. FZK examined the test section while AEA and RUB examined the upper plenum above the test section and the off-gas line which leads from it.

Like the single rod tests the bundle tests will have a pre-oxidising phase and it is on this part of the transient that FZK have concentrated their effort. Their calculations with SCDAP/RELAP5/mod3.1 clearly indicated that the outside of the shroud needed forced convection cooling if it was not to overheat. Counter-current argon flow provided the flattest temperature profile in the test section so this was adopted in the final design. An additional water loop will be used to cool the sensitive top part of the bundle.

AEA. assisted by RUB, have used the results from the FZK alculations as boundary conditions to assist in optimising the design of the upper plenum and offgas line. The principal design concerns were that:

(a) the steel and Inconel temperatures should not exceed the 600°C design limit governed by material strength and oxidation considerations.
(b) there should be no likelihood of condensation of steam on cold structure surfaces in the upper plenum. This could lead to rainback of water into the test section, giving possible premature and unprototypical quench from the top of the bundle. This imposed a minimum temperature of 150°C.

The calculational strategy followed four interlinked routes.

- SCDAP/RELAP5/MOD3.1 to provide a 1-dimensional model of the thermal response of the heat structures in the upper plenum and offgas line along with the thermal hydraulic response of the fluid (non-condensable gas and/or single or mixed phase water/steam),
- the finite-element code TAU to provide more detailed 2-dimensional heat conduction and thermal radiation modelling of the structural elements in the upper plenum.
- the computational fluid dynamics code CFX4 to give detailed flow modelling of the upper plenum and offgas line.
- Engineering calculations (performed at RUB-NES) to analyse independently the thermal loads on the offgas pipe structures during FZK quench test operation. These calculations aim to identify unintentional occurrences of hot spots threatening the pipe structure integrity as well as steam condensation processes leading to imprecise offgas measurements.

The calculations showed that the third and final design satisfactorily solved the earlier problems.

2.2 Air Ingress (USP)

A number of air-ingress tests will be performed as part of a sister project (OPSA) in the in-vessel behaviour cluster. As a support to that project, and also in preparation for future air-ingress tests in the Quench facility, USP have started an analysis to determine how such accidents could occur in reactor scenarios.

3. CODE DEVELOPMENT

The work programme on code development is proceeding according to schedule.

3.1 Quenching (CEA-IPSN, IKE, UP)

Both the ICARE-2 and KESS teams are involved in developing quenching models.

The ICARE2 team has started a cooperation with USP who have analysed the SCDAP/ RELAP5 quench model and proposed a number of corrections and improvements. It is planned to implement this model in ICARE2 for calculations of existing or future quench tests.

IKE's main task is to improve and to extend the KESS models considering the essential physical processes during quenching. This model development is based on the analysis of the FZK quench experiments. Four of the single rod tests have been analysed. The model development has concentrated on the quench and boil-off phase until rewetting of the specimen. The first comparison of the calculated temperatures with the experimental data shows that the cooldown rate of the rod specimen under film boiling conditions is underpredicted during quenching. This means that the heat transfer coefficient between the rod cladding and the fluid - calculated with the correlation derived by Bromley [1] - is not adequate for non-stationary flow conditions.

To solve this problem a new heat transfer correlation, depending on the assumed axial profile of the vapor film thickness and the resulting vapor velocity, has been applied. Using the modified thermohydraulic module a good agreement between the calculated and measured temperatures.

3.2 Transition to Debris Bed/Molten Pool Formation and Behaviour (CEA, ENEA, TUD)

Significant efforts have been made to improve ICARE's late phase modelling degradation:

- The effective conductivity of debris beds has been improved by:
  - considering the liquid fraction in porosities. This increases the equivalent conductivity.
  - modelling the dispersion effect of the cooling fluid which flows through a debris bed.
  - improving the modelling of heat exchanges by radiative transfer.
- improving steam properties at high temperature: The heat capacity of steam has been modified to take into account the dissociation of H_2O at high temperatures.
- improvement of molten pool modelling by:
  - taking into account natural convection caused by internal heat generation
  - improving the boundary layer model for molten pools.
  - enhancing the radiative exchange model in a cavity
  - modelling the heat transfer at the liquid-solid interface of the molten pool.

Many of these improvements were calibrated on the Sandia National Laboratories DC1 experiment (see section 4.4) and tested on Phebus FPT-0 and the Sandia MP tests.

Work on KESS concentrated on the control rod modelling. An analysis of the Phebus FPT-0 experiment indicated that the control rods may have contributed to an early formation of a debris bed/molten pool. Many post calculations of FPT0 test with KESSIII-Mod2 and single rod experiments in the DRESSMAN facility have been carried out in order to understand the process better.

3.3 Movement of Material to the Lower Head (UP, UPM, RUB)

TMI-2 investigations indicate the relocation of molten ceramic material from a molten pool in the core central region into the lower head via two main pathways

- a jet type flow of melt through the core bundle region
- a cascade type flow of melt through the peripheral core support assemblies

Bandini from ENEA has coordinated and the writing of a status report on the movement of material to the lower head on behalf of the Committee for the Safety of Nuclear

Installations (CSNI). Other members of the COBE project have participated. A first draft report has been issued and it is now under review by the Degraded Core Cooling (DCC) Task Group. As part of this work a review has been made of available code models. Both mechanistic (SCDAP/RELAP, ICARE/CATHARE, ATHLET-CD/KESS) and integrated (MELCOR, ESCADRE) system codes have been taken into consideration.

UP have used a finite element code, MARCUS, to simulate an immiscible and viscous corium flowing down in a simplified bypass-like geometry initially filled with water. A pseudo-concentration based technique is employed to track the moving corium-water interface. Only the isothermal case has been considered in the present computation.

Work is also underway on BWRs. UPM are modifying ICARE-2 to take into account BWR features such as control blades and will test this modified code on the XR2-1 experiment.

3.4 Behaviour in the Lower Head (RUB)

Like the MECO code developed by RUB, for ex-vessel melt spreading scenarios, MECI is based on the Navier-Stokes-Equations for two-dimensional, transient, viscous flows of incompressible fluids. A version for bottom head applications that can treat free surface melt jet impingement phenomena is under development.

3.5 Modelling Irradiated Fuel (JRC)

There is no model in ICARE2, nor indeed any other LWR system code, that takes into account the burn-up of fuel. Yet in several tests where irradiated fuel can be compared to fresh fuel large differences in behaviour have been observed.

A simplified approach was used to determine the influence of intragranular bubbles on swelling. Solving the Van-der-Waals equation of state for Xe (Olander, [2]) by assuming constant temperature and gas content, it was found that a bubble enlargement over two decades in the radius causes a swelling between 10% to 100%.

Some stand-alone codes can treat swelling mechanistically. Two of them, LAKU (Vaeth [3]) and VICTORIA (Heames et al. [4]), were chosen for further examination. When run in the "steady state" mode both codes suggested a 16% swelling in the cold rod regions (rod outer temperature about 1900K) and 12.5% in the hot rod regions (rod outer temperature about 2400K) for Phebus FPT-1. The comparison between transient and steady state calculations of LAKU are similar concerning the swelling but not for fission product release because both parts of the code have different porosity models.

4. CODE VALIDATION

The experiments used for the code validation part of this project are summarised in the table below. Nearly all the partners are involved.

Test	JRC	CEA-IPSN	ENEA	IKE	AEA	RUB	UPM	FZK	TUD
Quench		I	I/S	K	I&S	I/K		I&S	
MP1,2			I&S						
Phebus	I		I&S						K
XR2-1							I/M		
TMI2		I	I&S	K					
ICARE, K=KESS, M=MELCOR, S=SCDAP/RELAP5									

Analysis is concentrating on experiments that simulate the late phases of core degradation.

4.1 Quench Tests (IKE)

While waiting for results of the full bundle tests in the Quench facility the single rod tests have been studied. Calculations with KESS that do not take into account the cracking of the oxide layer underestimate hydrogen production.

4.2 Phebus-FP (JRC, ENEA, TUD)

Hydrogen production is a problem for Phebus too. A number of calculations of the test FPT-0 have been made and hydrogen generation was underpredicted by nearly all codes. It is thought that much of the missing hydrogen was generated by Zircaloy that was molten, liquified or relocated. The codes (except ICARE-2) assume that only intact solid Zircaloy can oxidise. JRC and ENEA have performed post-test calculations of FPT-0 and ENEA have started work on FPT-1 [5].

KESS calculations at TUD are concentrating on the impact of control rod behaviour on the melt progression. Results from Phebus indicate that the codes' assumption of no interaction between control rod material and cladding is false.

4.3 Three Mile Island (CEA-IPSN)

The Three Mile Island accident provides a performance indicator for the whole project. It includes quenching, molten pool formation and movement of material to the lower head. The accident can be divided in five phases: firstly a loss of coolant accident, secondly core heat-up and degradation, thirdly a partial reflooding with continued degradation, fourthly a corium relocation to the lower plenum, and finally a debris cooldown. Present analysis has concentrated on the molten pool formation during phase 2. CEA-IPSN are using ICARE2 V3 with boundary conditions supplied by the thermalhydraulic code CATHARE.

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Figure 1

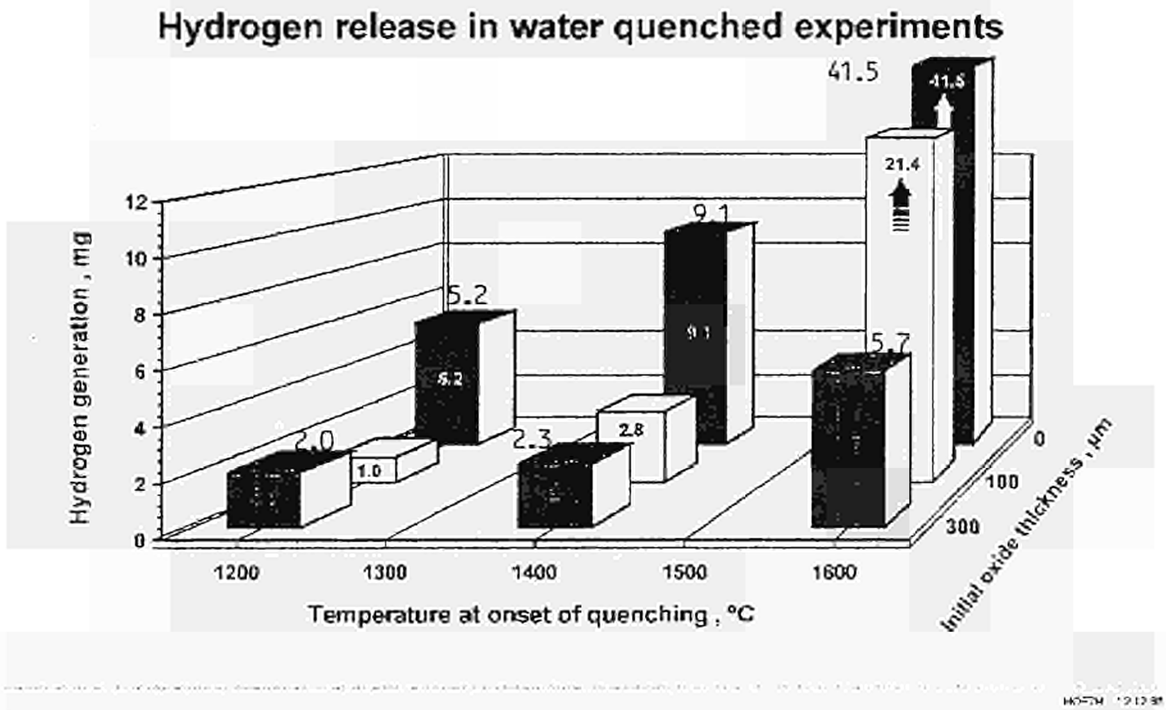
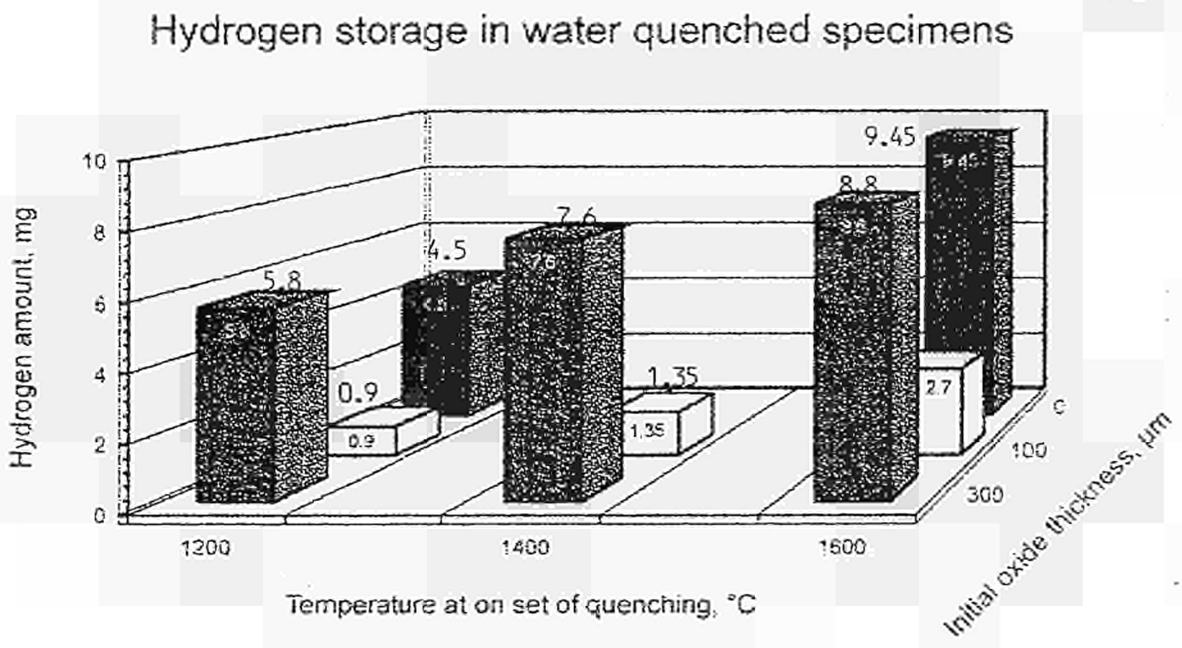


Figure 2



B.1.3-1 Core melt-pressure vessel interactions during a light water reactor severe accident - MVI

Contract No: FI4S-CT95-0007	**Duration :** 1 Jan. 1996 - 31 Jan. 1999
Coordinator :	**Prof. B.R. Sehgal, Univ. KTH (RIT) Stockholm/SE** Tel.:+46-8-7906541 - Fax.:+46-8-7907678 E-mail: sehgal@ne.kth.se
Partners :	**IVO Vantaa/FI, CEA-DRN Grenoble/FR, AEA Winfrith/GB, VTT Espoo/FI, ENEL Milano/IT, Univ. La Sapienza Roma/IT, ECN Petten/NL, Siemens-KWU München/DE**

A. OBJECTIVES AND SCOPE

The objectives of the MVI Project are to obtain data, and develop validated models, for the resolution of the various safety issues related to melt-vessel lower head interaction during a severe accident. The main objective is to assess whether melt can be cooled and retained within the vessel lower head. Data should be obtained for :

- the vessel wall ablation process due to the impingement of a melt jet,
- the melt coolability process in the presence of water,
- the in-vessel melt retention process in the presence of crust,
- the external cooling of the reactor vessel,
- the critical heat flux during external cooling,
- the lower head melting or creep-rupture process, and
- the vessel-hole ablation process.

B. WORK PROGRAMME

The project is divided into four main work packages :

B.1 RIT Experiments

The experimental programme at RIT is focused to study the interactions of simulant materials with the pressure vessel, coupled with scaling analyses towards applicability of the data obtained to prototypical accident conditions. It includes experiments on melt jet impingement, in-vessel melt coolability, lower head failure and vessel-hole ablation at the facility MIRA (RIT).

B.2 IVO Experiments

Several in-vessel melt retention experiments are performed at the COPO facility in order to evaluate the margins to melt-through of the reactor vessel (IVO).

B.3 CEA Experiments

These experiments are oriented to obtain data on the thermal hydraulic behaviour of a naturally convecting corium pool in the lower head of the vessel at the BALI facility, and to determine the critical heat flux for the configuration of a flooded reactor vessel or a core catcher under natural and forced circulation at the SULTAN facility (CEA).

B.4 Analysis Development Programme

The previous experimental programmes are supported with both simplified and 3-D fluid dynamic codes for providing pre-test and post-test analyses, developing understanding of the obtained data and validating models for phenomenological processes and prototypical situations (AEA, VTT, RIT, La Sapienza, ENEL, ECN, Siemens, IVO).

C. PROGRESS OF THE WORK AND RESULT OBTAINED

C.1. RIT Experiments

The work at RIT during the first year of the MVI Project consisted of the following topics:

C.1.1 Hole Ablation Experiments and modeling

Various melt and vessel wall simulants were employed in the test program, in order to, better, examine the influence of material properties upon the ablation phenomena. It was realized that the melt viscosity plays an important role in hole ablation. That is because the crust layer of melt which can form upon the ablating hole will undoubtedly exhibit a significant surface roughness. In lower viscosity melts the thickness of the boundary layer is small enough that the surface roughness governs the heat transfer, thereby minimizing any entrance effects, and producing a much more uniform (one-dimensional) ablation. In the experimental program, it was found that for the higher viscosity melts (binary oxide and paraffin oil) the final hole shape was very much two-dimensional, with a significantly wider "mouth", on upper surface, than the exit. Conversely, with the water and salt melt simulants the ablation profile may be characterized as one-dimensional. In all of the experiments, it was clear that the crust boundary condition prevails all the time, i.e., the temperature difference driving the ablation is the melt temperature and the melt liquidus temperature (i.e., $\Delta T = T_{melt} - T_{liq}$). Thus, even though the crust may be swept out by the melt flow, it re-establishes itself i.e., the time required for crust formation appears to be much less than the rate of crust sweep-out. A model named HAMISA (hole ablation modeling in severe accidents) has been developed and validated against data obtained at RIT and other laboratories.

C.1.2 Jet Impingement Experiments and Analysis

Experiments were conducted for jet impingement heat transfer with phase change. The motivation for this research is to gain a better understanding of the heat transfer phenomena associated with a, potential, relocating melt jet impinging upon a RPV lower head during a severe accident. Knowledge of such heat transfer rates will reduce the uncertainty of predictions regarding vessel failure, caused by direct contact of a corium jet with the vessel wall.

In the current experiments a variety of melt jet and solid plate simulants were employed. Temperature instrumentation in the stagnation zone of the impinging jet allowed for subsequent determination of the ablation heat transfer. The data were then compared to the available laminar and turbulent models for jet impingement heat transfer. This showed that the data is predicted quite well by the Saito correlation, previously obtained for jet impingement with phase change.

For conservative assessments in prototypical situations, where the jet diameter may be assumed to be relatively small and the pour times long, the majority of heat transfer will fall in the turbulent heat transfer regime, and, thus, the use of Saito's correlation is appropriate.

C.2. IVO Experiments on the COPO Facility

The COPO facility was modified to provide constant temperature boundary conditions and crust formation on all boundaries of the slice configuration. The first test carried out within the MVI project was called test L7. The test L7 was intended to be a base case test with the elliptical bottom version of COPO II facility to be compared with the results from BALI and from the COPO version with a hemispherical bottom. However, the run L7 partially failed due to gas leakage into the test section and consequential formation of a gas bubble under the upper cooler.

The test L8, which was basically a rerun of test L7, was carried out in September 1996, after modifications of the support system of the facility to prevent similar type of gas leakages as in L7. The modified Rayleigh number was $9.8 \cdot 10^{14}$. The test was run with a homogenous pool with all boundaries cooled.

The results showed relatively high heat transfer coefficients at all boundaries. The measured values are higher than predicted by the well-known correlation of Steinberner and Reineke and also higher than those measured in the ACOPO experiments at University of California, Santa Barbara. Also, at the side boundary, and at the lower boundary, the heat transfer coefficients were higher than expected and exceeded the values measured in COPO I. The reason for the discrepancy is under investigation.

The test L9 was carried out as the first test with a stratified pool configuration. The lower part of the pool consisted of water-zinc sulfate solution which was electrically heated as in the earlier experiments. A nonheated layer of distilled water was located on top of the heated layer. The two layers were separated by a thin, thermally conducting plate (aluminium with aluminium oxide coating).

According to the results, the ratio of heat fluxes to upward vs. sideward directions from the nonheated layer matched the ratio predicted by the well-known correlations of Globe & Dropkin and Churchill & Chu, correspondingly. However, the overall heat transfer from the lower, heated layer to the upper layer was disturbed by an apparent leakage at the intermediate plate, and by the consequential mixing of the two layers.

C.3 CEA Experiments

The CEA experiments are being performed on two facilities: BALI, in which data is obtained on the thermal loading imposed, on the vessel wall by a naturally-convecting corium pool in the lower head; and SULTAN, in which data is obtained on the critical heat flux (CHF) for external cooling of the lower head. The BALI uses a slice vessel of 100°C angle, at full scale, and represents corium by salt water, which is heated electrically. The Ra number reached is in the range 10^{16} to 10^{17}, i.e., the prototypic value. The SULTAN experiments employ 4 m x 0.15

m plates held at several angles to the horizontal. The plates are heated electrically and are cooled at bottom with water in forced and natural convection.

C.3.1 BALI Experiments

The first 6 months were devoted to the qualification of the facility. The first tests have pointed to a water leakage problem on the front and back windows: the initial solution (silicone seal) was not reliable and another one has been satisfactory tested. The first test campaign started in September. Up to now, seven tests have been run with 1.5 meter pool height, for different power densities and for one of them without top cooling.

The basic analyses show an under prediction of the upward heat flux when BALI results are compared with values extrapolated from the last COPO results.

The 1.5 meter height BALI tests have greater Ra number but have the same aspect ratio (H/R) as the 0.6 m height COPO tests. For these tests good agreement with Steinberner & Reineke correlation, was obtained.

At the end of the year, a 2 m height test was performed. During the test large deformation of the front and back windows were observed, caused by hydrostatic pressure and lateral ice crust formation. Waiting for appropriate test section modifications, the first test campaign has been momentarily stopped.

C.3.2 SULTAN Experiments

The first four campaigns have been completed:

1. Vertical position, channel gap = 3 cm
2. Inclined 10°/ horizontal, channel gap = 3 cm
3. Vertical position, channel gap = 15 cm
4. Inclined 45°/horizontal, channel gap = 15 cm.

The fifth campaign (inclined 45°, channel gap = 3 cm) is going on. It will be performed for a single outlet pressure of 0.1 Mpa.

The first three test reports and the document describing the test plan and a test campaign have been sent officially to participants in the MVI project. The test reports include:

- CHF conditions for all parameters
- pressure drop measurements, void fraction and temperature profile for P_{outlet} = 1 bar and T_{inlet} = 99°C.

All the available CHF results have been informally transmitted to the University of Rome to develop or qualify models of CHF.

C.4 Analysis Development Programme

This programme is being performed at RIT, VTT, AEA Technology. University of Rome, ENEL, Siemens and ECN. Each of these are described below.

C.4.1 RIT Programme: In-Vessel Melt Pool Formation and coolability

A model to describe the debris bed heat-up process, occurring in the lower head of an LWR vessel, during the course of a severe accident was developed. The model treats the case of a uniform composition, initially quenched, debris bed of zero porosity, which is slowly converted into a melt pool. The hemispherical lower head wall is included in the modeling, and its melt-through due to the thermal attack of the melt pool is calculated. The heat transport to the upper boundary is through the upward movement of plumes (or layers), whose average velocity is calculated to deliver the requisite heat flux at the upper boundary. The heat transport to the hemispherical boundary is through conduction, and, then, through a boundary layer created by the downward flow along the curved wall from the upper part of the pool. Thus, the temperatures within the pool are calculated. A melt pool is created after the liquidus temperature is exceeded. The vessel melting and melt-through is calculated by following the temperatures in the vessel wall.

This model named MVITA (melt-vessel interaction thermal analysis) was applied to a BWR lower head melt pool formation, and vessel melt-through, scenario as an illustration. The calculation showed that due to the relatively low heat conductivity of the core debris, the effect of the cold boundaries does not extend far into the debris bed.

It was found that the thickness of the crust (debris) around the pool does not affect the split of the heat generation into fractions going to the top and sideward boundaries. The crust simply acts as a boundary condition at the liquidus temperature to the melt pool.

The MVITA model was extended to a more complex debris configuration, which includes an overlying metallic layer. Effective conductivity, based on Eckert-type boundary layer correlations, and effective velocity, based on upward turbulent natural convection heat transport rates, represented the major heat transfer mechanisms in naturally- convected liquid pools.

The local heat flux distribution on the side wall of the metallic layer was found to depend on Ra_{VW}, i.e.,the input heat flux and the layer thickness. However, the results from the integrated analysis showed that the focusing effect of the metallic layer is significantly diminished in a reactor configuration. This is due to a thicker oxidic crust layer at the near-wall (debris corner) region, and hence, lower heat fluxes in this region, coupled with the two-dimensionality of the heat conduction process in the steel vessel.

C.4.2 VTT Programme: Failure Mechanisms of Vessel Wall and Penetrations

A model to calculate the creeping of pressure vessel and penetrations was added to the PASULA code. The formulae for calculating simultaneous linear-elastic, elastic-plastic, creeping and thermal expansion deformations were derived. The model is being tested and validated against data. BWR control rod penetrations are being analysed. A report was issued.

The effective thermal conductivity of a granular bed varies with particle size and temperature. A model was developed with particles represented as spheres touching or not touching each other. The effective conductivity was found to increase with temperature due to radiative heat exchange between the particles. A report of this work has been issued.

C.4.3 AEA Technology Programme

The AEA Technology work consist of model development, review and coordination of models developed by others in the MVI Project, and providing assistance in defining interfaces with models developed by other projects in the in-vessel, ex-vessel cluster. Close cooperation was maintained with RIT and later with VTT in order to integrate models together to produce an overall model for MVI.

A strategy for the MVI model development efforts was developed. A compilation of the corium properties, needed for the MVI development effort, was made. A review of corium relocation scenarios, needed as initial conditions for the MVI, was performed. All three of these efforts were documented, and distributed to MVI partners for comment. Additionally, three other reports were written based on joint work performed for the U.K Health and Safety Executive. These described:

- a hole ablation model for the LOWHED code,
- a jet impingement-ablation model for the LOWHED code, and
- review of the in-vessel retention by in- and ex-vessel flooding.

These reports have been made available to MVI partners.

C.4.4 University of Rome "La Sapienza" Programme

The work at University of Rome supports the experiments on SULTAN facility of CEA. A preliminary analysis of 129 experimental data points with forced convection and down-ward facing inclined surfaces was analysed. A correlation to predict CHF for these conditions was developed. Its predictions are quite reasonable: 89% within ± 20% and a r.m.s error of 12%. A paper based on this work has been submitted.

C.4.5 ENEL Programme

The ENEL work consist of supporting the MVI modeling work through its CORIUM-2D code. The radiation heat transfer model was improved. In addition work was started on other relevant topics, e.g., radiation within the particle bed, corium properties between liquidus and solidus, time required to reach steady state, corium mass relocation etc. A verification validation campaign for CORIUM-2D has been started. A review of the AEA Technology reports was performed.

C.4.6 Siemens Programme

The Siemens work is related to determining the influence (focusing effect) of the metal layer that may be present on the upper surface of a molten corium pool undergoing natural circulation in the lower head.

The whole system is represented in the 2-D ADINA code. It was found that many mesh points have to be included, thus, the calculations take a long time on the computer. A 6 minutes transient was completed. It is seen that crusts are being formed on the cold surfaces and the metal layer has started to heat up. The calculation has to be continued further to determine the existence or strength of the following effect.

C.4.7 ECN Programme

The work at ECN will support the BALI experiments at CEA. The work will start in 1997. No work was performed in 1996..

B.1.4-1 Behaviour of the reactor pressure vessel under mechanical and thermal loadings caused by core melt-down and steam explosion accidents - RPVSA

Contract No: FI4S-CT95-0002 **Duration :** 1 Jan. 1996 - 31 Dec. 1998
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A. OBJECTIVES AND SCOPE

The mechanical behaviour of the reactor pressure vessel strongly influences the environmental consequences of a core melt-down accident. If unfavorable large-scale failure of the vessel could be ruled out, this accident would lose much of its catastrophic character. Therefore, thorough investigations of the response at the most vulnerable parts of the reactor pressure vessel might contribute significantly to improve nuclear fission safety and to restore public acceptance of nuclear power.

B. WORK PROGRAMME

The project is divided into five main work packages :

B.1. <u>Lower head heated bymolten core material and loaded by quasi-static internal pressure</u>
The lower head representing the hottest part of the vessel may undergo large creep deformations. In order to overcome significant uncertainties in this field the creep experiments RUPTHER and KRAKATOA with cylindrical and spherical shells are carried out and benchmarks are defined for validation of computational models (CEA, ENEA, FZK).

B.2. <u>Lower head melt through failure caused by the attack of molten core material</u>
The lower head melt-through failure is being investigated by the experiment CORVIS where the contact of molten core material with the vessel wall is simulated under realistic conditions. The results are also used to validate computational models (PSI, FZK).

B.3. <u>Lower head region loaded by dynamic pressures during a postulated steam explosion</u>
The lower head region may also be loaded dynamically during an in-vessel steam explosion. Experimental and theoretical investigations are planned to investigate the formation of an upward moving molten fuel slug (FZK).

B.4. <u>Upper head region loaded by a molten fuel slug impact during a postulated steam explosion</u>
The upper vessel head may be hit by this slug. An upper head failure would severely endanger the containment integrity. Therefore, model experiments BERDA are carried out investigating the slug carrying capacity of the upper head. To avoid computational difficulties, the experiments BERDA fulfil essential similarity conditions allowing the direct transfer of the results to reactor dimensions. This activity will be supported by more simplified experiments FLIPPER and tests with the impact facility ORION as well as theoretical investigations (FZK, CEA).

B.5. <u>Material properties for the lower and upper vessel head problem</u>
The high temperature mechanical data base for vessel steels is being widen by carrying out creep tests with tensile specimens, dynamic tension tests and other additional tests (FZJ, JRC, FZK).

C. Progress of work and results obtained

Summary of main issues

Calculations of the natural convection in the corium pool formed by the lower head yielded useful results. On this basis the temperature distribution in the lower head can be determined. Uniaxial creep tension tests and the biaxial RUPTHER tests with pressurized tubes were performed in order to determine the parameters of advanced material models valid for the expected high temperature range. The tubes failed by local piercing. One RUPTHER test has been defined as a benchmark (task 1 and 5).

Several CORVIS experiments were done in order to investigate the contact of corium with the lower head. Realistic lower head thicknesses and temperatures were considered, but the vessel pressure was not simulated. Main results: A jet-like attack of overheated metallic melt is able to cause a fast lower head melt through failure. However, the contact of oxidic melt is less severe; oxide crusts occur which are able to protect the steel structure against melting (task 2).

First BERDA experiments allow assessment of the impact of molten core material which the upper head is able to withstand. If the geometry of a 1300 MW PWR is scaled down to 1:10 and if the temperature influence as well as the upper internal structures are neglected, the tolerable kinetic slug energy is about 0.5 MJ. In addition, similarity investigations were performed yielding scaling rules to transfer the BERDA results to reactor dimensions. Main problem: Are the high plastic deformations observed for the scaled down vessel head representative for the ductility of the real head? (task 4 and 5).

Progress and results

C.1 Lower head heated by molten core material and loaded by quasi-static internal pressure

The natural convection in the corium pool formed by the lower head was analyzed with the FE-codes TRIO and CASTEM 2000. Both homogeneous pool conditions and stratified conditions with an upper metallic layer and a lower oxide layer were considered. In addition, laminar and turbulent flow was assumed and the Rayleigh and Prandtl number were varied. In all cases very high temperature gradients occurred at the hemispherical boundary, the heat flux across this boundary reaching its maximum close to the upper pool surface. In the next step the resulting temperature distribution in the lower head will be determined. Moreover natural convection analyses will be done for the geometry of the CORVIS experiments (see task 2), and the results will be compared with the CORVIS observations in order to validate the applied method.

Biaxial RUPTHER tests were carried out to check the material model under biaxial loading conditions which are typical for the lower head loading. A tube of diameter about 100 mm is submitted to internal pressure and local annual heating using an induction coil reaching temperatures up to 1300 °C. Most of the measurements could be performed without direct contact to the hot specimen. As an example, Fig. 1 shows the axial temperature profile measured by an infrared camera. In particular, RUPTHER tests were done at 700 °C, 1000 °C and 1100 °C under rising pressure, and at 700 °C and 1000 °C under constant pressure up to failure. In all cases rupture occurred by local piercing after unstable bulging of the tubes.

In 1997 one RUPTHER test with a maximum temperature of 1000 °C will be done as a benchmark. Pre-calculations will be submitted by FZK, CEA, ENEA and PSI.

C.2 Lower head melt through failure caused by the attack of molten core material

These conditions are primarily related to core melt down accident conditions with reduced pressure levels (low pressure paths). Of particular interest are those reactors where the lower head carries penetrations of various designs. Melting tests are performed in a cylindrical steel container (test vessel), the bottom of which (test plate) represents a cut-out of an RPV lower head wall made of reactor steel. The test plate thickness and the diameter of the tube insertions agree with the corresponding reactor dimensions. The core melt substitute is an alumina/iron thermite, of which either the oxidic or the metallic part are used in the experiments. After pouring of the melt into the test section, the generation of the corium decay heat is simulated by a sustained heating with an electric arc submerged in the melt bath. The tests are performed at atmospheric pressure.

Three experiments were carried out in 1996. Fig. 2 shows typical temperature histories measured in the test plate. After the transient behavior during the first minutes, the temperature at the upper surface is almost constant for more than half an hour.

The most important findings are related to the plate failure:

- A jet-like release of overheated metallic melt from the core region certainly leads to a breach of the BWR drain line, when only a small portion of the core inventory has passed the tube. Failure of the RPV wall by erosion (jet impingement) is also possible.
- In contrast, accumulation of an oxide pool in the RPV lower head is propably no hazard to the drain line integrity, if the system pressure is low. However, the oxide melt can penetrate the drain line at least as far as there is no residual in the tube. If actually a tube breach occurred, a considerable amount of core melt can flow out.
- Oxide scales protect the steel structures against melting. It appears that a melt front in the steel cannot proceed as long as an overlying scale is not molten.
- Only a small amount of oxide melt can penetrate the narrow instrumentation tubes, especially if the tubes contain cable material or water. The decay heat is too small for a heating up of the tubes. Therefore, a tube breach by only melt penetration is unlikely.
- At low pressure, the RPV wall in the vicinity of a penetration cannot be destroyed within a short time if the oxidic melt is stagnant. The experiments showed that the test plate surface temperature does not exceed 1100°C (if not thermally insulated).

In 1997 the work will be concentrated on interpretation of the CORVIS results and on analysis.

C.3 Lower head region loaded by dynamic pressures during a postulated steam explosion

Some assessments were carried out and tests were prepared. Essential results are not yet available. (For this work EU funding is not available)

C.4 Upper head region loaded by a molten fuel slug impact during a postulated steam explosion

The impact of molten core material on the upper head is investigated by the model experiments BERDA simulating the reactor scenario 1:10 as closely as possible. The vessel head and its bolts are represented by models made of the original materials. They are precisely scaled down by 1:10. The molten core is simulated by liquid metal with about the same density but with a much lower melting temperature. It is contained in a crucible and accelerated against the model of the head using a pneumatic drive mechanism. Prior to the liquid slug impact the crucible is decelerated by a crash tube, thus the crucible does not participate in the impact process. To carry the dynamic loads, the whole facility is mounted on a heavy base plate of 38000 kg which is supported by springs.

In the first and second test lead spheres of 10 mm diameter and a total mass of 65 kg were filled in the crucible and accelerated against models of the head.

For comparison in the third test a steel projectile of 26 kg with a spherical surface was hurled against the head; radius of the spherical surface 160 mm; radius of the inner surface of the head 278 mm.

In the next four tests liquid metal having a mass of 80 kg was hurled against the head. During the fourth test the crucible failed and therefore, strong slug dispersion occurred reducing the impact force significantly. However, for the next tests compact liquid slugs occurred causing high load peaks and severe head deformations (Fig. 3). The strains reached values up to about 30 %, peak strains were even higher.

In the heads used for the tests also the holes for the control rod drive mechanicsms were simulated. Before the tests the holes were closed by plugs with threads similar as for German PWR. During the slug impact many plugs were expelled reaching velocities up to 70 % of the slug velocity.

Results of the different tests are compared in Fig. 4. Consider that the forces are linearly converted to a reference mass of 80 kg. (Thus for test 03 with a mass of 26 kg the force is increased by the factor 80/26 yielding a value much higher than the capacity of the head and the bolts.) The figure reveals that for slugs of lead spheres and liquid metal the impact forces are only about one quarter of the force from a solid projectile. From test 01 and 02 using lead spheres and test 05 and 06 using liquid metal it might be concluded that the relation between the maximum impact force and the impact velocity can be described approximately by a parabola. However, our computational model SimSIC recently developed for this problem suggests different and more complex relations for the lead spheres and the liquid metal. According to SimSIC the large plastic head deformations seem to be of considerable influence. This was confirmed by test 07 carried out just before the preparation of this paper.

In addition, simularity investigations were performed to obtain scaling rules for transfer the BERDA results to reactor dimensions. From the simplified impact experiments FLIPPER carried out on different scales it was concluded that for liquid structure impact problems the transfer to other scales follows approximately the known similarity rules. However, for certain materials the structural stiffness does not seem to follow known rules. Moreover it is not clear whether the high plastic deformations observed for the BERDA tests can also occur for real reactor components. Here further research is necessary.

C.5 Material properties for the lower and upper vessel head problem

For the investigations of both the lower and the upper head information about the mechanical material properties are required. Thus creep tensile tests were carried out at 700 °C, 730 °C and 750 °C. Based on the measured strain as a function of time the constants of the classical Norton creep equation could be determined. However, in this equation the strain rate is assumed to be constant for a given stress, while in the tests the strain rate varied. This will be overcome by the advanced material models developed under task 1.

Furthermore, dynamic tests at the Large Dynamic Test Facility (LDTF) in Ispra were prepared. Heating techniques to allow test temperature up to 800 °C were successfully tested. The procurement of the material for the specimens turned out to be time consuming. In order to allow the comparison of our test results with those to be obtained in the new EU project REVISA, the same heat of material should be used. Therefore, additional negotiations were required and a very large amount of material had to be ordered. Thus the milestone could not achieved. Now the first tests are scheduled for 1997.

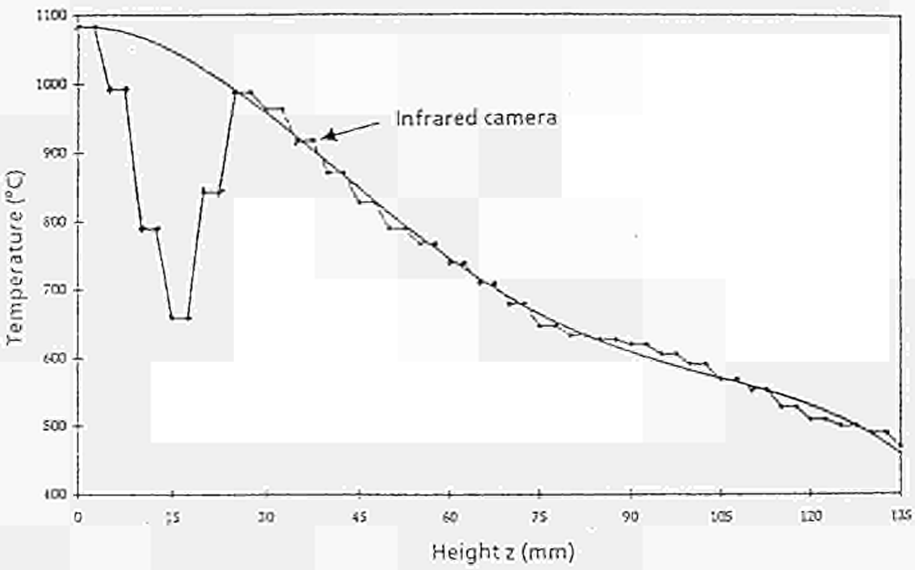

Fig. 1: Infrared camera image and corresponding axial temperature profile (the discontinuity is due to the induction wires)

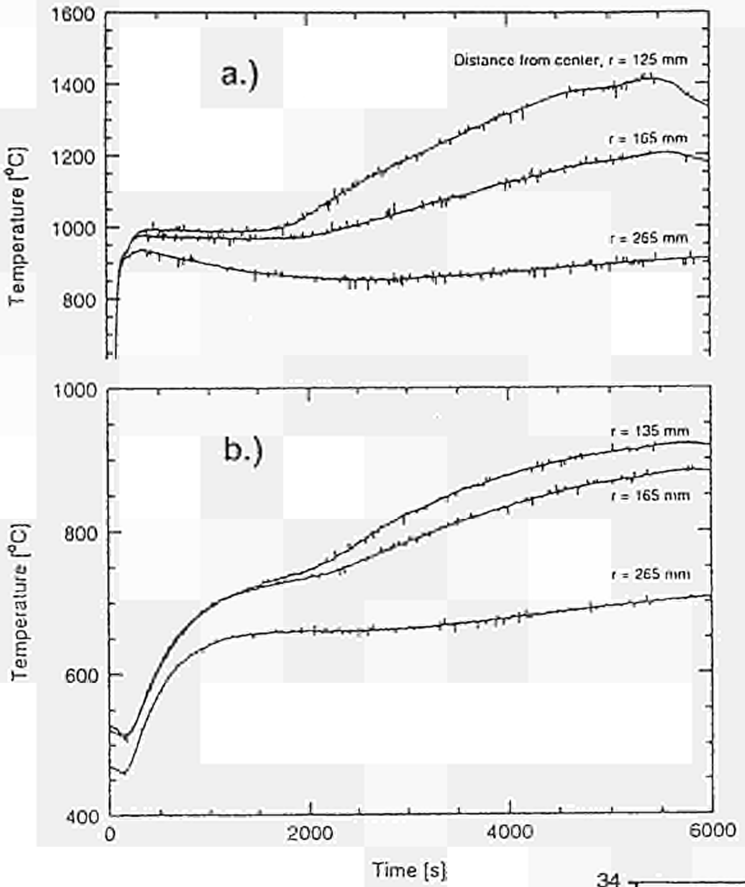

Fig. 2: Experiment 03/2.
Typical temperature histories at the test plate measured at different radius distances from the centre and at different levels above the test plate lower surface. Arc heater power 240 kW.

a) upper plate surface
b) lower plate surface

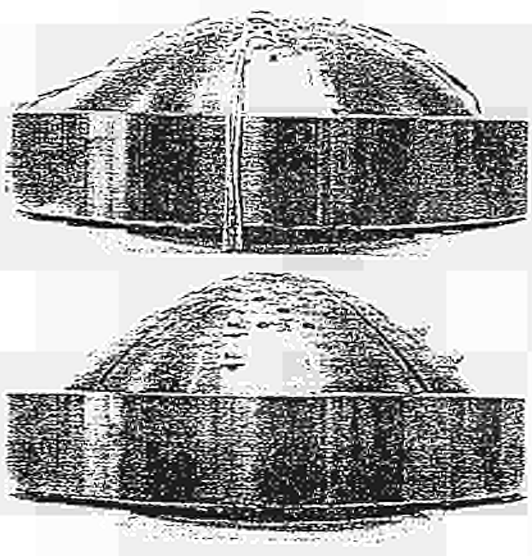

Fig. 3: Head of test 05 before and after the impact

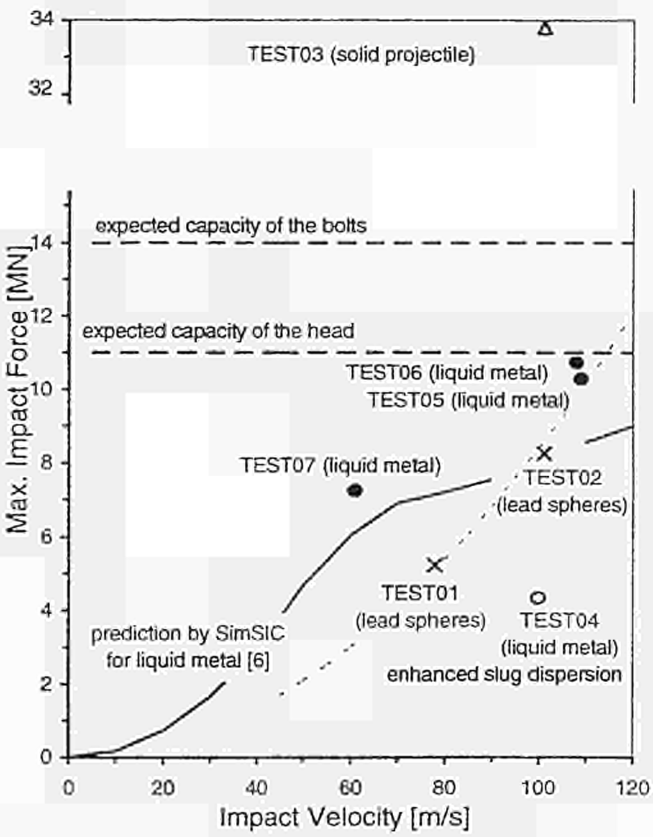

Fig.4: Results of BERDA tests without upper internal structures converted to a reference mass of 80 kg

B.2.1-1 Thermochemical modelling and data - THMO

Contract No: FI4S-CT95-0008	**Duration :** 1 Jan. 1996 - 31 Dec. 1998
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A. OBJECTIVES AND SCOPE

The overall objectives of this project are to provide experimental thermochemical data and develop further existing thermochemical calculational models to determine the composition of the melt and the quantities and chemical forms of the species released from the melt to the containment atmosphere during the ex-vessel stage of a severe accident.

Thermochemical methods, involving models and critically assessed thermodynamic data, play an increasing role in predicting fission product release and in source term applications. The thermochemical database is a vital part of the process for severe accident applications and is still being significantly upgraded by work from different European groups. Such groups have provided experimental data and models which are state-of-the-art to study these complex chemical interactions. The objectives of this proposal follow on from the previous work in providing new experimental data to reduce uncertainties, further development of the databases and assessments of the uncertainties in estimating the effect of ex-vessel interactions in severe accident analyses.

B. WORK PROGRAMME

The work programme is divided into four technical packages:

B.1. Experimental Determinations involving the critical assessment of thermochemical data for pure substances and gases relevant to melt interactions, followed by the experimental determination of thermochemical parameters that are considered inadequate or are based on estimated data (ECN).

B.2. Development of Models including critical assessment of thermochemical data for phase diagrams and the development on thermochemical models based on the assessed data. This part also includes the calculation for systems simulating stages during the ex-vessel interaction (AEA, ECN).

B.3. Validation of Models by experimental determinations for representative systems (AEA, ECN).

B.4. Sensitivity Studies of thermochemical data with respect to specific ex-vessel conditions, e.g. partial solidification, changing melt compositions due to dissolved basemat or protective material (Univ. Ruhr, KWU).

C. Progress of work and results obtained

Summary of main issues

Good progress has been made on both the experimental tasks and the assessment/modelling studies during the first 12 months of the project. A number of literature reviews have been completed and studies initiated following assessments of the available information. Thermodynamic parameters for important fission product phases have been determined experimentally and a number of thermodynamic models for relevant oxide and metal systems have been developed. The kinetic and thermodynamic parameters that impact on the uncertainty in the release of fission products from molten pools have also been reviewed. Links with groups in the In-Vessel, Ex-Vessel and Source Term Clusters of the Fourth Framework Programme have been established which will enhance the development of the project. The results from experiments performed by these groups on the behaviour of melts at high temperature could be used for the validation of the data and models. Similarly, the data and models produced in the THMO project could be used to interpret the experimental results.

Progress and results

C.1 Experimental determinations (Task 1)

The thermochemical properties for certain condensed and gas phase species of the radiologically important fission products are insufficiently known. In order to reduce the uncertainty in the predicted releases during MCCIs, thermochemical measurements (e.g. low-temperature heat capacity, enthalpy of formation and enthalpy increment) have been carried out on prepared samples. A variety of techniques have been used such as enthalpy-of-solution calorimetry, differential thermal analysis and electrochemical cell measurements. Condensed phases that have been studied experimentally, which could have an impact on the calculated releases, include $Sr_3MgSi_2O_8$, $BaCeO_3$, $SrCeO_3$, $La_2Zr_2O_7$, $Ce_2Zr_2O_7$ and α-$BaFe_2O_4$. For example, the derived enthalpy of formation, $\Delta_fH°(298.15)$, for $Sr_3MgSi_2O_8(s)$, $BaCeO_3(s)$ and $SrCeO_3$ are respectively -(4575 ± 5), -(1719.3 ±2.5) and -(1712.3 ±2.1) $kJ.mol^{-1}$. A number of publications of these results for the Journal of Chemical Thermodynamics are in preparation.

An assessment of the enthalpies of formation of the gas phase species BaO(g) and SrO(g) has been completed and parameters recommended. Preliminary experimental studies of the vaporisation behaviour of ruthenium in high temperature, high humidity atmospheres has been conducted using a transportation method to assess the importance of gaseous hydroxide species. Some experimental difficulties have been encountered at high temperature (i.e. 1673K) and the use of other experimental facilities is being assessed.

The low-temperature heat capacity of the cerium polymorphs have been studied by an adiabatic low-temperature calorimeter. The results indicate that the γ phase of Ce, and not the β phase, should be thermodynamically stable at room temperature. Along with this change, the entropy at 298.15K will change by approximately 20%. In order to be able to calculate the Gibbs energies of all three cerium polymorphs some additional enthalpy of solution measurements are being performed.

C.2 Development of models (Task 2)

Models that reproduce the available experimental phase relationships in the oxide and metal components of the ex-vessel melt have been further extended. This task involves the assessment of the thermochemical data and the binary and higher order phase diagrams of the components of the system. These models can then be used to calculate the equilibrium composition of the gas phase and melt for a range of input boundary conditions of importance during the early and late stages of the interaction (e.g. temperature, corium composition and concrete/basemat type).

As part of the task to extend the oxide database, a number of corrections to the data for the CeO_2 systems has now been implemented and data for the binary systems with Ce_2O_3 have been included to provide a full description of all the combinations of the oxide components. A literature review of possible oxide compounds in the FeO-oxide system has been performed and a number of intermediate compounds noted in the concrete systems Al_2O_3-Fe_2O_3, CaO-Fe_2O_3 and MgO-Fe_2O_3 and also in the fission product systems BaO-Fe_2O_3 and SrO-Fe_2O_3. There has been close interaction with ECN on this aspect since experimental measurements on the latter compounds, such as $BaFe_2O_4$ and $Ba_3Fe_{32}O_{51}$, where there is a lack of thermodynamic data, are required.

The extension of the thermodynamic database for the metal system has also been progressed. A literature review of the thermodynamic data for all the binary and ternary metal sub-systems has been performed and where available the published phase diagrams have been assessed. The sub-systems U-Zr-Si-Fe and U-Ba-Sr-La-Ce have been fully assessed and a solution database with the optimised interaction terms has been compiled. The U-Ba-Sr-La-Ce system is characterised by regions of large immiscibility and no intermetallic compounds. Of the other sub-systems which have not yet been assessed completely, there are a number of intermetallic compounds which have been identified from the literature and for which there are very little data available; these data are being estimated.

An interim report on the model development and calculations has been prepared and will be issued by the due date.

C.3 Validation experiments (Task 3)

Two types of validation experiments are to be performed in this task to assess the models used for the multi-component systems. The first involves the measurement of the extent of release of species from a simulated core melt at high temperature. Experimental data from other projects in the Fourth Framework Programme will also be used to validate the models. The second type will investigate the release of silica aerosol from fused mixtures of silicate and simulant core melts and characterise their interaction with the fission product simulants in the vapour phase.

Preliminary studies for mixtures containing various ratios of Si:SiO_2, Si:$CaSiO_3$ and Zr:$CaSiO_3$ have been completed. The powdered mixtures were heated to temperatures between 1500-1600°C for periods of 5-11 minutes in flowing helium gas. After each test any mass loss from the sample was determined and the filters inspected using a Scanning Electron Microscope to establish if any aerosol was transported by the gas flow.

The results from these tests clearly demonstrate the volatility of silica, as SiO(g), and its sensitivity to melt composition. In the absence of reducing agents, little vaporisation of silica occurs, however, in the presence of Si copious quantities of aerosol are generated. The silicate ($CaSiO_3$) did not reduce as readily in contact with the metal powder as silica and hence less aerosol was formed. Differences between tests also occurred as a function of the reducing agent (Si or Zr), however, further studies are required to examine this effect. When aerosol was generated it formed complex agglomerates with chainlike structures and average primary particle size of ~0.1 µm diameter. Particles in this size range are most persistent and likely to remain gasborne for long periods of time.

Thermodynamic calculations have also been performed to investigate the trends in the amount of aerosol released during the experiments performed and these were found to be in broad agreement with those in the experiments. For example, it has been shown that Si will reduce SiO_2 to form SiO(g), whereas $CaSiO_3$ is more stable in the presence of Si or Zr. Temperature is also a very important parameter influencing the amount of SiO(g) predicted to be formed.

C.4 Sensitivity studies (Task 4)

The studies are based on a coupling of thermochemical methods to calculate equilibria (CHEMSAGE code) and the kinetic parameters describing the diffusive and convective transport (RELOS code). This approach has been developed to study the uncertainty, for various boundary conditions, in the predicted release of radiologically relevant fission products from melt pools and the distribution of heat-generating radionuclides between metallic and oxidic melt phases.

A pure substance thermochemical datafile was compiled to perform the equilibrium calculations for the sensitivity study of the input boundary conditions and to provide the input data for mechanistic calculations with the RELOS code. Exploratory calculations have been performed to check the data and to identify inconsistent results.

The vaporisation release of fission products from the corium melt into the gas atmosphere above is dependent basically on the composition of the upper layer in the pool. Experimental observations show, that a core melt containing metallic and oxidic phases may undergo a rapid, density driven phase separation. Thus, possible melt stratification phenomena during the MCCI phase as well as within spreading area scenarios have been analysed. Possible mechanisms that may suppress the stratification process are also considered, for example, natural convection processes in pools, sparging of gas bubbles from the concrete during MCCI.

The distribution of fission products between these phases is also under investigation and is dependent on their chemical forms in the melt pool, which is determined by the oxygen potential of the system. Fission products in the upper layer of the molten pool can be transported directly from the pool surface into the gas atmosphere above whereas the release of fission products in the lower pool layer may be inhibited by the layer above. Thus, the calculation of fission product release from the pool surface into the atmosphere requires the coupling of the analysis of the stratification of the melt phases and the fission product distribution between them. In order to reduce the number of fission products which have to be taken into account for molten pool source term considerations, the most important radiological relevant fission products have been identified based on the annual limit of intake and the total core inventory of each fission product.

B.2.2.-1 Investigations on the phenomenology of ex-vessel core-melt behaviour - COMAS

Contract No: FI4S-CT95-0003 **Duration :** 1 Dec. 1995 - 31 May 1999
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A. OBJECTIVES AND SCOPE

The integrity of the containment must be ensured during a wide range of severe accident sequences. For new reactor designs this goal can only be achieved if strategies and technical solutions (core-catchers) will be implemented which guarantee long-term stabilization and cooling of a molten core. Since such solutions necessarily involve ex-vessel core-melt relocation processes, the theoretical and experimental investigation of such processes is an important topic of research. Consequently, the involved large-scale spreading experiments are fully in-line with actual scientific needs.

B. WORK PROGRAMME

The project is divided into five main work packages :

B.1. Pre-tests

Small-scale corium tests are carried out in an induction furnace in order to gain experience, clarify boundary conditions and check measuring devices (Siemens)

B.2. Facilities and Spreading tests

The experiments are carried out in the CARLA plant being used for the melting of radioactively contaminated metallic materials. For adaptation to the specific experimental boundary conditions, certain modifications with respect to additional safety equipment are implemented.

A large-scale spreading facility with adequate instrumentation has been developed and constructed. Eight experiments up to 3Mg of prototypical corium melts containing both a metallic (Fe) and an oxidic ($UZrO_{2-x}$) phase are being performed. Such mixture are - in respect to composition and temperature - similar to the corium expected at the time of RPV-failure in a PWR core-melt accident. The experimental programme investigates the spreading behaviour of molten corium, and its interactions with different surface materials. (Siempelkamp).

B.3. Instrumentation and measurement

The instrumentation and the recorded data focus mainly on the parameters of the melt (temperatures up to 2200°C, viscosity, density and composition), the spreading behaviour of the melt and the behaviour of the furnace (Battelle).

B.4. Material investigations

Solidified test specimens are being analyzed in order to determine among others the influence of melt parameters on the composition and structure of the melt as well as the formation of eutectical corium phases (Univ. Aachen, Siemens).

B.5. Theoretical work

The experimental data are used for verification of the computer codes, i.e. engineering models or numerical models as CORFLOW and MELTSPREAD (Ansaldo).

C. Progress of work and results obtained

Summary of main issues

The COMAS-EU tests performed so far, have demonstrated that the technical objectives of the project can be met. The first-performed, medium-scale pretests EU-1 and EU-2 yielded valuable information on the identification of reference substrata, and on safe and effective procedures for melt generation and pouring.

Based on this information, the large-scale test COMAS-EU 4 could then be performed succesfully. This test provided data for code verification and also highlighted phenomena that characterize the 2D-spreading of prototypic core-melts. The experiments performed so far confirm the results of earlier simulant material tests, namely that - with superheated melt of high enough flow rate - a uniform spreading and a clear separation of phases can always be achieved if only the average thickness of the spreaded melt will exceed a certain limit, being in the range of a few centimeters.

The pre- and post-calculations using the codes CORFLOW and MELTSPREAD-1 have pointed out some key phenomena which need further investigation and code improvements.

C.1 Pretests

Two first spreading 1D-tests, so-called pretests, with inactive materials were performed, both using one short 4 m spreading course, in order to check the experimental facilities for the real corium experiments.

In the PRETEST EU-1 350 kg molten iron on a temperature level of roughly 2000°C was spread over a 4 m concrete course. Concrete was chosen as substratum for covering the worst conditions with respect to steam and aerosol production, clearly detected during the spreading process as consequence of intensive melt-concrete interactions.

In the second test-run (PRETEST EU-2) a mixture of 560 kg molten iron and inactive oxide material (ZrO_2, Fe_2O_3, Cr_2O_3) with an oxide portion of approx. 10 % was spread over the 4 m course with a ceramic based substratum (mainly Al_2O_3). The maximum measured melt bath temperatures were in the range of 2000°C.

Measured melt front velocities were in the order of 1 m/s. Remarkable is the reflection behaviour at the end of the spreading channel. In both cases a significant wave propagation opposite to the incoming melt front could be detected.

C.2 Facilities and spreading tests

For the COMAS experiments, the equipment of the CARLA plant (Figure 1) has been especially adapted to the ultra-high-temperature regime (> 2000°C instead of ≤ 1600°C during normal melting procedures). Together with a manufacturer a newly developed material on ZrO_2-basis was prepared as furnace liner substituting the normally used Al_2O_3- or MgO-lining material.

Special emphasis was laid on the development of high quality W/Re-thermocouples for prolongation of the measurement period and therefore improvement of the signal accuracy. By the installment of totally three IR cameras the efficiency has been further enhanced.

Figure 2 shows the experimental arrangement for the 2D-spreading test COMAS EU-4 with cast iron as substratum. Due to symmetry reasons the real area of approx. 15 m² can be analytically expanded to an effective spreading area in the order of 30 m² equivalent to a 1:6 scaling of the present EPR design. The melt is heated up within the induction furnace and then poured into an intermediate basin. By lifting the plug system the melt is transferred to the spreading course with a maximum length of approx. 8 m.

As first of the planned large-scale experiments the 2D-spreading test COMAS EU-4 was performed. 2.2 Mg Corium R (composition see Table I) was heated up to 2175°C within the induction furnace. With a spreading temperature of approx. 2050°C, a spreading length of about 6.5 m along the direct line was reached (Figure 3), the front velocity started with approx. 2 m/s and held an average value of 1 m/s before solidification.

C.3 Instrumentation and measurement

In all three spreading tests the mass rate for the spreading process was measured via continuous weighing of the intermediate basin by means of force transducers.

Spreading velocity was monitored by use of IR cameras, a set of up to five video cameras and additionally by a system of burn wires immersed into the spreading course. These „thermocouples“ generate a signal when hit by the melt front and are destroyed shortly after.

Ni-Cr-Ni thermo-couples were embedded in the substrata in different depth levels to measure the instationary temperature field and, thus, determine the heat transfer from the melt to the substratum.

C.4 Material investigations

In order to determine the metallurgical structure and composition of all relevant materials samples taken during the process steps melting and pouring as well as after spreading by taking solidified test specimens have been analysed. Micro range analysis with the scanning electron microscope (SEM) and the attached X-ray micro analysator (EDX) were carried out for selected specimens. In addition, X-ray diffraction was performed to examine segregation effects of oxidic and metallic phases and by that determine the concentration of relevant elements with atomic numbers $Z > 4$ and a detection limit of < 0.2 wt.%.

In all cases the results of these examinations confirmed the mass balance and therefore the O/M-ratio. For the COMAS EU-4 test post-test examination of the solidified melt revealed a clear vertical separation between the metallic (bottom) and oxidic (top) phases. This is a noteworthy result, since the difference in densities was small (< 0.3 Mg/m³) and solidification occured quickly. No oxidic phases could be found between the metallic layer and the surface of the substratum.

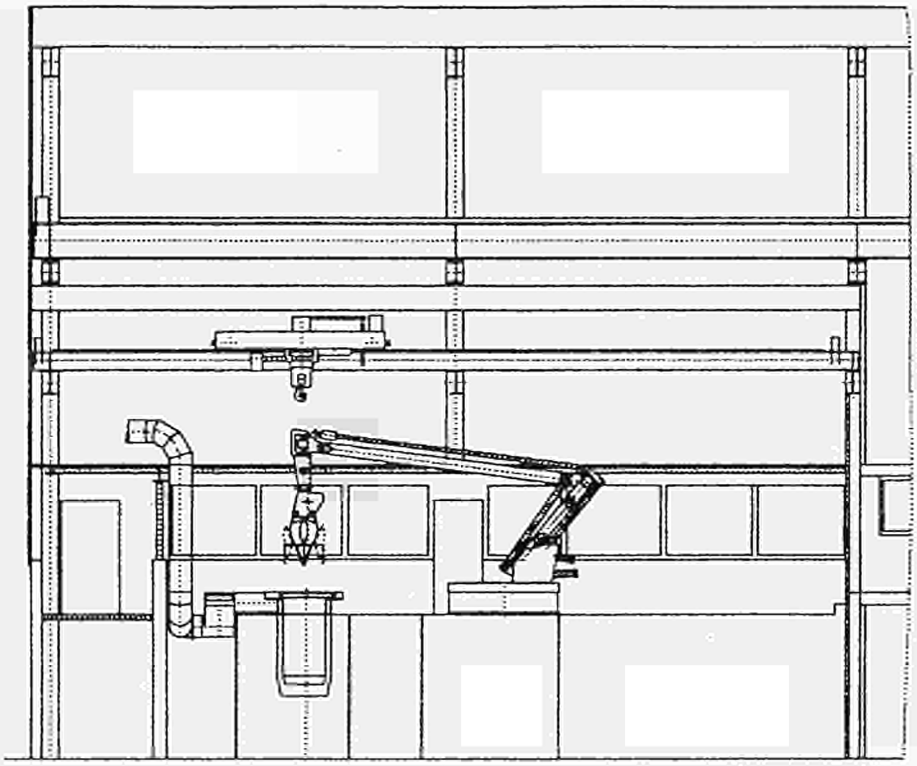
C.5 Theoretical work

The pre- and post-calculations using the numerical codes CORFLOW and MELTSPREAD-1 showed some deficiencies in the model assumptions. For further development of the CORFLOW code implementation of multi-component fluids, turbulence models and crust formation has to be required. For the improvement of the MELTSPREAD-1 code the inclusion of the intertial term at the spreading channel inlet has been identified. For both codes the melt-to-substratum heat transfer model needs assessment with view to the test data.

Table 1: Reference composition of prototypic Corium R

Corium R	
UO_{2+x}	29 wt.-%
ZrO_2	12 wt.-%
FeO	18 wt.-%
Cr_2O_3	2 wt.-%
Fe	39 wt.-%

high oxygen level



Main data:

Inventory	3.2 Mg	Max. Temperature	≈ 2200°C
Frequency	300 - 500 Hz	Melted Mass	10000 Mg
Power Input	2 MW	Melting Capacity	2000 kg/h

Figure 1: CARLA Facility

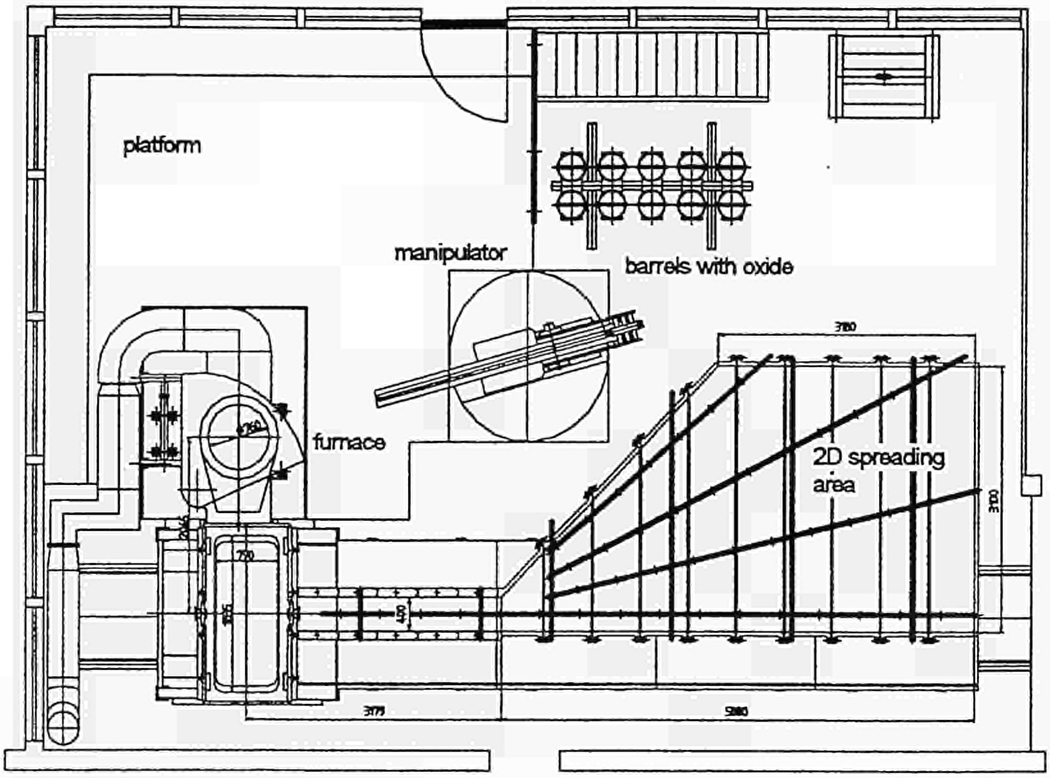


Figure 2: 2D-arrangement for corium spreading

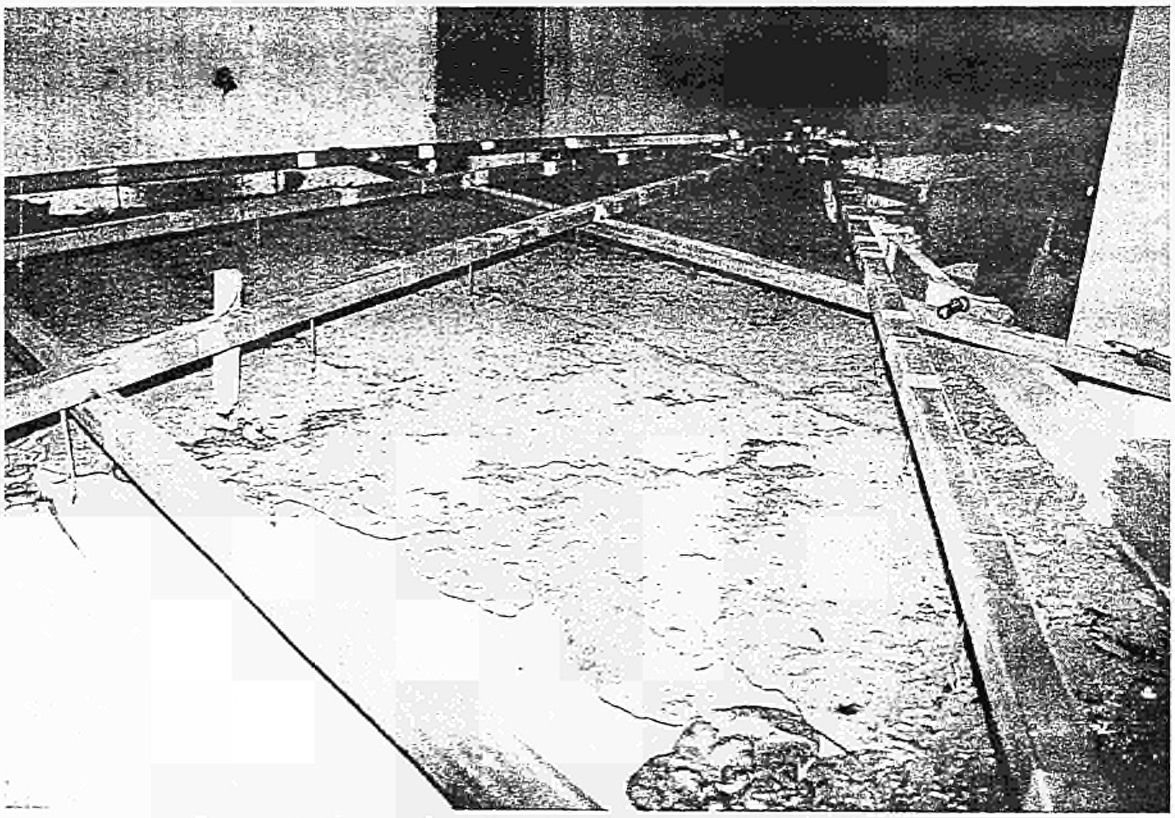


Figure 3: Spread corium R after the COMAS EU-4 test

B.3.1-1 Fission product release and speciation - RSP

Contract No:	FI4S-CT95-0006	**Duration:**	1 Feb.1996 - 31 May 1999
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Partners:	**CEA Cadarache/FR, ENEA Roma/IT, Universität Stuttgart/DE, JRC-ISIS Ispra/EC, Univ. Ruhr Bochum/DE**		

A. OBJECTIVES AND SCOPE

The overall objective of this project is to provide a database on the chemical identities (speciation) and kinetics of release of fission products evolved from irradiated fuel during a variety of severe reactor accident conditions. This experimental work will be integrated with theoretical studies to test capabilities in this area.

The chemical forms of fission products evolved from fuel during a severe accident will determine the radioactive source term to the containment and environment in terms of the volatility of fission products (eg. molecular iodine is far more volatile than caesium iodide), possible vapour reactions (eg. production of reactive caesium hydroxide compared with non-reactive caesium borate), and the nature and size distribution of any aerosols formed. However, because of the high temperatures involved and activities of irradiated fuel samples, few direct measurements have been recorded.

Therefore approximately 10 experiments are proposed to examine the release of fission products and other reactor materials from highly-irradiated fuel heated under a range of atmospheres to temperatures up to 2000°C. Mass spectrometry will be used to quantify the release rates and chemical forms of the major fission products. This technique will be supported by gamma-ray spectroscopy and thermal gradient tube/aerosol analysis methods to provide additional data. The main variables of the experimental programme will be the maximum temperature and rate of increase, pressure (from near-vacuum to 1 bar), oxygen potential (including steam, air and hydrogen - in particular a number of experiments will be conducted under oxidizing conditions where the available data are limited), and the effects of fuel, cladding and other core materials.

These experiments will be integrated with theoretical analyses in order to test and validate the fission product release codes. In this manner, the on-line results from the project can be compared with Phebus-FP data and other complementary separate-effect studies (eg. HEVA/VERCORS), where emphasis is placed on post-test analyses of aerosol deposits.

Specific objectives from this programme are to establish an experimental database on fission product release and speciation - notably for releases in oxidizing atmospheres, to test theoretical models for fission product release, and to validate thermochemical models used in state-of-the-art severe accident codes. This work will closely complement the Phebus-FP studies, providing separate-effects data with which to interpret these key integral experiments.

B. WORK PROGRAMME

B.1. Definition of the Test Matrix (Task 1)

The test matrix will be finalized by AEAT, CEA and JRC in conjunction with the other partners.

B.2. Experimental Work (Task 2)

The work will comprise scoping and sensitivity studies, oxidation studies and studies in support of PHEBUS-FP, and will be carried out by AEAT.

B.3. Code Analysis (Task 3)

This Task will be carried out by all partners.

C PROGRESS OF WORK AND RESULTS OBTAINED

Summary of Main Issues

The Fission Product Release and Speciation project has begun on schedule. The main activities have included definition of the test matrix, commissioning tests with the time-of-flight mass spectrometer and associated hardware, and pre-test calculations using a number of fission product release codes.

The programme will provide an understanding of fission product release from fuel in terms of both the rate (kinetics) of release and chemical identities (speciation) of the evolved fission products. This work is built around a test matrix of 10 or 11 experiments to follow fission product release from samples of irradiated fuel. These experiments are designed to provide a detailed understanding of the important processes affecting fission product release and speciation. In particular, the work will address the effects of temperature, pressure, oxygen potential, and other reactor materials on the release processes. These experimental studies are integrated with modelling work to test the fission product release and transport codes. In particular, the unique data generated on fission product speciation will allow the thermochemical models used within state-of-the-art codes to be tested and hence reduce major uncertainties in the source term. These studies closely complement the current Phebus-FP programme and also involve comparison with other related separate-effect studies.

The unique Fission Product Release and Speciation facility is at the heart of this project. This facility has been designed to determine the identities of chemical species present at high temperatures by conducting in-situ measurements. The main purpose of the facility is to characterise the vapour-phase species released from high burn-up fuel under a range of reactor accident conditions, although it is possible to study a diverse range of high-temperature vapour-phase systems. Up to 100 grams of highly irradiated fuel can be heated. The design of the facility is modular and the main features of the system are described below:

1. a precision furnace capable for attaining temperatures up to 1600°C, with plans to extend this to 2000°C in the next year;

2. provision for a range of atmospheres of different compositions;

3. a pressure reduction system that enables the vapour-phase species to be sampled on-line and at ambient pressure;

4. a Jordan reflectron time-of-flight mass spectrometer that combines sensitivity with scanning of the full mass range of species of interest;

5. fast data scanning and acquisition to optimise on-line measurements;

6. time-dependent sampling of material for analysis using γ-ray spectroscopy;

7. containment in a shielded high-active cell to enable handling of highly active samples.

Unlike a conventional quadrupole mass spectrometer, the state-of-the-art reflectron time-of-flight instrument has the benefit of being capable of scanning the whole mass range of interest, rather than measuring the intensity of pre-selected peaks in the spectrum. Therefore this feature, combined with comparable sensitivity, makes it less likely that important species will be omitted from the measurements. The flexibility of the system is such that it can be readily adapted to accommodate samples of different geometry.

As described below, an extensive commissioning programme has been conducted. The mass spectrometer is now fully operational and it is planned to install the facility within a cell later this year so that experiments with high burn-up fuel can begin.

C.1 Definition of the Test Matrix

A provisional matrix of experiments has been produced [1] and was discussed at the first meeting of the Project Group, held at Winfrith on 21 May 1996 [2]. The provisional test matrix is listed in Table I, and has been discussed in more detail elsewhere [1]. It comprises 11 experiments to be conducted in which the following variables will be studied:

(i) maximum temperature and rate of temperature increase,

(ii) pressure (from near vacuum up to 1 bar),

(iii) oxygen potential (a range of atmospheres to be studied including steam, hydrogen and air),

(iv) effect of fuel, cladding and other core materials (initial experiments to be conducted with unclad fuel, studies to increase in complexity to study the effects of Zircaloy cladding, control rod alloy, boric acid and stainless steel).

The initial experiments will be relatively simple, to assess the influence of atmosphere, pressure, temperature and cladding. These will be followed by experiments conducted in oxidising atmospheres (ie. CO_2 and air), whilst the final experiments will be performed under a variety of conditions pertaining to an accident in a PWR. The later tests will also be highly relevant to some of the Phebus-FP experiments, and will be performed under similar conditions. The matrix is flexible and expected to evolve during the course of the project in order to meet the requirements of the various fission product release models. Comments on the test matrix have been received from all the partners in the Project Group [3,4], and these will be taken into account, particularly with respect to the nature of the later experiments. It is envisaged that this will be a continuous process that will allow the test matrix to evolve and produce data that are of maximum benefit to model development.

C.2 Experimental Work

Initial effort was concerned with preparation of the mass spectrometer facility prior to in-cell installation. The mass spectrometer is fully operational, and a series of commissioning tests is nearing completion. The initial phase involved a number of alignment tests and handling trials, whilst the second phase of the commissioning programme comprises a number of different experiments that are designed to test the performance of the system in terms of sensitivity and resolution, and also provides an opportunity to mimic the in-cell experiments. The commissioning tests can be summarised as follows:

- experiments with gas mixtures (eg. air, inert gases)
- experiments with single fission product sources (eg. CsI, CsOH),
- experiments with complex systems (eg. simulant fuel, control rod material etc)

The aim of the experiments with complex systems is to simulate the experiments to be conducted in-cell with highly irradiated fuel. It is intended to conduct these experiments throughout the programme, using a non-active duplicate configuration. The results will provide information on the sensitivity of the system, and highlight any potential problems with the in-cell experiments. The results to date provide confidence that the required sensitivity and resolution will be achieved to follow the kinetics of fission product release.

Experiments have been conducted using simulants not only to determine the sensitivity and resolution of the instrument, but also to assess the deposition of fission products throughout the apparatus. Work has also been conducted to optimise the molecular beam produced in the instrument, with particular emphasis on the orifice design etc. Some problems were encountered with the data collection system, resulting in reduced sensitivity. The next stage of commissioning will involve examination of a more extensive range of simulant fission product species, and simulant fuel. The system will then be installed in-cell.

The parallel preparations for installation of the system in an active cell have also continued. The decontamination and re-commissioning of an existing cell are nearing completion. Various systems for in-cell operation have been designed and are currently being manufactured. These include a new set of vacuum chambers, that incorporates provision for both cumulative and time-dependent γ-ray spectroscopy measurements, and apparatus for remote alignment.

C.3 Code Analyses

Pre-test calculations have commenced using a number of codes. These calculations will enable the experimental programme to focus on specific phenomenon and processes, and hence produce highly usesful data. The work has progressed well, and is important as it aids with the definition of the experiment test matrix. Two approaches have been used: assessment and modelling of the results of previous experimental programmes, and calculations based on the experimental conditions listed in the initial test matrix. The results of both types of exercise will be used to identify key uncertainties, which will in turn aid in focusing the experimental programme. Calculations have been conducted with ELSA, VICTORIA, FIPREM/RES and RELOS/CHEMSAGE, with analyses of PBF-SFD, Phebus-FP and SASCHA tests, as well as the experimental conditions proposed for the current programme of experiments.

For example, a series of VICTORIA calculations has been performed using code version v92.01 to investigate the fission product release characteristics for some of the tests currently proposed (ie. role of inert (helium), reducing (steam/hydrogen), and oxidizing (air) environments). The key results for a 90% steam-10% H_2 atmosphere using peak final temperatures of 1200°C, 1600°C and 2000°C are given below [4].

The elemental release fractions of the noble gases, caesium, and iodine are similar to those predicted for the inert carrier gas calculations with the exception that the release of caesium at 1200°C is an order of magnitude less under steam/hydrogen conditions. At the lowest temperature the dominant caesium vapour species is predicted to be caesium iodide, with caesium hydroxide concentrations being of the order a factor of 2 less. At higher temperatures the importance of caesium hydroxide increases and, at 2000°C, it is predicted to be more than two orders of magnitude greater in concentration than caesium iodide. Elemental caesium is predicted to account for ~10% of the release at 2000°C.

The dominant iodine vapour species is predicted to be caesium iodide with equal contributions (~10%), at the lowest temperature, from hydrogen iodide and elemental/molecular iodine.

The importance of elemental/molecular iodine is predicted to increase with temperature and, at 2000°C, the contribution from this species is ~30-40% towards the end of the test.

Tellurium release from the fuel is predicted to be strongly dependent on temperature with less than 0.05% being released at 1200°C and ~27% at 2000°C. For the first ~70 minutes of the calculation for the low temperature scenario, the highest tellurium vapour concentration is predicted to be that of tin telluride which is an order of magnitude greater than the vapour pressure of elemental tellurium, this order is reversed towards the end of the test. As the test temperature is increased, other tellurium vapour species are favoured and the relative importance of tin telluride decreases; tellurium oxides, antimony telluride and hydrogen telluride fall into this category. The formation of tellurium iodides and iodates is not predicted to be significant under these test conditions.

The releases of barium and strontium are predicted to be reduced by a factor of ~2 at 2000°C and by a factor of ~40 at 1600°C compared with the predicted releases under inert conditions. Small amounts (5×10^{-6} - 2×10^{3}%) of molybdenum are predicted to be released. The behaviour of molybdenum is predicted to be complex with more being released at 1600°C than at either of the other two temperatures. The formation of caesium and barium molybdates is predicted.

The results of all the code analyses have highlighted some important areas of uncertainty, which will be addressed in the experimental programme.

<u>References</u>

[1] BENSON, C G, BOWSHER, B R and WILLIAMSON, R, Definition of Test Matrix, ST-RSP(96)-P003 (1996).

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[3] BENSON, C G, BOWSHER, B R, COPELAND, P S, DAVIDOVICH, N, KISSANE, M P, KOCH, M K, NEWLAND, M S, PALLER, A, SHEPHERD, I M, UNGER, H, WILLIAMS, D A and WILLIAMSON, R, Fission Product Release and Speciation Progress Report, 01/02/96-31/07/96, ST-RSP(96)-P004 (1996).

[4] BENSON, C G, BOWSHER, B R, COPELAND, P S, DAVIDOVICH, N, KISSANE, M P, KOCH, M K, KORTZ, CH, KOURTI, N, NEWLAND, M S, PALLER, A, SHEPHERD, I M, UNGER, H and WILLIAMS, D A, Fission Product Release and Speciation Annual Progress Report, 01/02/96-31/01/97, ST-RSP(97)-P007 (1997).

Table I: Preliminary fission product release and speciation test matrix

Test	Fuel Source	Atmosphere	Final Temp (°C)	Heat-up Rate	Comment
RSP-01	UO_2/unclad	Vacuum	600-1200	ramped temp staircase	Scoping expeirnents - effect of atmosphere, pressure and temperature
RSP-02	UO_2/unclad	Inert	600-1200	rampted temp staircase	
RSP-03	UO_2/unclad	CO_2/CO	1000	ramp to isothermal	Primarily GCR experiments, but potential to provide data on oxidizing conditions for PWRs
RSP-04	UO_2/unclad	CO_2/CO	1200	ramp to isothermal	
RSP-05	UO_2/clad	CO_2/CO	1400	ramp to isothermal	
RSP-06	UO_2/unclad	Air	600	ramp to isothermal	Transient accident and air ingress experiments (explore role of U_3O_8 formation on fp release) - relevant to Phebus-FPT-5
RSP-07	UO_2/clad	Air	1000	ramp to isothermal	
RSP-08	UO_2/clad	Air	1600	ramp to isothermal	Above critical tempertaure for U_3O_8 - impact of volatile fp oxides - relevant to Phebus-FPT-5
RSP-09	UO_2/clad/ control rod	As FPT0: Low H_2:H_2O ratio	2000	As FPT0	Simulation of Phebus-FPT0 (but with highly-irradiated fuel)
RSP-10	UO_2/clad/ control rod	As FPT1: Low H_2:H_2O ratio	2000	As FPT1	Simulation of Phebus-FPT1 (oxidising conditions)
RSP-11	UO_2/clad/ control rod/ boric acid	As FPT2: Low H_2:H_2O ratio, then H_2	2000	As FPT2	Simulation of Phebus-FPT2 (oxidising then reducing conditions plus boric acid)

B.3.1-2 Revaporisation of tests on samples from PHEBUS fission products - RVP

Contract No:	FI4S-CT96-0019	**Duration:**	1 May 1996 - 31 Aug. 1999
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Partners:	**JRC-ITU Karlsruhe/EC, VTT Espoo/FI**		

A. OBJECTIVES AND SCOPE

At a late stage of a severe accident in a light water reactor, fission products deposited initially in the reactor coolant system may experience increasing temperatures, due to heat transfer from the surrounding gas or radiating corium, or due to their own decay heat, leading to revaporisation. Revaporisation could become risk dominant if the containment is breached at this time and offers little attenuation to the source term. This phenomenon has been identified as a significant uncertainty in various probabilistic risc assessments and sensitivity studies. Addressing this key issue has become an internationally recognised priority.

The Phebus-FP experiments carried out by CEA/IPSN at their research centre at Cadarache/France provide a unique opportunity to acquire samples of real deposited fission products from irradiated fuel. This allows to investigate revaporisation in a supporting laboratory study under realistic conditions.

In order to carry out these investigations, the following work is being performed under this experimental programme :

- recovery of circuit portions from the Phebus-FPT1 test,
- characterization of the initial state of the deposits,
- revaporisation of the deposits,
- measurement of the simple kinetics of the revaporisation (weight change as a function of time,)
- characterization of the secondary deposits and the final state of the original deposits and, hence, characterization of the physics and chemistry of the emission.

This work is being supported by separate-effects tests to quantify the revaporisation phenomena and provide data from simple cases (both inactive and radiotraced samples) to interpret the releases from the more complex Phebus-FP samples.

B. WORK PROGRAMME

B.1. Separate-Effects Studies (Task 1)

The simulants studies will be made by AEA Technology; VTT and the JRC are responsible for the radiotracer studies.

B.2. PHEBUS-FPT1 Studies (Task 2)

This Task comprises Knudsen cell and thermal gradient tube studies and will be carried out by all partners.

C. Progress of work and results obtained
Summary of main issues

The Revaporisation Project has commenced as scheduled. Separate-effects experiments involving simulants have begun, and will provide a basis for interpretation of the revaporisation results from Phebus-FPT1 samples. Early tests have concentrated on the interaction and revaporisation of CsOH with stainless steel at high temperatures and at various oxygen potentials typical of those during a severe accident. Specimens have been analysed by Knudsen cell mass spectrometry, secondary ion mass spectrometry (SIMS) and scanning electron microscopy with dispersive x-ray analysis (SEM-EDX). In addition, thermal gradient tube apparatus, and its associated aerosol measurement systems, is being designed, built and tested for use in revaporisation studies of radiolabelled simulants and the Phebus-FPT-1 samples. The Phebus-FPT-1 test has now taken place and samples for the revaporisation experiments will be supplied to the project as scheduled.

The first meeting of the Project Group took place in Aix-en-Provence on 26 September 1996 [1], and a six-monthly Project Progress Report covering the period 1 May to 31 October 1996 has been issued [2]. Close interactions have taken place with the related Concerted Action concerned with comparing the modelling of revaporisation.

C.1 Separate-Effects Studies

The work involving fission product simulants has concentrated on the revaporisation of caesium. Thermal reaction tube apparatus has been set up and calibrated for a range of carrier gases: 4%H_2-argon, pure argon, and 4%H_2-argon with either 3%, 16% or 50% steam added. CsOH was vaporised at about 500°C into flowing gas and carried downstream to react with 304 stainless steel foil at 900°C. After deposition, Cs was revaporised by gradually re-heating the specimen (up to approximately 1400°C) in a mass spectrometer, either in the Knudsen cell or in a non-equilibrium configuration. Results indicate that the oxygen partial pressure and steam content of the gas during Cs deposition have no major effect on the Cs revaporisation characteristics, at least for the range of gases studied. The Cs release rate above 800°C was controlled by kinetic factors and kinetic rate constants for the release have been calculated between 800-1250°C. SIMS depth profiling prior to revaporisation has shown that the oxygen partial pressure during deposition influences the Cs concentration profile within the surface oxide. In another series of experiments, CsOH was deposited on stainless steel at 900°C in the thermal reaction tube in various carrier gases, and then revaporised in a second similar apparatus at 1100°C, in an environment with the same or a different oxygen potential. Atomic emission spectroscopy (AES) showed that most of the Cs deposited at 900°C had been revaporised after one hour at 1100°C, independent of the carrier gas used during deposition or revaporisation.

Two identical thermal gradient facilities are being prepared, one for use with synthetic radiolabelled samples, and the other for analysis of Phebus-FPT1 samples. A revaporisation measurement system and a test procedure have been designed that will optimise the accuracy of fission product vaporisation rate measurements. Particular care has been taken to produce test conditions close to those expected in a severe accident. The system will be operated as follows during the separate-effects tests. Synthetic radiolabelled samples will be put on oxidised stainless steel sample plates. These plates will be placed in the tube furnace over the second heating element. The vaporisation rate, which coincides with decreasing activity of the sample, will be measured from outside of the furnace with a germanium-gamma detector. Samples will be vaporised in a pure steam atmosphere at slightly under normal pressure. Downstream of the furnace, the gas flow will be cooled in a diluter with room temperature air. The fission product vapours will form aerosol particles during the cooling process, which will be collected in a particle filter. Any remaining gaseous iodine will be collected in active carbon filters. Before proceeding with the actual test programme, pre-tests are being conducted with inactive materials.

C.2 PHEBUS-FPT1 Studies

There has not yet been time to prepare and transport the vertical Phebus-FPT1 test samples to the participating laboratories. Once Phebus-FPT1 samples are available, revaporisation will take place in the thermal gradient apparatus described above. Sophisticated analytical techniques, such as γ-spectroscopy, inductively coupled plasma mass spectroscopy (ICP-MS), inductively-coupled plasma optical emission spectroscopy (ICPOES), x-ray diffraction (XRD), SEM-EDX, particle recognition and characterisation (PRC), electron microprobe analysis and SIMS, will be used to characterise the Phebus-FPT1 samples before and after revaporisation. Phebus FPT1 specimens will also be studied by Knudsen cell mass spectrometry. Deposits of single compounds or simple mixtures on stainless steel surfaces will be tested for comparison with the Phebus samples, and with the separate-effects studies.

References

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[2] BOTTOMLY, P D, BOWSHER B R, JOKINIEMI, J, NEWLAND, M S AND TUSON, A T, Revaporisation: Progress Report, 1 May to 31 October 1996, ST:RVP(96)-P4 (1996)

B.3.2-1 Iodine Chemistry - IC

Contract No:	FI4S-CT95-0005	**Duration:**	1 Jan. 1996 - 31 Dec. 1997
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Partners:	**CEA Cadarache/FR, Siemens AG Erlangen/DE, JRC-ISIS Ispra/EC, NNC Ltd Knutsford/GB, IVO Vantaa/FI, CIEMAT Madrid/ES**		

A. OBJECTIVES AND SCOPE

Iodine is one of the most important fission products which would be released in the event of a severe reactor accident. Plant assessments have shown that it contributes significantly to the source term for a range of accident scenarios. However, the considerable differences between the iodine behaviour observed in Phebus Test FPTO and that predicted by containment chemistry calculations indicate that the current models do not correctly treat all of the phenomena that could be important in a reactor accident. In particular, the presence of a large quantity of silver aerosol appears to influence very strongly the volatility of iodine from solution. This could have important consequences for active and passive safety measures and for accident management strategies.

To help clarify the phenomenology, and to increase confidence in the modelling of iodine behaviour in containment, an integrated programme of experiments and analysis is proposed involving the key European laboratories working in the field. This project will focus on understanding and quantifying the effects of silver on iodine behaviour, providing new experimental data that will be used to validate and improve the existing models and to stimulate code development.

The objectives of this project correspond to issues related to analysis of ex-vessel fission product behaviour. The work carried out should help to solve these high-priority issues.

B. WORK PROGRAMME

B.1. Experimental Studies (Task 1, CEA, AEA Tecn.)

Measurement of the rate of uptake of aqueous iodine onto silver particles in the absence of radiation. Investigation of irradiation induced effects including nitric acid formation and radiation stability of AgI on a silver surface. Experimental studies of iodine radiolysis at high temperature, focusing on the effects of silver on iodine volatility under irradiation.

B.2. Assessment, Analysis and Model Development (Task 2, lead organisation: AEA Techn.)

Assessment and analysis of new and existing data on iodine interactions with silver, leading to the development and incorporation of a suitable model into iodine chemistry codes (INSPECT, IMPAIR, IODE and ACT-WATCH).

B.3. Reactor Source Term Evaluation (Task 3, lead organisation: NNC)

Reactor calculations to evaluate the effects of the model developments on source term calculations.

C. Progress of Work and Results Obtained

Summary of Main Issues

The project progress to date is in line with the milestones defined in the contract. The literature review (Task 2.1) and definition of experimental results (Task 1.0) have been completed and reports issued. Experimental studies of nitric acid formation and silver iodide stability under irradiation (Task 1.2) have also been completed and data reports issued. Experimental studies of iodine-silver reaction kinetics (Task 1.1) and iodine radiolysis (Task 1.3), and analysis of the results (Task 2.2) are underway.

C.1 Experimental Studies

Task 1.0: The range of conditions to be used in the experimental programmes have been defined, based on the results of the Phebus-FPT0 test.

Task 1.1: An experimental programme is underway to quantify the kinetics of the reactions of I_2 and I^- with silver surfaces. The I_2 - Ag reaction is very rapid and may be mass transfer limited under the conditions of these experiments. The experimental results fit well to a pseudo-first-order kinetic law, with a rate constant of 1.3×10^{-4} m s^{-1} at room temperature. (Figure 1)

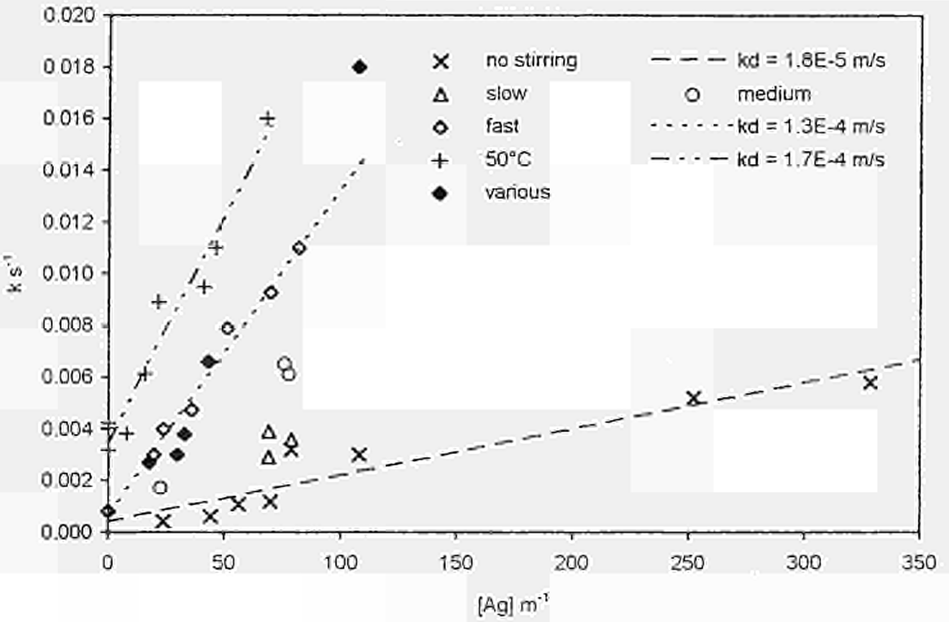


Figure 1: I_2 - Ag Reaction Kinetics

The I^- - Ag reaction is much less rapid, with an apparent first order rate constant of 3×10^{-7} m s^{-1} at pH 4.6 and 90°C. The reaction is slightly slower at higher pH, as shown in Figure 2. Different reaction behaviour is observed depending on whether the silver is present as fine mesh or particles (Figure 3); the reasons for this have not yet been established.

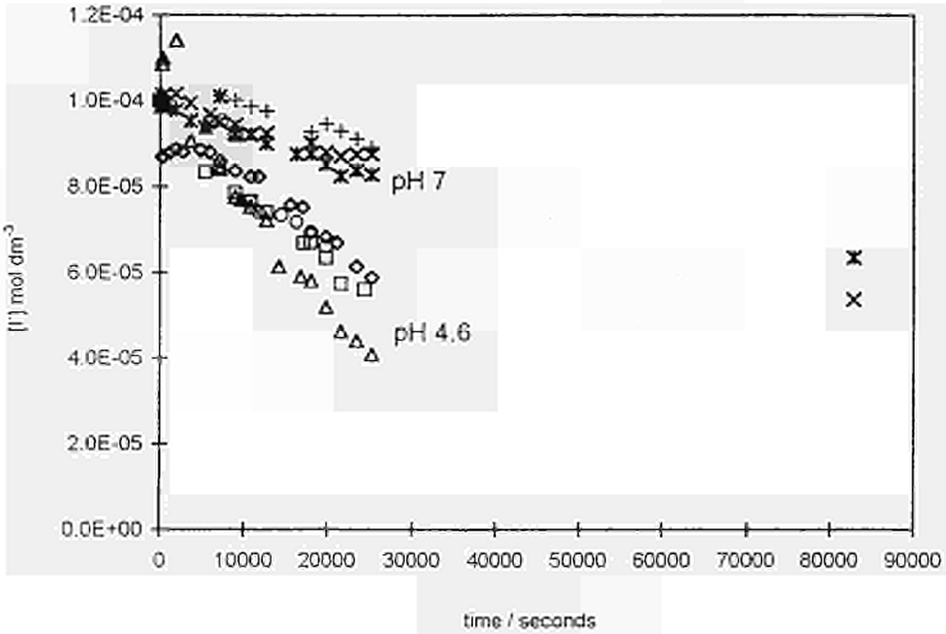


Figure 2: Reaction of I^- with Ag Mesh at 90°C

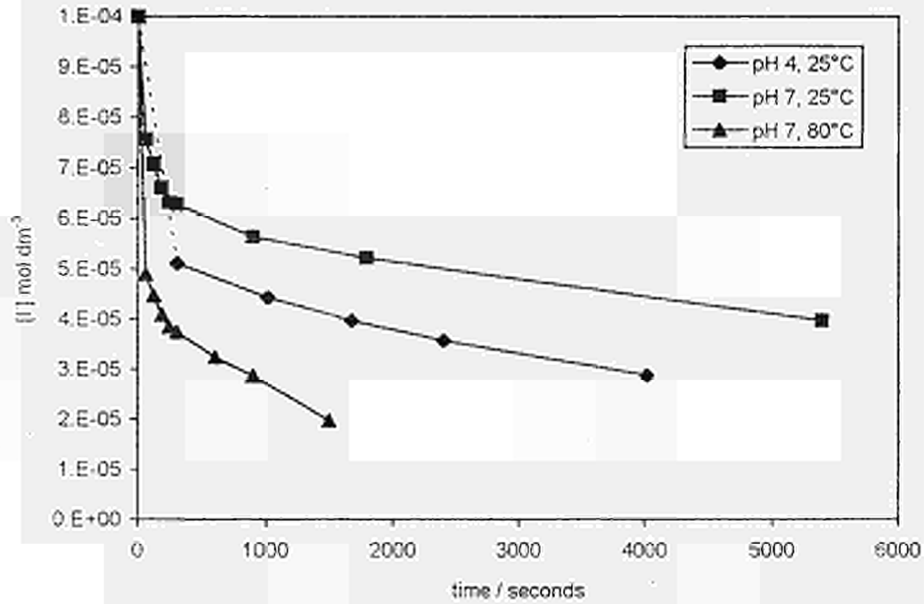


Figure 3: Reaction of I^- with Ag powder

Task 1.2: A series of experiments to quantify the radiolytic formation of nitric acid in air-water mixtures at elevated temperatures has been completed. This work has confirmed that nitric acid production is predominantly a gas-phase process. The G-value is approximately 2 /100 eV, and this is not influenced by the temperature (Figure 4).

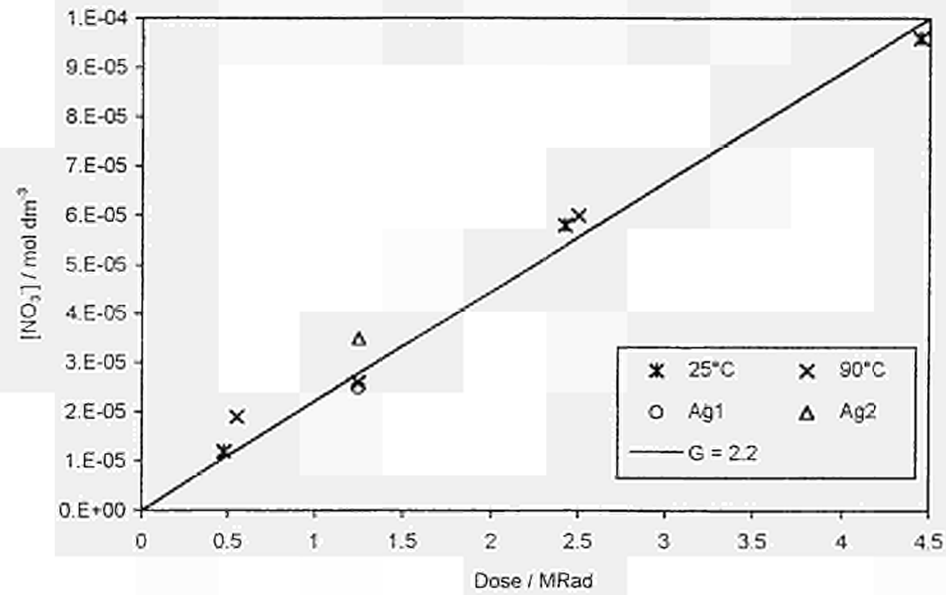


Figure 4: Nitric Acid Production in Irradiated Air-Water Mixtures

The effects of silver on the volatility of iodine from irradiated CsI solutions has been studied using a sparging technique. Iodine volatility is substantially reduced in the presence of excess silver (Figure 5). The reduction in volatility at pH 4.6 is consistent with the rate constant for the I_2 - Ag reaction determined from non-irradiated studies. Silver iodide is very largely stable to radiation at the dose rates used in this work.

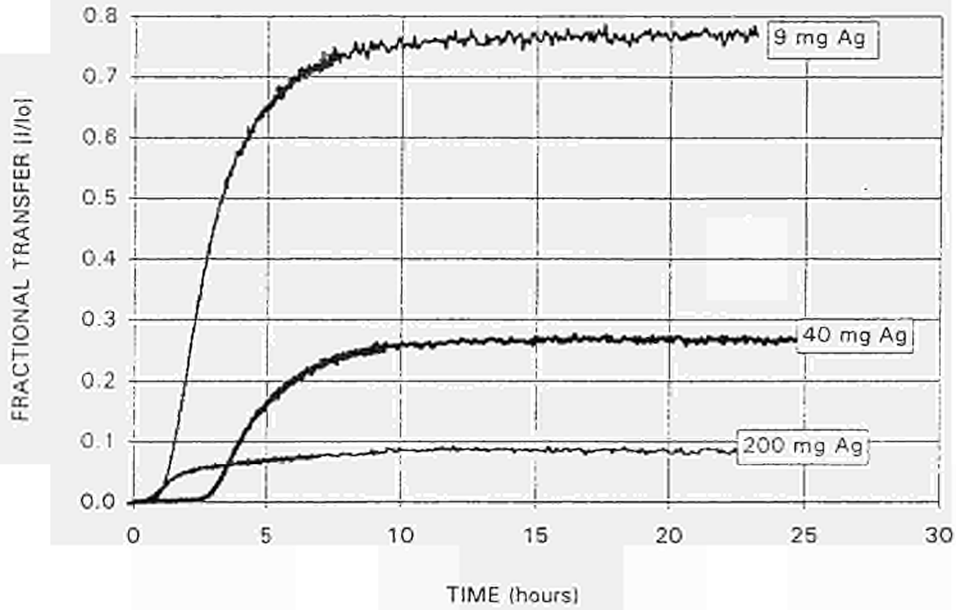


Figure 5: Effect of Ag on Iodine Volatility under Irradiation, pH 4.6

Task 1.3: The radiation stability of silver iodide formed by the reaction of I^- and I_2 with colloidal silver is being studied at elevated temperatures. Results obtained so far show that the product is largely stable at 60°C, particularly in the presence of excess Ag^+ ions (Table 1). Further experiments are planned at 90 and 110°C.

Initial species	$[Ag^+]$ (mg/l)	Test duration (h)	Initial pH ❶	Final pH	$\frac{A_{paint}}{A_{tot}}$ (%)
I_2	without	168	6.64	5.78	1.68
I_2	10^{-2}	168	6.65	6.03	0.33
I^-	without	168	6.21	5.92	0.19
I^-	10^{-2}	168	6.18	5.90	0.31

Note : ❶ the addition of colloidal silver into the iodine solutions of initial pH 4.6 leads to an increase in pH.

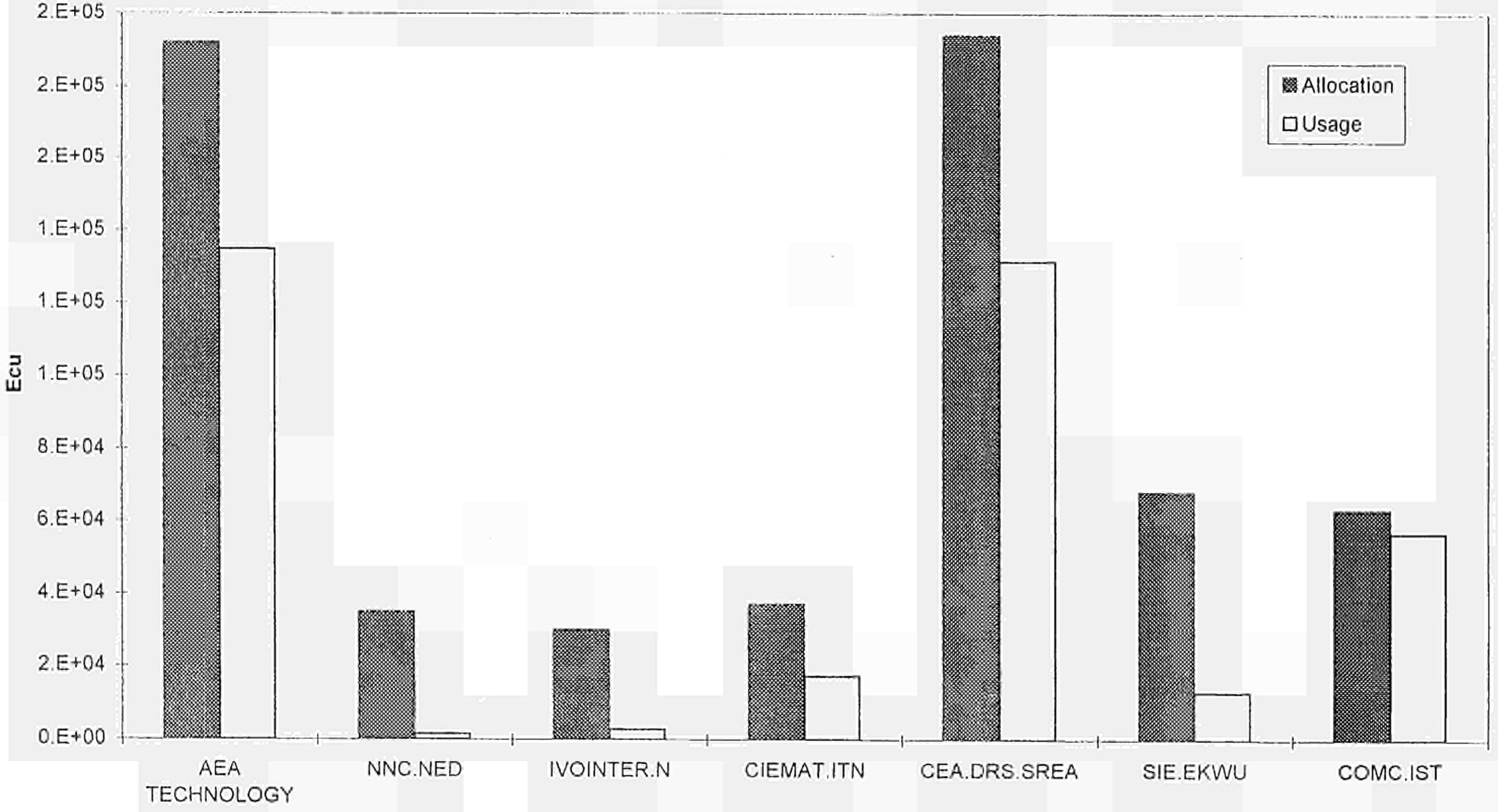
Table 1: Summary of Irradiated Tests with Silver Colloid

C.2 Assessment, Analysis and Model Development

Task 2.1: A literature assessment has been carried out to identify the areas in which existing data on iodine - silver reactions are sparse or contradictory. Based on this literature assessment, a set of research issues have been identified both for I - Ag reactions and AgI stability.

Task 2.2: Analysis of the kinetic data obtained in Task 1.1, together with data from previous experiments, is underway. A two-stage model for the reaction of iodide ions with silver particles has been developed, however the silver mesh used in the current tests appears to show different kinetics and the reasons for this are being investigated.

F14S-CT95-0005 Resource Usage to 31 December 1996



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B.3.2-2 Aerosol physics in containment - APC

Contract No:	FI4S-CT95-0016	**Duration:**	1 Jan.1996 - 31 Dec. 1997
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Partners:	**VTT Espoo/FI, IVO Vantaa/FI**		

A. OBJECTIVES AND SCOPE

In case of an hypothetical severe accident on a nuclear reactor, when the core melts, Fission Products (FP) are released, transported through the Reactor Coolant System (RCS) and they enter the containment building, mainly under aerosol form. In order to be able to estimate what would be the atmospheric release in case of a failure of this containment it is of primary importance to determine what are the characteristics of these FP aerosols.

The objective of the project is to improve the knowledge in this difficult part of aerosol physics, which is aerosol physics of multicomponent species, in a multicompartment, under 3 to 5 bar, at about 120°C with humidity ratios up to 100 % and with steam condensation occuring on the containment walls. The current data base on hygroscopic properties of FP aerosols will be implemented, existing models will be validated and new models will be developed as necessary. So, more accurate predictions of potential atmospheric release of FP aerosols should be obtained. Furthermore a better understanding of aerosol aspects in PHEBUS FP should also be achieved, through the findings of this project.

The work which will be carried out in this project to achieve the above objectives, consists of two complementary experimental programmes, the VICTORIA and the PITEAS experimental programme. They are complementary each other and will allow to establish a complete data set on the behaviour of hygroscopic/non-hygroscopic aerosols in well controlled conditions. For this the results of the AHMED and the early PITEAS tests will also be taken into consideration (all results given to European Partners in the previous Reinforced Cost Action). PITEAS tests carried out in this project give us data on diffusiophoretic deposition of aerosol which is a significant depletion phenomenon in the containment in case of an accident. The PHEBUS-FP test (results will be available within the PHEBUS project) give us data on realistic aerosol behaviour released from the reactor cooling system in single volume containment and VICTORIA tests complete this data set with aerosol behaviour (high and low aerosol concentrations and varying chemical composition to simulate real fission products) in multicompartment volumes.

B. WORK PROGRAMME

B.1. PITEAS Experimental Programme (Task 1)

The PITEAS programme consists of separate effect experiments to study diffusiophoresis of FP aerosols in conditions representative of an accident and will be carried out by CEA/IPSN.

B.2. VICTORIA Experimental Programme (Task 2)

The multicompartment experiments carried out by VTT and IVO will furnish data on a multicompartment geometry with possibly multicomponent aerosols and non-homogeneous thermal-hydraulic conditions in order to have a more global validation of the models included in the codes.

C. Progress of work and results obtained

Summary of main issues

During the 1996 year :

- the first PITEAS contractual experiment has been performed. Tree progress reports are issued : [1],[2] and [3],
- preliminary experiments have been performed in the Victoria facility and the first contractual experiment involving CsOH aerosol has been conduted. Two progress reports are issued : [4] and [5].

C.1 PITEAS experimental programme

1. PITEAS facility description

The experimental facility comprises (see Figure 1):

- a vessel which consists of a cylindrical shell with dished upper and lower heads with a total volume of 2.92 m3. The usual operating pressure ranges from 3.3 to 5 bar. The vessel includes 100 mm and 300 mm diameter penetrations necessary for instrumentation purpose. A vessel heating system using a thermofluid allows to maintain the PITEAS vessel at a given temperature.
- a vertical cylindrical condenser is fitted in the PITEAS vessel. The upper part is a condensing surface that can be thermically regulated by an organic liquid. The lower part, called « dry surface », is maintained at the vessel temperature (where no condensation should occur). Moreover the condenser contains a bottle to collect the water condensed on the upper part. All the external surfaces of this device are painted in order to simulate the internal surface of a real containment of nuclear reactors.

Concerning the classical thermal-hydraulic instrumentation, several thermocouples are positionned so as to measure the gas, the vessel's internal wall, and the condenser temperatures. The total pressure is measured at the top head of the vessel.

A new humidity measuring device has been mounted on the PITEAS vessel that permits to sample a given volume of the containment gas mixture and to condense the steam from the gas sample.Thus, it is possible to determine the steam partial pressure in the vessel and therefore the relative humidity.

The aerosol instrumentation which has been installed is composed of:

- an acoustic generator to produce CsI particles whose aerodynamic mass median diameter is about 2 µm.
- two sampling ports to measure the aerosol mass concentration with filters. Each filter is installed in between the PITEAS vessel and a 1 liter container. Before the sampling, this container is at the atmospheric pressure and reaches the PITEAS vessel pressure at the end of the sampling. After the test, the aerosol deposited on the filter are dissolved and analysed by atomic absorption spectrometry.
- one sampling port to determine the aerosol size distribution of the dry particles by mean of an Andersen Mark II cascade impactor. Because the PITEAS experiments are conducted at high temperature (120°C) and at high pressures (up to 4 bar) under steam atmosphere, a sampling system was built to allow the impactor to operate under these conditions.
- one sequential sedimentation sampling composed of 8 glass coupons allowing to evaluate the aerosol sedimentation in the vessel. After the experiment, the particles deposited on coupons are dissolved and analysed by atomic absorption spectrometry in order to determine the aerosol

deposition velocity. Moreover it is also planned to observe the deposited aerosol morphology by optical microscopy.

2. PITEAS test

The test protocol and the test parameters have been decided and have reported in the PITEAS experimental programme description [6]. The first diffusiophoresis test, PDICO1, was performed on December the 10th 1996. The experimental procedure was the following (see Figure 2 and 3):

- a preliminary phase, where starting from ambiant conditions, the PITEAS vessel and the internal condenser were heated up to 70°C to perform a first isothermal test. Then a second isothermal test was conducted at 120°C. In order to keep the PITEAS vessel at atmospheric pressure, the vessel was left open during this heating phase,
- a steam injection phase to obtain a relative humidity of about 65%,
- an aerosol injection phase in the containment : the CsI aerosol was injected until the total pressure in the vessel reached 3.9 bar. This injection phase lasted 1 hour. During this generation period, filter samplings were performed each 10 minutes, two cascade impactors sampling were also achieved. At the end of this phase initial conditions for the diffusiophoresis experiment itself were reached. So the condensing phase was started,
- the condensing phase consisted of cooling the upper part of the internal condenser down to 70°C. Condensed water was collected in the lower part of the condenser all along this phase. During this period filter samplings were performed each 5 minutes and three cascade impactors samplings were achieved. The analysis of the first thermal-hydraulic results shows that the condensation flowrate start from 0.6 g/s to 0 during the condensing phase.

The various samplings are currently being analysed and data reduction is in progress. The release of the final report for this test is scheduled on February 1997. The second diffusiophoresis test PDICO2 with a higher relative humidity (92%) is planned on March 1997.

C.2 VICTORIA experimental programme

1. VICTORIA facility description

Detailled facility description is presented in the report : Technical description of VICTORIA facility [7]. Here is presented only a short general description of the facility.

In these tests the vessel was divided into two compartments (upper and lower compartment). Thermohydraulic-data sampling system measures and records flow rates, pressures, temperatures and relative humidity (RH) values in the system. Temperatures are measured with T-type detectors (accuracy ~ 0.5 °C). RH-values are measured with several normal VAISALA humicap detector and with two (upper and lower compartment) new heated type VAISALA dewpoint detectors. In these new detectors RH-sampling head is temperature controled wich prevents water condensation into the detector head even when RH is over 100 %. RH can be however calculated based on the separate (near humidity sensor) temperature detector of the instrument.

2. VICTORIA test

The CsOH experiment (test number 58) is considered as the major test of this series and the relevant test conditions and test results are recalled in the this paragraphe. The relevant test conditions are :

- Aerosol generation from 0 to 79 min ; large generator (100 lpm air) ; exhaust open
- Vessel not preheated

- Exhaust closed at 84 min
- Steam feed 3 g/s from 84 to 105 min
- Exhaust open at 174 min (pressure difference 0 mbar)
- Impactor samples started at the time points : 82, 106, 196 and 556 min (impactors inside vessel). Sampling times were 1, 1, 3 and 10 minutes and flow ~ 25 lpm.

Measured temperature and relative humidity (RH) values in the upper and lower compartments and pressure values are presented in the figures 4, 5 and 6 (New temperature controlled RH meters).

Particles contain water and aerosol wet mass concentration is larger than dry concentration. Sampling flow from the vessel was ~ 3 lpm. The aerosol concentration in the lower compartment decreases fast during the steam feed.

The average particle diameter increases during the steam feed (hygroscopic growth) and after the diameter decreases as a function of time because large particles are settling faster.

Impactor samples (Cs) were analysed using ICP-MS.

All the corresponding data have been reported in LWR Containment Aerosol-Experiments at VICTORIA facility-Data Report 1/96 [8].

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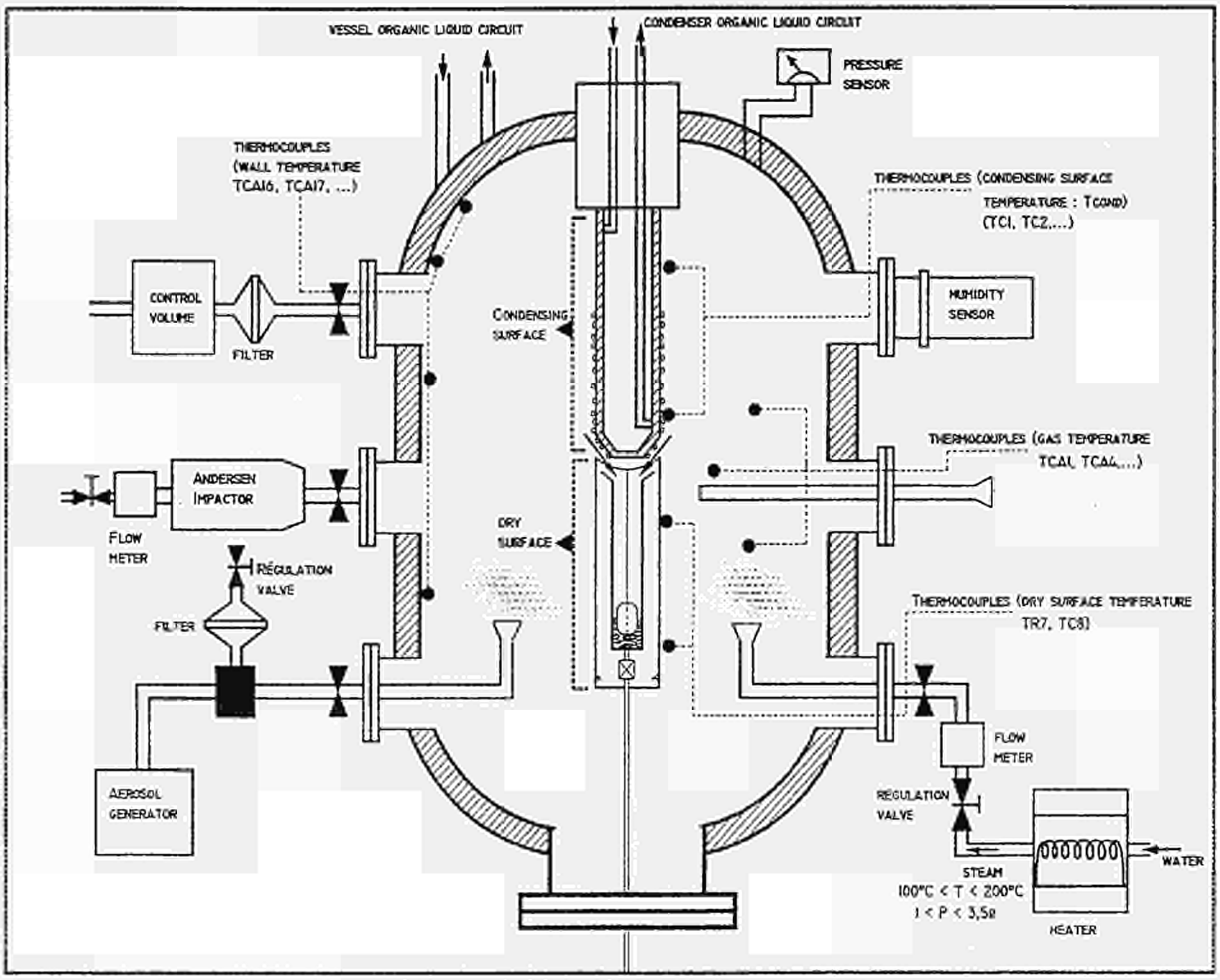

Figure 1: PITEAS experiment facility

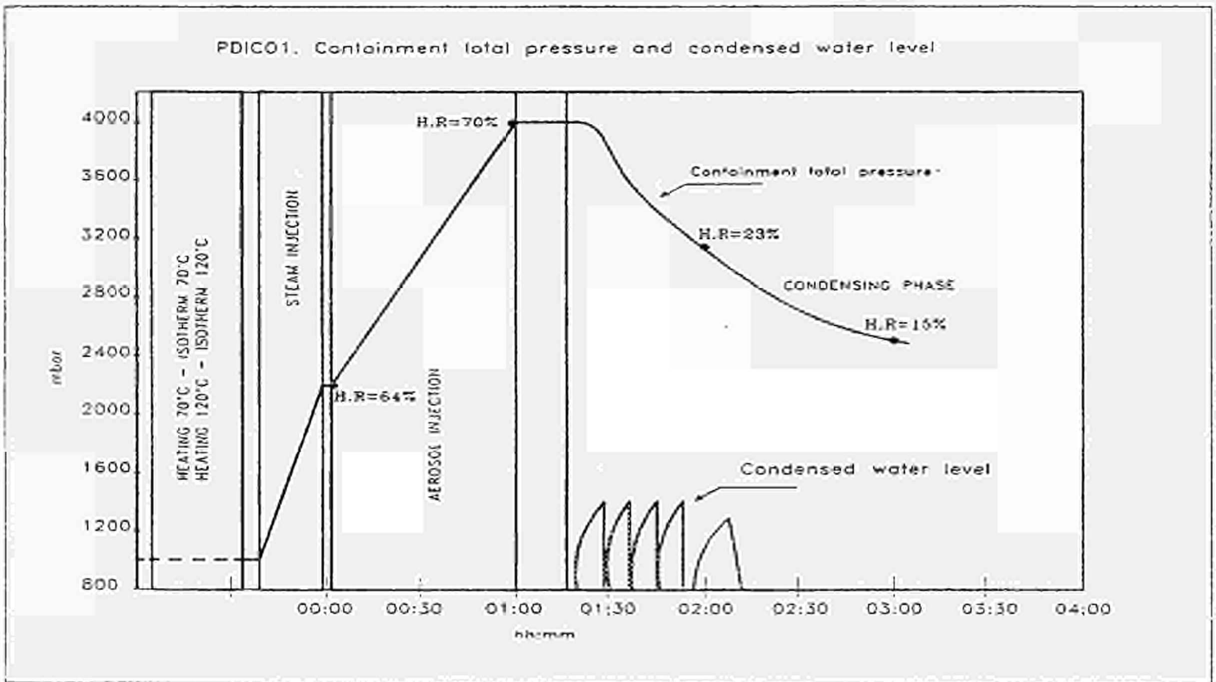

Figure 2: PITEAS total pressure and condensed water level

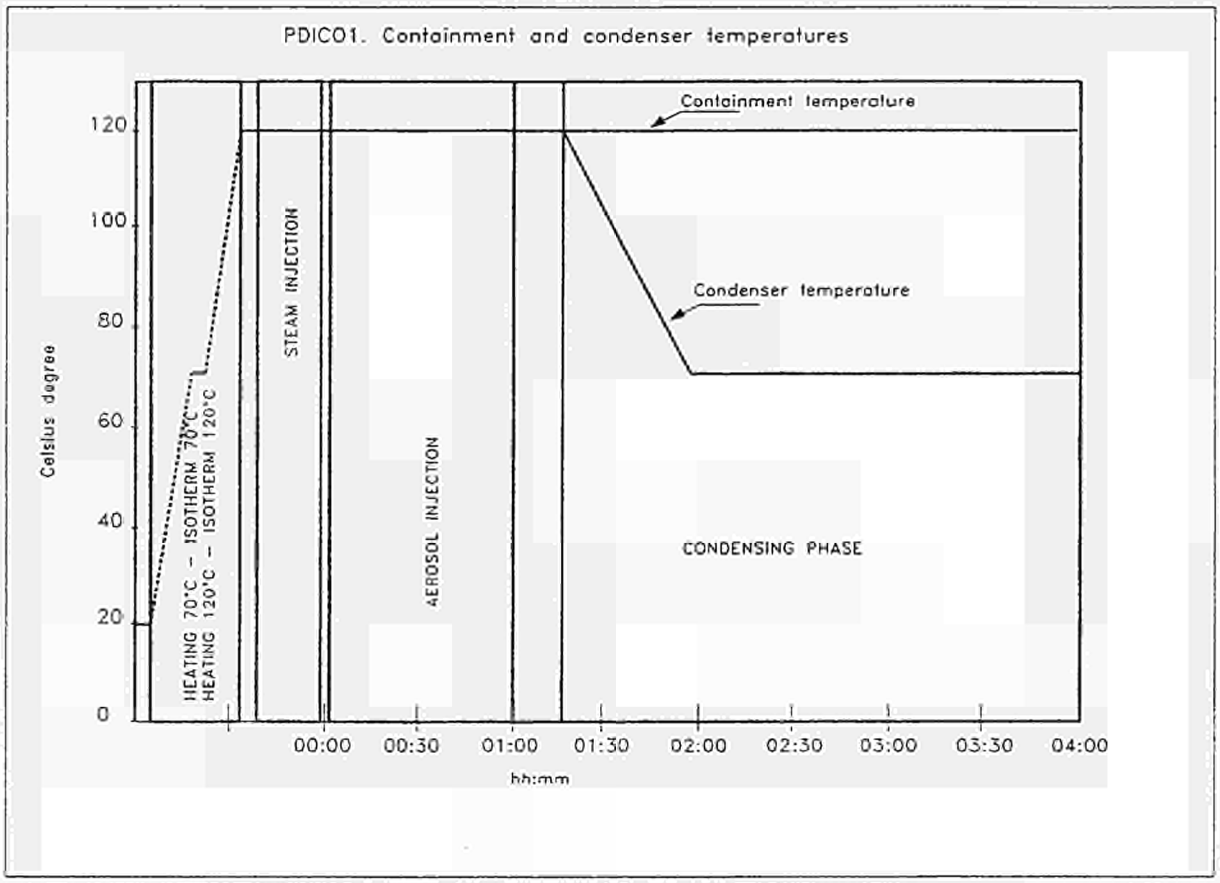

Figure 3: PITEAS containment and condenser temperatures

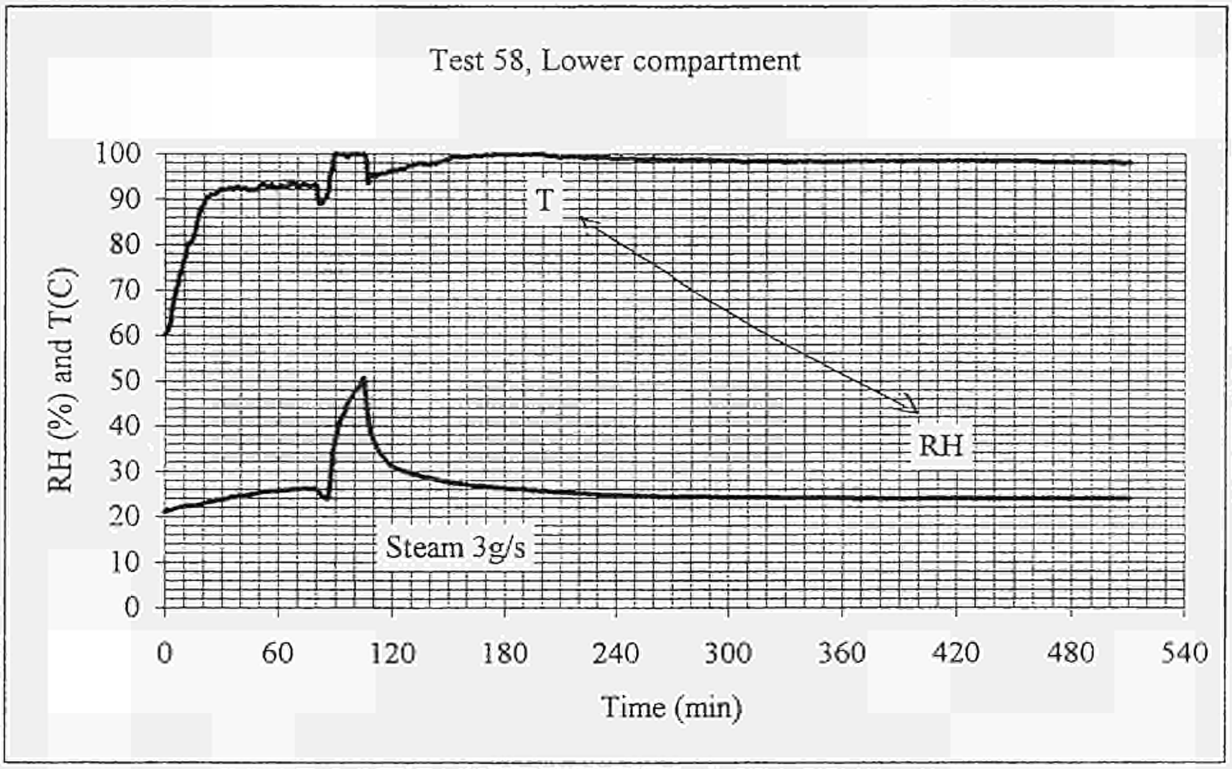

Figure 4: VICTORIA Lower compartment

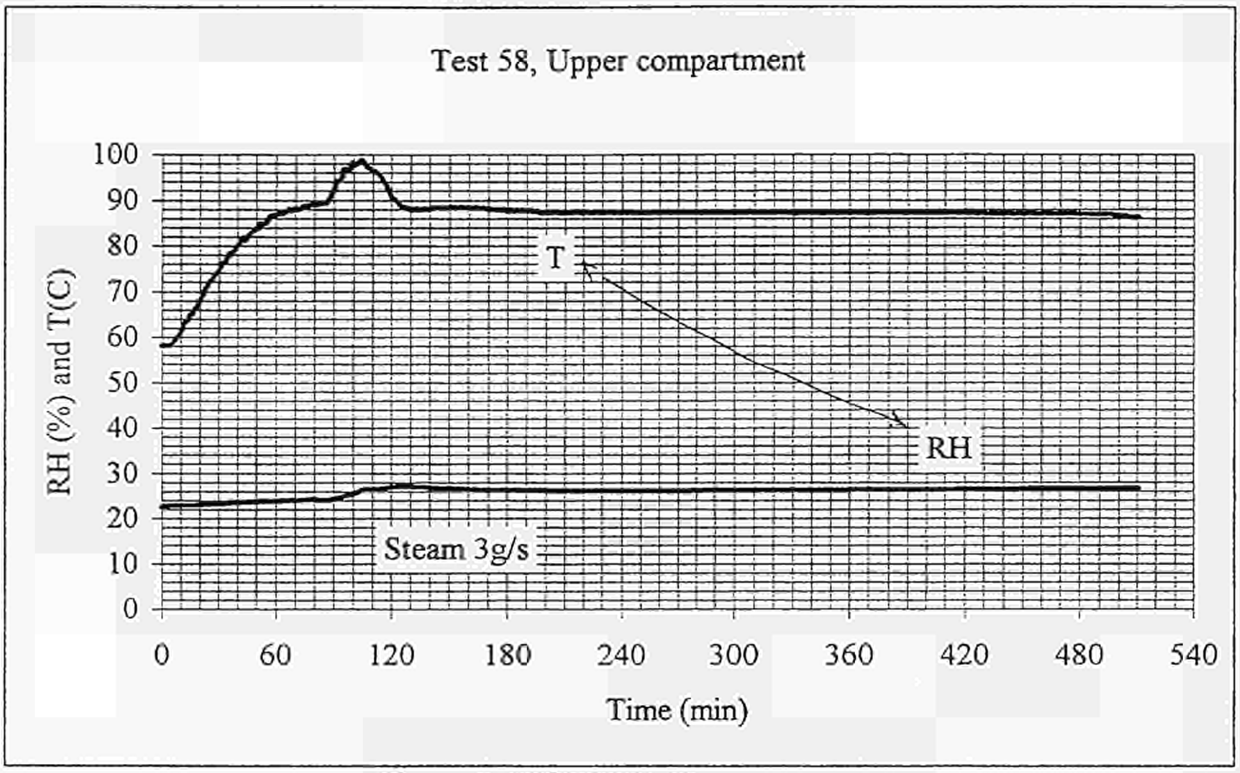

Figure 5: VICTORIA Upper compartment

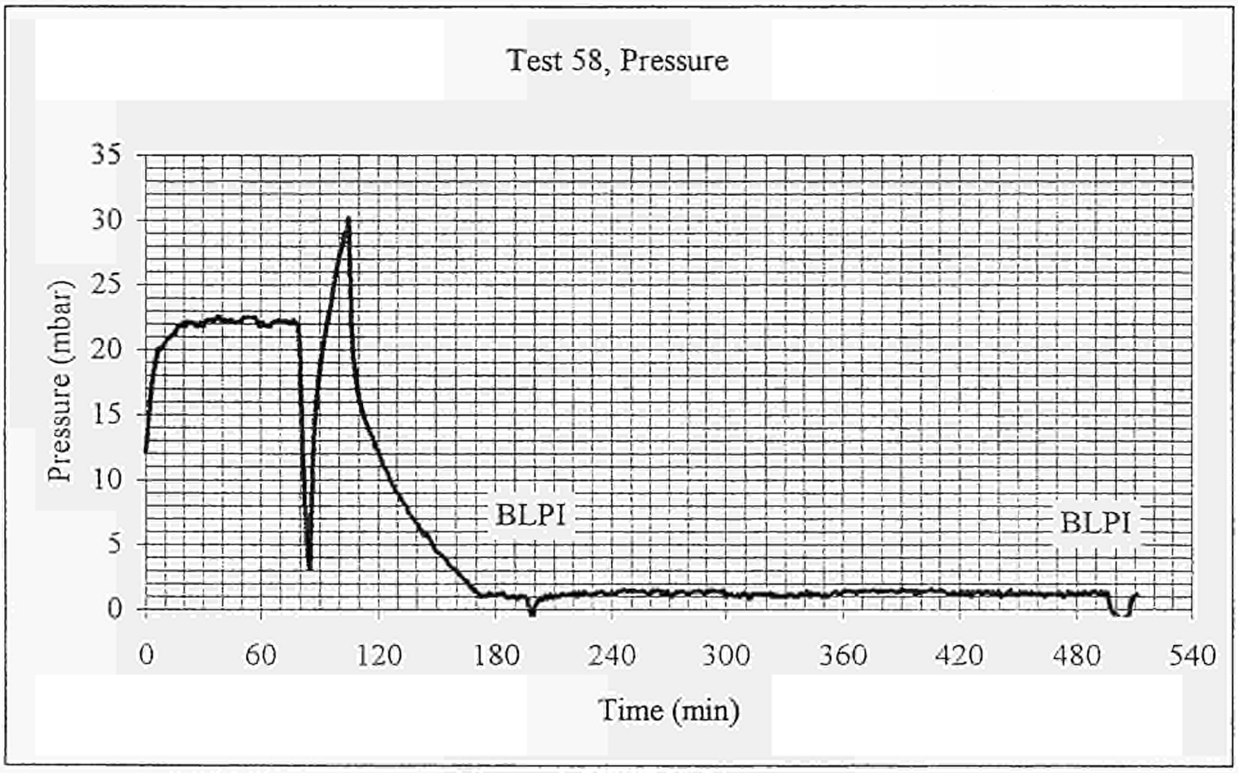

Figure 6: VICTORIA pressure

B.4.1-1 Multidimensional simulation of hydrogen distribution and turbulent combustion in severe accidents / H_2 distribution and combustion - HDC

Contract n°:	FI4S-CT95-0001	**Duration:** 1 Febr. 1996 - 31 Jan. 1999
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Partners:	**CEA-IPSN Saclay/FR, JRC-ISIS Ispra/EC, Univ. TUM Münich/DE, Siemens KWU München/DE**	

A. OBJECTIVES AND SCOPE

The proposed joint project aims at developing multidimensional, efficient, verified and commonly agreed physical and numerical models for the description of the governing physical processes in hydrogen distribution and combustion during severe PWR accidents. The main emphasis of the work will be on the prediction of accident consequences important for containment integrity, like pressure and temperature loads. The model development addresses also potential hydrogen mitigation techniques, like recombiners, ignitors, CO_2-inertization, and sprays.

The results will provide new advanced modeling techniques which can be used for containment simulation work with multidimensional field codes by the industry, safety authorities and research organizations in Europe. The proposed project will further result in improved understanding of hydrogen combustion loads and generate new load data.

The new physical and numerical models will also allow improved simulation of the effects of mitigation systems on the accident progression. It will become possible to better optimize mitigation systems on a mechanistic basis, like e.g. the position and distribution of spark igniters in a containment. In this way the operational safety of existing plants will be improved.

The present project will use CFD tools currently available to the project partners (GASFLOW, REACFLOW, FLUTAN, TONUS).

B. WORK PROGRAMME

B.1. Experiments on turbulent combustion and DDT (WP1)

FZK and TUM will conduct harmonized test series. Research in small scale facilities at Tum is focussed on understanding microscopic processes which determine the evolution of turbulent flames in complex geometries. At FZK, experiments investigate turbulent combustion and transition to detonation on a larger scale up to full reactor containment dimensions, to provide macroscopic results like pressure loads, flame position and burning velocities.

B.2. Modelling development (WP2)

Models will be developed especially for turbulent mixing and transport processes, heat transfer and mitigating systems. Key combustion problems to be treated are chemistry of H_2-air-steam flames and turbulent combustion for a range of flow regimes. The models will be evaluated in code systems existing at or developed by the partners organizations.

B.3. Model verification (WP3)

The models will be validated on commonly agreed experiments which include distribution without and with mitigation measures. Turbulent combustion experiments on different scales will be analyzed.

B.4. Model application for H_2 mitigation (WP4)

The new validated models will be used for analysis of distribution and combustion processes in future plants with the goal to optimize hydrogen mitigation techniques.

C. PROGRESS OF WORK AND RESULTS OBTAINED

The contract for this project started on Feburary 1, 1996. At the kick-off meeting and in two technical meetings at Karlsruhe and at Munich, the project gained significant momentum, and agreement on a more detailed working schedule was obtained. During the early phase of the project, priority is given to the experimental side which is essential in order to provide the data needed for model development and validation. A common test matrix for FZK and TUM experiments was agreed. Significant work was also performed on the modeling and validation fields. In addition, plant specific analysis was started.

Major achievements were the installation and/or upgrading of experimental facilities at TUM, FZK and Kurchatov Institute (RUT facility). Shakedown tests were performed and first experimental series were started. Two RUT experiments, the number agreed in the contract, were completed. Combustion parameters in simple geometries were measured on laboratory scale at TUM. Highly sophisticated techniques provide experimental data on various parameters which are important for modeling and validation.

Modeling efforts are underway for important phenomena including heat transfer, turbulent combustion for a wide parameter range, and numerical simulation of mitigating systems. A turbulent combustion model (Eddy Break-UP method) was implemented into the combined lumped-parameter and multidimensional code TONUS. The REACFLOW-code was extended by an axisymmetric solver, including axisymmetric version of the k-ε equations, and the Eddy-dissipation model for turbulent combustion was implemented. A new version of the GASFLOW code is being prepared by merging the FZK version with a version from LANL. Validation was continued by analysis of Battelle recombiner and igniter tests. The 3-D version of the COM code was applied to vortex problems and analysis of RUT experiments in order to evaluate the empirical model constant C_f.

C.1 Experiments on Turbulent Combustion and DDT

At FZK, a rectangular test tube was designed and constructed for visual observation of turbulent H2-air combustion around flow obstacles. After initial tests, the length of the tube was increased to avoid end wall reflection in the actual test section. A test series has been performed in the 12 m tube with different initial pressures, H2-concentration and obstacle configurations. The tube was then modified to allow inert tests in the shock-tube mode. This will allow to resolve and decouple turbulence and combustion models in two steps, firstly turbulence alone, and secondly, turbulence with combustion.

In the RUT facility, two large scale tests have been performed with H2-air-steam mixtures which resulted in a slow turbulent and in a fast turbulent deflagration. These tests allow upscaling to reactor scale.

At TUM, the experimental programme consists of detonation tube experiments with a sophisticated instrumentation to measure parameters like local flow velocities, flow fluctuations, chemical reaction rates, flame structure and velocity. In two detonation tubes („PHD-tube“, round ∅ 66 mm, length 6 m; „Glastube“, rectangular 60 mm x 100 mm, length 6 m) a DDT will be provoked by strong turbulent flame acceleration through periodically appearing obstacles, causing an unstable feed back mechanism between flame front and precursor shock front. In order to visualize the flame front in the tubes, they are examined with LDV, resulting in data of flow velocities and flow fluctuations, LIPF, showing the extension and shape of the reaction zone and high-speed Schlieren photography visualizing a global view of the flame and its shock system. The flame velocity is measured with UV-sensitive photodiodes, and light barriers along the tube axis. The pressure is recorded with several piezo-capazitive pressure transducers.

Experiments have been carried out successfully in measuring the flame acceleration shortly after ignition with high-speed Schlieren imaging in the „Glastube“.

C.2 Model development

Significant progress has been made in model development. These models have partly been implemented into the codes TONUS (IPSN), GASFLOW and COM 3d (FZK) and REACFLOW (ISPRA). Main topics in distribution phenomena were improvements of turbulence and heat transfer model systems and modeling of mitigation systems. Analysis of recombiner behavior and feedback on containment thermal-hydraulics is now possible with the GASFLOW code.

Modelling of fast turbulent combustion processes is in good progress. At IPSN, the standard Eddy Break Up-method was chosen which was coupled with a weakly compressible flow model, which should give acceptable results for a great variety of combustion regimes.

At FZK, the COM-Code was extended by two reaction kinetics models for different combustion regimes, namely an Eddy dissipation model and an Arrhenius model. The first is used when the chemical time scale is smaller than the turbulent time scale and the second in the opposite case. One global chemical reaction is used to describe the reaction progress.

In the equations for fast turbulent combustion models used by FZK, IPSN and JRC, a rate constant appears. Different approaches were chosen by the partners to determine this rate constant. A closer cooperation on specialist level was agreed in order to identify the most suitable approach for the range of combustion regimes to be covered.

C.3 Model verification

Work was started at FZK for validation of hydrogen distribution without and with mitigation. The Battelle test RX4 for the impact of sump vaporization on the natural convection process has been successfully analyzed with GASFLOW.

Confirmatory analysis of containment thermal-hydraulics was made for PHEBUS test FPT1. Battelle test MC3 with NIS recombiners was evaluated with the recombiner makro model, and analysis of test GX4 with smaller Siemens recombiners is underway. For the validation of the reaction kinetics models, FZK-tube experiments and large RUT-tests were analyzed. The calculations done so far did not result in a fully coherent set of rate constants. Additional detailed simulations are currently underway to resolve this question.

At IPSN, numerical tests have been done to check the feasability of calculations based on the weakly compressible reaction flow modeling in TONUS. For the case chosen, which consisted of a combustion process in a two room arrangement with well mixed atmosphere with 7.5 % hydrogen content, no major difference was found between laminar and turbulent combustion calculations.

Further validation of the various models will be performed by benchmark calculations, which are based on tests in the 12 m tube (FZK) and in the RUT facility.

C.4 MODEL APPLICATION FOR H2-MITIGATION

Analysis with the GASFLOW-Code for a containment design of the EPR assuming a dry hydrogen release from the IRWST was done with inclusion of igniters. A cyclic combustion removed hydrogen locally around the igniters when the concentration exceeded 4 % near the igniters. In the beginning, H2-concentrations were high enough in the IRWST for a fast combustion. In the long term, steady combustion was ongoing with continous removal of hydrogen. The simulation suggests that igniters can prevent the accumulation of larger hydrogen inventories in the containment.

B.4.1-2 Improved modelling of turbulent hydrogen combustion and catalytical recombination for hydrogen mitigation - HYMI

Contract n°:	FI4S-CT96-0017	**Duration:** 1 May 1996 - 30 April 1999	
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Partners:	**Univ. TUM Münich/DE, GRS Köln/DE, Univ. Politèc. Madrid/ES, BATTELLE Eschborn/DE, ANSALDO Genova/IT**		

A. OBJECTIVES AND SCOPE

The proposed project treats hydrogen combustion, initiated by deliberate ignition, and catalytical recombination of hydrogen and oxygen as two complementary mitigation measures to assess and control the hydrogen risk.

One objective of this project is, therefore, to better understand the complex process of turbulent combustion starting with slow deflagration flames turning into highly accelerating deflagration flames. DDT and detonation modes are not considered in this project.

The second objective of the programme focuses on catalytical recombiners and their response to transient feeding, feeding with inhomogeneous mixtures, and overfeeding with oxygen or hydrogen and thus addresses special effects, needing new computer models.

B. WORK PROGRAMME

B.1 Experiments and modelling of turbulent hydrogen combustion

Experiments are foreseen providing global data on turbulent flame propagation (flame shape, flame velocity and pressure history), with emphasis on the processes of molecular and turbulent heat and mass transfer, their interaction with chemical kinetics and the effects hereof on the effective turbulent burning velocity. The experimental programme will be carried out at Univ. Pisa and TUM and will cover the fields of slow deflagrations up to fast accelerated flames in order to fill a common data pool to be used by Univ. Pisa, GRS and UPM for modelling activities. Benchmark calculations are foreseen with the RALOC code. Further benchmarking of the RALOC code against the known integral code MELCOR on selected sequences.

B.2 Experiments and modelling of hydrogen catalytical recombiners

The work carried out on catalytic recombiners is aimed at investigating their interactions with the mixing phenomena in the containment atmosphere. Experiments will be performed in the 640-m^3 BATTELLE model containment with three recombiner units under various steam concentrations. Concerning the behaviour of catalytical recombiners under extreme conditions, GRS is going to improve models of the transient recombiner behaviour for CFD codes. The ANSALDO contribution will be focused on the development and assessment of a simulation methodology of mixing phenomena in a containment atmosphere in presence of catalytic recombiners, suitable for any multidimensional multiphase Eulerian code.

C. Progress of work and results obtained
Summary of main issues

The project started in May 96, during the 1996 many experimental efforts, in the field of hydrogen turbulent combustion, are devoted to the construction of the LargeVIEW and PuFlaG facilities.

The original LargeVIEW facility was damaged during a test in may and during the '96 a new, but geometrical equal, facility was constructed with more instrumentations.

The UPM, with the cooperation of GRS, start to use the RALOC code to be able in the future to make comparison between RALOC and MELCOR codes for sequences, hydrogen related, in real scale.

In the field of hydrogen catalytic recombination, the BATTELLE runs the experiments in the Battelle Model Containment; the analysis and the experimental data report are under publications. In the some time the GRS start to develop a recombiner model for FLOW-3D code wile the ANSALDO and UNIPI set up a procedure to implement a simplified recombiner model into a eulerian containment code.

C.1 Experiments and modelling of turbulent hydrogen combustion:

1. UNIPI

During the 1996 the work that has been carried out has been essentially devoted to the design and construction of a new test facility due to the fact that the former one was damaged during a test performed last May. The new apparatus, which has the same geometry and volume as the precedent one (Figure 1), has been notably reformed in the resistance (maximum pressure design). New instrumentation ensure a better knowledge of the distribution of the hydrogen concentration inside the test facility before the test.

For this peculiar aspect specific tests have been executed in other facilities in order to choose the type of hydrogen sensor that suits best for the existing experimental conditions. Additionally the method of atmosphere mixing between the two rooms has been modified, installing an internal system of ricirculation with fans in the region of the diaphragm to generate a ricirculation flow.

Moreover 4 rupture disks have been installed in the first room of the apparatus, to increase notably the level of safety and to protect the LDV system of TUM. Some LDV tests have been carried out by TUM on sample of the used glass supplied from UNIPI to investigate the effectiveness of the LDV-system in relation of the thickness of the glass.

2. TUM

During the 1996 the new test facility PuFlaG (Pulsed FlameGenerator) has being constructed and installed in Munich.

In this closed cycle of tubes, with a inner diameter of 80 mm, single flame fronts propagate against a steady counterflow, which is generated by a fan. Turbulence intensity and turbulent length scales are varied by a selection of validated turbulence grids. Advanced optical measurement techniques (photodiodes, light barriers, Laser-Doppler-Velocimetry and Laser-Induced-Fluorescence) for flow- and flame diagnostics are applied to the PuFlaG facility in order to obtain an overall view and deep physical insight into the turbulent combustion process.

The test section, with optical access on four sides, was built for the application of the optical measurement devices mentioned above.

All the necessary infrastructure like gas supply and evacuation system was installed. The basic instrumentation (pressure transducer and thermocouples) was calibrated and set into operation. A flame detecting system, based on a series of UV-sensible photodiodes to determine the global flame speed was developed and installed at the test facility. Due to the extremely small output, especially for lean hydrogen mixtures, it was necessary to develop a high-gain amplifier to reach appropriate signal outputs for measurements.

Parallel to that a light barrier with a He-Ne-Laser was developed and mounted to the optical test section to detect the flamefront. These two systems allow redundant measurements of the flamespeed.

The data acquisition system consists of a high speed 16 channel analog to digital converter-card installed in a Personal Computer System. As this card also consists of 8 digital output channels it is used to control the magnetic valves of the gas supply and evacuating system.

A suitable software to control the valves, the analog to digital converter and to evaluate the measured data was developed.

Some test measurements in the range of 8% to 16% hydrogen in air with and without counterflow were performed. Therefore, a Schlieren-camera was adapted to the PuFlaG facility to visualise the flame front (Figure 2) and to calibrate the photodiode and light barrier system.

Parallel to these measurements first tests were made in order to prepare the LDV measurements at the L.VIEW facility in Pisa. Therefore a small scale model of L.VIEW was installed in Munich (Figure 3) and adapted with a Schlieren-camera. The model with a diameter of 80mm is divided into two parts by a plate with a center-hole (hole-diameter 16mm). Four windows allow optical access from all sides over a length of 200mm.

Because of the sudden reduction of the free cross section in the channel a suction of the unburned gas as well as of the flame itself through the hole (Figure 3a) takes place. In the shear layer of the free jet areas very high turbulent intensities are created, which leads to a release of highly reactive combustion radicals, and in combination with the unburned gas to a spherical explosion (Figure 3b). The pressure increase leads to a backflow through the hole and to a fast outburn of the first chamber (Figure 3c)

3. GRS

In according to the contract, the GRS has transferred the latest version of the RALOC code to UPM Madrid. The code is supplemented by a detailed manual and an example for easier use of its main features. The example has been prepared in a way that all features which may be needed to perform the comparison analysis RALOC-MELCOR are included. These are among others the activation of the combustion model and some specific parameters, injection of heat, steam, hydrogen and others at predefined locations, distribution of condensed water through the compartments of the containment and the set-up of heat sinks like walls and equipment. Up to date the use of RALOC at UPM has to be considered provisional as the necessary licence agreement between GRS and UPM has not yet been signed. As soon as UPM has acquired some experience in applying RALOC the comparison dataset to MELCOR can be set up.

4. UPM

Although U.P.M. must contribute to the Project with RALOC MOD4.0 calculations, it was decided to start working with RALOC MOD2.2 due to the unavailability of the software at the start of the project. The idea was to get preliminary insights in the code usage, as well as to obtain a draft input-deck for the plant. However, due to the old nature of the code version, the present calculations were not studied in depth.

The nodalization of an actual Spanish-PWR containment (Westinghouse design) for RALOC MOD2.2 has been already carried out. In the same way, a Station Blackout sequence calculations using MELCOR V1.8.3. and RALOC MOD2.2 codes have been performed for this type of containment.

Nodalization

The containment building is cylindrical-shaped with an upper dome. The composing material is concrete, with an inner liner of carbon steel, and a little intermediate air chamber. Concerning thermal-hydraulics, the initial conditions of the problem are:

internal atmosphere:		- external atmosphere:	
components	air	components	air
pressure	128000 Pa	pressure	100000 Pa
temperature	25 °C	temperature	20 °C
relative humidity	100 %	relative humidity	40 %

The nodalization consists of 49 control volumes (including external atmosphere), 100 flow paths and 128 heat structures. Due to the three existing cooling loops in the primary circuit, the containment has been divided into three sectors, by means of the 120° symmetry, each containing a cooling loop.

The initiating event consists in a Station Blackout with no availability of the diesel electric generators. It was considered that the High and Low Pressure Coolant Injection System (HPCI and LPCI) will not be operative, neither the Auxiliary Feedwater System (AFW). The steam line to the turbine is isolated, just after the scram takes place. The Main Feedwater System costs down during the first 40 seconds after the accident start, allowing some additional water injection to the steam generators. In such conditions, the core heatup produces a rising of the primary system pressure up to the pressuriser relief valve setpoint. A correct Pressure Open Relief Valve cycling behaviour during the whole transient is assumed. An analysis of the effects on the primary system was performed in UPM with MELCOR V1.8.2. in the framework of previous works. The major results were:

- The steam generators sink is lost after 6000 s, so pressure increases up to the pressuriser PORV setpoint. Then steam to the containment is released and the pressure in the primary system stabilises.
- Just a moment before 12000 s, all core levels become totally dry, increasing the temperature rapidly. The core undergoes a severe degradation, when the melting temperature is reached. Before this happens, a strong hydrogen release is produced, due to the oxidation of the cladding zircaloy.

The hydrogen and water sources to the containment used for the present study have been taken from this previous analysis.

C.2 Experiments and modelling of hydrogen catalytical recombiners

1. BATTELLE

Work performed: In July and August 1996 two multi-recombiner experiments had been successfully performed in the Battelle multi-compartment model containment (560 m^3). Three industrial catalytic hydrogen recombiners, designed by NIS and Siemens, were installed at different positions in the containment. The aim was to investigate the interactions between the recombiners and the hydrogen-steam-air containment atmosphere.

Results: In the first experiment, an initially almost homogeneous steam-air atmosphere (40 vol% steam, 0.1 MPa total pressure) was established, and hydrogen was released close to the containment bottom at rates of 12 and 24 m^3/h until a concentration of 4.5 vol% of hydrogen was reached.

Recombination in the NIS device started already at concentrations below 0.2 vol% of hydrogen. The two Siemens recombiners started at 1.8 vol% of hydrogen.

For the second experiment, a stratified steam-air atmosphere was prepared with 60 vol% steam in the dome and 20 vol% steam in the lower compartments of the model containment. Hydrogen was released at an elevated position (at a level of 5.7 m in the 9.8 m high volume). As in the first experiment, the hot recombiner off-gas led to temperature rise in the dome compartment. Next Steps will be the further evaluation of the experimental results, reporting, and preparation of the data for code validation.

2. GRS

The work performed during the 1996 was devoted to the development of a Recombiner Micro Model. In order to establish a micromodel for a catalytic recombiner a literature study was made to get latest information on model techniques and reaction schemes on surfaces. This study revealed that sophisticated reaction kinetics models are available from mainly USA and Germany. These models pay often full attention to the chemical effects but reduce the influence of gas dynamics (convection and diffusion) by choosing very simple flow patterns like a stagnation flow field in front of a catalytic surface.

Available surface reaction schemes with strongly different sub-reaction rates coupled to flows can provide numerical problems therefore it has been decided to increase complexity of reactions step by step. A flow channel of a SIEMENS type recombiner has been modelled using CFX-F3D from AEA and a simplified reaction scheme implemented for the catalytic active surface to test grid quality and general convergence behaviour.

3. ANSALDO and UNIPI

In order to satisfy the contractual objectives, the work is organized as follows:

-Subtask a) To choose the Eulerian code for containment analysis.

-Subtask b) The empirical recombiner model setup.

-Subtask c) A preliminary sensitivity analysis.

-Subtask d) The empirical model setup at subtask b) is implemented into the GOTHIC code - in progress.

-Subtask e) Nodalization studies with the recombiner model activated - in progress.

-Subtask f) Check of the optimum model against recombiner test data - in progress.

Status of the progress.

-Subtask a) The GOTHIC code (version 3.4) has been chosen as the Eulerian code for containment analysis.

-Subtask b) The hydrogen recombiner component, introduced into the GOTHIC version 3.4, has the following characteristics:

1) It has to be located always on a junction;

2) It converts a specified fraction of the hydrogen flowing through the junction to steam and removes a corresponding amount of oxygen from the flowing gas;

3) For the volume downstream of the recombiner, source/sink terms for the hydrogen, oxygen and steam components have been implemented by modifications to main solvers of GOTHIC. The GOTHIC input subroutines have been also modified in order that they can take into account the empirical correlations, developed by PISA University, between the hydrogen recombination rate in a catalytic recombiner and hydrogen molar density.

-Subtask c) In this subtask the NIS recombiner and the associated experimental data have been assumed as reference in order to setup the methodology, since they constitute the widest set of free information about hydrogen recombiners characteristic.

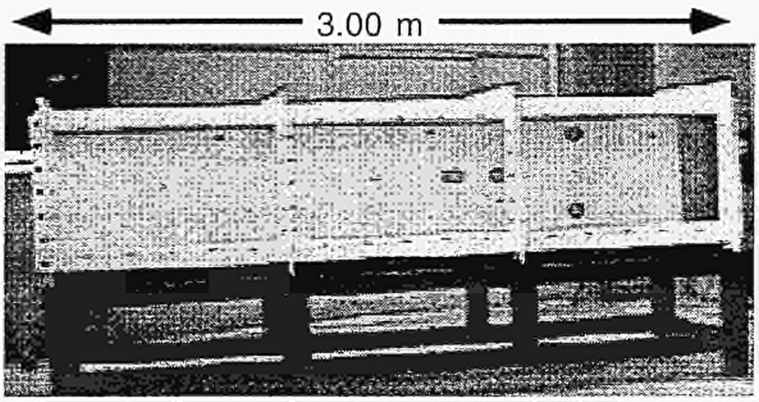


Figure 1: Picture of new LargeVIEW facility

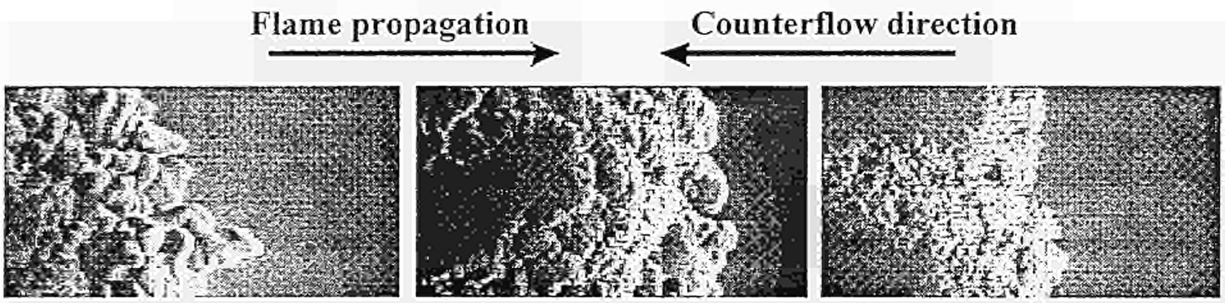
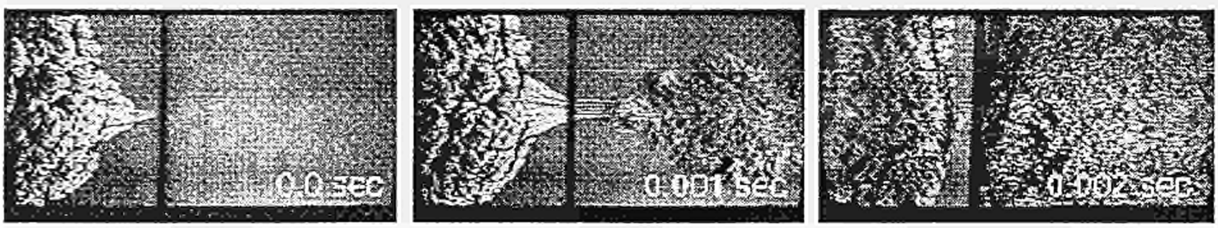


Figure 2: Schlieren-images of flames with different turbulence intensities of the counterflow in the new PuFlaG facility.



a) b) c)

Figure 3: Schlieren-images of a hydrogen-air flame in a small scale model in Munich of L.VIEW- in Pisa

B.4.2-1 INCON - Innovative containment cooling for double concrete containment

Contract n°:	FI4S-CT95-0011	**Duration:** 1 Jan. 1996 - 1 March 1998	
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A. OBJECTIVES AND SCOPE

The objective of this project is an exhaustive experimental and analytical programme to study the phenomenological aspects of innovative containment cooling for a rugged double concrete containment, which fully meets the European utility requirements (EUR). In particular, the desired solutions for the future generation plants have to be completely independent of any failure or operator error which could have caused a severe accident, should reduce the containment pressure, maintain a double barrier, be economical and compatible with the O&M and layout requirements. The containment cooling systems presently studied by the US and the European vendors do not meet completely all the above criteria.

The proposed solution involves natural circulating loops in the inside containment, in the outside containment, and across the two containments, whose phenomenological behaviour needs confirmation about both the stand-alone and the integrated functioning. There are two alternative solutions for the inner containment heat exchanger: one with a finned tube heat exchanger (HX), the other with a plate-type HX (water jackets with internal two phase reflux condensation). Two alternative solutions are also studied for the heat transfer mechanism to the environment. Both are based on external HX, immersed in a pool with natural draft cooling air.

B. WORK PROGRAMME

B.1 Experimental activities

The experimental activities of this project consist of :

- Functional and characterization tests of single and bundle finned tube heat exchangers in both natural and forced circulation conditions for different containment atmosphere composition and tube geometry (ENEL, PSI)
- Tests to assess the thermomechanical stresses of the plate type HX and its supporting structure under the most challenging conditions (ANSALDO)
- Intermediate loop tests to study its thermalhydraulic behaviour (ENEL)
- Integrated system tests to study the interaction among the different natural circulation loops and the system response time (ENEL)

B.2 Modelling activities

The analytical activities of this project consist of :

- Modelling of the finned tube tests ; assessment and development of heat transfer correlations (CIRTEN)
- Pre-test analyses using the GOTHIC and LEGO codes for the plate type HX experiments (ANSALDO)
- Pre and post-test analyses of the intermediate loop experiments using the RELAP5 code (EA)
- Develop and validate a model for the integral system experiments using the LEGO code (ANSALDO)

C Progress of work and results obtained

Summary of main issues

A good progress of the overall project has been accomplished in 1996 with an excellent contribution of all partners.

In particular the following tests (which have required activities of design, component procurement, test documentation, facility erection or modification, shakedown and tests matrix performance) have been completed:

- single tube tests at atmospheric pressure
- intermediate loop tests
- integral system tests

In addition the following analytical activities have been performed:

- the prediction of steam condensation on smooth and finned tubes, as interpretation of the data of single tubes at envinromental conditions
- the system computer code modellization with LEGO
- the intermediate loop simulation model with RELAP 5
- the model construction and the pre-tests of the plate-type inner heat exchanger

Also the design and procurement of the plate-type inner heat exchanger and of the small mock-ups of compact inner heat exchanger have been completed.

Progress and results

C.1 Experimental activities

The tests on the inner HX single tube at atmospheric pressure have been completed with the aims to investigate, at ENEL/CISE facilities, the overall heat transfer coefficient as function of the tubes slope and fin parameters (fin density for unit length, fin height). The drainage test is performed on a short section of a finned tube sample mounted inside a transparent tank to estimate the condensate bridging between adjacent fins; steam is supplied by electric heaters located on the bottom of the tank and the tube is cooled by water flowing inside.

Tests results show that the condensate drain and the build-up of noncondensables between the fins is not a concern for the fin density range used in the design (between 160 and 200 fins per meter).

The tests of the small mock-up (5 tubes)of the inner HX in forced circulation has been planed to be performed by ENEL at CISE in a dedicated new facility. The small mock-ups and facility component design and procurement have been completed. The test facility assembly is underway. Tests will start within January 1997.

The inner heat exchanger test with forced circulation will be performed at LINX facility at Paul Sherrer Institute. The new design for the next LINX-2 tests has been realised by PSI.The required equipment has been purchased. The modifications are in progress, and the LINX-2 facility will be ready in January 1997 to accomodate the inner heat exchanger mockup that will be provided by ENEL.

The intermediate loop test has been completed with the aim to analyze the thermal-hydraulic behaviour of the intermediate loop, including the internal side of both the inner and outer HX. More specifically, the main purposes are:

- to optimize the inner HX performances by changing tubes slope, recirculation pipes size and water inventory
- to verify the correct system behaviour during startup
- to evaluate the consequence of some degradation phenomena, due to the possible leakage of the downcomer (located inside the riser pipe) and to abnormal presence of air.

The test facility, located at CISE laboratories, is composed by scaled down models of the inner HX (3 electrically heated, full length tubes, and one recirculation pipe), by the outer HX (2 full length tubes, immersed in a water pool) and by the connecting pipes (riser and downcomer). Figure 1 represents the facility.
The performances are good, showing an internal HX tubes optimum slope and finding an optimum water inventory. The influence of the recirculation flow rate on the internal heat transfer coefficient in the internal HX tubes is found negligible. No particular problems or malfunctions result from startup tests. They evidence a good velocity in achieving full operational performance with no obscillating regimes and no instability phenomena.
The presence of non condensible gases in the intermediate loop affects the module performance, resulting in an increase of pressure and temperature of the internal containment atmosphere. However even very large amount of noncondensible gases does not stop the heat transfer mechanism.
The integral system test has been completed with the aim to verify the correct operation of the whole system, to analyze the interactions among different natural circulation loops (containment atmosphere, intermediate two phase fluid, pool water and external air flow after the pool water level decreases). The system response time and finally the air natural circulation start up have been investigated. The test facility is composed by the inner and outer HXs, connected to the pipes of the intermediate loop. The inner HX is placed in a vertical vessel, about 5 m3 volume, partially full of demineralized water. By means of electrical heaters, power up to about 90 kW is available to provide steam to the HX. The outer HX is placed in a full length/full height pool, filled at different levels with demineralized water. Cooling air in natural circulation was used; the draught is provided by a chimney 15 m high (500 mm internal diameter). The test facility is also equipped with a spray system. Both transient and steady state conditions test have basically confirmed the design assumptions and performances. Figure 2 shows the facility.
The distributed inner heat exchanger test the mock-up and its support structure has been designed, fabricated and delivered to the test facility by Ansaldo. The mock-up complete of internals with 250 cm height, 300 cm length, 12 cm depth outline dimensions and inner wall curved with 23 m radius, is a representative portion of the water jacket prototypical module. The mock-up is anchored inside a support structure (a welded boxed structure with removable front cover, filled with concrete at the rear, and bolted at the top and one side), designed to simulate as close as pratical the mechanical stresses induced by the rigid inner containment wall that limits its thermal expansion in the three directions (vertical, circumferential and radial outwards), during normal operation and accident conditions. Tests have been planed to be completed within March 1997.
The tests have the aim to:

- verify the water jacket feasibility,
- verify the tolerances achievable by good fabrication practice
- identify the relationship between imposed loads and measured stress levels.

C.2 Modelling activities

University of Pisa, on behalf of CIRTEN, performed the analysis of tests carried out at CISE laboratories on single tube at atmpospheric pressure, in relation to condensation on smooth and finned tubes in natural convection conditions.
The tests were modelled considering correlations in the available literature suitable for simulating the different heat transfer processes involved in the tests.
Heat transfer in the coolant side was predicted using the well known Sieder & Tate correlation [1] for both smooth and finned tube tests, with a correction for the entrance

effect following [2]. For heat transfer within the pipe walls a standard steady-state heat conduction relationship for cylindrical geometry was applied. The classical Nusselt theory for circular tubes [3] is used for heat transfer across the condensate film in the case of smooth tubes, with a correction by [4] accounting for the effect of tube axis inclination; for finned tubes, the thermal resistance due to the condensate film is accounted for only in the case of pure vapour tests, making use of a combination of the Beatty and Katz model [5] and of the Rudy & Webb [6] relationship for accounting the bridging effect.

The gaseous mixture side heat transfer is evaluated adopting the heat and mass transfer analogy. In the case of smooth tubes, classical natural convection relationships used for convective heat transfer and the analogy is applied in the form of the classical film model or as the more recent diffusion layer model by Peterson [7]. For finned tubes, a convective heat transfer correlation by Tsubouchi and Masuda [8] and the mentioned diffusion layer model are used.

The heat and mass transfer analogy is found to be suitable for efficiently correlating condensation heat transfer under natural convection conditions in the presence of noncondensable gas.

The results are generally good, with predictions within a 20% error band around the measured values.

A specific analytical activity related to inner HX modelling has been performed by ENEL/CISE on the basis of the test data obtained from small scale integral system test. As a matter of fact, many system tests have been performed with pure steam condensing outside the inner HX: this is a specially favourable situation to estimate the internal (boiling water) heat transfer coefficient of the inner HX itself, because the external heat transfer coefficient, due to pure steam condensation, is very high and can be calculated with good accuracy according to the classical Nusselt theory. Therefore, the external thermal resistance can be easily subtracted from the test data, providing a good evaluation of the internal coefficient.

The results have been compared with some literature correlations: a very good agreement (within a few percent) has been found using the well known Chen correlation for boiling heat tranfer.

A simulation model for the test facility of the two phase intermediate loop has been constructed by Empresarios Agrupados using RELAP5 code in order to analyse the behaviour of the subscaled system under different geometries, mass inventory and boundary conditions. Pre-test analyses has been performed to support the tests. Post-test analyses will be performed during 1997.

The innovative containment cooling system has been simulated by Ansaldo by means of the LEGO computer code library modules. The validation of the system model has been carried out the results of the small-scale tests at steady-state conditions. The computer outputs are in substantial agreement with the experimental results, but for the evaporation, which is slightly underestimated by the computer code.

The computer run with transient test parameters and the result comparison will be the next steps of the innovative containment cooling system model validation.

The validation completion is expected within February 1997.

Preliminary pretest analyses have been carried out by Ansaldo to provide inputs to the test definition and fabrication activities of the distributed inner heat exchanger. These analyses, carried out by means of the Gothic code coupled with the LEGO code, have highlighted the evolution of the containment atmosphere during the 3 days following the design basis event (large LOCA).

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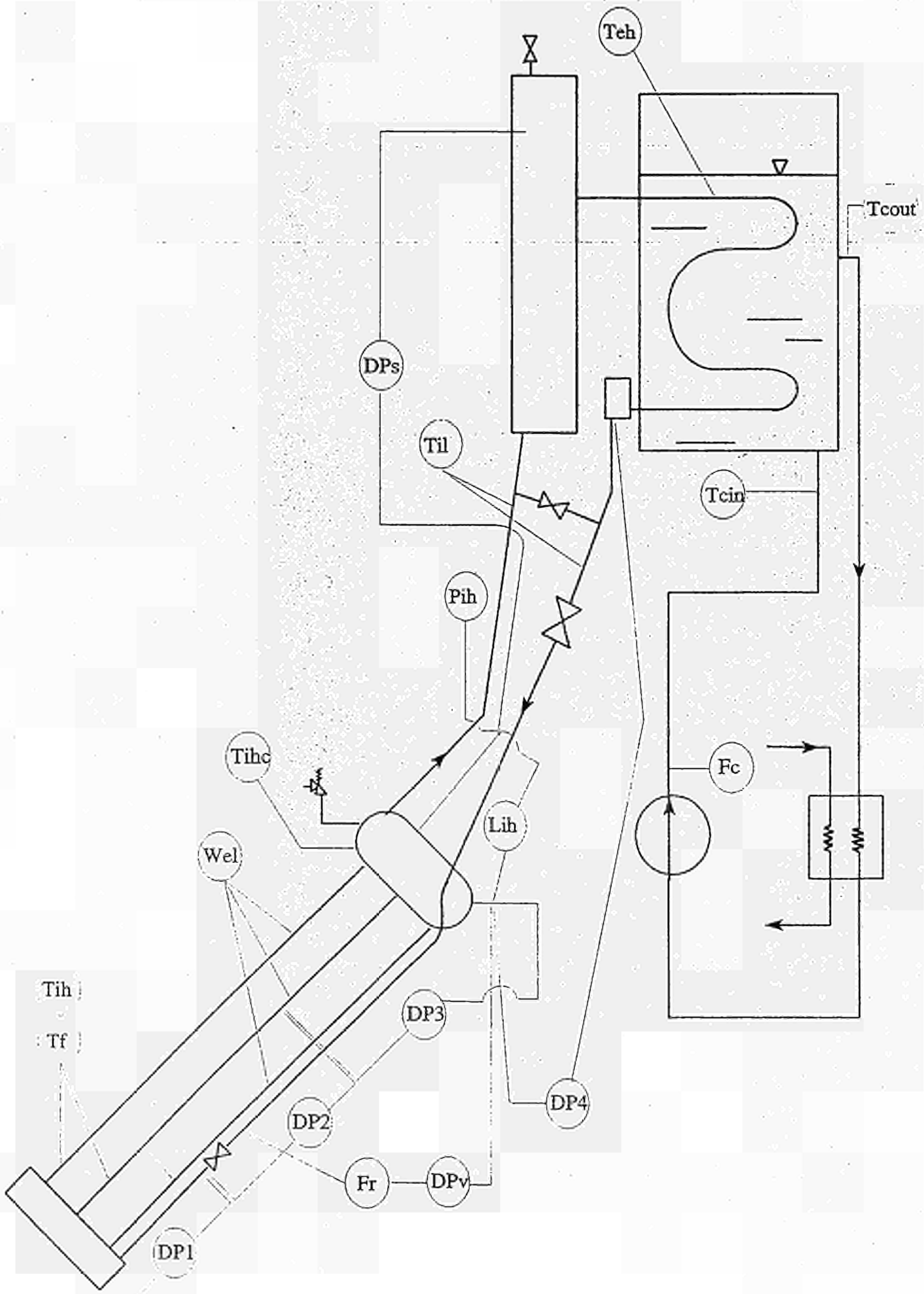

Figure 1: Intermediate Loop Test

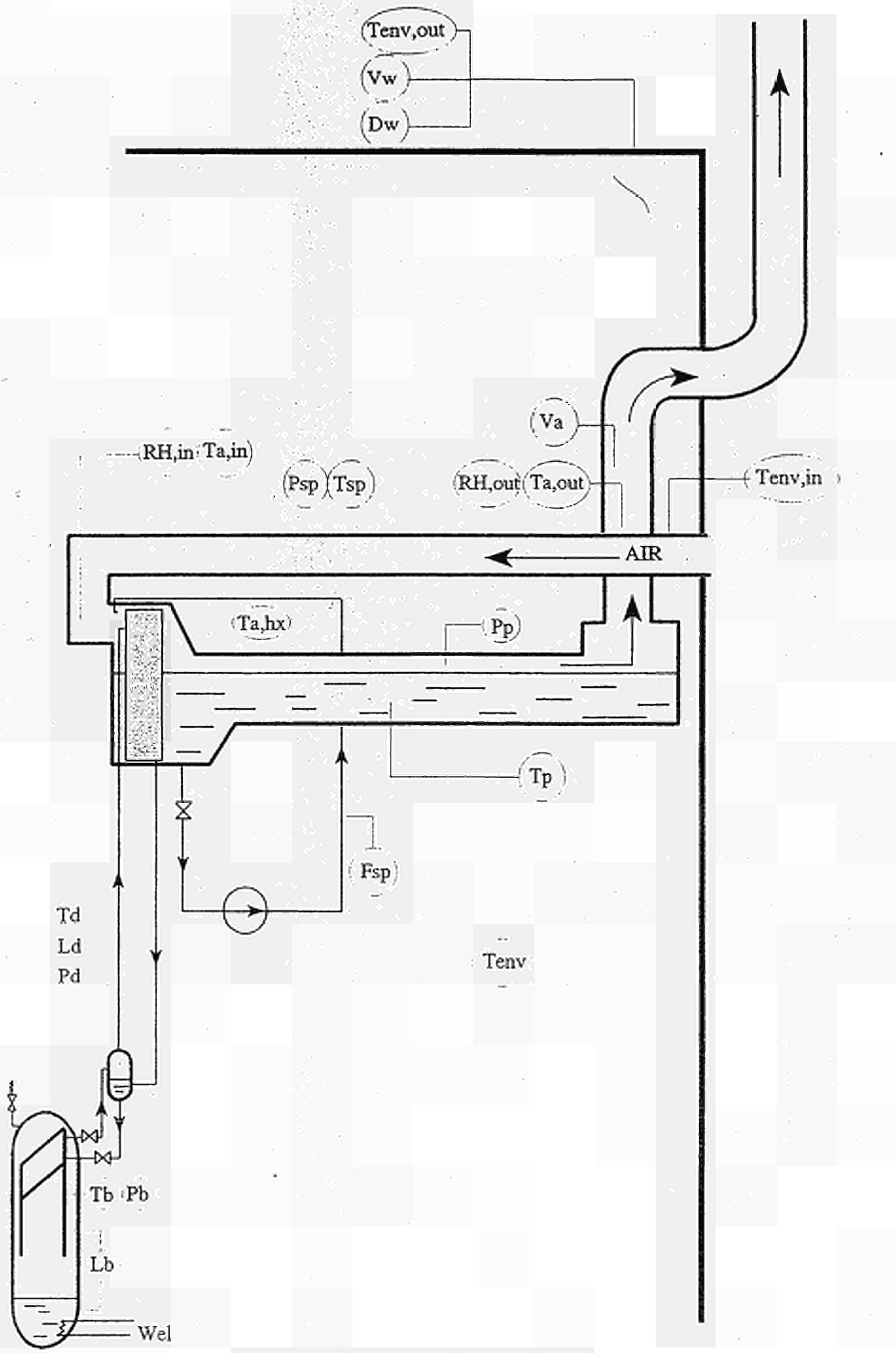

Fig.2: Integrated System Test

B.4.4-1 Benchmark on containment penetration sealing areas behaviour - ATHERMIP

Contract n°:	FI4S-CT95-0010	**Duration:**	1 Jan. 1996 - 1 April 1998

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A. OBJECTIVES AND SCOPE

The objective of the proposed project is to develop, and validate by tests, a theoretical approach and the related computer models to simulate the thermo-mechanical behaviour of the primary containment penetrations in which the leaktightness function depends on the performances of elastomeric material. This topic is particularly relevant because the requirements for next generation nuclear reactors, currently under discussion in Europe (EUR), require that the leaktightness of the containment be demonstrated for the severe accidents, and for the whole accident duration including the long term phase when the temperature and pressure decrease, and the elastomeric material mechanical characteristics are degraded by temperature and radiation loads. The modeling/validation activities are based on a full-scale test to be performed on a personnel airlock. The personnel airlock has been selected for the experimental activity in this programme because it is one of the most complex containment isolation devices from the thermal behaviour standpoint. In fact while the mechanical behaviour of the components (pressure and temperature loads) can be anticipated by finite element computer codes, the phenomena linked to the gasket leaktightness function are complex and depend on factors difficult to anticipate, as for example gasket elastic characteristics degradation due to radiological and thermal ageing, differential expansion of gasket material and steel, the synergy between temperature, radiation and pressure effects.

B. WORK PROGRAMME

B.1. Experimental activities

Small-scale tests allow only to evaluate separate effects; so only a full-scale test can provide realistic data to develop and validate theoretical models of containment penetrations, due to the gasket interaction between flange deformation and loss of elasticity induced by temperature and radiation. For this purpose a test facility has been accomplished around the personnel airlock of the abandoned Alto Lazio NPS. In this facility it is possible to simulate on the containment part of the airlock (i.e. external surface of the internal door and bulkhead) all the accident conditions in terms of temperature and pressure up to beyond severe accident conditions. The radiation effect is simulated by a temperature/time ageing performed according to a law coming from a complex experimental activity carried out on gasket samples. The facility is completely instrumented in order to get detailed thermo-mechanical maps of the whole component, to detect the displacement between door and bulkhead in the sealing area and to perform an accurate detection and measure of the leakages through the doors and through the gaskets of each door.

B.2. Modeling activities

These activities accomplish the main objective of the proposed project. It includes the computer model development and the validation by the experimental activities in order to have adequate tools to anticipate the response of any containment penetration sealing area with elastomeric gasket to any design accident conditions. It will provide inputs to component designers to evaluate future component modifications, if necessary.

In particular a computer model able to simulate the thermo-mechanical behaviour of the whole personnel airlock (doors, bulkheads, cylindrical part) considering all heat transfer mechanisms (conduction, convection and radiation) will be developed.

Furthermore a non-linear finite element code (FEA) will be employed and a gasket model will be developed to predict the behaviour of the elastomeric gasket between door and bulkhead during all the reference accident conditions (transient and steady mode).

To develop this model, ENEL makes available the results of a large experimental activity performed, out of the CEC ATHERMIP project, on gasket material samples in order to define the specific characteristics of the material (i.e. Poisson coefficient, Young module, thermal expansion, etc).

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The Test Plan has been issued on the last May.

About the Test Specification and Matrix, with reference to the project detailed schedule, two different Test Specifications/Matrix have to be issued for the two groups (TM and AC) of tests to be carried out. In particular, the TM Test Specification has been issued on December and a preliminary AC Test Specification has been prepared and it shall be updated based on the results coming from the SANDIA and ENEL experimental test on the simulation of the radiation ageing/damage

About the Test Procedures, each test will have its own procedure. At the moment, the Test Procedures for TM1, TM2 and TM3 tests have been issued.

With reference to the schedule included in the Technical Annex to the contract there was delay in the facility completion. It was mainly caused by technical problems and modifications decided in order to improve the facility flexibility and the accuracy of the main measures. Nevertheless the Technical Annex overall schedule is unchanged due to the good margins given during the planning phase for the experimental activity.

The thermo-mechanical model was developed and on the basis of the first results it is possible to conclude that the adopted models seem to be adequate for the foreseen analysis, the results of the calculations for the spherical head pressurized at 10 bars indicate that in the bulkhead reinforcement bars (with the supplied dimensions and geometry) the stresses may reach values well above the material yeld strength and the preliminary value of the gap in the gasket area (as calculated in the mentioned approximation) reaches values in the range 0.7 - 1 mm.

The model development activities have not yet been completed because the experimental activities on the gasket samples are more complex than foreseen and particular tests with very long duration have been added to have a complete knowledge of the gasket material characteristics. In this frame the law to reproduce the radiation ageing and damage by thermal parameters is going to be available.

C.1 Experimental activities (WP2)

The facility has been completed. In particular the test facility shakedown and preliminary tests have been completed during November, the Base configuration test (TM1) started on 4 December and it has been completed on 11 December and the facility preparation for Modification efficiency test (TM2) has been completed too.

During the TM1 test the Leak Detection System (LDS) has been successfully set up and the accuracy obtained make the next tests, particularly the AC tests, very interesting

C.2 Thermo-mechanical model (WP3.2)

This computer model able to simulate the thermo-mechanical behaviour of the whole personnel airlock (doors, bulkheads, cylindrical part) considering all heat transfer mechanisms (conduction, convection and radiation) was developed and will be validated by test results in particular to evaluate the internal and external sealing areas temperatures and the mechanical deformation of the inner door.

At present the calculations are performed by means of the ANSYS code, both by Empresarios Agrupados and CIRTEN; for the analyses carried on so far are considered the pneumatic test conditions , the pressurization of the spherical head (chamber A) up to 10 bars and the pressurization of the spherical head and cylindrical body (chambers A and B) at 10 bars.

Two main types of calculation models were foreseen and set up for the analysis of different parts of the structure: an overall model of the air-lock structure, to be used mainly to calculate the general stress distribution in the structure and the particular deformation/strain pattern at the interface between the cylindrical body and the bulkhead and a detailed model of the bulkhead/door structure, to be used to investigate in detail the relative displacements between the bulkheads and the doors.

C.3 Gasket model (WP3.4)

Two finite element models have been prepared with different purposes and scopes.

Model 1 is a 3-dimensional one covering about 120 mm on the closure areas and a quarter of the total area of the door. The zone of the housing of the rubber gaskets has been modelled in detail, but the gaskets themselves were not included. Eight-node SOLID type elements were used. The purpose of the model is to introduce calculated displacements into the nodes, for each load case and test, with the complete model of the test rig prepared by CIRTEN. This will enable us to verify the relative displacements and rotations between the bulkhead and the door of the closure area, and to select in which sections or parts they are greatest, where there is a risk of leaks through the sealing gaskets.

Model 2 is a 2-dimensional one, it is a detailed representation of a section of the bulkhead, the door and the rubber sealing gaskets. The basic objective of this model is to analyse the behaviour of the sealing gaskets for each section selected by Model 1.

A calculation process was developed, to represent the behaviour of the sealing gaskets inside their cavity, taking into account the non-linearity of the gasket material and the interaction between gaskets, gasket cavity and sealing surfaces

The behaviour of the gasket material is described using a hyper-elastic model. The model handles the material in terms of a deformation potential energy which defines the deformation stored in the material as a function of the deformation. The potential used in the simulation was the Mooney-Rivlin.

B.5-1 Benchmark exercise on expert judgment techniques in PSA level 2 - BE-EJT

Contract No: FI4S-CT95-0012 **Duration :** 1 Feb. 1996 - 30 Jan. 1997
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A. OBJECTIVES AND SCOPE

Probabilistic Safety Analysis (PSA) provides a systematic, coherent and self consistent basis for supporting decisions about the safety of Nuclear Power Plants. PSA addresses a wide spectrum of issues including the phenomenology and the management of severe accidents.

However, the complexity of the phenomena involved in severe accidents, the scarcity of data on component failures, human errors, etc., demand the use of engineering judgement at every level of the PSA. Experts from many different fields (fluid-dynamics, geology, seismology, structural analyses, economy, medicine, etc.) are involved for obviating to the lack of consolidate knowledge. Notwithstanding the methodological advancements of recent years, the PSA is strongly dependent on this expertise, in the form of Expert Judgment (EJ), and on its integration into the overall risk assessment process. Therefore the main objectives of this project are:

- The documentation of the different methods and techniques for the use of EJ actually adopted by European PSA practitioners.
- The benchmarking of the effectiveness of the different ascertained EJ approaches in increasing the consistency, completeness, repeatability, scrutability and legal defensibility of the PSA.
- The evaluation of the level of effort of the competing methodologies by means of cost-benefit analyses.

B. WORK PROGRAMME

B.1 Survey Phase

In this phase, the information concerning the different participants' approaches to EJ in PSA will be collected in a systematic way. Then a specific reference test from the JRC's FARO experimental facility will be selected to benchmark the different EJ approaches. A report containing the results of this phase will be produced.

B.2 Phase 1

The alternative approaches proposed will be benchmarked with reference to a parameter assessment of a phenomenological issue related to the selected FARO test. The following activities are planned :

- Identification of variables relevant to the experiment
- Application of EJ methodologies to the selected issue, and collection of results
- Evaluation of results against (i) pre-defined criteria and (ii) experimental data.
- Elaboration and publication of a report

B.3 Phase 2

The objective of the analysis is the evaluation of different EJ Techniques with respect to an APET (Accident Progression Event Tree) related issue, given a damage state for a nuclear power plant. Different methodologies for the solution of an issue related to the Level 2 PSA of an evolutive reactor will be benchmarked. The breakdown of activities will be similar to the one presented in B.2 above. A detailed description of these activities will be provided before starting this phase.

C. PROGRESS OF WORK AND RESULTS OBTAINED
SUMMARY OF MAIN ISSUES

During the year 1996 the Survey phase of the project was completed according to schedule and the work package B2 of the benchmark dedicated to a comparison and evaluation of Expert Judgement (EJ) techniques with reference to a parameter assessment problem was initiated. The forecasting of the result of JRC-ISIS FARO (Fuel melt And Release Oven) experimental facility having been selected as reference application. Some preliminary work has been performed on the definition and design of the second phase of the benchmark which is devoted to the comparison and evaluation of structured expert judgement techniques with reference to a scenario development problem (work package B3).

Material dealing with the national practices in the use of expert judgement techniques in PSA has been collected among the partners of the project and has been included in a survey report. A comprehensive set of comparison and evaluation criteria to be applied to the comparison of EJ methods has been worked out. To this end a collection of requirements and needs of PSA has been performed in order to set up a number of qualified characteristics as the basis of a comparison methodology which will be applied by a Peer review group composed of the members of the project.

C.1 Survey Phase

The aim of the Survey Phase is to collect, in a systematic way, all the information concerning the national participants' approaches to EJ in PSA, and to select the reference experiment for the phase 1 of the benchmark. The task has been accomplished on time and completed , as scheduled.

Contributions have been provided by the following partners of the BE-EJ: Enel for Italy, AVN (B), NNC for United Kingdom, STUK for Finland, GRS (D), FGUPM for Spain and HSK reflecting national or company practices. Table 1 summarizes the content of the contributions, limited to the PSA activities in the field of nuclear safety (NNC and GRS report also the use of formal expert elicitation in other nuclear areas, such as radioactive waste disposal studies). The individual contributions are collated in the corresponding report [1].

It should be noted that, when the use of EJ is shown in the table as "informal" (meaning that elicitation of experts is not performed with formalized techniques), the process is documented. In the case of the Italian entry, the documentation is available given the context in which expert judgement was used (ROAAM Risk Oriented Accident Analysis Methodology [2], which normally requires extensive documentation).

In addition, the Belgian contribution has identified several areas of PSAs performed for Belgian NPPs where informal expert judgement has been used, as well implicit (not declared, nor documented) as explicit (declared and documented).

As shown by the table, formal expert judgment has been employed in PSA by NNC, STUK-VTT and GRS based on original methodologies especially developed for PSA issues. In addition the large use of informal application of expert judgment in PSA has been identified, leaving room for a more extensive use of formal EJ approaches in PSA future applications.

During the kick-off meeting of the project [3] held on February at EC-JRC ISIS, located in Ispra, Italy, the main objectives and goals of the project were finalized and the project was initiated. In addition the new low pressure test at 0.5 MPa of FARO was selected as reference for the first phase of the benchmark. The experimental facility FARO was visited and the boundary and initial conditions of the tests at 2 MPa and at 0.5 MPa were presented to the participants, together with the results of a benchmark of calculations just terminated for the case at 2 MPa. The respective technical information describing the facility and the details of the tests were provided, as background material, to the participants in the project BE-EJTs by the ISIS researchers running and analyzing FARO experiments [4-8].

Table I: Use of EJ in PSAs or PSA-related activities by partners' organisations or in their respective country

Partner or Country	Italy	Belgium	UK	Finland	Spain	GRS	HSK
Use	Informal	Informal	Formal	Formal	Formal/Informal	Formal	Informal
Context	ROAAM	Rules (e.g.,screening)	ROAAM	HRA	Ranking PSA Data	Input parameters	APET
Scope	Level 2	Level 1/2	Level 2	Level 1	Nuclear Waste Management/ Level 1/2	Uncertainty Analyses	Level 2
Method	-	Documentation	NNC	VTT Bayesian update	Delphi/ Documentation	GRS	Documentation

C.2 Benchmark of EJ techniques on a parameter assessment issue

Sited at JRC-ISIS Ispra, the FARO plant is a large multi-purpose test facility in which nuclear reactor accidents can be simulated by out-of-pile experiments [9]. A large quantity of up to 150 Kg and more of oxide fuel ($UO_2/ZrO_2/Zr$), is melted up to more then 3000 °C in the FARO furnace, possibly mixed with metallic components, and delivered to the test section, a full scale water depth (up to 2 m), with high pressure and high temperature vessel (up to 10 MPa, 300 °C). Since 1990, a series of corium-melt water quenching experiments has begun in collaboration with USNRC, EPRI, and ENEL. The test situation simulates that of an in-vessel core meltdown accident when jets of molten corium penetrate into the lower plenum water pool, fragment and settle on the lower head. This issue suffers from a lack of data on the water quenching potential, the thermal loading and melt-structure interactions.

The objective of the test series is to determine, under a variety of conditions, the steam generation rate associated with the melt quenching, the thermal load on the bottom structures, and the debris configuration.

For the parameter assessment phase of the BE, the issue consists in the prevision of quantities relevant to the first experiment at 0.5 MPa. For the following list of parameters, the expert judgment teams were requested to give the 5th, 50th and 95th quantiles [10] (Note: hereafter pressure stands for relative pressure (DP= P-P0):

1. Peak pressure within the first three seconds of the experiment.
2. Time to maximum peak pressure (from time of melt entering into the water region), within the first three seconds of the experiment.
3. Pressure at 15 seconds.
4. Long term maximum pressure and related time.

5. Final percentage of debris fragmented (ratio of fragmented mass vs. the total mass of fragments and contiguous cake).
6. Final mean diameter of the fragments.

Optional
7 and 8. Water and steam average temperatures at 5 seconds.
9 and 10. Water and steam average temperatures at 15 seconds.
11 and 12. Long term water and steam average temperatures at the time of maximum pressure.

As an outcome of the survey phase, a number of structured expert judgement approaches to be benchmarked in the first phase, the one related to parameter assessment, have been reported. In the first phase, the NNC, STUK-VTT, GRS and JRC-ISIS new approach will directly contribute to the project with an expert judgement methodology. The following EJ methodologies were bechmarked in Phase 1 of the project:

- Type 0: Totally informal, no guidance, no specific expert training. In order to create a common understanding of the questions among the experts, a very brief explanation of the meaning of the three asked quantiles was given to all the domain experts during the kick off meeting. For each parameter, domain experts provided written individual answers for three points of the distributions they obtained using any methods they wished. The experts were asked to devote a limited effort to the task, i.e., one or two man days of work. All the 19 experts subsequently involved in the benchmark in the various panels provided type zero individual answers, without the use of any specific method for structured expert judgement. Actually 17 individual answers were collected since the NNC experts provided only a global answer since individual expert estimates were not possible within the NNC quality assurance rules. The type zero approach constitutes a minimum baseline for the benchmark and could provide a reference point for the numerical comparison of the estimates provided by structured approaches.
- Type 1: NNC (UK) methodology for Expert Judgement. It is applied by the NNC team to an internal panel of at least 3 experts/analysts. This approach [11], based on quality assurance methods of the sources of information and of the problem solving process, is based on individual estimates. It implies some interaction between experts from different disciplines, substantial recording, checking, review and revision.
- Type 2: GRS (D) methodology for Expert Judgement [12] . It is applied by GRS to a panel of three experts working in Germany. The method is based on extensive use of Fuel Coolant Interaction codes, and on sensitivity and uncertainty analysis of the code applications.
- Type 3: STUK-VTT (SF) methodology for Expert Judgement [13]. The method highly structured is based on the NUREG-1150 general framework with in addition the use of advanced Bayesian aggregation techniques. It is applied by the Finnish team to a national panel of four domain experts of Fuel Coolant Interaction phenomena.
- Type 4: Highly structured approach, NUREG-1150 type [14], including training of the experts, review discussions, individual elicitations, composition and aggregation of the opinions, review by experts. Experts are asked for three points of distributions. The approach is supported by JRC and is applied by Decision Insights from USA, working for JRC as consultant, to a multi-national panel of six domain experts provided by JRC.

- Type 5: JRC-ISIS Knowledge-based approach. A new approach to Expert Judgement based on Knowledge Engineering techniques [15]. The approach has been developed at JRC-ISIS in collaboration with Università di Brescia and Università di Bologna. The approach is applied by JRC to a multi-national panel of three experts.

Domain experts have been given a couple of weeks to provide individual level 0 estimates, i.e. unstructured answers. In addition the experts have provided personal Curriculum Vitae and have filled in the questionnaire explaining the methods employed for providing the level 0 estimates. All the material has been assembled in a draft report containing all the level 0 baseline estimates [16].

Results of the structured expert judgement approaches will be collected at the beginning of 1997, i.e. before the quick look reports of the FARO L24 reference test are circulated among the scientific community. The comparison and evaluation procedure will then be initiated.

The comparison criteria, having to take into account the specific requirements of the PSA, such as scrutability, defensibility of the results, etc., are derived from a number of documents [17-23]. A general framework for the assessment criteria has been produced within the project specifying the need for structured expert judgement within PSA and defining suitable quality characteristics for expert judgement in PSA. Quality characteristics have been divided into two groups, namely: internal characteristics (i.e. independent from the specific application of the methodology) and external characteristics (requiring the application of the methodology) [24]. Qualitative ratings procedure have then been designed for all the internal characteristics while quantitative metrics have been designed for rating external characteristics including the comparison of the EJ results with the FARO experimental ones. With this aim in mind proper statistical scoring rules and "surprises" indicators will be employed to count for example how many experimental physical variables fall outside of the 90 % confidence interval provided by the EJ methodologies. Table II summarizes the internal expert judgement quality characteristics that will be considered together with a brief description and the rating set that will be employed on the basis of the procedure presented in [25]. A "peer review group" composed by representatives of the CA participants, is foreseen for the implementation. The Peer Review group is supposed to terminate the comparison and evaluation of methods by autumn 1997. A final report will document the evaluation and comparison of the EJ methods as performed by the Peer review group.

C.2 <u>Benchmark of EJ techniques on a scenario development issue</u>

The objective of the analysis is the evaluation of different EJTs with respect to an APET (Accident Progression Event Tree) formulation, given a damage state for a nuclear power plant. Different methodologies for the construction of the event tree and dealing with the severe accident phenomena will be benchmarked. The discussion on the selection of the study case to be solved has been initiated since the kick-off meeting of the project. General requirements and background material to be supplied to the participants by a utility or a vendor to perform an APET study were discussed by the participants during the June 1996 progress meeting. Given the large effort devoted to Phase 1 of the project and in order to perform the Phase 2 within the time limits of the concerted action and the resources of the participants, it was decided during the June 1996 meeting to focus on a single significant issue related to a reference NPP to be selected. All participants expressed an interest in performing Phase 2 with reference to the Evolutive European Pressurized Reactor. Contacts have been made with Framatome, and Siemens in order to devote phase 2 to assessment of Hydrogen issue in an evolutive PWR design. The task will be officially initiated in spring of 1997 and will be terminated by the beginning of the year 1998.

Table II: Internal Quality Characteristics for Structured EJ approaches in PSA

Internal Characteristics	Description	Documentation required (a)	Ratings (b)
Applicability	Suitability to be applied to a sufficiently large set of cases (issues in the PSA field) without substantial modifications	Boundaries and conditions for application. Explicit evidence that the methodology can be applied in other EJ cases in the PSA field and even outside PSA field.	C1
Robustness	Capability to provide quality results even in presence of disturbing causes, such as bias, lack of co-operation, insufficient resources, time constraints, etc.	Tolerated faults, boundary of applicability. Explicit evidence that the methodology can be applied in presence of biasing factors of domain and normative experts, lack of cooperation and differently experienced normative experts	C2
Traceability	Capability to support explicit representation of the application in order to allow easy inspection for control, certification, diagnosis, and recovery	Assumptions, explanation of terminology, clear description of analysis. Formal or informal process generated by the application of the methodology.	C2
Justifiability	Capability to provide explicit justifications for the conclusions reached	Justification of assumptions. Understandable informal/formal justification of the conclusions reached, according to user background and expectations	C2
Incrementalism	Capability to support incremental or staged development through step-wise refinement, according to the resources actually used	Explicit evidence that the methodology can support a staged application, incrementally improving quantity and quality of results	C3
Knowledge integration-support	Capability to support integration of knowledge derived from different domain experts	Explicit evidence that the methodology can support acquisition, representation and integration of integrable knowledge and identification and assimilation of non- integrable knowledge derived from different sources.	C2
Uncertainty management support	Capability to support an explicit management of the uncertainty affecting domain knowledge	Procedure and method used to handle all kinds of uncertainty. Evidence that the methodology can support representation of uncertainty of the results etc.	C2

(a): The necessary documentation allowing evaluation of the characteristic

(b): Rating set C1: {poor, acceptable, good, excellent}; C2: {poor, acceptable, good, very good, excellent}, C3: {poor, good, excellent}

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B.5.1-1 Development of methodology for the evaluation of severe accident management strategies - SAMEM

Contract No: FI4S-CT95-0015 **Duration :** 1 Jan. 1996 - 31 Dec. 1998
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A. OBJECTIVES AND SCOPE

Accident management (AM) strategies with the potential to terminate or mitigate degraded core accidents are currently being developed and implemented at nuclear power plants worldwide. Decisions on their implementation are however, not straightforward as the actions may cause potential adverse effects and also involve phenomena that are not well understood. Current research emphasis has centred mainly on achieving a better understanding of the phenomena. Apart from the phenomenological issues, each accident management strategy also requires consideration of other key interrelated issues like operator actions, and equipment/instrumentation availability and performance.

The qualitative assessment and quantification of these thus entails a high degree of uncertainty and should be addressed in an integrated fashion. The objectives of this project are twofold. The first objective is to further develop integrated AM models for the assessment of the feasibility and effectiveness of potential severe accident management measures. The second objective is, by application in case studies, to contrast the unique features and to understand the limitations of these models such that they may be appropriately applied in relevant situations.

B. WORK PROGRAMME

B.1 Review of methods and case studies
Detailed review of different models and their recent applications in the assessment of the potential impact of severe accident management actions. Some examples of the models and applications to be reviewed are : ROAAM, Influence diagrams, event tree methods and human reliability assessment.

B.2 Formulation of Criteria
Due to differences in national requirements, different criteria have been adopted for the assessment and implementation of severe accident management measures. This task examines the basis and type of criteria adopted by the partners in this project.

B.3 Development of Integrated AM models
This task involves the development of integrated accident management models reviewed in B.1, for both preventive and mitigative measures, to consider the key issues such as severe accident phenomena, operator response, and systems and instrumentation availability.

B.4 Demonstration of methodology
On the basis of case studies agreed and specified by the partners (e.g. severe accident sequences, the phase of severe accident progression and the accident management actions to be evaluated), this task involves the demonstration of the methodology based on reference reactor designs (PWR, BWR, VVER).

B.5 Evaluation of results
An evaluation of the benefits and drawbacks of the different methodologies, as a result of the case studies, will be carried out. These case studies will provide a way to demonstrate the capability of the different methodologies.

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

Tasks B.1 and B.2 were completed during this period and two reports were issued (AMM-SAMEM(96)-P002 and AMM-SAMEM(96)-P003). A summary and the preliminary conclusions from these two studies are described next. The milestones and deliverables for this period were met. Tasks B.3 and B.4 are currently in progress and some aspects of the work are summarised in AMM-SAMEM(96)-M002.

C.1 Review of Methods and Case Studies

A basic understanding of the plant capabilities and limitations during severe accidents is normally achieved through a plant-specific probabilistic safety assessment (PSA) study. Invariably the PSA is further extended to examine engineering options to further enhance the plant capabilities in coping with severe accidents. Recent evaluations of several severe accident management (SAM) strategies have been conducted within the realm of a Level 2 PSA and using Level 2 PSA methods. The containment event tree (CET) approach and the phenomenological fault tree approach and their applications were reviewed by NNC and Vattenfall/Sydkraft respectively. It is however also recognised that, whilst the Level 2 PSA models provide the means of establishing the likelihood in the development of different accident scenarios and allow some representation of the uncertainties, the models are seen as inadequate when the resolution of severe accident issues is to be considered. A more fundamental methodology referred to as Risk Oriented Accident Analysis Methodology (ROAAM) has been developed to address specifically this aspect of resolution of severe accident issues. This ROAAM approach and some applications were reviewed by IVO. The necessity to consider a large number of SAM issues in Level 2 PSA decision tree models could potentially result in very large and complex structures. An influence diagram approach to provide more compact problem presentation and clearer representation of probabilistic dependencies of the different issues has been applied recently in the evaluation of a number of SAM strategies. This approach was reviewed by NNC.

The assessment of operator response is an essential component of a methodology to provide a realistic evaluation of SAM strategies. The human reliability analysis has either not been addressed or received only cursory treatment in some of the case studies reviewed. GRS provided a review of a model for human reliability analysis for preventative measures developed for the PSA for the French 900 MWe and 1300 MWe nuclear power plants. The data were specific to French plants and derived primarily from simulator training. The key elements of this methodology were reviewed and the potential for extending this methodology to other nuclear power plants and to SAM measures was examined.

Summary of Results

The methods referred to earlier are intended to provide essentially a systematic and structured framework for the appraisal of rare, high consequence events entailing complex physical processes. A summary of some key features is provided in Table 1. The varying emphasis on the different aspects of quantification reflects the intended application, for example, for the purpose of PSA, issue resolution and more recently severe accident management. As is illustrated in some of the case studies reviewed in the study, the methods can be applied in a complementary manner, depending on the objective and scope of a study. For example, in the US NUREG-1150 studies, the use of established PSA methods such as CET has highlighted the potential impact of a number of key severe accident issues and placed their relevance in the context of an integrated PSA model. Some of these issues, in particular the phenomena associated with early containment failure, entail a high degree of uncertainty. This has led to the application of ROAAM to allow the resolution of these issues in a more transparent and acceptable way.

More specifically, the treatment of phenomenological issues, as exemplified by the direct containment heating phenomenon, is also examined in the review. Overall, it can be concluded from this example, that the controlling physics has been consistently identified and considered, albeit being addressed in different ways and depth in the various approaches. It is however, difficult to provide further comparison on details of the key steps involved, such as for example the rationale for issue decomposition and probability representation. The studies also led to similar conclusions regarding the impact of direct containment heating on the containment integrity.

It is clear from the review of case studies the emphasis has been placed largely on the quantification of phenomenological issues. In some studies, although accident management actions and the related issues (eg. system availability, system recovery, equipment survivability) have been accommodated within the models, the treatment has either been cursory or taken in the form of simple sensitivity studies. The objective of the next phase of this project is to further develop these methods to allow a more realistic analytical approach to these issues.

An essential component of a methodology to provide a realistic evaluation of severe accident management strategies is the evaluation of operator response. No specific model for the quantification of human reliability of severe accident management has yet been developed. A model derived from the French 900 MWe and 1300 MWe PSAs has been reviewed. This model was developed relying largely on simulator studies. Some of the key features of the French model will be incorporated in the development of a methodology based on the HCR (human cognitive reliability) approach in the next phase of this project.

C.2 Formulation of Criteria

For nuclear operation certain safety goals imposed by the licensing authorities have to be fulfilled. A safety goal commonly adopted in various countries is that the activity releases to the environment must be small event in case of a severe accident. Except in cases with a very low probability acute fatalities from radiation and long term contamination of the ground around the plant is to be avoided. To fulfil the safety goal described above two criteria have to be satisfied:

- The core damage frequency is below a given limit.
- Given core damage, the frequency of large releases is sufficiently low.

The core damage frequency is determined in PSA level 1 studies and the frequency of containment failure and large fission product releases in PSA level 2 studies.

Implementation of Severe Accident Management (SAM) strategies is a major process in enabling the utilities to meet such criteria. In this study, a contrast is provided by the partners on their approach in the implementation of SAM strategies, given the differences between the national safety goals. The discussion provided by IVO, Vattenfall/Sydkraft and NNC considered three key aspects:

- the safety goals and their interpretation relevant to SAM implementation

- the approach to SAM implementation to fulfil the criteria

- the role of PSA and supporting analysis

The study provided by GRS deals more specifically with the criteria defined to initiate an action and is exemplified by the feed and bleed procedures adopted in the German PWRs.

<u>Summary of Results</u>

Criteria can be formulated for different levels concerning issues related to SAM. In three of the contributions the discussion centres on the use of criteria in defining the SAM strategy and in justifying the extent of implementation. The starting point is the definition of the national safety goals and additional definition provided by the utilities in the interpretation of the safety goals. The common goal is that the probability of containment failure causing large activity releases to the environment is low.

If the safety criteria are not met there are basically two possible reasons for this:

- A vulnerability in the plant has been identified
- The violation of criteria is related to conservatism in the analysis.

There are also cases where the present knowledge is not sufficient to decide which of these factors is the most important.

From the contributions mentioned above a number of observations can be made:

- In some cases, given the uncertainties in the analysis, an unambiguous demonstration in meeting the criteria may not always be possible, if the criteria are to be interpreted as absolute values.

- Fulfilment of criteria is related to a successful implementation of accident management strategies.

- Further development in order to reduce the uncertainties in the analysis whether the criteria are met or not is important. Results from PSA level 2 studies are sometimes difficult to use in the evaluation of criteria. There are different alternatives to improve the situation. The PSA method can be further developed and the uncertainties in input to the PSA studies can be reduced by further research. Another possibility is to use the ROAAM methodology more extensively.

The fourth contribution (from GRS) deals more specifically with the criteria adopted in the actual implementation of the bleed and feed procedures in a German PWR. These criteria concern the initiation and control of the effectiveness of the bleed and feed measures. In addition the use of simplified criteria for the assessment of the capability of the instruments is also considered. For the preventive phase of an accident, the evaluation was focused on sensors in the primary circuit and containment. It has been found that needed information is available either from direct or indirect measurement of the operational, safety or wide range plant instrumentation. If the fuel is approaching melting temperature, all thermocouples are assumed to fail. The information and related instrumentation to perform successfully the preventive Bleed and Feed Measures are thus available. As long as the temperature in the core stays well below the melting temperature, most of the instruments can provide the necessary information.

Table 1 Some Key Features of the Reviewed Methods

	Models			
	Containment Event Tree CET	Phenomenological Fault Tree PFT	ROAAM	Influence Diagram
Application	used directly in PSA, used in assessment of AM actions	used directly In PSA	designed for issue resolution, integrated ROAAM for plant SAM strategy development	used in assessment of AM actions, used as a complementary analytical tool to a PSA study
Issues Modelled				
Type	phenomenological, system recovery, AM actions, equipment survivability	phenomenological	phenomenological, AM actions, system availability	phenomenological, AM actions
Issue decomposition to a level corresponding to controlling physics	limited, can be improved by using decomposition event tree (DET) or PFT	yes	yes	limited
Time window of accident	extended to duration of accident sequence	narrow	narrow or extended	narrow
Uncertainty representation	normally by discrete probability, with upper and lower limits typically defined for sensitivity analysis	normally by discrete probability, with upper and lower limits typically defined for sensitivity analysis	splintering, decomposition, probability distributions	normally by discrete probability, with upper and lower limits typically defined for sensitivity analysis
Peer Review	more restricted in most commercial PSAs performed to date, USNRC funded NUREG-1150 study was extensively peer reviewed	more restricted in most commercial PSAs performed to date	extensive in order to achieve convergence in views amongst experts	limited in the case studies performed so far

B.5.2-1 Dosimetry and irradiation programmes of AMES european network - AMES

Contract No:	FI4S-CT95-0004	**Duration:**	1 Jan. 1996 - 30 June 1997
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Partners:	**JRC-IAM Petten/EC, ECN Petten/NL**		

A. OBJECTIVES AND SCOPE

The neutron exposture of a reactor pressure vessel (RPV) and its internals is one of the key factors that has to be quantified reliably when assessing the lifetime of such a component. Therefore the determination of a neutron exposure level for a RPV or any other component at a critical location should have at least the same level of accuracy as other limiting factors such as fracture potential based on known or projected mechanical properties.

Although great progress has been made in understanding the neutron induced degradation of the mechanical properties of RPV steels, many aspects of this are not fully understood yet. Therefore a European Network, Aging Materials Evaluation and Studies (AMES), was set up in 1993 to bring together the main institutions in Europe working in this field. The objectives of AMES are:

- to define and to carry out research projects;
- to act as a European Review Group;
- to provide technical advice to regulatory bodies and a basis for the development of common European standards;
- to participate in cooperations with the Eastern European Countries and with the Newly Independent States (CIS).

The main objective of this project is to support the activities of AMES by:

- establishing and organizing the dosimetry of the AMES projects;
- comparing the measured and calculated neutron fluxes at certain positions in the RPV of a Spanish nuclear power plant and in the High Flux Reactor (HFR) in Petten;
- specifying the irradiation programmes of the AMES 1 and AMES 2 projects.

The work to be done consists of:

- the establishment of the dosimetry and the irradiation programmes of the AMES1 and AMES2 projects;
- the creation of a neutron spectrum library to be used for the conversion of damage data obtained in a certain spectrum to data valid for a reference spectrum;
- the analysis fo the impact of the uncertainties of the neutron exposure parameters on the evaluation of the mechanical properties;
- a benchmark exercise. An intercomparison of results based on the same reaction rates and neutron spectrum valid for a reference location in a Spanish NPP.

B. WORK PROGRAMME

B.1. Dosimetry AMES1/AMES2

ECN is responsible for the recommendation of the dosimeters and Tecnatom will recommend the type of transport calculation method to be used in this project.

B.2. Irradiation AMES1/AMES2

The JRC is responsible for the establishment of the Irradiation Programmes.

B.3. Improvement and Harmonization

This task, including a benchmark exercise, will be performed by all partners jointly.

C. Progress of the work and results obtained

Summary of main issues

AMES Dosimetry project is intended to support the projects AMES 1 and AMES 2. The first one concerns the validation of surveillance practice and mitigation methods, the second one the effects of irradiation on reactor internals. Among the activities being carried out are:

- Making recommendations for the use of specific type of dosimeters in accordance with reactor irradiation conditions.
- Analyse the impact of the uncertainties of the neutron exposure parameters on the evaluation of the mechanical properties. Particularly, with respect to embrittlement trend curves.
- Review the dosimetry capabilities within the AMES member organisations together with the existing databases from previous work-programmes.
- Act as a link between AMES and the following dosimetry groups: The European Working Group on Reactor Dosimetry (EWGRD), the Working Group on Reactor Dosimetry for VVER reactor (WGRD-VVER), and the OECD Dosimetry Group.
- Design the irradiation programmes of the AMES projects setting up the irradiation conditions (temperature, neutron spectrum, fluence rate, fluence, specimen arrangement, etc).
- Optimise the irradiation programmes preventing duplication of efforts between AMES members. Analyse the quality of untested and irradiated material coming from previous national projects that may be an in kind contribution to the AMES projects.
- Give advice and make assessment with respect to the design of equipment for irradiation experiments and associated activities (handling, transport, etc.)

C.1 Dosimetry AMES1/AMES2.

The necessity has been recognized to use a very well calibrated dosimeter, or a set of dosimeters covering the full range of energies, in all AMES irradiations in order to reduce uncertainties that could affect neutron damage assessment. The make of this "AMES Common Reference Dosimeter" (AMES-CRD) may depend on the irradiation conditions. In order to develop the specification of the AMES-CRD dosimeter, a necessary first step is to collect information from the AMES members. A questionnaire for the item of recommendation of dosimeters was prepared by ECN and sent to relevant organizations (including Eastern countries organizations). Questionnaire is relevant for work programmes B.1 and B.3. After having evaluated the questionaire returned, ECN will prepare the specification of the AMES-CRD dosimeter.

There is some delay to reach the first milestone, that means, to establish the dosimetry of the projects AMES 1 and AMES 2, since it is necessary to collect first all the answers to the questionnaire. The consequences of this delay will be evaluated at the next project meeting in March 1997.

C.2 Irradiation AMES1/AMES2.

The document [1], now in second draft version, contains the main results of work programme B.2. Detailed information on LYRA facility and its potential use for AMES irradiations have been provided by JRC, and will be also taken into account in the final version of the report [1]. That information includes also results of new nuclear calculations with the Monte Carlo code MCNP 4A and a cross-section library based on the data file JEF2.2. For the AMES 2 project, the JRC has proposed to develop a new irradiation rig LIMA. Details were provided in annex 3 of document [2].

The AMES Reference Laboratory ECN-JRC has prepared a list of materials/specimens available in Petten for investigation (irradiation and testing). They may be used in the projects AMES 1 and AMES 2.

The document [1] discusses and proposes an irradiation programme for the project AMES 1. The programme emphasizes close cooperation between Eastern and Western European countries, and it is focussed on the Eastern countries since embrittlement issues are of great concern for VVER reactors.

The programme for AMES 1 is addressed to analyze the effects of irradiation temperature, neutron dose and spectrum, but different experimental techniques that require validation or improvement (reconstitution, fracture toughness testing of small specimens, micro indentation, etc.) may be introduced in the programme.

Relevant facilities for AMES irradiations are surveyed in the report [1]. Namely,

- Budapest Research Reactor
- HFR Petten
- Korpus RBT-6 Test Reactor (RIAR-Dimitrovgrad)
- LVR-15 Research Reactor (NRI-Rez)

The report [1] has been prepared with the collaboration of members of the Task Group AMES 1F. Three different irradiation programmes have been proposed by AEKI (Hungary), Kurchatov Institute (Russia) and the Nuclear Research Institute Rez (Czech Rep.) [3]. These irradiation programmes propose complementary irradiations among the experimental reactors mentioned above, optimizing in this way resources and enhancing collaboration between Eastern and Western European countries.

Section 6 of the document [1] gives complete information about the programmed irradiations in the HFR Petten reactor, using the LYRA irradiation facility, along the period of 1996-1999. The costs for irradiation in the different facilities are also mentioned, and the final section tries to outline and give recommendations for a final proposal.

It is considered that the second milestone, to establish irradiation programmes for the projects AMES 1 and AMES 2, has been reached.

C.3 Improvement and Harmonization.

The third milestone, the improvement and harmonization of dosimetry practices, is planned to be reached at the end of the project (June 1997).

The creation of a neutron spectrum library for conversion of damage data obtained in different spectra is in progress.

A procedure for analyzing samples (retrospective dosimetry) is being prepared by ECN.

A specific task on retrospective dosimetry has been planned by ECN for the Reactor Pressure Vessel Internals Project (part of the AMES 2 Project). This task involves activity measurements of ^{54}Mn, ^{58}Co, ^{60}Co and $^{93}Nb^{m}$. Since the amount of some of the target materials (Co and Nb) is not well specified, small amounts of component material have to be re-irradiated in a reference neutron field (e.g. HFR-Petten). The extra activity induced will be compared with well known amounts of material irradiated in the same field and will give information on the composition of the component material. To develop this procedure, the method will be tested using samples from irradiated steel specimens irradiated in the HFR which are monitored with activation monitor sets. Advantage of the use of these materials is that the irradiation conditions (irradiation time, change in fluence rate during irradiation, temperature) are recorded. This will exclude the extra problem of lack of information on these quantities for an actual internal of a NPP. The results of the retrospective method applied on these samples will be compared with the results of the available monitor data. This will lead to further elaboration of the method.

The data needed for the benchmark exercise between ECN and Tecnatom have been collected by Tecnatom and provided to ECN. The data included are:

- Activities of dosimeters samples of an in-vessel irradiation capsule removed recently from a Spanish reactor.
- Detailed information on the dosimeters used (geometry, sample mass, etc.).
- Theoretical neutron spectrum at the dosimeters position.
- Irradiation history

Some preliminary calculations have been performed by Tecnatom and ECN.

The report [4] was prepared and presented at the 9Th International Symposium on Reactor Dosimetry, Prague September 1996. The objective was to initiate the establishment of links between AMES and the European Working Group on Reactor Dosimetry EWGRD.

The document [5] has been prepared by Tecnatom within work programme B.3. This is one of the contractual deliverables of the project. Very few publications address the problems of the influence of uncertainties regarding the chemical composition of vessel materials and neutron fluence on the determination of the values of increase in the Reference Temperature, RT_{NDT}, and of decrease in the Upper Shelf Energy, USE. The dependence e.g. of uncertainty in ΔRT_{NDT} on the uncertainties in the chemical composition and fluence is analyzed for correlations of the form:

$$\Delta RT_{NDT} = CF \cdot FF$$

with CF and FF being the chemical and fluence factors and containing these uncertainties respectively. This document quantifies the impact of such uncertainties on the resulting values of ΔRT_{NDT} and ∇USE.

With the view to estimating the influence of uncertainties associated with neutron fluence and chemical composition, the second section of the document [5] reviews a series of formulae developed in different countries for the evaluation of the embrittlement caused in the vessel materials by neutron irradiation. These formulae predict the increase in the Reference Temperature depending on residual elements (mainly Cu, Ni and P) and the neutron fluence.

When consideration is given to the effect of uncertainty in only neutron fluence on ΔRT_{NDT}, two different behaviours are observed depending on the form of the fluence factor of the correlation used. In correlations with a potential type fluence factor, the tendency of the uncertainty regarding the fluence factor, and consequently ΔRT_{NDT}, is to increase with increasing fluence. When a fluence factor with a saturation effect is used, however, there is a value of fluence for which the uncertainty is maximized, and another value of fluence for which the corresponding uncertainty in the fluence factor will be zero. Figure 1 depicts clearly this behaviour.

As a general conclusion, the greater or lesser contribution made by the fluence factor term to the global uncertainty in ΔRT_{NDT} depends on the content of residual elements, on the associated uncertainties and on the ΔRT_{NDT} correlation used.

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[2] BALLESTEROS, A, DEBARBERIS, L, VOORBRAAK, W, Six-months Progress Report, AGE-AMES(96)-P2 (1996).
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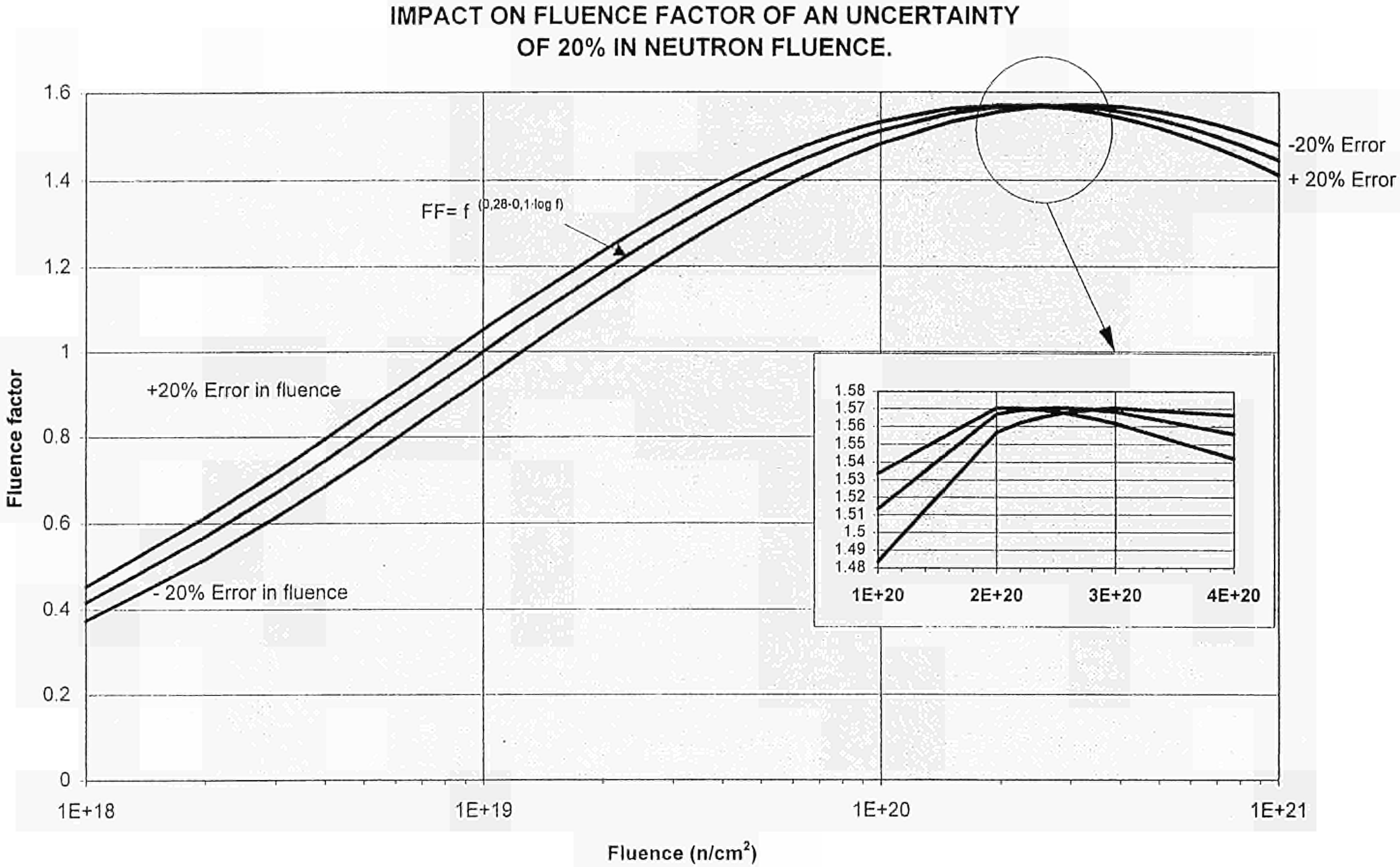

Figure 1. Fluence factor according to R.G. 1.99 rev.2

B.5.2-2 Relation between different measures of exposure-induced shifts in ductile-brittle transition temperatures - REFEREE

Contract No:	FI4S-CT95-0009	**Duration:**	1 Jan. 1996 - 31 Dec. 1998
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Partners:	**SCK/CEN Mol/BE, VTT Espoo/FI, JRC-IAM Petten/EC**		

A. OBJECTIVES AND SCOPE

Demonstration of the continuing integrity of reactor pressure vessels requires the prediction of the Ductile-to-Brittle Transition Temperature (DBTT) of the vessel materials. Conventionally, irradiation induced changes in DBTT are monitored by the shifts in the transition behaviour measured in impact tests. The assumption has then frequently been made that such shifts are equal to shifts in quasi-static fracture toughness.

However recent experimental results have shown that, for some materials, these two measures of the transition shift are not equal, and that in these circumstances impact shifts are often more closely equal to shifts in dynamic fracture toughness.

This project is designed to obtain high quality data on different materials, so that a full comparison of Charpy impact shifts, dynamic fracture toughness shifts, and quasi-static fracture toughness shifts can be made. A wide range of reactor types of interest within Europe will be covered by this project and a comparison of properties between western and eastern materials will be possible.

The data will be compared with less complete datasets already available on a wider range of materials with the objective of obtaining a European consensus view of the behaviour of different materials. Micro-mechanical and micro-structural modelling will be employed to obtain a physical understanding of the relation between the different measures of the DBTT shift, and in particular to explain material-specific differences.

The work carried out is expected to contribute to the development of predictive tools for use in practical applications.

B. WORK PROGRAMME

B.1. Preparation of Material Samples (Task 1)

Specimens will be prepared from pedigree materials held by the partners as follows: Magnox RPV steel/Magnox Electric, Chooz A steel/(SCK/CEN), Loviisa steel/VTT and Reference steels/JRC.

B.2. Irradiation at the HFR (Task 2)

The specimens will be loaded into instrumented sample holders and irradiated at the High Flux Reactor Petten. Responsible for this Task is the JRC.

B.3. Post Irradiation Examination (Task 3)

The irradiated specimens will be tested to produce data on quasi-static fracture toughness, dynamic fracture toughness, Charpy impact transition as well as static tensile data. The PIE will be carried out by all partners.

B.4. Data Analysis and Final Report (Task 4)

The interpretation of the data in comparison with other datasets supplied by AMES Members, using micro-mechanical and micro-structural modelling where appropriate, will be performed jointly by all partners.

C. Progress of work and results obtained

Summary of main issues

Manufacture of specimens for both irradiations at HFR Petten (Task 1, s. B.1) has been completed. Delivery of specimens to HFR (Milestone 1) has been done successfully, to time, for the first irradiation, and the specimens for the second irradiation will be delivered to HFR in time for loading. One Deliverable which should have been delivered by now is a report on sample preparation and characterisation. Substantial progress has been madé, but it is not available yet. However, this is not an important slippage in the programme, because the information is not required until the project comes to analyse the test results on irradiated materials.

Delays in the original schedule for the irradiations, which are to be carried out in HFR, Petten (Task 2, s. B.2), were due to staff reductions and budget difficulties at HFR. Once the situation stabilised, the irradiation start date was revised to January 1997. All parts for the rig and the capsules have now been made, and the first specimens loaded in the capsule. A minor rig modification was needed, causing a further month's delay to the irradiation start, but no change is needed to the planned delivery date of the first specimens to the test laboratory.

The testing and analysis phases (Tasks 3 and 4, s. B.3 and B.4) cannot begin until after the irradiations, but it appears likely that the delays in the irradiation programme can be accommodated within the testing programmes, i.e. without any adverse effect on the finish time of the project. The participants are keeping this under review.

C.1 Preparation of Material Samples (s. B.1)

This milestone has been passed successfully. Manufacture of specimens for both irradiations at HFR Petten has been completed. For the first irradiation the specimens are made from Magnox-specific materials, and are mainly Charpy-sized, with some additional tensile and compact-tension specimens. The specimens for the second irradiation are mainly Charpy-sized, and are made from material specific to a PWR, from a VVER-specific material, and from PWR-type reference steels. The specimens for the first irradiation have been loaded in the capsule ready for irradiation. A drawing of the capsule is shown in Figure 1.

Progress has been made on the characterisation of the materials in terms of chemical composition, microstructure, heat treatment details, and initial mechanical properties. Pre-existing information is also being collected on the irradiation response of the materials. This information will be presented in a report on the sample preparation and characterisation.

C.2 Irradiation at the HFR (s. B.2)

The irradiations wiill take place at HFR in a new facility called LYRA, of which the design and construction are complete. A schematic diagram is shown in Figure 2. The nuclear characteristics of the rig have been calculated, and thermal analysis carried out. These calculations permit the definition of the conditions required to obtain the specified irradiation dose and temperature. Safety clearance for the operation of LYRA has been progressed.

Delays were experienced in the original schedule for the irradiations, due to staff reductions and budget difficulties at HFR. Once the situation stabilised, the irradiation start date was revised to January 1997. Following a further delay to permit a minor rig modification, the expected start date is now in late February or early March 1997. No change is expected to the planned delivery date of the first specimens to the test laboratory.

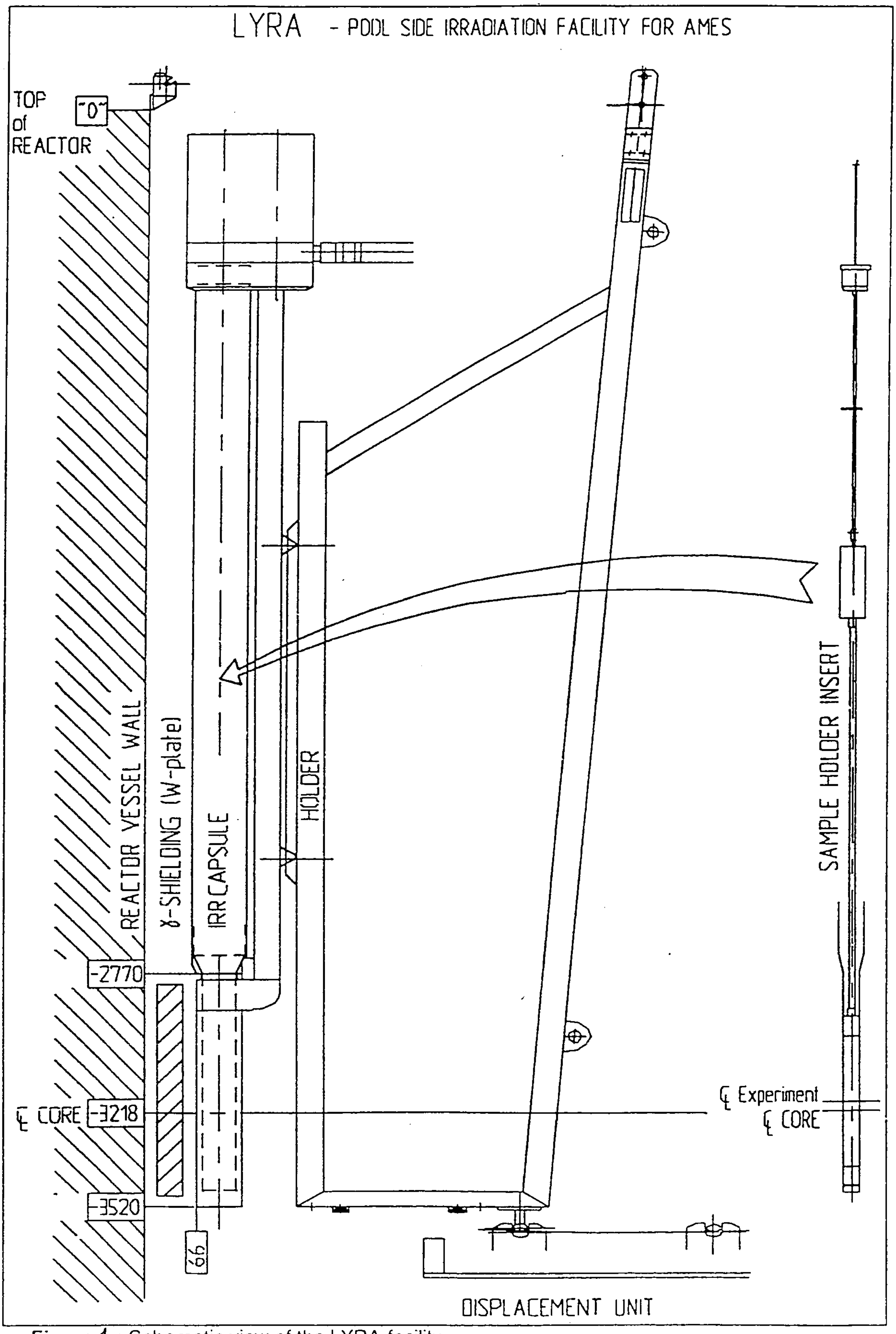

Figure 1 : Schematic view of the LYRA facility.

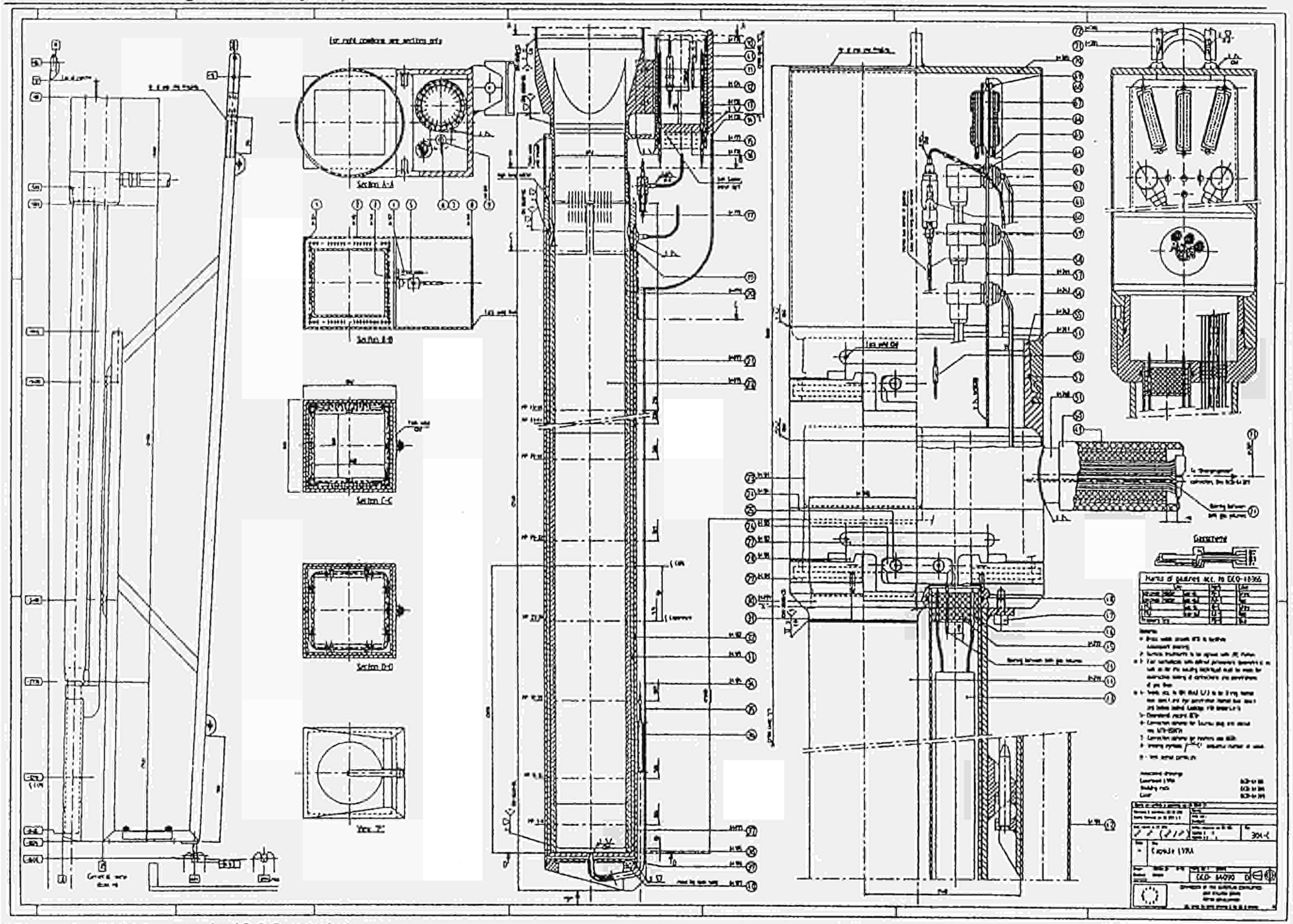

Figure 2 : LYRA capsule (GCO-64090)

B.5.2-3 Evaluation of techniques for assessing corrosion cracking in dissimilar metal welds - DISWEC

Contract No:	FI4S-CT95-0018	**Duration:**	1 Jan. 1996 - 30 June 1998
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Partners:	**VTT Espoo/FI, STUDMAT Nyköping/SE, Univ. Sheffield/GB, GKSS Geesthacht/DE, Rolls-Royce Derby/UK, CIEMAT Madrid/ES, JRC-IAM Petten/EC**		

A. OBJECTIVES AND SCOPE

Dissimilar metal weldments (DMWs) are used in a variety of locations in light water reactor plants, including pipework connections to the reactor pressure vessel, pressuriser and steam generator shell (safe end welds) and vessel penetrations, e.g. for the control rod drive mechanism and instrumentation. Because of their metallurgy these weldments are particularly prone to localised corrosion and especially environmentally assisted cracking (EAC). Despite this potential EAC susceptibility and the occurrence of environmental cracking in practice, only very limited data on crack initiation and propagation rates are available in the literature for DMWs. Analysis suggests that this may be due in part to the fact that the test techniques in this area are not well developed or standardised. The objective of this proposal is to identify and evaluate techniques suitable for assessing environmentally assisted cracking of dissimilar metal welds. It consists of two parts:

1. The first being a comparison of a variety of screening test techniques designed to investigate the susceptibility of the various dissimilar metal weld constituent materials and local microstructural regions to corrosion cracking.
2. Having established the regions of the dissimilar metal weld (DMW) most sensitive to corrosion cracking, a second series of experiments will be performed, targeted specifically at generating quantitative data on crack growth rates.

The testing programmes will be performed on specimens cut from a specially prepared weld length having a specification typical of a DMW found in the nuclear industry. The tests will be performed in a simulated light water reactor environment but in order to guarantee corrosion cracking and hence permit a comparison of the capabilities of the individual testing techniques to screen for corrosion cracking, the water chemistry will be enhanced to favour EAC conditions.

The work to be carried out is a precursor to other research activities in this area of Corrosion Cracking of Dissimilar Metal Welds which will lead to the performance of a Large Scale Integrated Benchmark Project.

B. WORK PROGRAMME

B.1. Review of plant experience and existing experimental data associated with DMWs (Task 1)

Responsible for this task is Sheffield Hallam University (SHU).

B.2. Preparation of the test weldment (Task 2) and its general characterisation (Task 3)

A length of dissimilar metal weld will be manufactured under the leadership of STUDSVIK, its general characterization will be headed by SHU.

B.3. Screening Tests (Task 4) and Crack Growth Rate Testing (Task 5)

A variety of smooth and notched specimen tests will be carried out under the responsibility of ROLLS-ROYCE, the crack growth testing will be co-ordinated by GKSS.

B.4. Evaluation (Task 6)

The Group Leader of this task is VTT.

C. Progress of work and results obtained

Summary of the main issues

During the reporting period considerable ground work has been undertaken. Progress of the project is close to the planned schedule despite some problems associated with the weld procurement. Three intensive meetings of the project steering committee have been held during the reporting period at which extensive planning of the project Work Packages has been realised. The weld block has been procured, examined and, despite some identified problems with weld defects, the weldment has been deemed to be suitable for the programme. Therefore the weldment has been cut into sections and distributed to partners, who are well advanced with specimen preparation. A series of preliminary experiments have been performed which confirm the susceptibility of the weld to EAC in the proposed environment and full testing will commence shortly.

C.1 Review of plant experience and existing experimental data for dissimilar metal welds

During the reporting period the literature review has been prepared, reviewed and submitted to the EC in fulfilment of Deliverable D1.

Dissimilar metal welds (DMWs) are found in a variety of locations in Light Water Reactor plants, other high temperature energy conversion systems, petrochemical plants etc. The present review is aimed at the commercial experience and experimental data of environment-assisted cracking (EAC) in DMWs. The review has been restricted to dissimilar metal weldments using nickel based metal consumables, following the selection of this class of material for the test weld.

The main points arising from the present review are summarised as follows:

1. For PWRs, the majority of incidences of cracking in DMWs have been on external surfaces, and then usually in weldments using austenitic weld consumables. However, a number of failures have been observed in BWR conditions, in weldments using both austenitic and nickel based weld consumables. The crack locations in DMWs vary from case to case, depending on constituent metals, manufacturing processes and post weld heat-treatment procedures.

2. Cracks have also frequently been detected in heat affected zones of ferritic reactor pressure vessel steels and sensitised stainless steels. With regard to the latter, Nuclear Grade Type 316L stainless steel (316NG) has been increasingly used to replace sensitised Type 304 or 316 stainless steels or nickel alloys such as Alloy 600, in order to eliminate intergranular stress corrosion cracking (IGSCC). However, up to date, there is limited service experience concerning the performance of 316NG.

3. The review has confirmed that there is a considerable lack of experimental data on the nickel based materials which are targeted by the present project. The available literature does not reveal any laboratory work that has been conducted on the specific material combination (low alloy steel/Alloy 82/316L) employed in this study. Thus, the review has been carried out using information relating to all combinations of the materials of interest.

4. Laboratory tests indicate that the presence of adverse water chemistry (e.g. sulphate contamination) or a creviced condition accelerates the SCC of DMWs as for most systems subject to SCC. Evidence is provided by slow strain rate tests, constant load or displacement tensile tests and crack growth tests, for DMWs and/or nickel alloy weld metals such as Alloys 182 and 82.

5. The available data for Alloy 82 indicate that gas tungsten arc welding results in a greater susceptibility to SCC than submerged arc welding; there is also a dependence of crack path on welding process and heat treatment.

6. Although potential measurements do not suggest that the galvanic effect between dissimilar metals would provide a significant driving force for the SCC of the DMWs concerned, unpublished laboratory data do indicate a propensity for localised pitting in the low alloy steel at the interface, which may subsequently result in initiation of near interface cracking. It is expected that the aggressiveness of water chemistry and loading conditions, metallurgical factors such as Cr content, Cr depletion and carbon stabilising elements, the influence of welding etc., will all play vital roles in the EAC of DMWs.

C.2 Preparation of test weldment and its general characterisation

Selection of the weld system

During preliminary discussions between the project participants, two classes of dissimilar metal weld system were identified of relevance to the nuclear industry, those utilising filler metals based on stainless steel and those based on nickel alloy consumables. A preliminary decision was made in favour of a nickel based system for the experimental work, based on the preference of the majority of the project partners and on the known SCC susceptibility of these systems. Two potential nickel based alloys were identified: Alloy 82 and Alloy 182. Initial group experience suggested that Alloy 182 was prone to weld metal SCC cracking in oxygenated, BWR type environments, whilst for Alloy 82, cracking was more commonly associated with the weld interface, although the availability of data was limited. Since SCC of the weld interface is the main purpose of the current project, the system A533B-Alloy 82-SS316L was selected for the experimental study. For this system the ferritic to filler metal interface was considered to be the most susceptible to SCC, so this will be the main area of study.

Consideration of the dimensions and number of specimens for the programme defined the need for a plate thickness / weld depth of 80mm and a weld length of 1500mm. Low alloy steel and stainless steel plate widths of 200mm were considered necessary in order to facilitate the manufacture of the weld with representative heat input conditions. To facilitate specimen extraction, a vertical weld interface was specified for the ferritic to weld filler metal interface. In accordance with the usual heat treatment practice of Alloy 82 DMWs, an intermediate heat treatment was specified after the lay-down of the butter layer, with no final post weld heat treatment.

NDE examination of the DISWEC Weld block

Upon receipt of the weld block from the manufacturer, a full NDE examination was performed, consisting of an X-ray analysis of the full weld length supported by additional detailed manual UT examinations. The evaluated results of the X-Ray and UT inspections revealed a high number of registrations characterising porosity and lack of fusion defects, which had to be taken into account in the weld cutting plan. However, the important zone adjacent to the ferritic / butter layer interface was largely free from defects and so the weldment was considered suitable for the present study.

Weld characterisation

Weld characterisation consisted of macro-metallurgical sectioning, micro and macro hardness scans and chemical characterisation across both weld interfaces. This work is currently in progress, but initial results indicate a high interfacial hardness discontinuity at the ferritic/butter layer interface. There is also significant metallurgical variability in the A533B/Alloy 82 interface region, with appreciable discrete martensitic zones (Figures 1 and 2). Since such an extent of martensite is not normally associated with nickel based weld consumables, this suggests poor control of the welding process.

Vickers hardness scans performed both across and along the weld interface exhibited peak values of 500 H_{v25g} in the regions of transformed martensite with values between 400-450 H_{v25g} being common. Such a microstructure would be considered to be highly susceptible to EAC. The non-martensitic regions exhibited a lower peak hardness (up to 370 H_{v25g}) and would be less susceptible although probably not immune to EAC. Considerable variability in test results is to be expected as a result of this microstructural variability and it will therefore be essential to adopt a multiple specimen testing philosophy, with post test specimen characterisation being performed to check the individual specimen microstructures.

C.3 Screening tests and crack growth rate testing

During the first year of the project, three main actions have been undertaken relating to the test programme: selection of the testing environment, establishment of a preliminary test plan and the completion of a short series of preliminary tests to confirm that the chosen environment would lead to EAC.

Environment

The environment chosen for testing was air saturated pure water (conductivity less that 0.1μS cm^{-1}) to which sulphate is added in the form of sulphuric acid to increase the conductivity to 1.0 ± 0.2μS cm^{-1}; thus increasing its propensity for inducing SCC in the tests materials. The use of air saturation produces ~8ppm dissolved oxygen in the feedwater and thus an electrochemical potential ≥0mV SHE. The chosen test temperature was 288°C requiring high temperature autoclave testing facilities.

Preliminary tests

A series of preliminary tests were performed in order to ascertain whether the proposed test environment could induce environmentally assisted cracking (EAC) in the low alloy steel/Alloy 82 weld metal interface of the test weld, and thus confirm the suitability of the test weldment for the programme. These comprised a series of four slow strain rate tests which were performed at a nominal extension rate of 1×10^{-6}mm s^{-1}. Two tests were performed in the proposed test environment and a further two in an inert (argon gas) environment, both at 288°C.

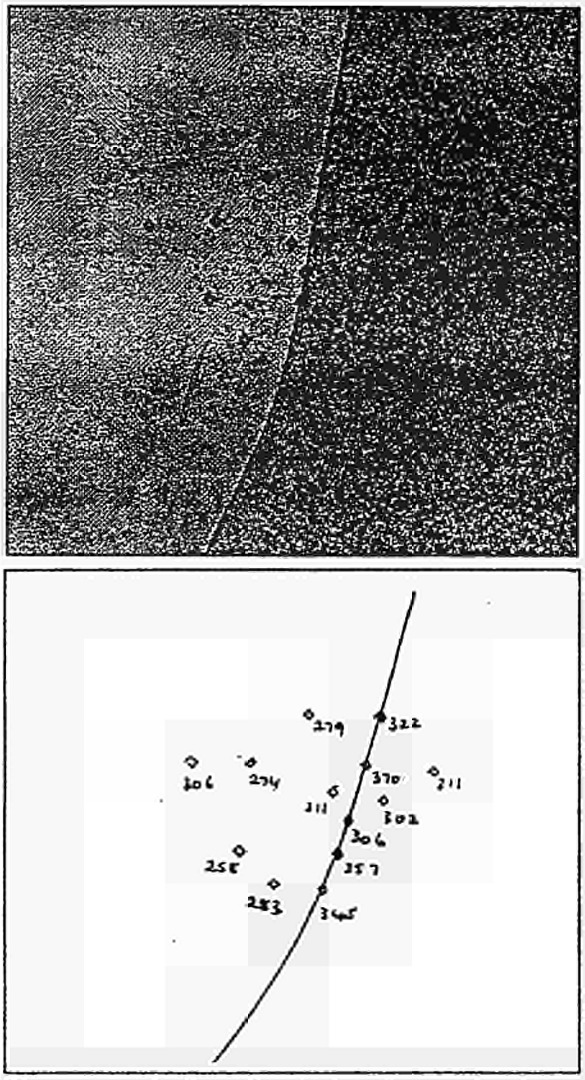

Figure 1 : A533B - Alloy 82 interface: region free of martensite

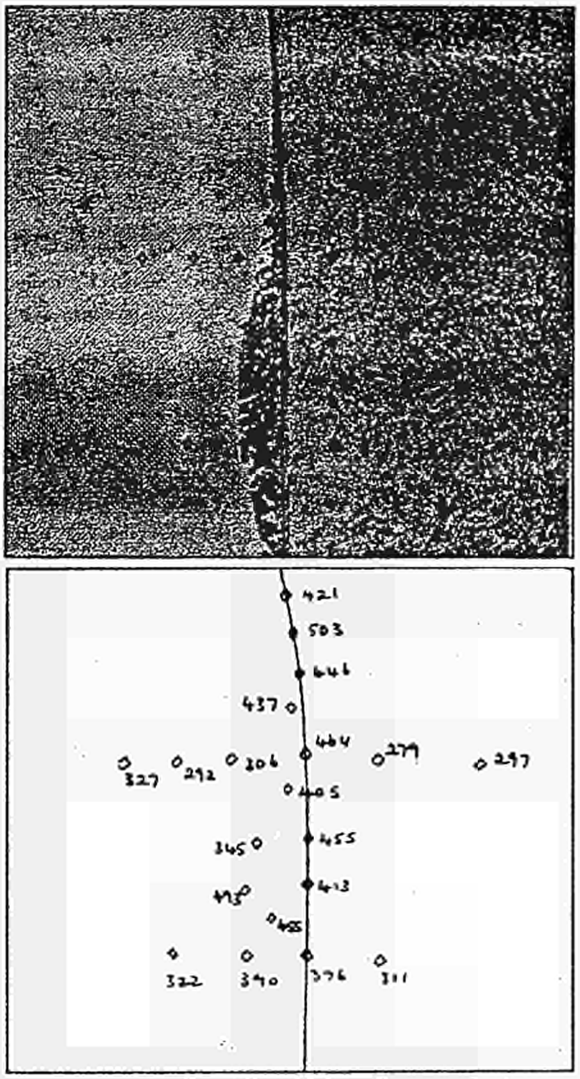

Figure 2 : A533B - Alloy 82 interface: region containing martensite

The specimens tested in environment failed in approx. 18 and 20 hours, failure being accompanied by minimal specimen ductility, as measured by the reduction in area at the fracture site, less than 10%. Metallographic sectioning showed the fracture path to be adjacent to the low alloy steel/Alloy 82 interface, and principally within the low alloy steel. There was evidence of intergranular separation on the fracture surfaces and rapid load reduction upon the load/time and load/extension plots from these two tests. By contrast the specimens tested in the inert environment failed in the Alloy 82 weld metal ~3-6mm away from the low alloy steel/alloy 82 interface, after periods of approx. 50 and 70 hours. The fracture plane for the air tested specimens was at an angle to the axis of the specimen, and appeared to exhibit a typical ductile morphology. Both specimens exhibited significant failure ductilities; the reduction in area figures for the specimens being 64% and 69%. Although analysis is not fully complete, it can be concluded that the specimens tested in environment failed by an environmentally assisted cracking process and thus the material is susceptible to EAC and so is suitable for the test methods evaluation study which is the main part of this project.

Preliminary test programme

Preliminary testing plans have been established for both the screening (WP4) and the crack growth measurement (WP5) activities (Tables 1 & 2). These plans are currently being reviewed following the results of the preliminary tests.

Table 1 : Summary of proposed testing plan for WP4

WP	*Description*	*Participants*	*Tests Conditions*
4.1	Slow Strain Rate Tensile (SSRT)	AEAT	Strain Rates of 10^{-5}, 10^{-6}, 2.10^{-7} & 5.10^{-8}
		GKSS	Work programme to be shared
		RRA (prelim)	Pitted specimens to be considered
4.2	Constant Displacement Tensile	AEAT	Loadings of 70, 80 & 105% of YS & hold for 14 days
		RRA	Could consider pitting or increased loading if no SCC observed.
4.3	Tensile Ripple Load	AEAT	80% YS ± 10% 2 frequencies 1 cycle/min and 1cycle/hour; Δ wave form
4.4	Constant Displacement Bend	Studsvik	Load 75, 105, 125 & 150% YS (duplicate tests) 4pt bend ; specimen geometry 2 x 15 x 80 mm
4.5	Slow Strain-Rate Notched Tensile	Studsvik	2 notch geometries 3mm dia. Notch depth .25mm 2 notch radii 0.75mm (K_T~1.5) & 0.1mm (K_T~3.1) Strain Rates of 10^{-5}, 10^{-6}, 2.10^{-7} & 5.10^{-8}
4.6	Keyhole CT	SHU	Slow displacement control ramp (rising load) Test duration 1 month
4.7	Constant Displacement Notched Tensile	AEAT	Same 2 notch geometries as in WP4.5 Loadings of 70, 80 & 105% of YS & hold for 14 days
		RRA	Could consider Ripple loading if no SCC observed

Table 2 : Outline test programme proposed for WP5

WP	*5.1*	*5.1*	*5.2*	*5.3*	*5.4*
Partner	CIEMAT	GKSS	SHU	VTT	GKSS
Specimen Type	1T CT	½ T CT	1T Keyhole CT	10x10x55 Charpy	DCB
Type of Test	Constant Load	Rising Displacement	Ripple Load & Constant Load	Displacement Control Rising Load	Constant Deflection
Environment	as specified	as specified *	as specified	as specified	as specified
Specimen Preparation	fatigue	fatigue (air → ΔK_{TH})	pitting	-	fatigue
Detection of Crack Initiation		PD	PD	-	-
Crack Growth Monitoring	PD	PD	PD	-	
Post Test Evaluation	SEM, a_o, Δa	SEM, a_o, Δa	SEM, a_o, Δa	-	SEM, a_o, Δa
Deliverables	K_{ISCC}, da/dt	da/dt=f(K)	K_{ISCC}, da/dt	-	
Variables	K, crack position	Displ. Rate	K, loading	-	K_{init}, crack position

C4. Evaluation

Work on this WP has not started, since this awaits the results from the testing work.

AREA C

RADIOACTIVE WASTE MANAGEMENT AND DISPOSAL AND DECOMMISSIONING

C. RADIOACTIVE WASTE MANAGEMENT AND DISPOSAL AND DECOMMISSIONING

INTRODUCTION

The main objectives of this area are the further development and integration of the efforts undertaken by the Community and the Member States in the field of the management and disposal of radioactive waste, as well as in the decommissioning of disused nuclear installations. The understanding of the phenomena and processes in and around a geological repository is a prerequisite for evaluating the efficiency of the multi-barrier concept and the safety of the disposal system as a whole. The work in this area is intended to develop a consensus in the safety evaluation methods, to provide data on waste forms and host rocks and models for the various phenomena which control the radionuclide release and movement from the repository into geosphere and then into the accessible biosphere, to demonstrate the feasibility of disposal concepts (e.g. in underground laboratories).

C.1 Safety Aspects of Waste Disposal

A consensus on the approaches and methodological aspects for evaluating the long-term safety of disposal systems should be developed. The question on the time horizon up to which safety analyses have to be elaborated will be considered. The safety evaluation of the direct disposal of spent fuel as well as of the waste retrievability are also considered.

C.1.1 Disposal of spent fuel: evaluation of the performances of a disposal system for spent fuel including review of packaging policies and properties and near-field aspects.

C.1.2 Retrievability: possible goals of retrievability and on the corresponding time scale options. Long term safety evaluations, including the operational and the retrieval phases and the safeguards aspects.

C.2 Underground Research Laboratories

Investigation and demonstration of the feasibility of disposal concepts for vitrified high level waste, long-lived waste and for spent fuel and collection of data on the performance of repository barrier components under representative disposal conditions. The following topics are covered.

C.2.1 Testing and demonstration of disposal concepts: demonstration of specific repository construction and waste emplacement techniques for the disposal of vitrified high level, long-lived waste and of spent fuel.

C.2.2 Backfilling and sealing of repositories: demonstration and testing of backfill and sealing concepts and constructions in boreholes, galleries and shafts.

C.2.3 Long term behaviour of repository components: in-situ experiments for testing of models to predict transport processes in host rocks and interactions between them under representative disposal conditions.

C.2.4 Groundwater flow analysis and radionuclide migration: in-situ experiments for testing of models to predict transport processes in host rocks and in the engineered barriers.

C.3 Research on basic phenomena

Better understanding of the phenomena which control the release of radionuclides from the waste packages and their migration through the various subsequent barriers up to the accessible environment. Modelling of the phenomena and testing of the models.

C.3.1 Waste volume minimization: waste decategorisation with exhaustive decontamination processes to reduce the waste to be disposed of in underground repositories and enabling their disposal in surface repositories.

C.3.2 Characterization of waste forms and matrices: Characterization and evaluation of repository component materials and of the waste forms and matrices (cement, spent fuel, glasses) and their behaviour under conditions encountered in deep repositories in clay, crystalline rock and salt. The research covers:

- **Cement as containment and barrier material** including interactions with inorganic and organic waste constituents.
- **Spent fuel** direct disposal into clay, crystalline rock or salt formations (experiments and models).
- **Glass matrices** for HLW and MLW (experiments and modelling of phenomena). Creation of a data base, effects of the corrosion products from metal canisters on migration in the near-field.

C.3.3 Quality control of nuclear waste packages and waste forms: to promote and facilitate collaboration in the development, application and standardization of quality checking for waste packages to identify R&D requirements; to collaborate in the development of new test methods. Measurement on waste forms and round robin tests and establishment of Quality Assurance and Quality Control procedures.

C.3.4 Geomechanical behaviour of engineered barrier materials and host rocks:

- **Conceptual model development** for thermo-hydro-mechanical and geochemical properties of various engineered materials and host rocks deduced from experimental investigations.
- **Benchmark exercise** of coupled models to simulate the thermo-hydro-mechanical and thermo-hydro-chemical properties of engineered materials and host rocks (near field) and their interfaces as a function of time.

C.3.5 Gas generation and transport: further development of a common methodology for both laboratory and field testing and modelling of the various aspects to gas generation and its release and transport properties from waste packages through various repository components and host rocks:

- **Theoretical and experimental studies of gas generation, release and transport** through engineered barriers and the near field host rocks into the far field.
- **Verification and testing of conceptual and numerical models** of gas generation and transport considering also models of natural gas flows.

C.3.6 Radionuclide migration: theoretical studies, laboratory and field experiments to supply further information and data for performance assessment of geological disposal:

- **Complexation with natural organics** by considering significant physico-chemical parameter variations for critical radionuclides and mixed complexation under natural aquatic conditions.
- **Colloid transport** and generation studies in shallow and/or deep aquifer system to verify and test current conceptual and numerical models for aquifer systems supported by large scale column experiments and natural analogue studies.
- **Transport and retardation processes** studies on water/rock interactions to quantify significant RN transport and retardation processes through porous and fractured rock systems.
- **Geochemical modelling** of radionuclide transport through the engineered barriers, near field/far field host rock interface and the geosphere.

C.3.7 Natural analogue/systems studies:

- **Natural analogues of repository materials,** studies on the longevity of the near field chemical environment and sorption properties of engineered barrier corrosion products.
- **Natural analogues of radionuclide release and transport processes** in various porous or fractured aquifer systems, site specific matrix diffusion quantification, evidence for long-distance colloid transport in relevant aquifer systems.
- **Natural analogues of thermo-hydro-mechanical and thermo-hydro-chemical** properties of host rocks.

C.3.8 Palaeohydrogeology and geoforecasting: studies should provide information on site evolution over geological times. Information on ancient flow regimes is indicative of past rates of uplift, subsidence or erosion, and of climatically or tectonically induced changes in groundwater behaviour:

- **Palaehydrogeological aspects** on fractured basement rock environments by using the information to assess long-term predictions of flow and transport models.
- **Model testing** related to new methods and techniques to quantify flow and transport and palaeofluxes through argillaceous rocks.

C.4 Decommissioning of nuclear installations

The dismantling of nuclear installations is part of closing the nuclear fuel cycle. R&D in this field particularly concerns the solution to issues of environmentally compatible disposal of radioactive dismantling wastes, the minimisation of radiological impact and the reduction of costs, i.a. by the application of innovative techniques. The work is intended to develop relevant technology, to collect and analyse relevant data as well as demonstrate and evaluate decommissioning strategies and techniques.

C.4.1 Development of innovative dismantling techniques: the projects are mainly aimed at demonstrating decommissioning strategies. These will mainly address the dismantling of reactor pressure vessels and core internals.

C.4.2 Collection of technological performance data: it concerns data of dismantling techniques at decommissioning projects in Europe (and abroad, if possible) in a systematic way, e.g. in the EC-DB-TOOLS database.

C.4.3 Collection of data on specific waste arising, doses and associated costs: EC-DB-COST (costs, occupational doses, wastes arising from decommissioning). The inclusion of data from current and future decommissioning projects is of interest to the Community as a whole.

The following "CLUSTERS" have been set up:

- C.1- Safety evaluation of spent fuel disposal - SES;
- C.2- Underground research laboratories - URL;
- C.3- Partitioning experiments and waste minimisation - PART MIN;
- C.3- Waste form characterisation - WFC;
- C.3- Performance of cement - PERCENT;
- C.3- Non-destructive assay - NDA;
- C.3- Destructive assay - DA;
- C.3- Geochemical behaviour of clay barriers - GEOMEC;
- C.3- Gas generation and gas flow - GAS;
- C.3- Radionuclide transport / Retardation process - RATREP;
- C.3- Palaeohydrogeology - PALHY;
- C.4- Decommissioning - DEC.

C.1.1-1 SPA : Spent fuel performance assessment

Contract No: FI4W-CT96-0018 **Duration:** 1 June 1996 - 30 April 1999
Coordinator: P. Escalier des Orres, CEA/IPSN Clamart/FR
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Partners: ECN Petten/NL, ENRESA Madrid/ES, GRS Köln/Braunschweig/DE, SCK/CEN Mol/BE, VTT/ET Espoo/FI

A. OBJECTIVES AND SCOPE

The project concerns the direct disposal of spent fuel in various host rock formations (clay, crystalline rocks and salt formations) and will be sharing the experiences, assessment tools and databases in use and under development in the different national programmes to come to a common understanding and harmonization of the assessment methodologies and practices in the Member States dealing with the safe disposal of spent fuel. The project will focus on various issues such as :

- review policies, packaging plans and repository designs for direct disposal of spent fuel in various host rock formations;
- develop models for the simulation of processes in the near-field of the spent fuel repository;
- evaluate the performance and safety of different disposal concepts by carrying out total system PA of spent fuel disposal in various host rock formations.

B. WORK PROGRAMME

B1. Waste packaging and disposal concepts (All partners)

- Review of packaging policies and properties
- Repository design

B2. Near-field aspects (All partners)

- Waste form and activity inventory
- Near-field modelling and data

B3. Performance Assessment (All partners)

- Scenario analysis
- Total system models and data
- Deterministic approach
- Stochastic approach

C. Progress of work and results obtained

Summary of main issues

This project will last 36 months: it has been started in May 1996 and it will be carried out until April 1999.

For Work Programme 1, the different sources, types and amounts of spent fuel which may be disposed of as waste in various host rock formations under consideration (clay, crystalline rock and salt) will be reviewed. Waste packages and repository design will be issued from the different national programmes dealing with the direct disposal.

For Work Programme 2, a first selection of data in terms of waste form and activity inventory will be established for the reference calculations from the expected spent fuel previously mentioned. A review of the main components of the source term and of the far field modelling to improve the performance assessment will be specified.

The main deliverables of the project are included in Work Programme 3 "performance assessment" of different alternative canister and repository designs for spent fuel disposal. The two first tasks, scenario analysis and total system models and data, form the basis for the evaluation. A total system analysis of the spent fuel disposal covering the near field, far field and biosphere, will be performed. This work will be realized using deterministic and stochastic calculations for normal and selected altered scenarios for each site. The results obtained will be used for sensitivity analyses. The final phase will lead to a hierarchized list of most influential elements (scenarios, phenomena, radionuclides, ...). This list can contribute to the definition of future orientation of the R&D programmes.

Progress and results

C.1 Waste packaging and disposal concept

A large part of the year 1996 was devoted to the Work Programme 1. This Work Programme is divided into two sub-tasks: packaging policies and properties, and repository design.

The first step of the project is to review the sources, types and amounts of spent fuel which may be disposed of as radioactive waste. The direct disposal concerns mainly UOX (uranium oxide) and/or MOX (uranium and plutonium mixed oxide) spent fuel.

For many countries involved in this project, the direct disposal is an alternative policy to reprocessing; an estimation of the amounts and types of spent fuels intended for the geological disposal has to be defined in agreement with the national nuclear power supply cycles and the radioactive waste management policies:

- Belgium and France report that reprocessed and recycled plutonium in MOX fuels would use a part of the plutonium breeding from the nuclear reactors: the amounts of spent fuel which should arise from the actual nuclear PWR plants in both countries have been estimated to be 4,226 and 15,000 tHM for UOX fuels, 670 and 5,000 tHM for MOX fuels respectively,
- Germany does not consider reprocessing: the unloaded spent fuels (only UOX fuels) are designated to be disposed of with a cumulated waste in the repository of 25,000 tHM.

For Spain and Finland, spent fuel is considered as nuclear waste and direct disposal has been studied since many years. The amounts estimated to be disposed in the repository are 11,600 PWR elements and 8,400 BWR elements for Spain, and about 2,600 tHM including 1,870 tHM (10,700 fuel assemblies) of BWR fuel and 740 tHM (6,200 fuel assemblies) of VVER-440 type PWR fuel for Finland.

The containers are designed for the interim storage, transport and final disposal and should protect the fuel assemblies during the transient thermal period. No other long-term barrier function is required except for Finland: the Finnish disposal concept is based on a copper canister which has a very long expected lifetime in the repository conditions.

Repository designs for the different host rock formations and types of waste are not yet definitively established for most of the participating countries except for Finland, where the repository design (ref. [1]) is of the KBS-3 type. For France, UOX and MOX fuels will be disposed in separate modules in either galleries or boreholes taking into account thermo-mechanical constraints (for clay and granite). For Belgium, the repository design in clay is based on a gallery disposal concept with 4 disposal tubes for UOX fuels and 1 for MOX fuels. Germany considers that the containers will be emplaced in drifts in the case of the salt site disposal and in boreholes in the case of the granite disposal. The Netherlands have elaborated a disposal concept in salt consisting of open horizontal galleries, and the disposal concept in clay is not yet done. In Spain, granite, clay and salt are considered as potential host rocks. The disposal concept foresees the emplacement of canisters in a steel tube surrounded by the buffer materials, along the axis of the drifts.

C.2 Near field aspects

During the period covered by this report, a part of the Work programme 2 was treated. This Work Programme is divided into two main sub-tasks: waste form and activity inventory, and near field modelling and data.

Waste form and activity inventory have been established following the different assumptions of Work Programme 1. The reference UOX fuels are quite equivalent for the different countries involved in the project: 3.7% U-235, burn-up 45 GWd/t for France, 3.6% U-235, burn-up 45 GWd/t for Germany, 3.3-3.6% U-235, burn-up 36 GWd/t for Finland, and 4.1% U-235, burn-up 40 GWd/t for Spain. For Belgium, the activity inventory for UOX fuels might aggregate the different inventories of UOX fuels (burn-up of 36, 45, 50 and 55 GWd/t) to avoid considering too many different ones. The reference MOX fuels considered by France and Belgium assume an average plutonium abundance of 8.2% and 7.05% with a depleted uranium (0.25% of U-235) and an average burn-up of 43.5 and 45 GWd/t respectively.

The source term modelling has important implications for the total performance assessment of radioactive waste disposal. Co-operation among organisations studying different aspects of the nuclear waste disposal will be expected in the framework of this project: the source term modelling will be elaborated in close contact with the project "source term for performance assessment of spent fuel as a waste form" (ref. [2]). From the point of view of the release mechanisms, the different radionuclides involved in performance assessment can be roughly grouped into different regions as shown in Figure 1:

- The fuel pellets: the major part of the activity is bound to the UO_2 grains. From fuel matrix, some of the volatile fission products have migrated to the gap between the fuel pellets and the cladding, or have migrated to the grain boundaries of the fuel matrix,

- The claddings, made of zircaloy, and the structural parts of the fuel assemblies (stainless steel): they may also contain activation products and some radionuclides which have migrated from the fuel pellets.

For performance assessment analyses, a common source term model and alternative models, as conceptual model uncertainty, will be used.

Within this common source term model, the approach which will be considered as the reference source term model is indicated in Table I.

The alternative source term models used by Belgium, Finland, France, and Spain are based on the following assumption. Inventories of radionuclides in the fuel are divided into three source compartments: gap, grain boundaries and fuel matrix. The release of radionuclides from the gap is instantaneous, delayed from the grain boundaries and the dissolution rate of the fuel matrix is assumed to be proportional to the α-activity of the spent fuel ("self oxidation" model) with a congruent release of radionuclides. For the fuel matrix, the "self-oxidation" model leads to a degradation period of some millions of years. For Germany, gap contains both actinides and fission products. No distinction is made between grain boundaries and fuel matrix (dissolution rate of 600 years corresponding to the grain boundaries dissolution).

The far field models are at different steps: for France, a specific far field modelling will be included in the MELODIE far field code (explicit modelling of the engineered barriers, repository concept, ...) for mid-1998. Finland will use the far field model REPCOM in which the far field of the canister is divided into several compartments. Belgium will use a similar approach and has incorporated the Finnish release model into its clay code NUCDIS. The far field modelling for Spain will be based on the diffusion of the radionuclides through the bentonite, immediate transfer to the EDZ (Excavation Damage Zone), advection in water conducting features equivalent to a single pathway. Germany will review the far field models for salt and develop a near field model for granite based on the repository concept which was worked out in a national project.

C.3 Performance assessment

This Work Programme is in progress and no results are yet available.

References

[1] TVO, TVO-92. Safety analysis of spent fuel disposal. Report YJT-92-33 E (1992).

[2] EC, Project on "source term for performance assessment of spent fuel as a waste form" (contract FI4W-CT95-0004).

Table I: Common approach on the source term modelling

	GAP RELEASE	GRAIN BOUNDARIES AND FUEL MATRIX	STRUCTURAL PARTS AND CLADDING
% inventory of the fuel	**Best estimate value:** 5%: Cs, I, Rb, Zr and Cl 2%: Tc, Pd, Sn, C 1%: Se, Sm 0.5%: Cm, Am, Pu, Pa, Th, U, Ra, Ni, Mo, Nb **Range:** Factor 2	100% diminished of gap inventory	100 % activation products
Alteration rate	Instantaneous release	Constant absolute rate **Best estimate value:** 10^{-6} year^{-1} **Range:** 10^{-4} to 10^{-7} year^{-1}	Constant absolute rate **Best estimate value:** 10^{-3} year^{-1} **Range:** 10^{-2} to 10^{-4} year^{-1}
Distribution	Uniform distribution	Log triangular	Uniform distribution

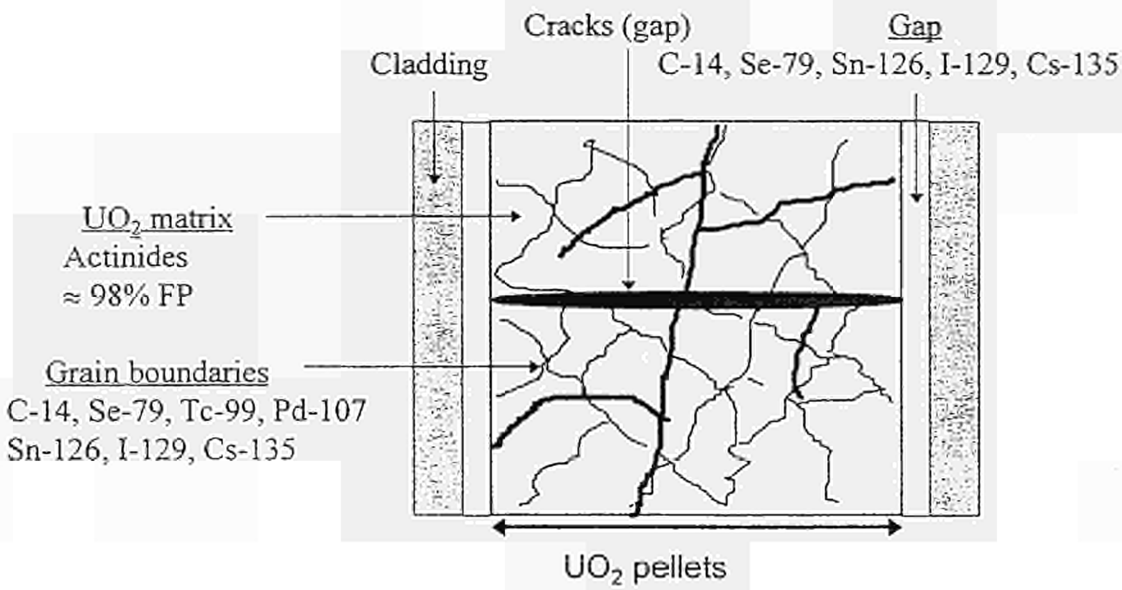

Figure 1: Main radionuclides sources in a spent fuel: the cladding, the fuel gap, the grain boundaries and the UO_2 fuel matrix.

C.2.1-1 FEBEX, Full-scale engineered barriers experiment in crystalline host rock

Contract No:	FI4W-CT95-0006 **Duration:** 1 Jan. 1996 - 30 June 1999
Coordinator:	**F. Huertas, ENRESA Madrid/ES**
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Partners:	**CIEMAT Madrid/ES, AITEMIN Madrid/ES, G3S Palaiseau/FR, ANDRA Chatenay-Malabry/FR, GRS Braunschweig/DE, NAGRA Wettingen/CH**

A. OBJECTIVES AND SCOPE

To asses the safety of an underground repository for the isolation of high-level waste (HLW), it is necessary to understand the behaviour of the engineered barrier system (EBS) and the host rock within the near-field zone. There is also a need to demonstrate the feasibility of the repository concept under conditions similar to those in reality, i.e. same scale, natural hydration conditions and under a heat load similar to the one produced by the waste thermal decay.

Therefore a full-scale test is being performed in a drift at the Grimsel Test Site (CH). This test is being complemented by a large-scale "mock-up" test at CIEMAT - Madrid (ES) and by a series of laboratory tests.

The objectives of these tests are :

- To demonstrate the feasibility of handling and construction of the EBS components, including industrial manufacturing of highly-compacted bentonite blocks, quality assurance, and system monitoring. It is expected that experience will be gained and standards developed for the waste disposal techniques, even during dismantling, by observing the EBS..
- To study the thermo-hydro-mechanical processes in the near-field, mainly in the buffer material.
- To develop, verify and partially validate existing codes and constitutive relations for non-saturated swelling clays.
- To identify and model alteration processes in the buffer. Investigation of gas generation and transport, is also being made.

B. WORK PROGRAMME

B 1. Full scale "in-situ" experiment (all partners)
   * Test area characterization.
   * Component Design, manufacturing and installation.
   * Heating and monitoring.

B 2. "Mock-up" test (ENRESA - CIEMAT)

B 3. Laboratory tests on buffer materials (ENRESA - CIEMAT - GRS)
   * Characterization tests.
   * Laboratory tests for the improvement of THM constitutive laws.
   * Geochemical tests.

B 4. Modelling (ENRESA - CIEMAT)
   * Inventory and improvement of codes and constitutive laws.
   * Thermo-hydro-mechanical (THM) modelling.
   * Thermo-hydro-geochemical (THG) modelling.

B 5. Coordination and final assessment (ENRESA - NAGRA)
   * Coordination and integration of work.
   * Implementation of QA/QC programme.
   * Public information.
   * Final assessment.

C. Progress of work and results obtained

Summary of main issues

The main issue of B 1 was that the full-scale "in situ" test was installed, thereby completing most of the first objective of FEBEX: the demonstration of the feasibility of constructing an engineered barriers system (EBS). The test area was characterized and the demonstration made, including the fabrication, handling, and emplacement of highly-compacted bentonite blocks (buffer) at semi-industrial scale, as well as the design, manufacturing, and installation of other components. Thus, the physical part of B 1 is complete and heating will start in early 1997.

The main issue of B 2 was that the other large-scale test, the "mock-up" test, was completed. This included all the physical components (confining structure, heating and hydration systems, buffer, and instrumentation) having been designed, manufactured/procured, and installed. The data acquisition system (DAS) is being connected and the hydration, as well as the heating, will start in early 1997.

The buffer material laboratory tests, B 3, are only partially completed as the tests will be conducted throughout the entire experiment, including the dismantling stage of the large-scale tests.

All of B 4 has been started and the inventory completed. Preliminary thermo-hydro-mechanical (THM) and -geochemical (THG) modelling have been made. Improvement of the THM and THG models and constitutive equations are on-going.

B 5 is also on-going; however, the implementation of the appropriate Quality Assurance/Quality Control Program (QA/QC) has been effected in all the work accomplished in 1996. The integration of the work is, of course, on-going as is the dissemination of public information.

Progress and results

C.1 Full-scale "in situ" test (Figure 1)

The "in situ" test is installed in a drift, which is 73 m long (of which 17 m are the test section), has a diameter of 2.28 m, and is in a predominately granite rock mass.

Test area characterization - the geological and hydrogeological characterization have been completed. The preliminary three-dimensional modelling of the FEBEX area, geological mapping of the drift, extensive core descriptions, interpretation of the hydro-mapping of the test section of the drift, hydraulic characterization of the boreholes within the drift, three-dimensional interpretation of the hydraulic tests, and hydrochemical characterization of the water took place.

Design, manufacturing, and installation of components - was completed. The main components (heaters; buffer, constructed of highly-compacted bentonite blocks; concrete plug; and instrumentation) have been installed and both the heating system and instrumentation checked for functionality. The basic features of each heater are shown in Table I. The number and weight of the bentonite blocks manufactured for the buffer are shown in Table II. The dry density of the blocks averaged 1.70 t/m^3. To construct the buffer, 5331 blocks (116.2 t) of bentonite were used. With construction gaps of 5.53%, the resulting average dry density of the buffer was 1.6 t/m^3 with a water content of 14.3%. The sensors (instrumentation) used are shown in Table III. In addition, both conservative and non-conservative tracers were used. The monitoring and control system provides access functionality, both locally and remotely (Madrid).

C.2 "Mock-up" test (Figure 2)

The components were designed, manufactured, tested, and installed. The main components are the following: confining structure, heaters, hydration system, buffer, sensors, and monitoring and control system. The confining structure consists of two cylindrical bodies of carbon steel assembled together, with a useful length of 6.00 m and an internal diameter of 1.62 m. It has been designed to withstand a working pressure of 9.0 MPa. The heaters are two carbon steel cylinders, each 1.625 m long with an external diameter of 0.340 m, designed to withstand a pressure of 9.0

MPa. Three electrical resistances, able to supply a power of about 1000 W each, are wound around a carbon steel core. The temperature on the surface of each heater will be continuously controlled. The hydration system consists of two water tanks of 600 l each, pressurized by nitrogen and supplying synthetic granitic water to the confining structure. The tanks are placed over pressure cells to determine, by weighing, the evolution of the water intake with time. For the buffer, bentonite has been compacted into blocks with a dry density of 1.75 t/m^3 and these blocks were placed in two concentric rings around either a heater location or a core of bentonite blocks. In the "mock-up" test were emplaced 22.5 t of bentonite blocks, resulting in an average dry density of 1.64 t/m^3 with a water content of 14% and construction gaps of 6.7%. Installed were 328 temperature, 48 total pressure, 20 fluid pressure, and 40 relative humidity sensors. The monitoring and control system consists of two independent units with an uninterrupted power supply (UPS). There are 380 channels for temperature and 120 for humidity and pressure.

C.3 Buffer material laboratory tests

The initial tests indicated these basic results.

The bentonite is 90 to 95% calcic-magnesic montmorillonite with an ion exchange capacity of 105 to 113 meq/100g. The swelling pressure ranges from 4.7 to 6.3 MPa for dry densities from 1.60 to 1.65 t/m^3, and the corresponding saturated hydraulic conductivity is about 5.90 * 10^{-14} m/s.

Vertical strain (ε) induced by soaking and the resulting void ratio (e) after soaking, of a specimen of bentonite (compacted at an specific dry weight, γ_d) under an applied load (σ) expressed in MPa, can be approximated by the following expressions:

$$\varepsilon = -0.4693-0.1935\log_{10}\sigma + 0.0373\,\gamma_d \qquad \textit{and} \qquad e = 0.842-0.327\log_{10}\sigma$$

For a water content lower than 20%, the water retention curve may be represented by the equation:

$$\omega(\%) = 40.68-14.26\log_{10}s \qquad \text{(s, suction, in MPa)}$$

A preliminary state surface may be derived for the case of suction reduction with small changes in stresses (σ, s, p_a, p_{atm} are the vertical stress, the suction, the air pressure, and the atmospheric pressure, all expressed in MPa):

$$e = 0.8169-0.1268\log_{10}(\sigma-p_a)-0.04968\log_{10}(s+p_{atm})+0.05409\log_{10}(\sigma-p_a)\log_{10}(s+p_{atm})$$

The thermal conductivity varies between 0.5 and 1.24 W/m*K for dry and saturated compacted bentonite and the linear expansion coefficient ranges between $5 * 10^{-5}$ and $2 * 10^{-4}$ 1/°C.

Laboratory tests for the improvement of THM constitutive laws and geochemical tests - are on-going.

C.4 Modelling

The inventory of existing codes has been completed. The improvement of the codes and constitutive laws is on-going.

The development of a constitutive model for joint behavior and implementing it into the existing code of CODE-BRIGHT has been undertaken during 1996. The model being developed includes thermal, mechanical, and flow transfer capabilities at the joint and is compatible with the current THM formulation of CODE-BRIGHT. A finite element formulation has been made for the theoretical framework. A number of numerical examples have been solved to verify the model.

THM modelling - sensitivity analyses were made on: effect of buffer dry density on development of pressure on the heaters, effect of rock desaturation on hydration times, effect of gas instrumentation on the behavior of the "in situ" buffer, and effect of hydration pressure in the "mock-up" test.

THG modelling - has resulted in developing a new ULC code (TRANMEF-3D) for solving groundwater flow, multicomponent solute transport, and heat transport in fully three-dimensional confined porous and fractured media by the Finite Element Method (FEM) using an Eulerian scheme. It can solve the water flow, solute transport, and heat transport equations simultaneously, or any of them separately. It can handle up to six different types of elements used arbitrarily together in any problem. Moreover, these elements may have different dimensions, a capability which will allow modelling 1D and 2D fracture networks within a general 3D porous medium.

C.5 Coordination, integration, and final assessment

Coordination and integration of work - required:

- Communication between modellers, field staff, and EBS engineers to optimize the system characterization/sensor layout.
- Iterative evaluation of EBS emplacement design and procedures for both large-scale tests in the light of observations and scoping tests in the laboratory, in a large-scale test pad (Toledo), and in Grimsel.
- Coordination of material and equipment production in several countries and their shipment to Switzerland was accomplished by rapid communication between partners.

Implementation of QA/QC program - both large-scale tests, "in situ" and "mock-up", have been subjected to a Quality Assurance Program, which was officially issued at the beginning of 1996. Some of the areas where QA/QC has been of particular importance are: issuing of administrative procedures for the project by the different participating organizations; independent design verification or reviews of the principal components design; fabrication of principal components subjected to Quality Control Plans; implementation of measuring and test equipment control by the applicable participating organizations; installation of both large-scale tests subjected to Quality Control Plans, including density analysis, sensor calibrations and checks, and functional tests; implementation of software Quality Assurance by the modelling organizations; and implementation of independent inspections, surveillances, or audits by Quality Assurance Department of ENRESA.

Tables

Table I: Basic features of the heaters

Length	4.54 m
Diameter	0.90 m
Wall thickness	0.10 m
Material	Carbon steel
Unit weight	11.5 t
Electrical supply	400 V AC
Power	3 * 4000 W

Table II: Number and weight of bentonite blocks

Block Type	Unit weight (kg)	Produced units	Total weight (Kg)
BB-G-01	22.1	2,898	64,046
BB-G-02	21.8	2,310	50,358
BB-G-03	21.3	1,614	34,378
BB-G-04	23.1	562	12,982
BB-G-05	18.0	184	3,312
Totals :		7,568	165,076

Table III. Number and type of sensors installed in the "in situ" test

Parameter to be measured	Sensor type	In charge	Number in area				Total sensors
			Granite	Bentonite	Heaters	Service	
Temperature	Thermocouple	AITEMIN	62	91	36		189
Rock mass total pressure (3D)	Flat jack/vib. wire.	AITEMIN	4				4
Rock contact total pressure	Flat jack/vib. wire	AITEMIN	30				30
Total pressure on heater	Flat jack/vib. wire	AITEMIN		6			6
Rock mass hydraulic pressure	Piezoresistive	AITEMIN	62				62
Pressure of borehole packer	Piezoresistive	AITEMIN	62				62
Pore pressure	Vibrating wire	AITEMIN		52			52
Humidity	Capacitive	ANDRA		58		1	59
Humidity	Psychrometer	AITEMIN	28	48			76
Humidity	TDR	NAGRA	4	20			24
Rock displacement (extensometer)	Vibrating wire	AITEMIN	6				6
Heater displacement	Vibrating wire	AITEMIN		9			9
Bentonite expansion	Vibrating wire	AIT		8			8
Bentonite expansion	LVDT	G.3S		6			6
Tilt (inclinometer)	LVDT	G.3S		6			6
Fissure displacement (fissuremeter)	LVDT	G.3S	1				1
Gas pressure	Piezoresistive	GRS		4			4
Gas flow	Manual reading	GRS		6			6
Atmospheric pressure	Piezoresistive	AITEMIN				1	1
Ventilation air speed	Hot wire	AITEMIN				1	1
Current-heating element	Elec. converter	AITEMIN				6	6
Voltage-heating element	Elec. converter	AITEMIN				6	6
Totals			259	314	36	15	624

Figures

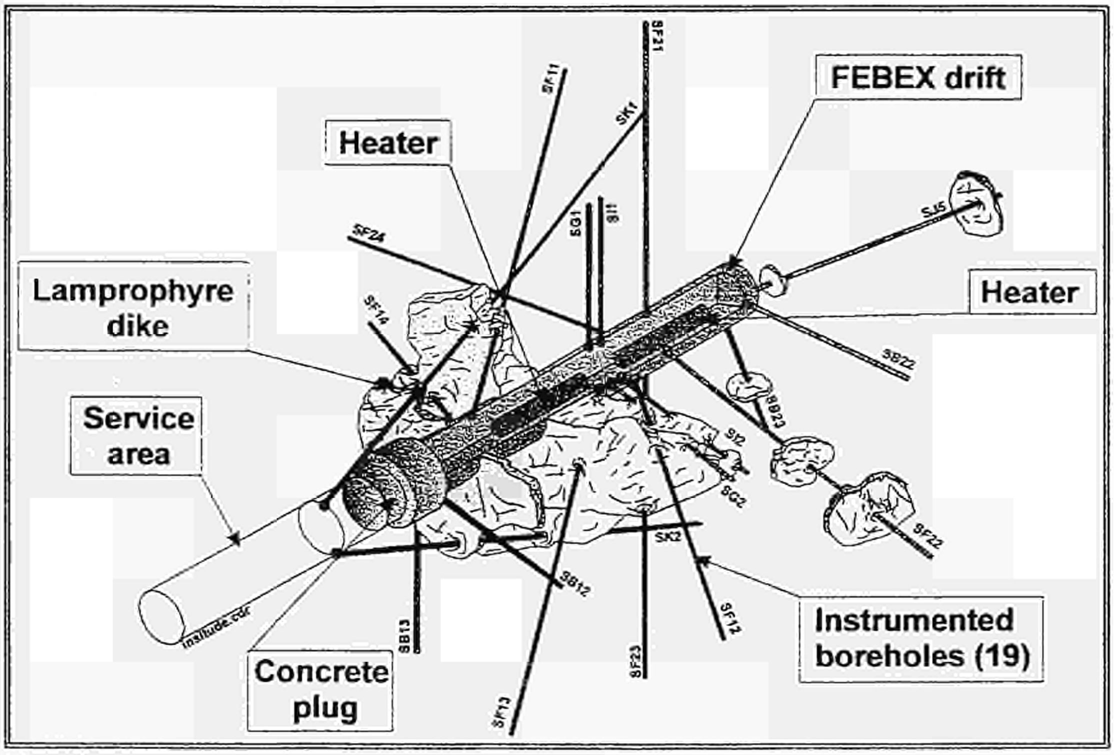

Figure 1: Schematic diagram of the "in situ" test.

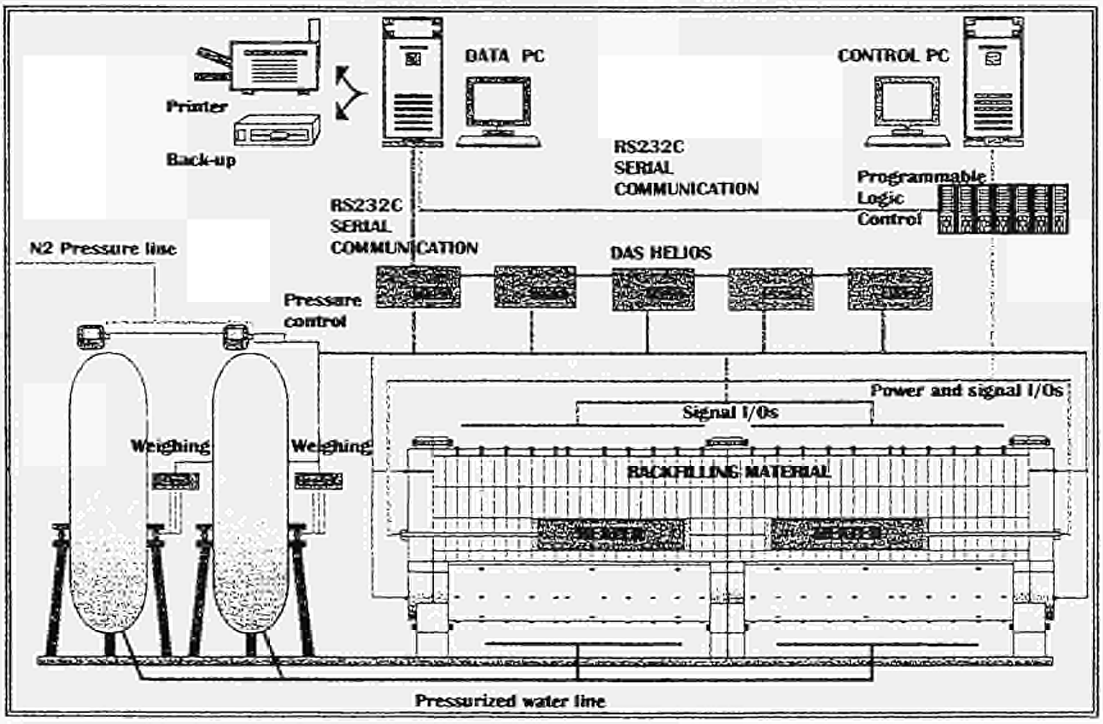

Figure 2: Schematic diagram of the "mock-up" test

C.2.2-1 BAMBUS : Backfill and material behaviour in underground salt repositories

Contract No: FI4W-CT95-0009 **Duration:** 1 Jan. 1996 - 31 Dec. 1998
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Partners: BGR Hannover/DE, ECN Petten/NL, ENRESA Madrid/ES, GRS Braunschweig/DE, G.3S Palaiseau/FR, FZK/INE Karlsruhe/DE, Univ. UPC Barcelona/ES

A. OBJECTIVES AND SCOPE

In the concepts for disposal of radioactive waste, backfill is an important component in the multi-barrier system. Two different emplacement concepts were developed for disposal of heat generating waste in a salt repository. According to the drift emplacement concept, self shielded heavy casks are deposited in drifts. Immediately after deposition of the casks the empty drift volume is backfilled. In the borehole emplacement concept waste canisters are deposited in boreholes about 300 m deep. The annulus between waste canisters and borehole wall is backfilled. In order to isolate the waste canisters from the drift above, after filling of a borehole the remaining space above the canister stack is also backfilled.

Crushed salt was identified to be the most suitable sealing material.

The objective of the project is to investigate important phenomena and processes in backfilled drifts and boreholes and to increase the data basis required for repository design and safety assessments. The data acquired by the tests will be used to validate and develop further the constitutive equations used in computer codes to predict the mechanical and hydraulic behaviour of the backfill material in the repository and to assess their predictive capability.

B. WORK PROGRAMME

B 1. In-situ investigations (GRS, BGR, ECN, FZK/INE)
- 1.1. Backfill in drift emplacement.
- 1.2. Sealing of emplacement boreholes.
- 1.3. Backfill in emplacement boreholes.
- 1.4. Convergence of non backfilled emplacement boreholes.

B 2. Laboratory investigations
- 2.1. Backfill compaction tests in triaxial cells (GRS, FZK/INE, G.3S).
- 2.2. Backfill permeability tests (GRS, G.3S).
- 2.3. Backfill thermal property measurements (G.3S).
- 2.4. Creep tests on rock samples (BGR).

B 3. Benchmarking Crushed Salt (BGR - coordinator)
- 3.1. Verification exercises on theoretical examples.
- 3.2. Validation exercises on specific laboratory tests.
- 3.3. Validation exercises on in-situ tests.

B 4. Model calculations (GRS, ECN, G.3S, FZK/INE, UPC, ENRESA/CIMNE)
- 4.1. Borehole convergence and backfill compaction.
- 4.2. Hydraulic conditions in backfilled boreholes.
- 4.3. Temperature distributions.
- 4.4. Thermo-hydro-mechanical calculations.

C. Progress of work and results obtained

Summary of main issues

In Work Package 1 the in-situ investigations related to drift emplacement and to non backfilled emplacement boreholes were continued according to the time schedule by measurements in the experiments already operating. The activities related to sealing of emplacement boreholes and backfill in emplacement boreholes were started by test-field preparation, design calculations, and measuring equipment design and fabrication. In the preparation of the experiment „Development of Borehole Seals for High-Level Radioactive Waste“ (DEBORA 1, Task 1.3) a delay of several months occurred due to unexpected problems during the preparatory phase. The experiment will be started in January 1997.

In Work Package 2 laboratory investigations on backfill compaction were performed at different boundary conditions in laboratories of three participants. Thermal conductivity measurements were performed in one laboratory. Backfill permeability and rock creep tests will be started in 1997.

In Work Package 3 (Benchmarking Crushed Salt) the work in the first stage (Verification exercises on theoretical examples) was completed. The results will be presented in 1997. For Stage 2 (Validation exercises on specific laboratory tests) the tests, to be carried out in different laboratories, were specified. Six participants are taking part in this exercise.

In Work Package 4 model calculations were carried out for the tests on borehole convergence and backfill compaction and on the hydraulic conditions in backfilled boreholes. For the operating drift emplacement experiment, temperature and THM calculations were performed.

Progress and results

C.1 In-situ investigations (Work Package 1)

The general objectives of Work Package 1 are to

- improve understanding of thermomechanical processes in backfilled drifts, boreholes, and in the host rock,
- quantify properties and behaviour of backfill and rock and the interactions between them,
- develop constitutive models for repository performance assessment,
- qualify numerical models used for repository performance assessment.

The Work Package consists of four tasks as named in Section B.

The geotechnical measuring programme of the TSS test (Thermische Simulation der Streckenlagerung, Thermal Simulation of Drift Emplacement) (Figure 1) has been continued. The TSS test was started by turning on the heaters in the six heater casks in two test drifts on 25 September 1990. Since the renovation of the heater control system in 1995, the heaters are operating without any problems with the designed thermal power output of 6.4 kW for each heater cask.

The temperatures at the heater cask surface are ranging between 165°C and 175°C. Temperatures of up to 100°C are recorded at the drift walls and up to 93°C at the roof above the casks. In a depth of 0.3 m below the heaters, temperatures of approximately 145°C are measured, decreasing to about 108°C in 1.2 m depth. In the pillar between the heated drifts the actual temperatures are reaching 81°C in the pillar centre. In the almost non-heated area 12 m away from the heaters the temperatures have increased up to 47°C.

The drift convergence rates, which in the beginning of the experiment in 1990 had been accelerated to a maximum of 3.5 %/a, are further reduced due to the increasing support by the backfill. In the heated area convergence rates in vertical direction are now approximately 0.6 %/a, whereas horizontal rates of about 0.5 %/a are measured. In the non-heated area actual rates are about 0.4 %/a and 0.3 %/a in vertical and horizontal direction, respectively. To compensate for failed convergence measuring gauges a replacement of horizontal convergence measuring gauges will be carried out.

The initial backfill porosity of about 35 % has been reduced to 25.5 % to 27 % in the heated area as a result of drift closure. In the non-heated sections the actual backfill porosity ranges between 31.4 % and 32.8 % (Figure 2).

The pressure between backfill and surrounding rock is continuously increasing due to drift convergence. In the heated area the maximum pressure at the roof is now 3.2 MPa. This value corresponds to 26 % of the initial vertical stress, which has been estimated at about 12 MPa in the test field. In the non-heated backfill the current pressure ranges between 0.13 MPa and 0.20 MPa.

The rock pressure is recorded since 1995 by additional stress monitoring units which are replacing some initial gauges after they had failed. After the improvement of the gauge inclusion in the host rock by a subsequent injection of epoxy resin in spring 1996, the pressure is rising continuously. Current values range between 3.6 MPa to 8.5 MPa in the pillar and 1.8 MPa to 3.6 MPa above the heated drifts. For the major part of measurements the gauges had not to be replaced. There the current pressure ranges between 6 MPa (inclined, 45°) and 14 MPa (vertical).

Gas concentrations in the backfill pores are being measured regularly in both drifts. Before heating was started, at the ambient temperature of approximately 36 °C the concentrations of the major gas components hydrogen, methane and carbon dioxide in the backfill were 28 to 44 vpm, ≤ 4 vpm and 35 to 75 vpm, respectively. Significant gas release started immediately after the heaters were switched on. Within six months the gas concentration increased to 550 vpm hydrogen, 40 vpm methane and 3000 vpm carbon dioxide. As a result of atmospheric pressure changes and variations in the mine ventilation the gas concentrations are constantly diluted due to the high porosity and permeability of the backfill. Consequently, the concentration of hydrogen, methane and carbon dioxide decreased after one year of heating, indicating that the gas production was lower than the escaping amount of gases. In order to determine the total amount of gas generated, one of the two test drifts was sealed at its entrance in February 1996. The last measurements in October 1996 showed an increase to 7830 vpm carbon dioxide and 334 vpm hydrogen.

A humidity of 35 g water per m^3 air was measured in the air from the backfill pores. In the access drifts a humidity of 7.5 g water per m^3 air is measured at 1096 hPa and 36 °C.

The investigations related to sealing of emplacement boreholes (Task 1.2) will be performed in the DEBORA 2-experiment which according to the actual time schedule will be started in the first half-year of 1997. Work for the test preparation is on schedule.

Investigations related to backfill in emplacement boreholes (Task 1.3) will be performed in the DEBORA 1 experiment (Figure 3). The preparation works for the conduction of the in-situ test in the Asse salt mine were started in January 1996. First, the liner was withdrawn from the borehole B2 in the former HAW test field. Afterwards, the diameter of the lower part of the heater borehole was enlarged from 500 to 600 mm and the inclined borehole for gas injection into the lower part of the borehole was drilled. The liner was provided with the instrumentation for borehole convergence, backfill pressure, and backfill temperature measurements. Prior to the re-installation of the instrumented liner, the pressure and temperature transducers in the gas injection volume were installed. The equipment for the flow experiments, the gas supply and gas flow monitoring station, and the data collection system were installed in November 1996. Calibration of the measurement systems was performed in December 1996.

In this test the corrosion behaviour of five candidate materials for long-lived HLW/Spent Fuel disposal containers will be investigated under conditions prevailing in the repository operating phase. Three material conditions will be investigated: hot rolled and normalized, TIG-welded, and EB-welded. The material specimens were prepared and characterized by gravimetry, surface profilometry, and metallography. The specimen carrier was constructed and, together with the 72 specimens, fixed on the borehole liner.

Up to now a total delay of approximately eight months is to be noted in the DEBORA 1 schedule so that the test can not be started before January 1997. The complete testing strategy, however, can be kept unchanged because the time span until the end of the project in 1998 is long enough to conduct the test as planned.

The investigations on convergence of non backfilled emplacement boreholes (Task 1.4) were continued by measurements at four levels in a 500 m deep borehole. This borehole was drilled in early 1994 in the Asse salt mine, far away from former production rooms. Thus, the rock pressure near to the borehole can be assumed to be equal to the lithostatic rock pressure. The measurements were compared with extrapolation calculations by use of different rock mechanical theories.

C.2 Laboratory investigations (Work Package 2)

The general objectives of Work Package 2 are to specify in detail specific backfill material parameters and to support interpretations of the in-situ measurements. The work package consists of four tasks (see Section B).

Backfill compaction tests in triaxial cells were performed by FZK/INE, G.3S, and GRS. Their aim was to get additional data for a better understanding of the material behaviour at higher temperatures and compactions, and at slow consolidation rates. The data are needed to establish a constitutive law for crushed salt as used as backfill material in repositories.

FZK/INE used a specially designed testing device with a cubic test cell with 250 mm side length. The triaxial stress is applied by six hydraulic pressure pads, the maximum temperature is 200°C. Three tests were performed at different temperatures, pressures, consolidation rates, and initial porosities. Comparison of the measured and calculated consolidation rates shows a good agreement (Figure 4). In one of these tests 1% water was added to the crushed salt. In this test consolidation rates were higher by factors from 60 to 600 as compared to dry material. The stress exponent of the consolidation rate is 2 (compared to about 5 for dry material).

G.3S performed a test series in three different test cells. This series comprised measurements of the ratio lateral stress/axial stress, relaxation behaviour of the crushed salt, and deviatoric stress at different initial porosities. This first test series was performed at room temperature. The results show that the ratio lateral stress/axial stress remains almost constant and equal to 0.4 at porosities greater than 10% and that relaxation depends highly on porosity.

GRS performed short term uniaxial compression tests for determining specific material parameters in the constitutive equation developed by Hein [1] which is used for design calculations for the in-situ experiments. The measured parameters were implement in the material law and used for design calculations. According to these calculations the borehole convergence is considerably smaller than predicted using the parameter values proposed by Hein. For testing crushed salt containing larger grains, the big triaxial cell (sample diameter 280 mm, sample length 700 mm) was prepared. First tests were performed up to a confining pressure of 50 MPa.

Backfill thermal property measurements were performed by G.3S at different stages of compaction. According to preliminary results the evolution of thermal conductivity with porosity seems to be linear in the range of investigated compaction (porosity greater than 10 %). (Figure 5)

C.3 Benchmarking Crushed Salt

A benchmarking exercise called „Comparative Study on Crushed Salt“ (CS^2) is being performed in order to validate and qualify the constitutive models developed by the participants. The exercise consists of three stages (see Section B).

In the reporting period, Stage 1 (Verification exercises on theoretical examples) was performed and completed. Six participants verified their codes and constitutive models (Table I) on the basis of two different theoretical tasks in which the backfill stresses and porosities over time should be calculated under different conditions in backfilled boreholes. The backfill performance could be derived from laboratory data. The comparison of the calculational results revealed a few deviations caused by use of different types of constitutive models. Some deviations were caused by different interpretation of the laboratory data. These deviations could be eliminated after a discussion in the working group.

C.4 Model calculations

Calculations on borehole convergence and backfill compaction were performed by ECN, GRS, and UPC. ECN used the code ANSYS for a pre-test analysis of the entire test field including galleries and boreholes. The calculations were aimed at the structural analysis including convergences, temperatures, and stresses over time for the test field which was excavated in the years 1985 to 1988 and in which one borehole was heated from 1988 to 1993. The calculational results show a good correspondence with the in-situ measurements. GRS used the code SUPERMAUS for pre-test calculations of the experiments DEBORA 1 and 2. Their aim was to predict borehole wall displacement and backfill porosity with different material parameters. In these calculations the material parameters measured in the laboratory investigations (Work Package 2) were used. The results will be used to choose the heater power for the experiment. By UPC the CODE BRIGHT was used. The calculations were aimed at predicting rock temperatures and backfill porosities over time and depth from the gallery floor.

Hydraulic conditions in backfilled boreholes were calculated by GRS and UPC. The transient gas flow in the DEBORA-1 and DEBORA-2 borehole was calculated by GRS with the code MUFTE. These calculations served as a basis for determining mass flux rates, total gas masses, and duration of the experiment. In addition, calculations for estimating the accuracy of the measurements to be performed during experiment operation and the impact of different experimental parameters on these measurements were performed. UPC calculated permeabilities over depth from gallery floor and time at different initial conditions. The calculations show that the impact of the heat transport by the gas is negligible for backfill porosity development.

Temperature distributions in the TSS test field were calculated by FZK/INE with the finite-element code FAST including a new three-dimensional test-field model. Temperature-dependent heat conductivities and heat capacities of backfill material and rock salt were used. The calculational results show a good agreement to the in-situ measurements. Therefore, these calculations will be finished.

For thermomechanical calculations on drift emplacement performed by FZK/INE, the finite-element code MAUS was applied with a more precise element discretisation of the drift shape and a larger rock volume around the experimental drifts than in the models used before. The calculational results for drift closure, backfill compaction pressure and backfill porosity show some discrepancies to the measuring results. Their reason will be analysed. For this purpose sensitivity analyses are being performed. ENRESA performed a preliminary analysis of the drift convergence with CODE-BRIGHT. A new mesh including the entire test field has been developed for these calculations.

References

[1] HEIN, H J, Ein Stoffgesetz zur Bestimmung des Thermischen Verhaltens von Salzgranulat, Thesis RWTH Aachen, 1991

Table I: Participants, codes, and constitutive models in Stage 1 of the benchmarking exercise

Participant	Code	Constitutive Model
GRS (subcontractor DBE)	MAUS-2/ -5	Hein (deviatoric) Korthaus (hydrostatic)
ECN	ANSYS EMOS_ECN MARC	Spiers
FZK/INE	MAUS ADINA	Hein (deviatoric) Hein (deviatoric)
G.3S	GEOMEC	Cam-Clay modified
UPC	CODE BRIGHT	FADT + DC
BGR	ANSALT I	Zhang/Heemann (deviatoric)

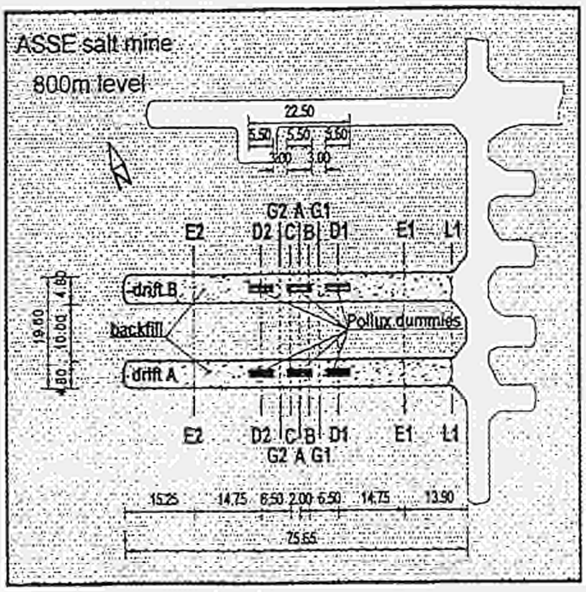

Figure 1: Plan view of the TSS test field with two test drifts with three heater casks each

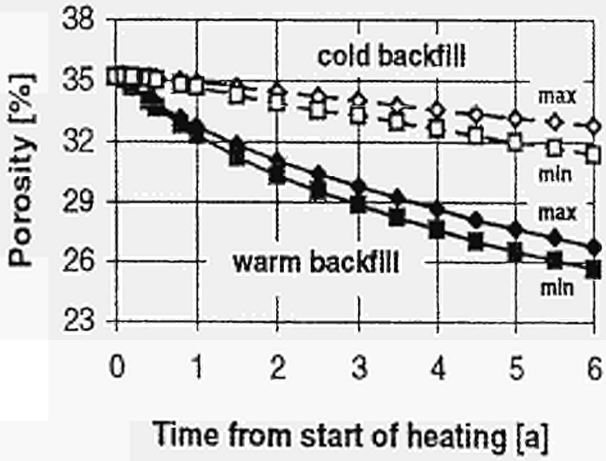

Figure 2: Backfill porosity in TSS test drifts over time

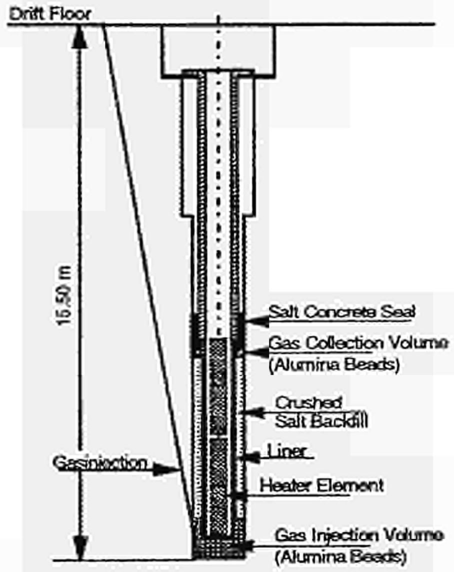

Figure 3: Section through borehole DEBORA 1

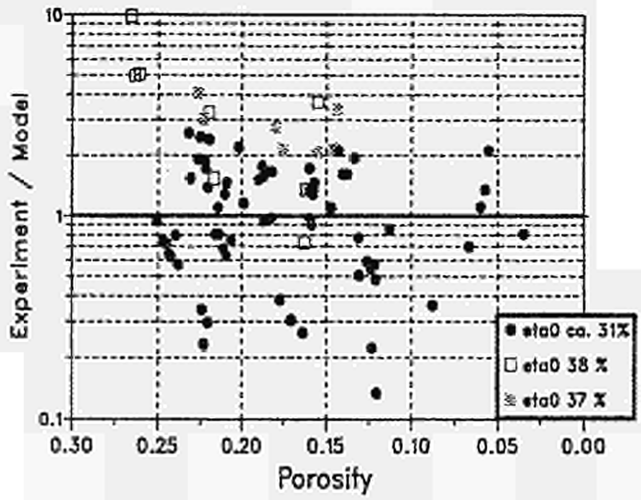

Figure 4: Comparison of measurements and calculations of backfill consolidation rates

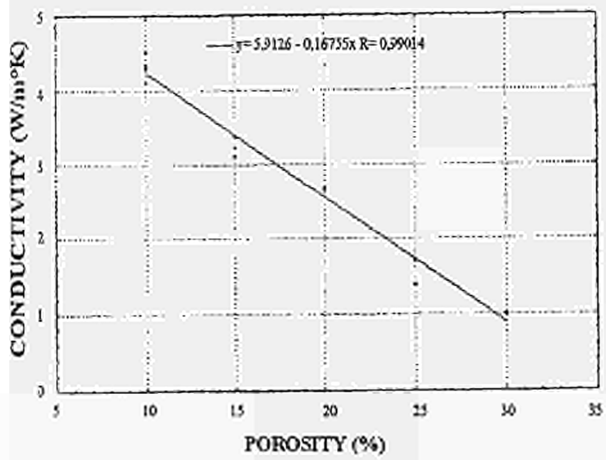

Figure 5: Backfill thermal conductivity over porosity

C.2.2-2 RESEAL : A large scale in-situ demonstration test for repository sealing in an argillaceous host rock

Contract No: FI4W-CT96-0025 **Duration:** 1 May 1996 - 31 Oct. 1999
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Partners: **ANDRA Chatenay-Malabry/FR, ENRESA Madrid/ES**

A. OBJECTIVES AND SCOPE

For the long term performance of a HLW repository, effective backfilling and sealing of the shafts and connection galleries is needed to avoid preferential pathways for the migration of water, gas and radionuclides. Therefore the demonstration of the feasibility of this sealing on a representative scale in-situ is essential and will be performed in the HADES facility at Mol.

The objectives of this large scale in-situ test are :

- to demonstrate the installation technique(s) for the backfilling and sealing of a shaft on a representative scale;
- to demonstrate that a low permeability seal to avoid preferential migration of water or gas along or through the shaft lining or even through the excavation damaged zone of the host clay formation, can be realized;
- to validate models for the transfer of gas and water through the sealing system including geomechanical aspects;
- to demonstrate the feasibility of the sealing of a borehole;
- to demonstrate the mechanical stability of the seal under accidental overpressure conditions.

B. WORK PROGRAMME

B 1. Laboratory experiments

* Definition of a procedure for the production and installation of the seal (ANDRA-CEA).
* Measurement of the transport properties of candidate sealing materials (SCK/CEN - ANDRA/CEA).
* Investigations on gas migration in clay based backfill and sealing materials (SCK/CEN - ENRESA/CIEMAT).
* Measurement of geomechanical properties (ANDRA/CEA - ENRESA/CIEMAT/UPC).

B 2. In-situ borehole experiments

* Preliminary small scale in-situ borehole sealing tests (SCK/CEN).
* Further use of the BACCHUS 2 experiment (SCK/CEN).

B 3. The large scale in-situ sealing demonstration test

* Design, instrumentation and installation (SCK/CEN - ANDRA/CEA - ENRESA/CIEMAT).
* Host rock instrumentation (SCK/CEN).
* Follow-up of the test (SCK/CEN).

B 4. Modelling

Hydro-mechanical modelling will be performed by all partners using codes such as NOSAT, CATSEM 2000 et al.

C. Progress of work and results obtained

Summary of main issues

A method for the production of the granular sealing material has been defined and successfully tested on a French and a Spanish bentonite. With this method regular pellets with a density of about 2 g/cm³ were obtained.

Several experimental set-ups have been constructed to measure the hydro-mechanical and gas transport properties of the candidate seal materials. The laboratory experiments have been started and the first results were already obtained. The hydro-mechanical behaviour of a saturated sample of a pellet/powder mixture is similar to this of a saturated sample of pure compacted powder.

Water injection tests were performed on two central filters of the BACCHUS 2 in situ experiment. The measured hydraulic conductivity's are respectively 2.2 10^{-11} m/s and 2.3 10^{-11} m/s indicating the homogeneity of the saturated Boom clay pellet/powder mixture.

A first draft of the design of the small scale bore hole sealing test and the large scale shaft sealing test has been drawn up and is currently being discussed with the modelling groups. Special attention has been given to the development of the in situ instrumentation.

In preparation of the simulation of the new in situ tests, UPC has continued its effort to simulate the three phases of the BACCHUS 2 experiment. The ANDRA/CEA modelling group has started to incorporate the constitutive laws for the hydro-mechanical behaviour of unsaturated swelling clay into the CATSEM 2000 code.

C.1. Laboratory experiments

Definition of a procedure for the production and installation of the seal

The CEA/DESD/SESD has performed a series of dynamic compaction tests to produce pellets with a rectangular (candylike) shape 24mm long to 15 mm wide. Tests were performed for different initial water contents on both candidate sealing materials i.e. the FoCa clay (a French bentonite) and the Serrata clay (a Spanish bentonite from Almeria). The obtained densities are given in Table I.

Transport properties of the candidate sealing materials

The CEA/DESD/SESD has started a first X-ray tomography hydration test on a FoCa clay pellet/powder mixture. In this test X-ray tomography is applied to measure the progression of the hydration front.

Gas migration in clay based backfill and sealing materials

CIEMAT has built an experimental set-up (Figure 1) to measure the gas permeability as function of saturation degree and dry density. The gas permeability of Serrata clay has been measured and a logarithmic relation between the gas permeability and the saturation index was obtained ($\log k\ (m^2) = -0.1042\ Sr\ (\%) - 6.725$ with $R^2 = 0.9018$).

The SCK•CEN is installing a new gas migration laboratory which includes a climatized room. Due to the moving of the laboratory equipment to the new laboratory no experiments could yet be performed.

Geomechanical experiments

At the CEA/DESD/SESD a large (120 mm diameter) swelling pressure cell has been constructed to perform tests on the powder/pellet mixtures.

CIEMAT has started suction controlled oedometer experiments on Serrata clay powder. In these experiments the suction-stress path, expected for the seal, is applied. An oedometer cell for samples of dimensions upto 100 x 100 mm in which water inflow/outflow, deformation and/or swelling pressure, and interstitial pressure can be measured has been designed and constructed by CIEMAT. This cell will be used to test the pellet/powder mixture.

The SCK•CEN has performed a first permeability/swelling pressure test on a 50/50 pellet/powder mixture of FoCa clay with a dry density of 1.6 g/cm³. The test was performed using a swelling pressure cell with 80 mm internal diameter. The obtained results (hydraulic conductivity 1.3 10^{-13} m/s and a swelling pressure of 3.6 MPa) correspond very well with those previously obtained by the CEA on compacted FoCa clay powder.

C. 2. <u>Work package 2 : In situ borehole experiments</u>

Preliminary small scale in situ bore hole sealing test

The preliminary design of the test has been continued and information concerning the filter tubes and sensors has been obtained.

Further use of the BACCHUS 2 experiment

Before the start of the water permeability measurements, first a thermal pulse test was executed to see the evolution of the thermal conductivity since the last measurement. The results show only small changes in the thermal conductivity. A first permeability test on the second filter section (filter PW09C, length 20 cm) of the central tube was performed from 30.09.96 to 07.10.96. The obtained hydraulic conductivity is 2.2 10^{-11} m/s while at the beginning of this year it was 3.2 10^{-11} m/s. Apparently some further consolidation of the backfill has occurred. After re-equilibration of the hydrostatic pressures a permeability test was performed on the upper filter section (filter PW11C, length 75 cm) of the central tube from 10.11.96 to 25.11.96. The obtained hydraulic conductivity is 2.3 10^{-11} m/s. This value is very close to that obtained on filter PW09C, indicating the homogeneity of the saturated backfill.

C.3. <u>The large scale in situ sealing demonstration test</u>

Design, instrumentation and installation

The CEA is continuing its efforts to qualify sensors for water content and total stress measurements. This includes mechanical, reliability and water tightness tests.

Host rock instrumentation

As the host rock instrumentation needs to be installed about 1 year before the seal installation starts, the SCK•CEN has concentrated its effort in this work package on the design of the host rock instrumentation. A system of radially installed mini piezometers with total stress sensors and of radially installed displacement transducers has been designed (Figure 2). The design of the piezometer is based on the BACCHUS 2 central tube but with a smaller diameter. The displacement transducer system is a new development and needs advance testing.

C.4. Modelling

UPC is continuing its analysis of the BACCHUS 2 test. It is attempted to simulate the three phases of BACCHUS 2 (natural hydration, artificial hydration and long term pore pressure equilibration) with the same data set. It is now clear that the deformation response of the host clay plays an important role in the third phase. A numerical back analysis of the pore pressures measured during this phase was done to obtain a better approximation of the mechanical parameters for the host clay. Figure 3 shows the time evolution of the pore pressure measured in situ in the backfill versus the back analysis results. Also the influence of the compaction state in relation with the swelling pressure exerted by the barrier was found to be important. Therefore suction controlled swelling pressure tests with measurement of lateral stress were performed on Boom clay at a dry density of 1.4 and 1.7 g/cm^3. The results of these tests are now contemplated to study the relationship between swelling pressure and long term stresses in the seal.

The ability of the numerical model to treat both the unsaturated and saturated conditions is enhanced by adapting the treatment of the gas mass balance equation, when saturated conditions are reached locally in the mesh and by assuring the continuity of the stresses between both states. The modifications are tested on simple cases.

ANDRA has chosen the CEA/DMT/SEMT as modelling group. This group will use the CATSEM 2000 code. First this code will be used for hydraulic and hydro-mechanical design calculations. The constitutive laws for the hydro-mechanical behaviour of unsaturated swelling clay will be integrated in the code.

Table I Results of the tests of the pellet production method on FoCa and Serrata clay

clay	water content (%)	apparent density (g/cm^3)	dry density (g/cm^3)
FoCa	7.73	2.09	1.94
FoCa	12.9	2.16	1.92
Serrata	8.06	1.97	1.82
Serrata	13.06	1.91	1.69

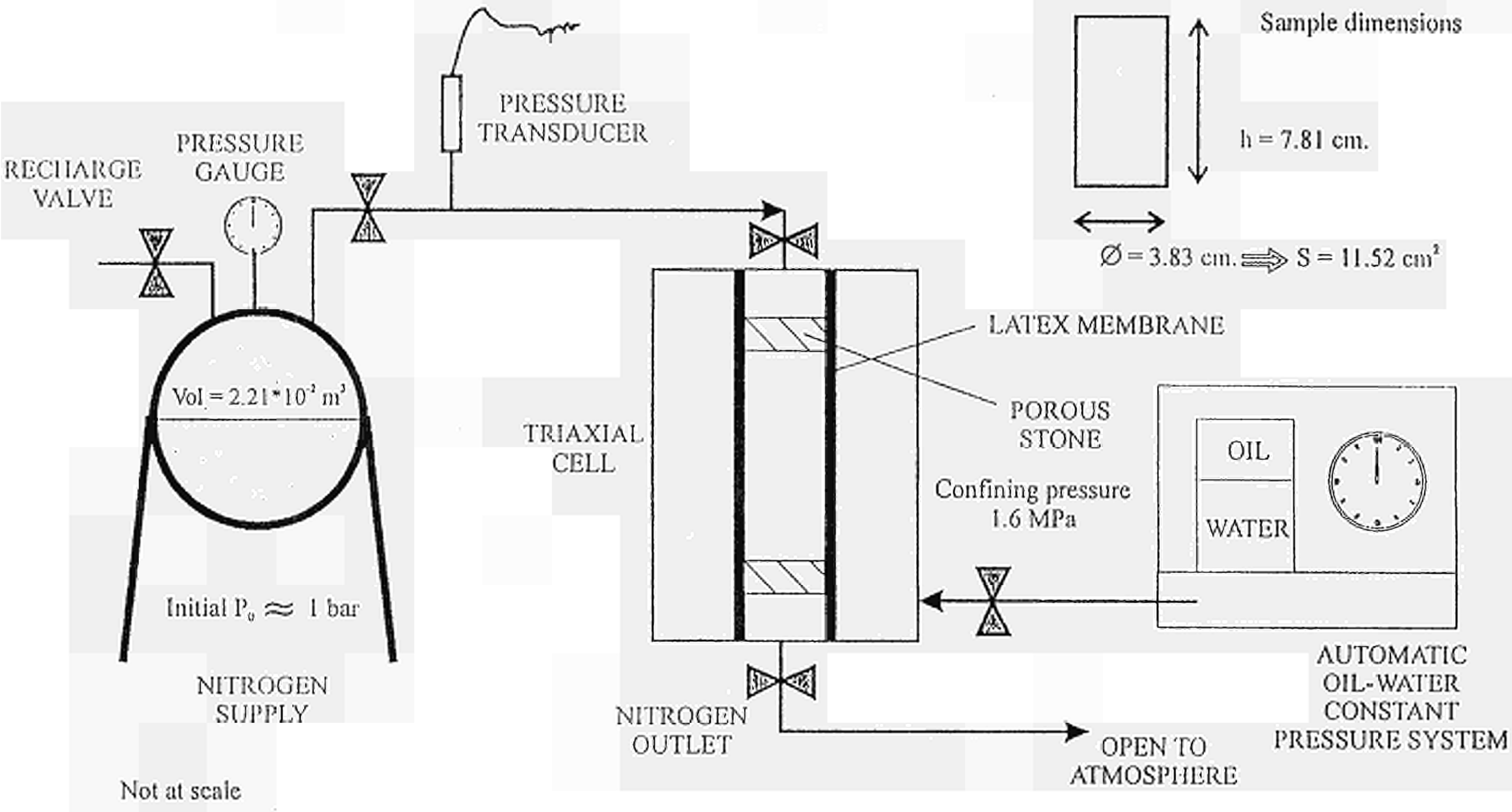
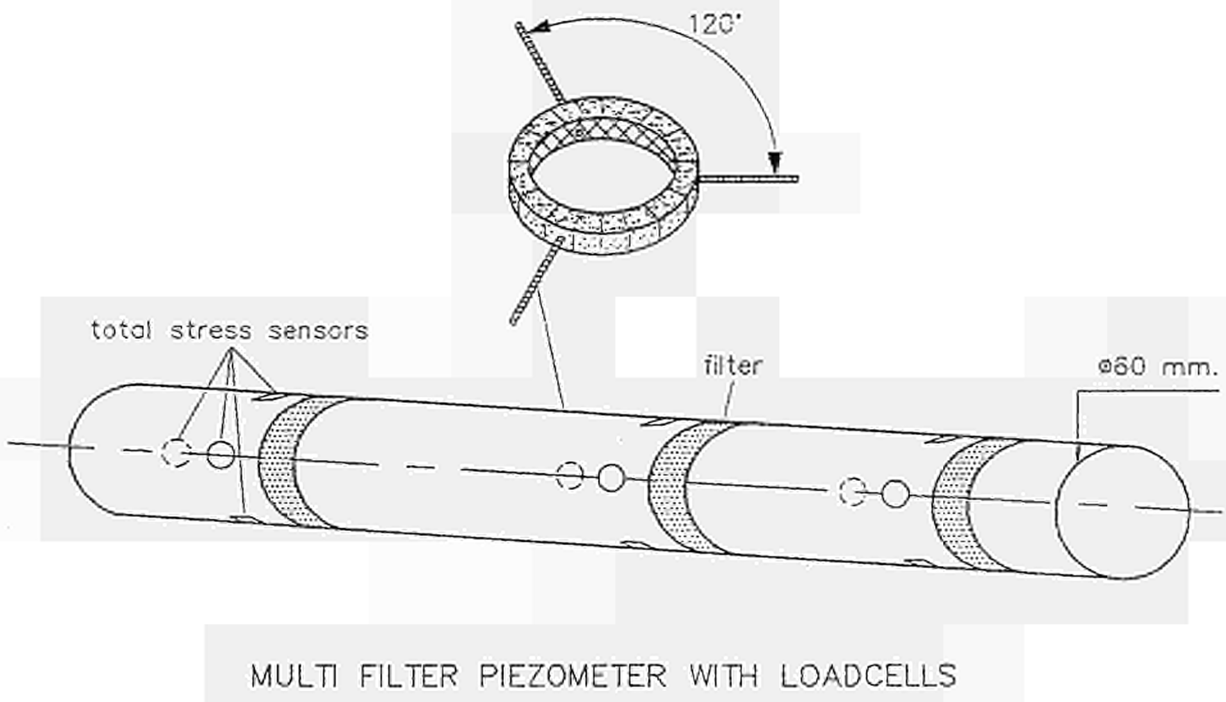


Figure 1: Scheme of the experimental set-up for the air permeability measurements.



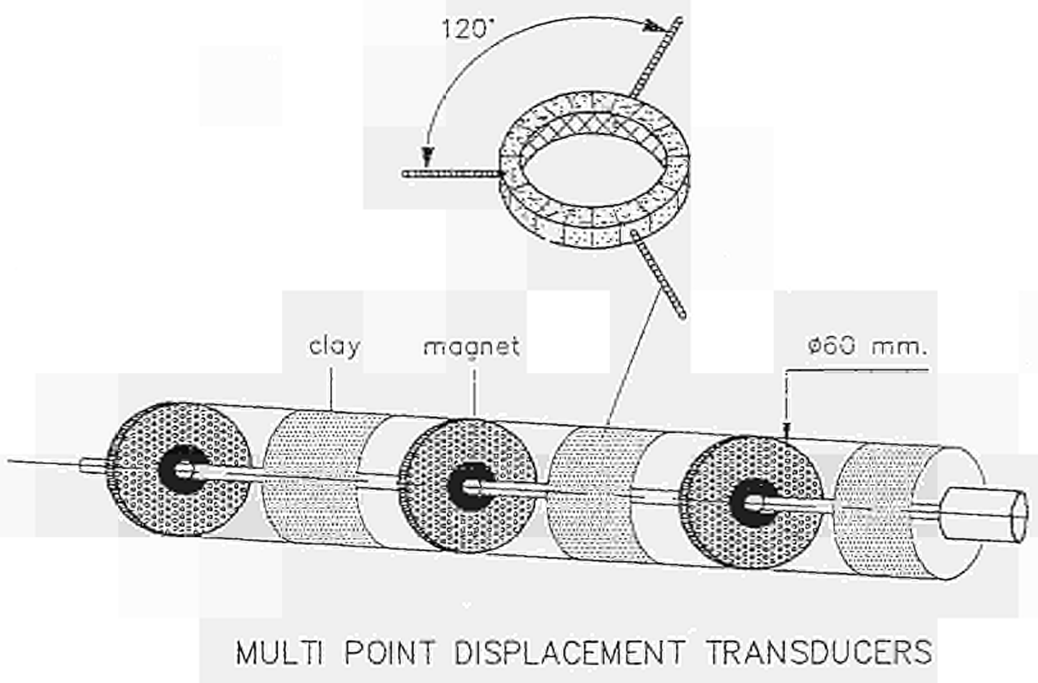


Figure 2: Design of the in situ host rock instrumentation for total stress, pore water pressure and displacement measurements.

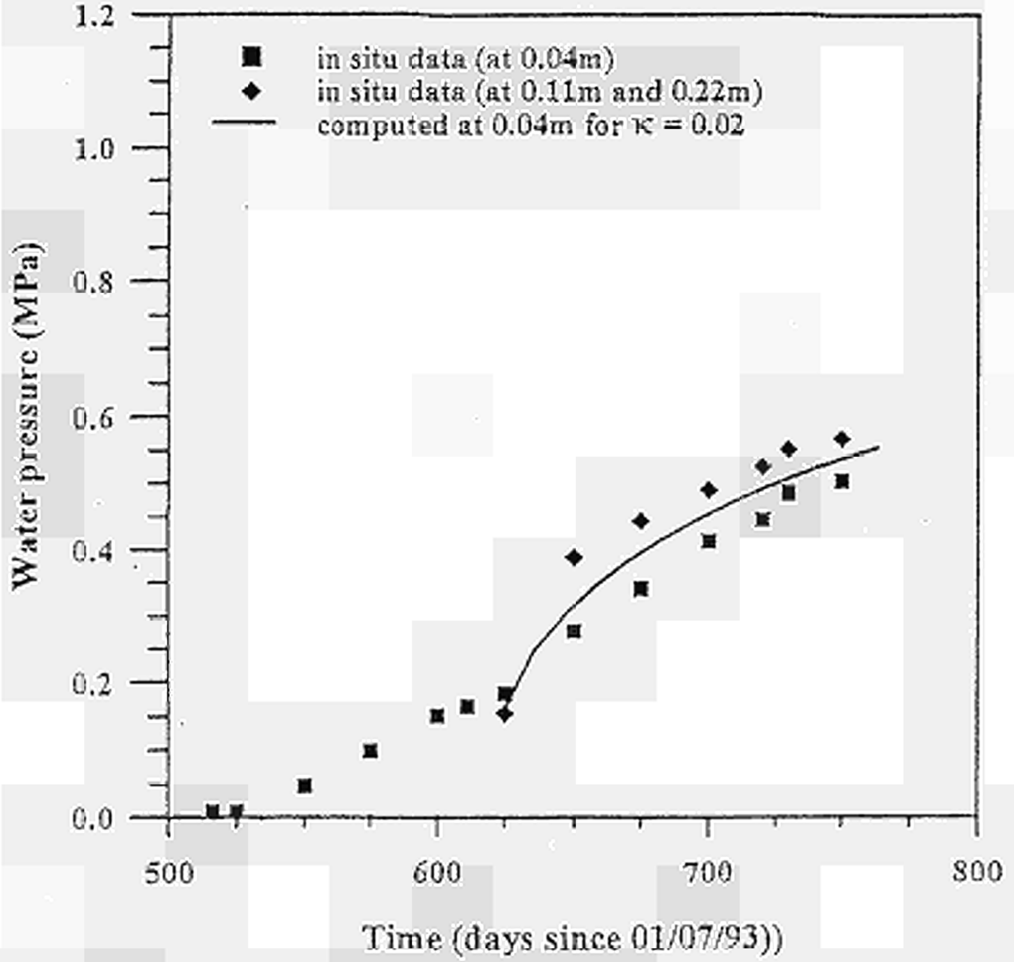


Figure 3: Experimental resluts versus the back analysis results for the pore pressure evolution in the backfill during the third phase of the BACCHUS 2 experiment.

C.2.3-1 CERBERUS Test Phase III : Study of the effect of heat and radiation on the near field of a HLW or spent fuel repository

Contract No: FI4W-CT95-0008 **Duration:** 1 Jan. 1996 - 31 Dec. 1998
Coordinator: **L. Noynaert, SCK/CEN Mol/BE**
Tel. +32/14.33.34.34 Fax: +32/14.32.03.13 e-mail: lnoynaer@sckcen.be
Partners: **ETREMAT Civaux/FR, Univ. La Coruña Madrid/ES, CEA Fontenay aux Roses/FR**

A. OBJECTIVES AND SCOPE

The SCK/CEN launched in 1989 the CERBERUS project aiming at the study of the effects of heat and radiation on the near field of a geological repository for HLW in clay.
A Cobalt 60 source of 397 TBQ and 6 heating elements with a nominal power of 500 W each were emplaced in a borehole in the HADES facility at Mol. In October 1994, the heaters were switched off and the Co-60 sources retrieved.
Moreover in the HADES facility heating tests have been performed such as CACTUS, Atlas and in connection with corrosion loops.
An extensive sampling programme around the before mentioned tests will be undertaken with a view to investigate long term behaviour of engineered barriers and host rock under repository conditions.

B. WORK PROGRAMME

B 1. Analysis of the behaviour of clay material under representative in-situ conditions and at various temperatures

* physical analysis of clay samples (SCK/CEN).
* chemical analysis of water samples (CEA).
* mineralogical analysis of clay samples (ETREMAT).

B 2. Synthesis of near field effects on argillaceous material and engineered barriers

Qualitative assessment of behaviour of waste matrix, canister material, buffer material and clay host rock (all partners).

B 3. Thermo-hydro-geochemical modelling

* improvement of existing models for simulating water flow, heat transport and multicomponent reactive solute transport (Univ. La Coruña-CEA).
* modelling of the CERBERUS test (Univ. La Coruña).

B 4. Preparation of data sets for performance assessments

Development of a well funded consensus on data sets to be used in performance assessments (all partners).

C. Progress of work and results obtained

- Summary of main issues

The sampling programme follows the planning of the *in situ* experiments. It concerns:

- 4 clay samples coming from the near field of the corrosion loops at temperatures of 80-90 °C during 4.3 to 7 year were analysed;
- 4 interstitial clay water samples coming from the near field of the *Atlas* test (Allowable Temperature Load of a Argillaceous Storage) at a temperature of 35 °C;
- 4 gas analyses of water samples taken around *Cerberus* test (Control Experiment for the BElgian Underground Storage), *Atlas* and piezometers.

Up to now, the results are in good agreement with the literature and particularly with the observations made in the framework of the *Archimede* (FI2W-CT92-0117)project. The extensive sampling programme around the *Cerberus* and the *Atlas* is planned for the second trimester 1997. Results of the analysis will be available during the second semester 1997.

The literature survey of the temperature effect on the behaviour of clay materials is almost completed. For the other near field effects, the study is still on going. We intend to present the synthesis in the next report.

The development of the thermo-hydro-geochemical model is on going. The results obtained up to now concern the thermo-hydraulic modelling of the *Cerberus* test and the conceptual development of the THG-model (i.e. surface complexion sub-model).

- Progress of work and results obtained

C.1. Analysis of the behaviour of clay material under representative in-situ conditions and at various temperature.

The sampling programme follows the planning of the *in situ* experiments. It concerns:

- 4 clay samples coming from the near field of the corrosion loops at temperatures of 80-90 °C during 4.3 to 7 year were analysed;
- 4 interstitial clay water samples coming from the near field of the *Atlas* test (Allowable Temperature Load of a Argillaceous Storage) at a temperature of 35 °C;

4 gas analyses of water samples taken around *Cerberus* test (Control Experiment for the

Excepted for one clay sample, the values obtained for the hydraulic conductivity, swelling pressure and volumetric swelling are similar to the values observed for unheated clay samples. Differences between two samples were observed for swelling argillaceous minerals, kaolinite and mica. Up to now, it seems that the heating at 90°C during 4 or 7 years did not induce significant transformation of the interstratified illite/smectite (R=0). Therefore one can conclude that the results are in good agreement with the literature and particularly with the observations made in the framework of the *Archimede* project.

Water samples were taken in the near field of the *Atlas* test to highlight the presence of major species and trace elements known as essential parameters of the equilibrium state (sulphur species, Si, B, Al). Samples were taken twice at the *Atlas* piezometer screens 85 and 93. Differences have been observed between 2 samples taken at the same screen, specially for the Cl, SO_4, Na and Ca contents. The analysis were repeated three times and the same results were obtained. The sampling will be performed again to avoid the possibility of a sampling aretefact. A deposit of grey colour was also observed and must be analysed with the ICPMS at SCK•CEN.

Hydrogen and methane analysis of water samples have been performed. No hydrogen was observed around the *Atlas* test or the *in situ* undisturbed clay host rock, but around the *Cerberus* test a concentration up to 7.55 μg H_2 kg^{-1} H_2O has been measured (supplementary agreement n°3 to the contract FI2W-CT90-0003). The amount of volatile organic carbon (mainly methane) in the *Cerberus* water reaches 2.2 ppm carbon, i.e. only 2 to 5 time higher than the values observed on blank samples.

The extensive sampling programme in the near field of the *Cerberus* test and the *Atlas* experiment is planned for the second semester 1997 i.e. respectively at the end of the hydration phase or the heated phase. Comparisons of the results obtained with clay samples and interstitial clay water submitted to various *in situ* conditions will highlight the behaviour of clay material under representative waste disposal conditions.

C.2. Work package B: synthesis of near field effects at various repository conditions

A significant effort has been made to review all the available information about the general hydrodynamic, thermal, and hydrochemical aspects of the *Hades* Project. Concerning the thermo-hydro-mechanical behaviour of the Boom clay, the work performed at SCK•CEN in the framework of a PhD on the influence of the temperature on the behaviour of clay materials will be used. The synthesis of the near field effects is still on going.

C.3. Work package C: thermo-hydro-geochemical modelling

The activities on thermo-hydro-geochemical modelling can be grouped in two parts. The first one deals with the formulation of the thermo-hydro-geochemical conceptual model, and the second with the extension and improvement of previously available ULC reactive solute transport codes.

Conceptual model development has been particularly concerned with the identification and evaluation of the most relevant geochemical processes taking place around the *Cerberus* experiment. They are thought to be the following:

- Dissolution/precipitation of mineral phases present in the Boom Clay;
- Cation exchange between Boom clay and pore water solution;
- Cation adsorption onto clay crystal surfaces;
- Evaporation/condensation of pore water;
- Increase in the ionic strength of the solution due to evaporation;
- Clay dehydration (close to the Co source and heaters);
- Gas generation (H_2 in particular) due to water radiolysis;
- Corrosion of some carbon steel parts.

Some of these processes have already been observed (H_2 production, for example) and others are common processes occurring in clayey environments. However, the extent of some of the processes might not be evaluated before the end and post-mortem analysis of the experiment. On the other hand, the effect of γ -radiation onthe clay rock (i.e. mineralogical transformations) is not thought to be relevant considering the low ionising energy of the Co source. Only temperature effects on the minerals are expected. Geochemical effects due to the degradation of organic matter present in the Boom clay as well as those by microbial activity are and, probably will remain, uncertain.

As stated in the Work Plan of the Project, ULC codes (mainly the TRANQUI code) for water flow and reactive solute transport need to be improved and extended in order to be applied in a meaningful manner for modelling the *Cerberus* test. The following aspects have been addressed during the first year of the Project:

- The mechanisms of water and ionising radiation interaction (radiolysis);

- Modelling of radioactive decay series;
- Improvement of the understanding and quantification of adsorption processes through the implementation of realistic surface complexion models. The Double Layer Model has been implemented in the TRANQUI code;
- Improvement of the thermal and hydrodynamic coupling.

Radiolysis has been proposed to be a significant source of H_2 which potentially could buffer the redox potential in the surroundings of the *Cerberus* experiment. Conceptually, radiolysis is a quite complex phenomenon, with generation of reducing (H_2) and oxidising species (H_2O_2) and needs to be understood in detail prior to account for in the model. Cobalt-60 decay does not result in important daughter products. Nonetheless, it is interesting to build into the modelling code a versatile tool to cope with more complicated (and realistic) radioactive systems. Surface complexion models (double and triple layer) offer a powerful approach to the understanding and modelling of surface reactions. The conceptual and mathematical basis necessary to implement double and triple layer models into the modelling code has been obtained. Work has also been done on identifying the list of model parameters required for the thermo-hydro-chemical (THG) modelling of the *Cerberus* test. This list includes the following parameters:

- Basic Boom clay parameters such as total porosity, bulk density, mineralogical composition (qualitative and quantitative);
- Detailed hydrochemical characterisation of clay pore waters, including pH, Eh, electric conductivity and major and trace elements;
- Hydrodynamic parameters such as laboratory and field hydraulic conductivity's (horizontal and vertical), specific storage coefficient (this could be derived from clay compressibility), and water flow boundary conditions (near-field and regional hydrogeological conditions in terms of water pressures);
- Solute transport parameters which include molecular diffusion coefficients for various chemical species, cinematic porosity, porosity structure of the clay (values of inter-aggregate or “mobile porosity” and intra-aggregate or “immobile porosity”), and solute dispersivities (longitudinal and transverse);
- Thermal parameters such as the isotropic thermal conductivity, clay heat capacity, heat dispersivities and initial temperature distribution;
- Chemical parameters including selectivity coefficients for major cations, cation exchange capacity, organic matter content and composition, sorption parameters (specific surface, equilibrium constants for surface complexes, double layer parameters), and a list of most likely redox processes.

C.4. Work package D : Preparation of a data sets for performance assessments

This activity is planned to start in 1998.

C.2.4-1 TRANCOM-Clay : Transport of radionuclides due to complexation with organic matter in clay formations

Contract No: FI4W-CT95-0013 **Duration:** 1 Jan 1996 - 31 Dec 1998	
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Tel. +32.14.33.32.21 Fax: +32.14.32.35.53 E-mail : mput@sckcen.be	
Partners:	**ARMINES Paris/FR, Univ. KUL Leuven/BE, Univ. Lough. Louhborough/GB**

A. OBJECTIVES AND SCOPE

Organic Matter (OM) may form stable complexes with many radionuclides and these complexes may serve as transport agents for the radionuclides. To quantify this influence the main objectives of the project are to obtain reliable transport models and migration parameters directly usable for the Performance Assessment calculations taking into account the influence of the OM on the migration of radionuclides in the Boom clay formation at the Mol site.

The study will investigate the migration of OM in the Boom clay by the use of C-14 or I-125 labelled OM extracted from the interstitial water of the formation and will consist mainly in laboratory and in-situ experiments followed by the modelling of the results obtained.

B. WORK PROGRAMME

B1. Laboratory work

WP 1 : Extraction, purification and characterization of OM (SCK/CEN, KUL)
WP 2 : Radiolabelling of the OM with C-14 or I-125 (Univ. Loughborough)
WP 3 : Laboratory batch sorption experiments with labelled OM (KUL)
WP 4 : Interaction with radionuclides with OM under reducing conditions (KUL)
WP 5 : Laboratory migration experiments with single and double labelling (SCK/CEN)

B2. In-situ validation experiments

WP 6 : Large scale in-situ injection experiments with C-14 labelled Boom clay OM (SCK/CEN)
WP 7 : Reference large scale in-situ injection experiments with non retarded tracers (SCK/CEN)

B3. Modelling

WP 8 : Modelling of batch sorption experiments with labelled OM and with radionuclides (ARMINES)
WP 9 : Migration experiment modelling (ARMINES)

C. Progress of work and results obtained

Summary of main issues

Organic matter has been concentrated from interstitial Boom Clay water. Characterisation of the concentrate is started. The concentrate has been covalently labelled with ^{125}I and ^{14}C. The stability of the label has been tested in absence and presence of Boom Clay, by batch and migration experiments. The ^{125}I label is unstable under Boom Clay conditions and will not be used in migration or batch experiments. The ^{14}C label showed a small instability and further testing of the stability is necessary.

The first distribution measurements of Eu and U under reducing conditions are started. Because of the anaerobic and reducing conditions, laboratory work will take longer than planned.

All single labelled migration experiments with ^{14}C-organic matter are started. The double labelled migration experiments will soon follow and preparations for the *in situ* migration experiment are well progressed.

Concerning the modelling, a coupled migration-transport model is currently adapted for diffusion driven transport of radionuclides. A first calibration of the model with a long term ^{134}Cs migration experiment is done.

C1. Laboratory work

WP-1: Extraction, purification and characterization of organic matter (KUL, SCK•CEN)

At SCK•CEN, different batches of organic matter (OM) from Extinction Gallery Bottom Shaft clay water (EG/BS) are concentrated by means of DEAE-cellulose [1]. The following batches were prepared in the framework of the TRANCOM-Clay project (see Table I).

Intact Boom Clay cores were also drilled, by SCK•CEN, from the underground installation. The samples were packed in PE/Al/PET foil (UCB) to avoid oxydation and confined in a stainless steel container at 4°C in the dark to avoid microbial growth.

At KULeuven, different methods to determine the functional group content of OM are tested:

1. The use of index cations (cobaltihexammine, cetylpyridinium and hexamethonium) is abandoned because these cations failed to flocculate humic acids.
2. An acid-base titration procedure to determine the " titratable acids" in the Boom Clay interstitial water is proposed. Validation of this method is under way.

Classical acid-base titrations are performed on purified humic acids, which were concentrated from the interstitial clay water.

WP-2: Radiolabelling of the organic matter with ^{14}C and ^{125}I (LU, SCK•CEN)

Labelling campaigns

To date three samples of concentrated clay water, have been radiolabelled at LU according to the procedure developed by Warwick *et al.* [2]. The first was labelled with ^{125}I (TROM-0) but the second (TROM-1) and third (TROM-6), for reasons detailed below, were labelled with ^{14}C.

The ^{14}C labelling procedure and the stability of the product were tested at the pore water pH of 8.5, rather than at the more routinely used value of 6.5.

Parallel experiments were also conducted to investigate possible oxidative degradation of the HA at pH = 8.5. Storage under nitrogen showed no obvious benefits.

Stability testing

Stability tests at SCK•CEN, both batch tests and percolation tests, showed that the ^{125}**I** label was not stable and continuously released at pH ~9. Different types of experiments have been set up to test the ^{14}C stability: batch experiments in absence (LU) and presence (SCK•CEN) of the Boom Clay, and percolation experiments through Boom Clay cores (SCK•CEN).

-Stability testing in absence of Boom Clay

A long term stability study was conducted on TROM-1. The sample was stored in the dark at 5°C and at pH 8.5 for 113 days. No evidence of label dissociation was observed.

-Stability testing in presence of Boom Clay

The ^{14}C activity of the percolated samples is below the detection limit of the HPSEC system. Labelled OM is set in contact with Boom Clay in a batch experiment under *in situ* conditions. The supernatant was analysed, and contained approximately 9% unbound ^{14}C methylamine and 91% ^{14}C labelled OM. Since 79% of the OM sorbed on the Boom Clay, it was surmised that the unbound ^{14}C methylamine fraction corresponded to 2% of the total activity. The second sample that resulted from an attempt to recover $^{14}CH_3NH_3^+$ sorbed on the clay, showed a small ^{14}C methylamine peak. At present it is not clear what causes the instability of the label in presence of Boom Clay.

WP-3: Laboratory batch sorption experiments with labelled OM (KUL)

Postponed until next semester.

WP-4: Interaction of radionuclides with organic matter under reducing conditions (KUL, SCK•CEN)

-Predictive calculations (KUL)

Based on literature data for the Eu^{3+} oxide (clay mineral) interaction and the Eu^{3+}-OM interaction, the Eu^{3+} speciation in the Boom Clay formation is predicted to be entirely due to complexation with humic substances.

-Distribution experiments (KUL, SCK•CEN)

The distribution of Eu^{3+} between Boom Clay and the solution in equilibrium with the clay was measured at different time intervals. Experiments were performed in anaerobic conditions in glove boxes, which have an atmosphere of nitrogen (94.6 %), hydrogen (5 %) and CO_2 (0.4 %). The partial pressure of CO_2 is similar to the one in the Boom Clay. The results are presented as *Kd* versus *absorbance* in Figure 1. It is clear that the Eu distribution between the solid and the liquid phase depends on the concentration of the dissolved organic matter in solution, which is leached from the Boom Clay. Increasing organic matter concentrations lead to decreasing Kd values because of the complexation of Eu with organic matter in the supernatant solution.

Attempts were undertaken to assess the solubility of U under Boom Clay conditions. If the solubility is unknown, one cannot make a distinction between precipitation and sorption from the distribution experiments. At present, Uranium is successfully reduced from U(VI) to U(IV) by means of a Walden reductor.

WP-5: Laboratory migration experiments with single and double labelling (SCK•CEN)

Migration experiments are performed in a room at 25°C and real clay water is forced through intact clay cores at a hydraulic pressure difference of about 1.5 MPa. The clay cores are confined in a stainless steel core holder, screwed with a momentum of 150Nm or consolidated at 2.7 MPa in Wyckham Oedometers.

-Single migration experiments

From the pulse injection experiments with ^{125}I-OM it was concluded that the iodine label is not stable at alkaline pH.

Pulse injection experiments with ^{14}C-OM of different size are started at this point (full range, MWCO < 1 000, MWCO > 100 000). The percolation cells are percolated with real clay water in equilibrium with a partial pressure of CO_2 equal to 0.04 %.Double labelled experiments

Before starting the double labelled experiments with ^{241}Am, we wanted to assess the maximum soluble concentration of americium in the solution. The experimentally determined solubility for Am^{3+} in Boom Clay interstitial water has a mean value of $1.4 \pm 0.1.10^{-6}$ M.

C2. *In situ* validation experiment

The *in situ* experiment with ^{14}C labelled OM (WP-6) is actually being prepared.

The large scale migration experiment with HTO, called CP1 (WP-7), is running for 9 years now [3]. The results correspond well with the MICOF model predictions.

A second reference *in situ* experiment called TRIBICARB (WP-7) is also started at SCK•CEN. Both HTO and $H^{14}CO3$- were injected.

C3. Modelling

To model the batch sorption experiments, or the distribution experiments (WP-8), it is decided to follow a phenomenologcial approach. Rather than characterising the solid phase in terms of the different constituents, different proceses will be modelled.

To model the migration experiments (WP-9), the computer model HYTEC will be used. HYTEC couples a migration model (including key-processes such as diffusion and radioactive decay) with a full geochemical module CHESS [4]. A more detailed description of the model is given in [5].

HYTEC has been extended and tested for diffusion driven transport, which is the main migration mechanism in clay systems. Radioactive decay has been introduced in the model. A first attempt has been made to apply the model on the existing data of a SCK•CEN experiment of ^{134}Cs diffusion in a Boom Clay plug.

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[5] van der Lee, J. (1997, January). HYTEC, un modèle couplé hydro-géochimique de migrqtion de polluants et de colloïdes. Technical Report LHM/RD/97/02, CIG, Ecole des Mines de Paris, Fontainebleau, France.

Table I: Batches of concentrated organic matter and their use

Batch number	Use
TROM-0[a]	Stability tests of ^{125}I labelled OM
TROM-1	Stability tests of ^{14}C labelled OM
TROM-2	Determination of functional group content: WP-1
TROM-3	Determination of functional group content: WP-2
TROM-6	Laboratory experiments: WP-3 and WP-5

a: The acronym TROM stands for TRancom Organic Matter

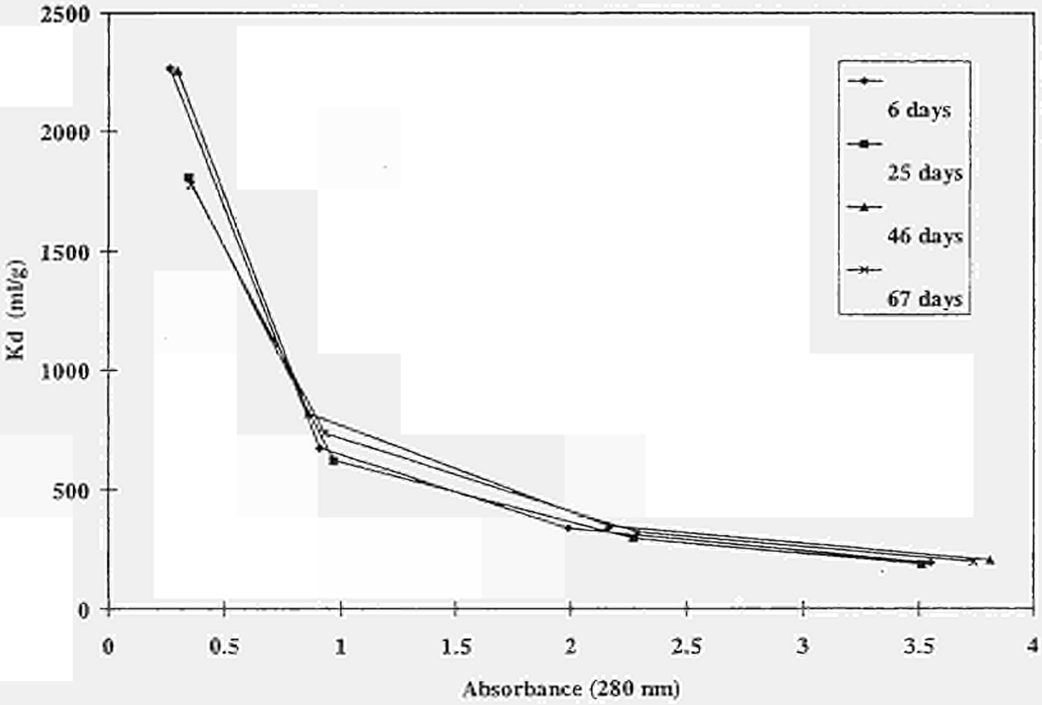

Figure 1: *Kd* versus *UV absorbance* (*A*) at different time intervals on unoxidised Boom Clay samples

C.3.1-1 Wet Oxidation Mobile Pilot Plant Demonstration on European Organic Radioactive Wastes

Contract No: FI4W-CT95-0005 **Duration:** 1 Jan. 1996 - 30 Jun. 1997
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Partners: **BELGOP Dessel/BE, GNS Essen/DE, NUCELEC Barnwood/GB**

A. OBJECTIVES AND SCOPE

The design construction and operation of an active mobile pilot plant using a hydrogen peroxide based on wet oxidation process for the treatment of radioactive wastes containing organics were carried out within the previous EC Research Framework Programme.

The objectives of the present project are to extend the active demonstration of the mobile wet oxidation pilot plant to a wider range of active organic wastes from other EU countries. An important aspect of the programme is to demonstrate that the plant can be set-up for operation on a new site with a minimum of delay, can be readily decontaminated between campaigns, and easily transported from site to site.

B. WORK PROGRAMME

B.1 Rig modifications

They are principally associated with improving techniques for pH and volume measurement, re-routing pipework, upgrading the heating system and the datalogging system. This work will be carried out by AEA Technology.

B.2 Operations with German waste

GNS will transport active resins arising from LWR to Winfrith in the UK, where AEA Technology will process them by wet oxidation. The final product will be cemented and returned to Germany for disposal, once the cemented waste has met German waste acceptance criteria.

B.3 Operations at a Nuclear Electric site

Demonstrate the mobile option through operations at a Nuclear Electric host site and determine typical support costs for the host site through actual operations. The waste to be used is approximately 1.5 m^3 of resin.

B.4 Operations at Belgoprocess, Dessel

The description of the task is the same as B.3, but the waste to be treated is up to 5m^3 of active effluents of different compositions.

B.5 Data assessment and final report preparation

C. Progress of work and results obtained

Summary of main issues

This programme of work has been considerably delayed due to the time necessary to obtain approval for the transfrontier shipment of radioactive waste. The waste, comprising three drums of ion exchange resin from Krümmel Nuclear Power Plant (NPP) in Germany, is required for treatment in WORK PACKAGE B.2, 'Operations with German waste'.

WORK PACKAGE B.1, 'Rig modifications', has been completed. This has involved the upgrading of several plant systems including reaction vessel contents monitoring, pH measurement, heating capacity, waste loading and emptying, and datalogging and process control. The mobile plant has been successfully re-commissioned and will commence operations with the ion exchange resins from Krümmel upon receipt.

Preparation for subsequent WORK PACKAGES B.3, 'Operations at a Magnox Electric site' and B.4, 'Operations at Belgoprocess, Dessel' is progressing. Safety documentation for operations at Magnox Electric's Oldbury power station has been prepared. Samples of aqueous organic effluents from Belgoprocess have been sent to AEA Technology for laboratory trials.

C.1 Rig modifications (s. B.1)

The first phase of the programme to enable the objectives to be met was the modification and re-commissioning of the ModulOx™ mobile wet oxidation plant. The plant had functioned effectively during the previous (3rd Framework) design and development programme, however it did not achieve its full design capability [1]. A number of modifications were therefore carried out to improve plant performance and to enhance operability. WORK PACKAGE B.1 took longer to complete than originally anticipated as decontamination and removal of the reaction system containment box was found to be necessary to allow some of the modifications to be completed. The removal of the containment required the disconnection of all electrical and pipework links to the reaction system.

The existing capacitance probe system which was used to measure the level of the contents of the reaction vessel was replaced with four load cells, one on each of the reaction vessel supports. This will remove errors in level measurement due to variations in the capacitance of the reaction vessel contents and enable better control of the reaction.

A new pH measuring system has been installed. This includes a new electrode that is better suited to operating at temperatures of up to 130°C. The electrode is mounted in a new stainless steel housing fitted in the flange originally used for the capacitance probe. The probe is connected to a new pH meter that incorporates a diagnostic system to monitor the status of the electrode. This system is also capable of calibration by sampling which eliminates the need for removal of the electrode during operational campaigns.

The plant feed system has been extensively modified to provide improved capability for loading 'difficult' materials into the ModulOx™ plant. The existing hopper and screw feed loading pump have been removed. A large peristaltic pump has been installed within the containment box and this will be used to load waste from an external feed station. This feed station is located at the open end of the ISO container and is contained in a purpose built ModuCon housing. The ModuCon containment is connected to the containment box ventilation system. Drums of waste will be placed on a roller - conveyor and moved manually into the ModuCon housing. The drum will be located in the feed station which will have lead shielding installed as required to reduce operator dose. Glove ports in the housing enable the drum lid to be removed by an operator standing outside the ModuCon. The feed system is then lowered pneumatically onto the drum. This system is equipped with a pneumatically operated stirrer and facilities for water addition and waste loading. The mechanism allows the drum to be lidded whilst the stirrer and loading lines are gradually lowered into the waste drum.

The reagent dosing systems have been modified to prevent blocking of the condensate line by lime slurry. All reagent lines have now been separated and no longer feed into the reagent vessel at the same point. Acid / catalyst and antifoam lines now enter the reaction vessel via the flange originally used for the pH probe. Each line is fitted with a short dip tube to ensure direct addition to the reaction vessel contents. Lime is added via the original feed line, however, a dip tube has been run through the condensate pot into the reaction vessel. These modifications separate the lime from the acid / catalyst, antifoam and condensate systems, removing the potential for blockage. In addition, an improved lime formulation has been identified. Kalic milk of lime has a very fine particle size and very slow settling rate. Its flow characteristics enable it to be handled as a liquid rather than a slurry and will reduce the possibility of blockages.

The steam boiler has been up-graded by the installation of a 30kW boiler to replace the existing 15kW system. This boiler is of the same make, design and operating pressure as the original boiler. The increased boiler capacity has resulted in reduced start-up times and improved control of the reaction product composition by distillation. A new steam control valve has been installed and will enable better control of reaction vessel temperature, particularly during batch addition.

The computer control logging system has been replaced by a PC / Windows based system to provide improve operator information whilst greatly simplifying the operation of the plant. Where possible, plant processes have been automated to reduce the requirement for operator intervention. In addition, the change to a PC / Windows platform will enable the system to be backed up to allow rapid recovery on computer failure.

In addition to the changes to major plant systems a number of minor modifications and general maintenance operations have been carried out. These have included the installation of sampling ports to enable secondary wastes to be analysed prior to discharge, the re-calibration of plant control and analysis systems and changes to the off-gas discharge arrangements.

Following the programme of modifications and maintenance, the plant systems were re-commissioned to confirm their operation. This functional checking programme took longer than originally anticipated due to the requirement to check all systems following removal and replacement of the reaction system containment box.

The plant has been re-commissioned operationally using non-radioactive powder ion-exchange resin combined with polyamide filter aid. This material was chosen as it was similar to the material identified for WORK PACKAGE B.2. This trial enabled the new plant systems to be tested under operational conditions.

C.2 Operations with German waste (s. B.2)

The completion of WORK PACKAGE B.2 has been delayed due to the unanticipated period required to obtain regulatory approval for the transfrontier shipment of three drums of radioactive ion exchange resin from Krümmel NPP from Germany to the UK. All necessary approvals have now been obtained and shipment of the waste is expected in the first half of February 1997.

C.3 Operations at a Magnox Electric site (s. B.3)

Treatment operations in WORK PACKAGE B.3 have not yet started due to the delay in the completion of WORK PACKAGE B.2. A suitable plant location has been identified at Oldbury power station and safety documentation for this phase of the work has been prepared.

C.4 Operations at Belgoprocess, Dessel (s. B.4)

The start of operations at Belgoprocess' Dessel site have been delayed. Preliminary preparation for this phase have started, including the supply of samples for laboratory trials.

References

[1] TWISSELL, M A, & HOLT, N S, Final Report, EUR 16776 EN, Luxembourg (1996)

C.3.1-2 REBONDIN - Reuse of concrete from decommissioned nuclear installations to produce radioactive waste storage packages

Contract No.: FI4W-CT95-0015	**Duration:** 1 Jan. 1996 - 31 July 1999
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Partners: KEMA Arnhem/NL, BNFL Sellafield/GB	

A. OBJECTIVES AND SCOPE

In a previous research contract No. FI2D-0015, it was demonstrated that coarse concrete constituants can be separated from fine ones and that particles > 1 mm can be reused as aggregate for concrete production; the mechanical properties of this concrete showed no deviations from normal concrete.

The objective of this research project is to determine the optimum recycling conditions for the fine aggregate fraction (about 50 wt % of the concrete mass) by heat treatment capable of producing a hydraulic binder comparable to cement, and to assess the decontamination effect obtained (up to 80 % is expected). Possibilities for reutilising the material in the nuclear industry include the filler, backfill or encapsulation material for waste containers in near-surface storage sites; the fabrication of concrete for certain radiological protection shields; the fabrication of waste storage containers; the construction of new facilities under certain conditions (possible task sequences are given in Figure 1).

B. WORK PROGRAMME

B.1. Concrete characterizations (CEA, BNFL) of the major types of concrete (limited to 9) dismantled in France and in the UK involving sampling in contaminated and radioactive areas.

B.2. Optimisation of the concrete separation based on 25 kg concrete lots of each designated type (KEMA)

B.3. Hydraulic binder reconstitution (CEA, KEMA)
Based on a fraction of each lot (B.2) and comparison with the usual concrete requirement for binders and also with specific ANDRA requirements.

B.4. Improvement of test procedure (KEMA, CEA), using the outcome from the tests conducted during steps B.2 and B.3.

B.5. Determination of radioactivity balance (BNFL) on radioactive samples and assessment of suitability for the thermal treatment (B.2) notably with regard to the volatile elements Cs and Pu.

B.6. Industrial fabrication specification (CEA) will be established, based on utilisation requirements of nuclear waste repositories, notably for possible use by the French ANDRA.

C. PROGRESS OF WORK AND RESULTS

Summary of Main issues

C.1 Concrete characterization (BNFL, item B.1)

Due to the large number of buildings on the Sellafield site, approximately 430 in total, the investigations were limited to the main *'ACTIVE'* buildings within 'the Separation area', 'the 'Calder reactors site', and the 'Thorp site'. Only buildings primarily constructed from concrete have been considered, cladded steel portal frame buildings have been discounted. Information related to concrete that has been collated in this period was obtained from the following sources;

(1) Specification for the concrete found in 'Engineering drawings'.

(2) Historical data cards from the 'Old concrete laboratories', for the testing of concrete cubes.

(3) Results from the Concrete laboratories computerised 'Concrete analysis system, version 2.03, Contact mix design'.

The classification of contamination within the buildings on the Sellafield site was limited to the broad definitions i.e. alpha / beta / gamma activity and was not concerned with particular radionuclides or their intensities. The volumes of contaminated concrete contained within the buildings have only been estimated for approximately 25 buildings due to the time and costs involved.

Concrete Survey

(1) The specifications for concrete mix design have changed 3 times since construction first started on the Sellafield site in 1947. The site was then known as the 'Windscale' site. The original specification was Code of Practice CP 114, which was replaced by CP 110, which in turn was replaced with British Standard's BS 8110 in association with BS 5328

(2) The most commonly used concrete on the Sellafield site from the 1960's to the mid-70's was concrete to CP 114 and CP 110, mix designs 1:2:4 for general construction or 1:1.5:3 for water-retaining constructions. These were 3000 $lbsf/in^2$ characteristic compressive strength grade at 28 days.

(3) The most commonly used concrete on the Sellafield site over the last 15 years is concrete to BS 8110 and BS 5328, grade 35B and 40B for general construction or grade 35BW for water-retaining constructions. These are 35 or 40 N/mm^2 characteristic compressive strength grade at 28 days.

(4) The vast majority of information available on the types of concrete that buildings were constructed from, came from the timespan 1980 to date.

(5) Very little information could be found on the concrete used to construct older buildings on site. This was the result of poor record keeping in concrete test data and Engineering drawing's either being lost or of so poor quality, that they cannot be read.

(6) Most of the concrete information retrieved from engineering drawings cannot be verified, as no laboratory test data can be found.

(7) For older buildings on site, the only information available is for the size of aggregate used; the type of aggregate is usually not specified.

(8) From historical data, it is known that the aggregate used in concrete on this site till the 1960's was either Granite or Whainston (an igneous rock, BASALT). After this time, 99 % of all the aggregate used was limestone.

(9) In the 1970's, as a result of the domination of the 'Ready-mix' concrete companies in designed mixes, there was an increase in the 28 day strength. This lead to micro cracking due to the higher rate of heat evolution during hydration. The introduction of Pulverised Fuel Ash (p.f.a.) or Ground Granulated Blastfurnace Slag (g.g.b.f.s.) as part

of the cementitious product elevated this problem. The first major building to use p.f.a. in the concrete was B 355 Pond 5.

(10) The only other addition to the aggregate used in early buildings was iron shot where high density concrete (typically 325 lbs/ft^3) was required, such as B 30 caves.

See Table I.

C.2 <u>Optimization of the Concrete separation</u> (KEMA, item B.2)

Cementstone agglomerates and particles of aggregates are present in the fine material (KEMA, 1994). The milling unit is mainly developed to separate the cementstone from the aggregates. In the fine material < 1.0 mm stone agglomerates and small particles of quartz were found. For the separation of the quartz and the cementstone agglomerates, further clinkerization can be important for the reuse of the cementstone.

From KEMA's research of an additional processing technique to separate and minimize the quartz aggregate content from the fine material, it was found that separation by density of the two different materials (cementstone, quartz) will be difficult. The cementstone and quartz have almost the same density, and through agglomeration of cementstone the separation efficiency could be influenced strongly (Table II).

<u>Electrostatic separation</u>:

There are three methods to charge and to separate the particles:

- corona charging (Ion bombardment)
- conductive induction charging
- contact or piezo charging.

<u>Corona</u>

Separation by means of a corona charging is most common. With corona charging, all particles are charged with the same polarity. By moving the particle on a grounded (neutral) surface, the conductive particles are neutralised and the separation can be realised between conductive and non-conductive particles.

<u>Conductive induction</u>

A conductive particle which is located on a grounded plate under an electric field will be charged by the polarity of the electrode. This charged particle lifts from the surface attracted to charged electrode. Non-conducted particles are not charged.

<u>Contact charging</u>

With contact charging or piezo charging the particle are charged by moving the particle along each other. When the particles enter a separator with a potential difference (between a negative and positive electrode), the particles are attracted to the electrode of opposite potential.

The current knowledge is inadequate for prediction of the charging of cementstone and quartz particles. Industrial implementation is necessary to assess the influence of the humidity, temperature and the size of the electric field (the polarity) and the extend of the charge.

From the results and observations, separation with corona charging (ion bombardment) and conductive charging are not a real option for further investigations. A small range in the granulometry improves the separation of different particles (cementstone and quartz) on specific gravity (density) and the dielectric value.

The observations have shown that the additional milling to break the cementstone agglomerates the particles by contact charging.

Generally, it can be concluded that contact charging is a favourable and potential option. Further experiments in combination with optimisation of contact charging by milling could result in a separation (efficiency) of the quartz and the cementstone from fine material (Figure 1).

C.3 Hydraulic Binder Reconstitution (CEA, item B.3)

Element Analyses

Element analysis is done by flame atomic absorption spectrometry after fusion of the untreated material with lithium metaborate and dissolving in acid (Table III).

Comments :

The K1 material, a DECO process material, shows the following characteristics :

- ⇨ Very low loss on ignition (1.60 %) indicating the absence of calcium carbonate, the result of heating at around 700°C
- ⇨ High silica content (77.23 %)
- ⇨ Low calcium and aluminum contents

These global figures, when placed in a ternary diagram (Figure 2) where the values of SiO_2, CaO and $Al2O_3$ are totalled to 100%, show that the positions of the K1 samples are way outside the limits for hydraulic or pozzolana binders.

The position of K1 in this diagram corresponds to that of a glass. Thus a high temperature treatment will melt the silica and give vitrified products with no hydraulic properties.

X ray diffraction analysis

Examination of the constituents by X-ray diffraction shows the presence of crystalline compounds (Table IV).

X-ray diffraction examination of composition shows :

- a high proportion of quartz in both samples
- the absence of calcium carbonate

Analysis of the soluble phase of K1 samples

Analysis of the soluble phase enables to estimate the quantity of cement in the samples. This is done based on their respective rates of soluble silica or by a differential calculation (Cement = 100: insoluble + loss on ignition), taking account of the insolubles and the loss on ignition which represent respectively the insoluble phase silica and the carbonates. The second calculation is used when the loss on ignition is low, as it is for the K1 sample.

Examination of the constituents of the soluble phase and their proportion enables us to see whether they have potential hydraulic qualities, this being related to the nature of the constituents, their proportions and their chemical structures (fig. 3).

The results of the previous analyses show the cement proportions (Table V).

The absence of water of hydration in the K1 previously heated to 700°C should be noted.

The position of the soluble phases in the ternary diagram shows that K1 is found in the hydraulic binder zone (figure 3) (Table VI).

These results show the need for separation of the constituents of concrete so as to isolate cement part or at least a part with a very high cement content. This would enable the temperature of clinkerization to be attained without vitrification of materials, as is shown in figure 3.

On another hand, a BNFL DECO-processed material gave the results as shown in Table VII.

D. MILESTONES AND CONCLUSION

Task B1 could be behind schedule. Getting the three active samples does not look so easy. If the Pu-contaminated sample is available, drilling the concrete of the ponds however to get other samples is difficult for safety reasons because their are all still in operation.

Nevertheless, BNFL hopes to get all three samples by mid-97.

B2 and B3 are for the moment on schedule.

Table I : Volumes of contaminated concrete contained within certain main buildings at Sellafield site (m^3).

Description	'P' Volume	'H' Volume	'M' Volume	'L' Volume	'I' Volume	Total
Pond 1	0.00	0.00	885.60	885.60	0.00	2793.63
F.H.P. Pond 50	0.00	0.00	35622.59	35622.59	2467.35	41207.17
Pond 2 & 3	0.00	0.00	1835.12	1835.12	1093.80	4941.41
W.V.P.	0.00	5094.84	8263.89	8263.89	13493.27	29796.44
H.A.L.S.T.	0.00	0.00	1318.53	1318.53	2648.15	24510.67
M.E.P.	0.00	0.00	1899.88	1899.88	3240.33	7896.08
2nd Primary Sep.Plant	64.03	0.00	1630.44	1630.44	1085.18	8838.57
THORP	0.00	3032.50	13454.46	13454.46	61717.92	87649.58
M.B.G.W.S.	0.00	0.00	5714.08	5714.08	2644.54	8912.90
Finishing Line 6	553.43	58.59	0.00	0.00	5515.73	6127.76
R & D Labs	766.56	63.91	0.00	0.00	5288.35	6962.67
D.G.P.	No Demolition	No Demolition	No Demolition	No Demolition	No Demolition	No Demolition
Tit. Evap.	No Demolition	No Demolition	No Demolition	No Demolition	No Demolition	No Demolition
Butex Recovery Plant	0.00	2268.35	2594.60	756.12	341.67	5960.73
E.A.R.P.	0.00	628.84	4116.12	1653.47	1208.98	7607.41
Pipe Bridges	0.00	0.00	239.98	6220.25	271.08	6731.31
L.A. Drain	0.00	0.00	942.30	2571.00	0.00	3513.30
S.I.X.E.P.	0.00	0.00	919.06	10244.25	8533.29	19696.60
Sea Tanks	0.00	0.00	0.00	4748.56	0.00	4748.56
W.E.P.	0.00	0.00	2888.20	0.00	11815.16	14703.36
Finishing Line 4 & 5	1199.02	0.00	0.00	455.32	680.35	2334.70
W.T.C.	663.34	0.00	0.00	1247.12	948.41	2858.86
Tank Farm	0.00	0.00	4941.20	256.07	0.000	5197.28
Pond 4	0.00	0.00	403.02	2323.75	1997.60	4724.37
Oxide Fuel Pond	0.00	0.00	0.00	5547.50	0.00	5547.50
UO_3 Store	0.00	0.00	0.00	0.00	5297.83	5297.83
Grand Total	4960.95	11147.02	69184.22	108962.53	142390.83	336645.56

The categories listed above are in accordance with current Decommissioning Section assumptions and modelling strategy :

Prefix	Category	Volume	Waste Route
P	Plutonium contaminated cells	100 % ILW	Interim store / Nirex
H	Highly active Beta Gamma cells	100 % ILW	Interim store / Nirex
M	Medium Active Beta Gamma cells	25 % ILW	Interim store / Nirex
		75 % ILW	Drigg
L	Low active Beta Gamma cells	100 % ILW	Drigg
I	Inactive areas	100 % ILW	Drigg (Free Release *)

*Low Level Waste (LLW) and what is understood to be Very Low Level Waste (VLLW) which includes building materials, steelwork, office furniture etc . originating from inside the separation area is currently sentenced to Drigg for final disposal.

Table II : Results of separation of concrete from BNFL (weight in g)

concrete BNFL	run 1	run 2	run 3	run 4	run 5	run 6
weight	33.094	33.589	34.164	32.941	33.976	33.713
after heating	30.491	30.930	31.636	30.493	31.415	31.220
weight loss (dry) %	8.54	8.60	7.99	8.03	8.15	7.99
after milling and sieving						
< 0.500 mm > 0.500 mm	6.353 23.412	6.359 24.730	5.908 25.246	7.034 23.924	6.707 24.930	6.182 25.000
% < 0.500 mm % > 0.500 mm	21.34 78.66	20.45 79.55	18.96 81.04	22.72 77.28	21.20 78.80	19.83 80.17
total	29.765	31.089	31.154	30.958	31.637	31.182
loss loss in %	0.726 2.44	-0.159 -0.51	0.482 1.55	-0.465 -1.50	-0.222 -0.70	0.038 0.12

Table III: K1 values after atomic absorption spectrometry

SAMPLES	**K1 (%)**
Loss on ignition at 1000°C	1.6
Total silica (SiO_2)	**77.23**
Iron oxide (Fe_2O_3)	1.37
Alumina (Al_2O_3)	**3.34**
Calcium (CaO)	**14.57**
Sulfuric anhydride (SO_3)	0.71
Magnesia (MgO)	0.55
Chloride (Cl)	0.012
Sodium oxide (Na_2O)	0.13
Potassium oxide (K_2O)	0.43

Table IV: Presence of crystalline compounds after X-ray diffraction

Crystallized compounds	**K1**
Quartz	++++
C_2S (calcium silicate)	+
Calcite	+
Feldspath	+
Ettringite	

Table V: Cement proportion of untreated material

Samples	CEMENT PROPORTION % of the untreated material
K1	**25.55**

Table VI: K1 presence in hydraulic binder zone

REBONDIN		Origin Kema 95 first sending	2nd sending > 0.063 mm	> 0.063 mm
insoluble		72.85	78.00	49.35
soluble	SiO_2	6.25	5.00	11.9
in	Fe_2O_3	0.75	0.60	1.25
perchloric acid	Al_2O_3	1.70	1.40	3.1
	CaO	14.65	12.70	26.6
	SO_3	0.70	0.70	1.55
	S	0		
	MgO	0.45	0.40	0.8
	Cl	0	0	0
	Na_2O	0.04	0.04	0.08
	K_2O	0.08	0.06	0.16
cement %		25.55	20.8	47.2
Total		97.47		94.79

Table VII: BNFL material composition

BNFL DECO Processed material	%
Loss on ignition at 1 000°C	4.40
% insoluble quartz	47.30
% soluble SiO2	**10.18**
% Fe_2O_3	1.57
% Al_2O_3	3.49
% CaO	31.45
% MgO	0.74
Ca/Si***	3.10
Si/Al***	2.90
Al/Fe***	2.20

Cement / insoluble	1.00

% cement **	48.50
% insoluble	47.30
% loss on ignition	4.40
Total	**100.14**

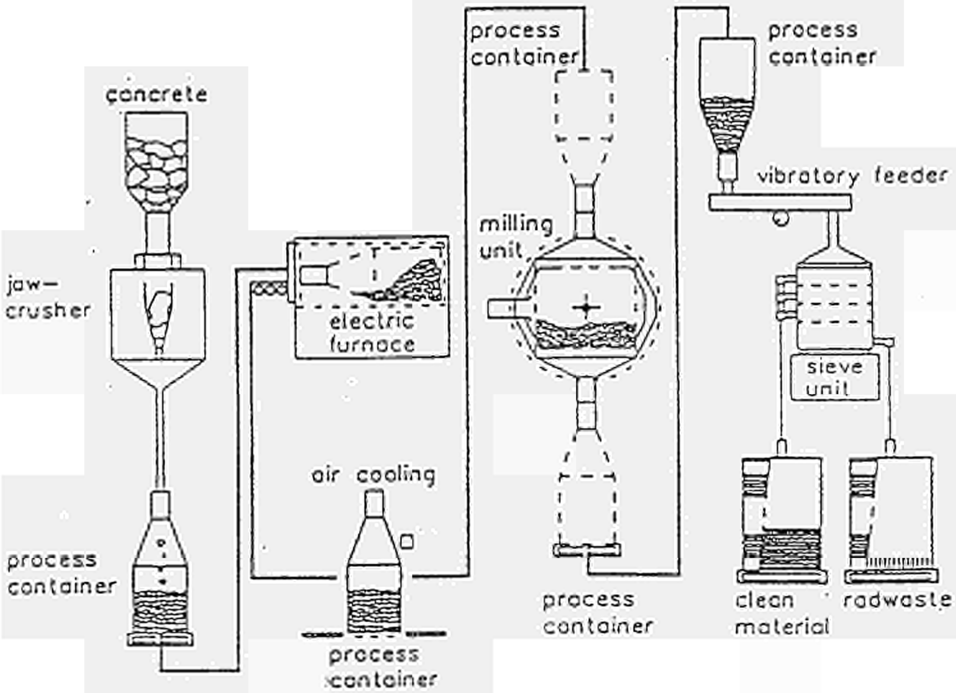

Figure 1 Process flow diagram of the DECO test installation

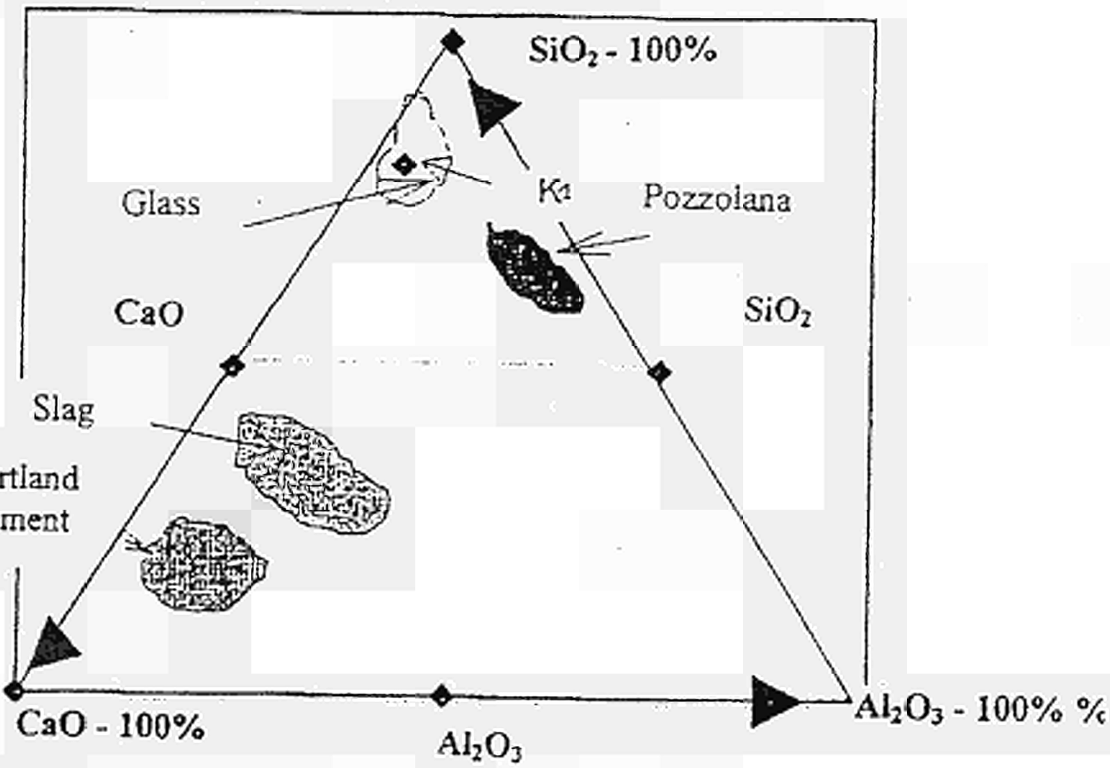

Figure 2 : Ternary diagram for material K (global composition)

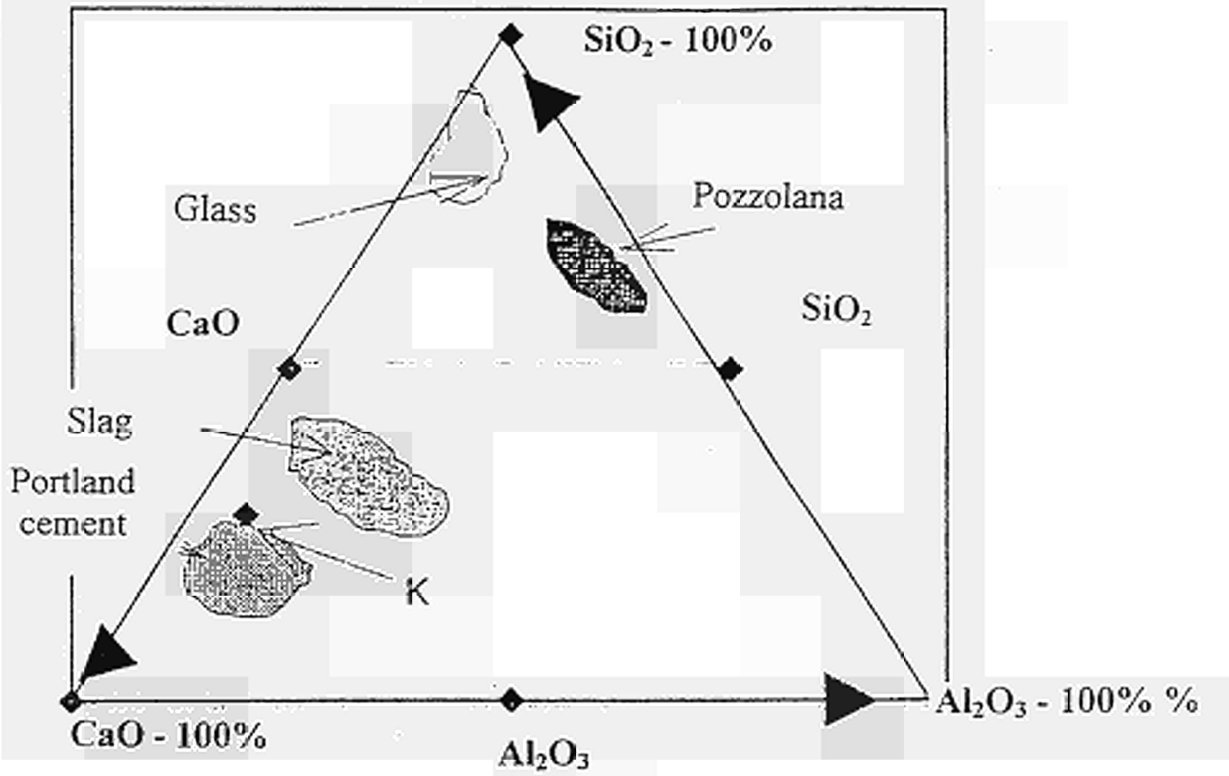

Figure 3 : Ternary diagram of material K - Soluble phase (Cement)

C.3.1-3 Waste volume minimisation using novel highly selective inorganic ion exchange materials

Contract No:	FI4W-CT95-0016	**Duration:**	1 Jan. 1996 - 31 Dec. 1998
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Partners:	**CROSFIELD Warrington/GB, IVO Vantaa/FI, Univ. Helsinki/FI, Univ. Salford/GB**		

A. OBJECTIVES AND SCOPE

The objective of this proposal is the long-term disposal of radioactive wastes at minimum volume. It will be achieved by the development of efficient methods for the decontamination of aqueous nuclear waste effluents from actinide elements, fission products, activation products and heavy metals using highly selective inorganic ion exchangers designed for use in column operations.

Measurable objectives are: a better understanding of exchanger selectivity; planned synthesis of novel well-characterised crystalline ion exchangers at the laboratory and pilot scale and their systematic evaluation for the separation of the most common nuclides in liquid wastes; synthesis of all organic granular material for column use; highly effective column operations for industrial applications; and compatibility with final waste forms.

B. WORK PROGRAMME

B.1 Review of exchanger materials
B.2 Laboratory scale preparation and structural characterisation of materials
B.3 Product screening
B.4 Evaluation of ion exchange performance with variation in macro-components
B.5 Evaluation of ion exchange performance in simulated waste
B.6 Tests with actual waste solutions
B.7 Radiolytic stability and leaching
B.8 Encapsulation tests
B.9 Scale-up of materials production
B.10 Preliminary process design and costing

C. Progress of work and results obtained

Summary of main issues

The project has now been running for twelve months, during which time all milestones and achievable targets have been met. An extensive literature review of inorganic ion exchange materials identified a number of groups of materials worth further investigation. Several materials have now been synthesised by the research groups at the University of Helsinki and the University of Salford. Most of these materials have now been characterised by X-ray diffraction (XRD). The ion exchange capacities of these materials for selected radionuclides has been screened by determining their distribution coefficients (Kd). Certain materials have been identified as having potential for removing specific radionuclides or a variety of radionuclides from typical waste solutions generated in the nuclear industry. More effort will be concentrated on these more promising materials, in terms of their synthesis and post-treatment methods.

C.1 Review of exchanger material

A number of materials were identified as having the potential to remove radionuclides from waste solutions. These materials can be divided into four main groups [1-3]:

- Group 1: Crystalline materials synthesised from octahedral/tetrahedral co-ordination polyhedra, e.g. 'MOS' materials built from Ti and Mo polyhedra.
- Group 2: Layered oxide materials including those with inorganic pillars Incorporated between the layer. The pore size can be controlled by the nature of the pillaring agent and charge density of the host material.
- Group 3: High silica materials, including those with large mesoporosity, e.g. the 'Mobil Crystalline Materials' (MCM) type molecular sieves.
- Group 4: Microporous materials with interstitial cations other than Si or Al in tetrahedral co-ordination, e.g. those based on large-pore VPI or similar frameworks.

This work package was completed in Quarter 1 of 1996.

C.2 Laboratory scale preparation and characterisation of materials

Materials were synthesised following experimental conditions given in literature and patents [1-6]. Where necessary, experimental procedures were modified to obtain the desired material. The synthesised materials were analysed using X-ray diffraction. It was decided that all crystalline materials synthesised should exhibit clean XRD patterns, in agreement with literature and/or database patterns. Samples with low crystallite sizes (*ca.* 50Å) may be X-ray amorphous and in these cases should be analysed by scanning electron microscopy (SEM) or transmission electron microscopy (TEM). For samples where heat treatment is required to remove organic templates, thermal analysis (TGA, DTA) as well as XRD analysis should be carried out on the as-synthesised and calcined materials, in order to optimise the process and verify loss of organic component and retention of the framework structure. To-date, approximately 50 variants of 10 main products from Groups 1, 3 and 4 have been prepared and characterised as described above. This work package is progressing according to schedule [7] and will continue through until the end of 1998.

C.3 Product screening

An experimental protocol was agreed upon in order to assess the efficiency of the synthesised materials in main waste solution categories, by distribution coefficient

(Kd) measurements [2, 4, 5, 6]. The key radionuclides to be investigated were ^{51}Cr, ^{54}Mn, ^{57}Co, ^{59}Fe, ^{65}Zn, ^{85}Sr, ^{110m}Ag, ^{134}Cs, ^{236}Pu and ^{241}Am. The 5 "model" solutions were 4 M HNO_3, 0.1 M HNO_3, 0.1 M $NaNO_3$, 0.1 M $NaNO_3$/0.1 M NaOH and deionised water. Batch factors of 100 with an equilibration time of 1-3 days were used, after which time samples were centrifuged, filtered and the activity (β and γ) counted. Blank determinations were also carried out in order to make corrections for adsorption on contact vials and filters and to check for possible precipitation of radionuclides. A mica product performed extremely well, giving high Kds for a number of radionuclides, over a pH of 1-13. A molybdophosphate material with a novel structure gave a very high Kd for ^{134}Cs in 4 M HNO_3. Further testing of this material showed that radionuclide uptake is still strong in 8 M acid but decreases in 16 M acid. OMS-1 type materials were selective for ^{137}Cs in 0.1 M HNO_3 and even in 4 M HNO_3 to a lesser extent. All of the titanosilicates were very selective for ^{89}Sr and ^{137}Cs in neutral to mildly alkaline solutions. Zorite and K-pharmacosiderite gave very high Kds for ^{137}Cs in 0.1 M HNO_3. A titanoborosilicate material gave a high Kd for ^{110m}Ag in mildly alkaline to acidic solutions. Other materials tested also showed promise, although over a much narrower pH range. This work package is progressing according to schedule [7] and will continue through until the end of 1998.

C.4 Evaluation of ion exchange performance with variation in macro-components

The performance of some of the more promising materials, that pass the screening stage, were further evaluated by carrying out tests with variation in common macro ions present in nuclear waste solutions (Na^+, K^+, Li^+, H^+, Ca^{2+}, Mg^{2+}) [5, 6]. So far some of the micas and molybdophoshate materials have been tested. For the micas, the effect of H^+, Ca^{2+} and Mg^{2+} has been tested on ^{85}Sr uptake. Mg^{2+} has very little effect on ^{85}Sr uptake, whereas Ca^{2+} interferes strongly. However, compared to another Sr-selective material, the capacity of the mica was considerably higher in the presence of Ca^{2+}. Uptake of ^{85}Sr is strong in neutral and slightly acidic solutions, in contrast to other Sr-selective materials, e.g. Na-titanate and silicotitanate. This work package is proceeding as scheduled and will continue for the duration of the project.

C.5 Evaluation of ion exchange performance in simulated waste

A number of specific waste streams common to nuclear power plants, reprocessing plants and decontamination activities have been identified [8-11]:

- NPP primary circuit decontamination solutions (IVOINTER)
- Medium active salt free concentrate from Purex-style processes (BNFL)
- Mildly alkaline sludge recovery wastes from decommissioning operations (BNFL)
- Decontamination streams based on inorganic acids such as tetrafluoroboric, nitric, sulphuric or phosphoric (BNFL)
- Decontamination streams based on organic complexants such as citrate, EDTA and detergents (BNFL)
- Acid dissolved sludge and alkaline supernate from high level waste tanks (Hanford, USA)

The actual waste streams evaluated will depend upon the performance of the exchangers in the simulated streams. This work package is an on-going task and will continue through until the end of 1998.

C.6 Task end-points

This project will officially end on 31.12.1998. Task C.2 and C.3 should end in the 2nd quarter of 1998. Task C.4 should end in the 3rd quarter of 1998 and Task C.5 at the end of 1998 (see GANNT Chart of Technical Annex) [7].

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C.3.1-4 Extraction and selective separation of long-lived nuclides by functionalized macrocycles

Contract No:	FI4W-CT96-0022	**Duration:**	1 May. 1996 - 30 Apr. 1999
Coordinator:	**J.F. Dozol, CEA-Cadarache/FR**		
	Tel. +33/442.25.74.97	Fax:	+33/442.25.35.45
		e-mail:	
Partners:	**ECPM Strasbourg/FR, Univ. Mainz/DE, Univ. Parma/IT, Univ. Twente Enschede/NL, Univ. Strasbourg/FR, Univ. Valencia/ES, Univ. Liège/BE, Univ. Autonoma Madrid/ES**		

A. OBJECTIVES AND SCOPE

The objectives of the project are to synthesize and test macrocyclic compounds for the selective extraction of radionuclides with two purposes:

- Decategorization of waste by selective removal of strontium and actinides.
- Separation of trivalent actinides from lanthanides. This could be achieved by a slight modification (nature of donor or other macrocyclic compounds (coronands, cavitands)) of the calixarenes able to effectively extract both lanthanides and actinides synthesized in the previous contract.

B. WORK PROGRAMME

B.1 Synthesis of new extractants

B.1.1 Synthesis of strontium extractants

Synthesis and study of the complexation of 1,3-cyclohexyl DC 18 C 6 derivatives with two sites of complexation and that present negative allosteric cooperativity (Univ. Valencia).

Synthesis of calix[n]arenes bearing amide groups (Univ. Parma and Madrid).

B.1.2 Extraction of actinides

Efforts to anchoring phosphine oxide binding units on calix[6]arenes or calix-crowns in 1,3 alternate conformation (Univ. Parma).

Preparation of extractants in which C.M.P.O. or diamide groups are attached to the upper rim of calix[4]and[5]arenes and intends to modify the C.M.P.O-structures in various ways, for instance to (NR-CO-CH2-PO-R2, to -CH2-NH-CO-PO-R2 or to -PO-CH2-CO-NR2 with the phosphorous atom being attached to the calixarenes). (Univ. Mainz).

Synthesis of C.M.P.O. like compounds built on very rigid calix[4]arenes (Univ. Parma and Mainz).

Efforts to the preparation of calix[6]arenes functionalized in 1, 3, 5 positions, either at the lower rim or at the upper rim, these compounds are endowed with the appropriate sizes and shapes for trivalent cations (Univ. Madrid).

B.1.3 Separation of trivalent actinides / lanthanides

Synthesis of ligands with soft and hard donor groups derived from calix[4] or [6] arenes (Univ. Parma).

Development of resorcinarenes with C.M.P.O. or diamide units (Univ. Mainz).

Study of the synthesis of macrocyclic ligands bearing phosphoryl groups with cavities commensurate to the trivalent elements (Univ. Liège).

Elaboration of calix[6]arenes with different functional groups (phosphine oxides, phosphonic or sulfonic acids, amides and ureas). (Univ. Madrid)

B.2 Complexation extraction and transport measurements

Fast screening of the new compounds synthesized in the framework of the contract and provides thermodynamic parameters (Univ. ECPM Strasbourg).

Distribution coefficient measurements and transport experiments of strontium lanthanide and actinide cations through supported liquid membranes (S.L.M.) with the new extractants synthesized. Figure 1 shows the S.L.M. device used for these experiments (CEA Cadarache)

At the end of the project, CEA Cadarache will test the most promising extractants on real waste.

B.3 Molecular modeling NMR and X-ray structures of the complexes

B.3.1 Molecular modeling

Studies of the complexation of strontium and trivalent elements and devotes its efforts particularly on C.M.P.O. like calix[4]arenes and calix[4]arene-crowns containing phosporyl moieties (Univ. Strasbourg).

Molecular mechanics [M.M.] and molecular dynamics [M.D.] calculations on ligands relevant for the extraction of trivalent cations (Univ. Twente).

B.3.2 NMR and X-ray structures of the complexes

The University of Parma provides X-ray crystal structure on complexes between different synthesized calixarenes and cations (Strontium, lanthanides).

The University of Liege provides X-ray, N.M.R. structures of some complexes of macrocycles with lanthanides or actinides.

C. PROGRESS OF WORK AND RESULTS OBTAINED

The role of each group in the present project is summarized in figure 2.

Except the University of Parma which has prepared several extractants, the other groups are still synthesizing the foreseen produtcs. E.C.P.M. Strasbourg and C.E.A. Cadarache study the first molecules synthesized.

Many extractants are functionalized calixarenes, obtained from basic calix[n]arenes (n=4, 5, 6, 8) represented figure 3.

C.1 Synthesis of new extractants

C.1.1. Strontium extraction

The University of Valencia is synthesizing a cyclohexyl 18 crown 6 derivative containing two carboxylic groups. In acidic medium the crown ether adopts the diequatorial conformation, the most stable for complexing the cations; in neutral medium, for instance in demineralized water, the carboxilic groups become diaxial, producing a conformational change that favors the release of the cation complexed in acidic medium (allosteric effect).

The University of Parma has already prepared several calixarenes able to complex strontium:

Calix[8]arenes with amide groups;

Calix[4]arenes (de-tert butyated or not) bearing two tertary and two primary amides

The University of Madrid developes calix[6]arenes hexa di ethyl hexamide and calix[6]arenes di octyl hexa amide.

C.1.2. Actinide extraction

The University of Parma has sent to the groups of Cadarache and Strasbourg tert-butylated calix[6]arenes comprising three amide and three phosphine oxide moieties.

The University of Maiz continues to synthesize C.M.P.O.-like calixarenes, these products are exceptional extractants and carriers of actinides whatever their valencies and prepares a complete series of calix[4]arenes tetraphenyl ethers bearing one to four C.M.P.O. functions.On the other hand, the University of Mainz studies a new strategy of synthesis in order to be able to vary the residues attached to the phosphorous

C.1.3 Separation of trivalent actinides / lanthanides

The University of Parma has also prepared ligands with a mixture of hard donors (amides) and soft donors (pyridine or bipyridine);

The University of Mainz tries to favor the extraction of actinides by rigidifying the C.M.P.O. like calixarenes.

The synthesis of some extractants described in B.1.3. are in progress at the Universities of Liége and Madrid.

C.2 Complexation, extraction and transport measurements

Most of the extractants synthesized at Parma have been tested by E.C.P.M. Strasbourg and C.E.A. Cadarache.Among these extactants the most promising is the calix[8]arene bearing eight amide moieties, it appears to be an excellent extractant of strontium and barium and displays a very high selectivity towards sodium (D Cs/Na higher than 20000).

C.3 Molecular modeling , N.M.R. andX-ray structures of the complexes

The calculations of molecular mechanics being complex, results cannot yet be reported.The first X ray structures of lanthanides with calixarenes have been achieved at the University of Liége.

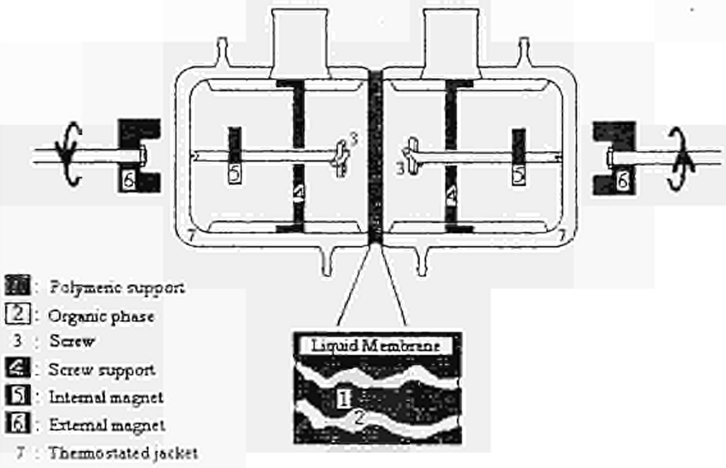


Figure 1 - Flat sheet supported liquid membrane device for transport experiments

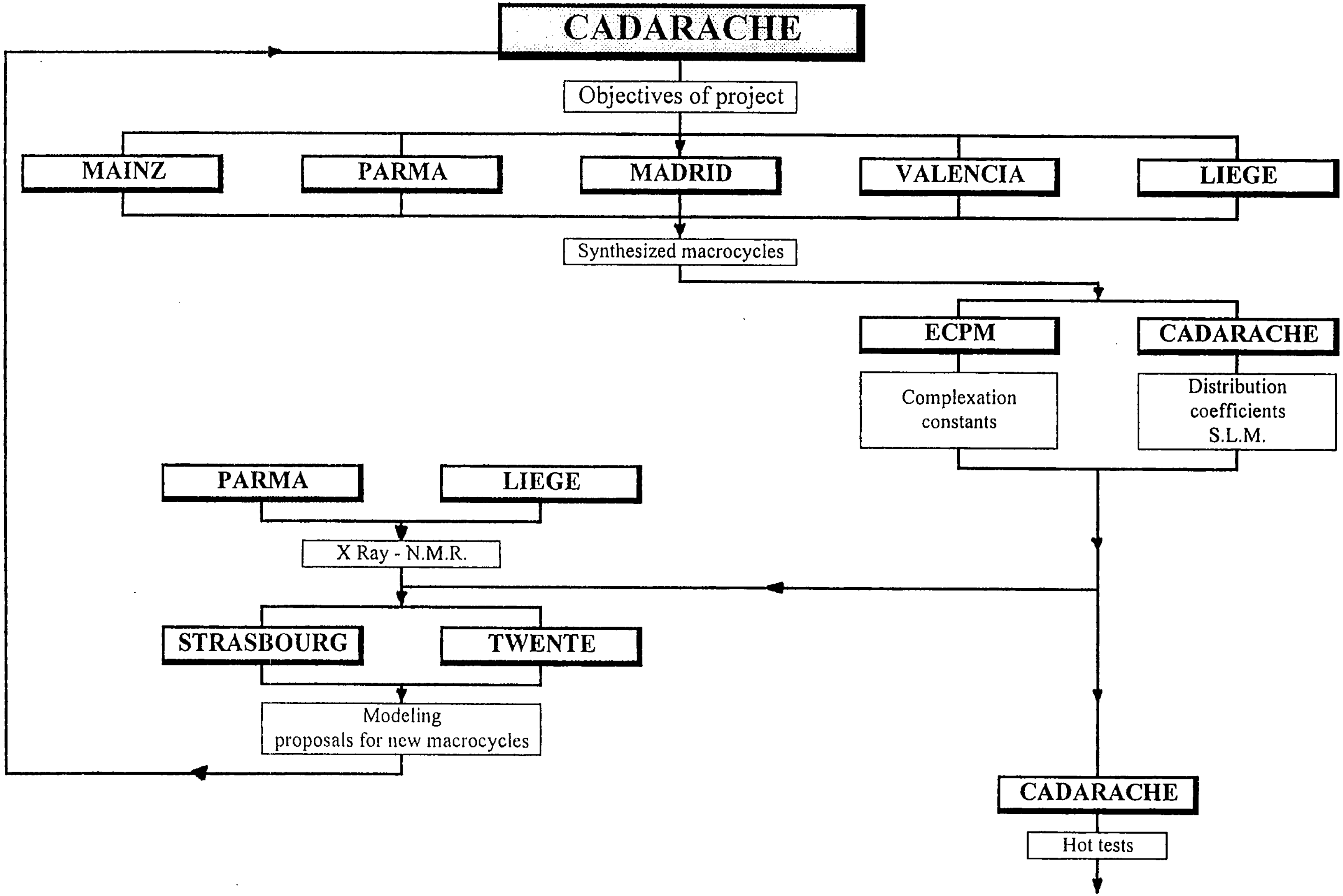
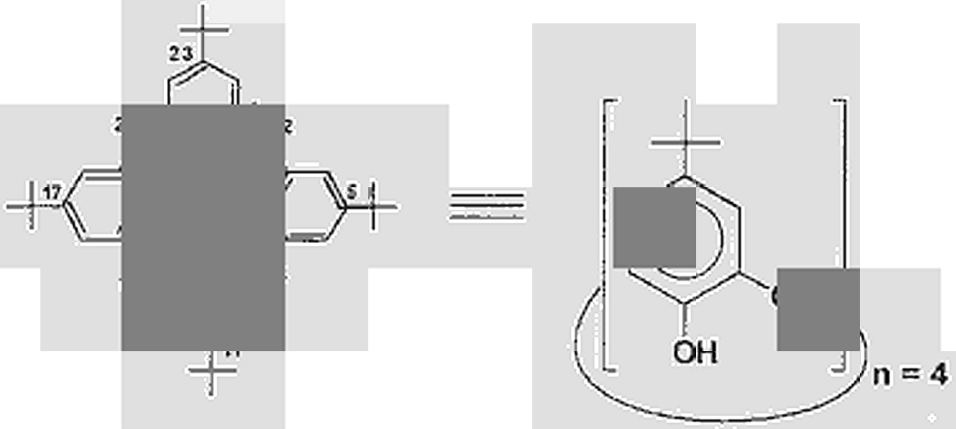


FIGURE 2 - PROJECT ORGANIZATION



Fi[redacted][4]arene

C.3.2-1 Experimental and modelling studies to formulate a source term of nuclear waste glass in representative geological storage conditions

Contract No: FI4W-CT95-0001	**Duration:** 1 Jan. 1996 - 31 Dec. 1998
Coordinator: **T. ADVOCAT, CEA Valrhô/FR**	
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Partners: **ONDRAF/NIRAS Brussels/BE, FZK Karlsruhe/DE, PSI Villingen/CH**	

A. OBJECTIVES AND SCOPE

The proposed work consists of key experiments on long-term glass corrosion, radionuclide release as a consequence of glass alteration and radionuclide partition between groundwater and secondary solids. This experimental work will support a modelling approach to produce a source term for nuclear waste glass in representative geological disposals. The modelling will be also based on previous experimental results obtained from one of the most studied R7T7 glasses in the past during worldwide laboratory studies and previous EC-projects.

The investigation will simultaneously follow four major axes, combining experimental and modelling investigations: (1) review of the state of the art and data bank development on R7T7 glass leach test data to formulate a coherent experimental data-set for modelling validations, (2) experimental work related to nuclear waste glass dissolution kinetics especially under saturation conditions, (3) experimental work related to the behaviour of safety relevant radionuclides during glass alteration and (4) modelling for interpretation of experimental results, scale up, long-term extrapolation and repository performance assessment.

B. WORK PROGRAMME

B.1 Data-bank development

The current state of knowledge is being reviewed and summarised to develop a data-bank of R7T7 glass leaching data. A programme *Versail* is being developed to process the data from a wide range of parameters covered by the experimental conditions, the leaching data and the experimental results (CEA). A literature review is underway of French, German and Belgian data of inactive and active laboratory results, which will be incorporated into the data-bank (CEA, FZK, ONDRAF/NIRAS, SCK/CEN).

B.2 Experimental Work on Nuclear Waste Glass Dissolution Kinetics

The mechanism of 'long-term reactions' is investigated with relation to the presence of other engineered materials, to the geochemical conditions in the repository and to the glass composition.

2.1 Determination of the pH-dependency of the saturation concentration of silica (FZK).
2.2 Determination of the kinetic effects of the dissolved glass forming elements (CEA).
2.3 Investigation of particular processes in the gel layer (ONDRAF/NIRAS, SCK/CEN).

B.3 Experimental work related to radionuclide behaviour during glass dissolution

The experiments will be conducted with non-radioactive and radioactive glass specimens (containing notably technetium and actinide elements). The alteration products (gels) containing actinides (U, Pu, Np, Am) will be synthesised and characterised and their radiation stability will be assessed.

3.1 Determination of the sorption isotherms and/or coprecipitation (FZK).
3.2 Alteration of highly active CEA R7T7 glass in NaCl brines (FZK).
3.3 Alteration of highly active CEA R7T7 glass in the presence of pre-corroded steel (FZK).
3.4 Parametric studies of R7T7 glass alteration in silicate water with complexing ions representative of groundwater (CEA).
3.5 Alteration of a-doped R7T7 glasses in synthetic groundwater (CEA).
3.6 Gel synthesis (CEA).
3.7 Interaction of non-radioactive and a-doped R7T7 glass specimens with clay-water (ONDRAF/NIRAS, SCK/CEN).

B.4 Modelling for interpretation of experimental results, scale up, extrapolation and repository performance assessment

This task will implement enhanced geochemical codes (EQ3/6, KINDIS) and mathematical codes (Monte Carlo, GLADIS) with data common to all the partners. The objective will be to allow for the corrosion reaction progress, to describe the glass matrix corrosion kinetics and to specify the behaviour of the major glass elements and radionuclides (precipitation, sorption, coprecipitation) for specific disposal environments. The database (the first research domain) will provide the parameters for predictive modelling.

4.1 Code intercomparison (FZK, CEA, ONDRAF/NIRAS, SCK/CEN, PSI).
4.2 Geochemical modelling for $MgCl_2$ and NaCl rich brines (FZK).
4.3 Mechanistic modelling using LIXIVER and geochemical modelling using KINDIS (CEA).
4.4 Geochemical modelling of interactions between glass, clay-water and moist clay (ONDRAF/NIRAS, SCK/CEN).
4.5 Mathematical and geochemical modelling of glass/clay materials (CEA, PSI).

C. Progress of work and obtained results

Summary of main issues

The architecture of the *Versail* database, designed to compile R7T7 glass leaching behavior data, has been finalized. *Versail* is now installed on a CEA/SCD server and is ready for entry of the experimental data. Access to the partners' database is now being arranged.

The behavior of the principal radionuclides affecting long-term safety (Np, Pu, Am, Tc) during glass alteration was assessed in two types of experiments. Leach tests in flowing solution with Pu, Np or Tc-doped glass specimens showed that radionuclide retention in the alteration film cannot be interpreted exclusively by equilibrium precipitation of actinide hydroxides, carbonates or phosphates; sorption phenomena should also have a major role. Additional experiments were performed to quantify radionuclide sorption phenomena using aqueous solutions doped with Th, U and rare earth elements (simulating trivalent and tetravalent actinides) in contact with nonradioactive simulated R7T7 glass alteration gels. R_D-values independent on the initial Eu concentration were found in the pH range 2 to 6. clearly characterizing sorption behavior. At higher pH-values, however, R_D-values are not constant as a function of initial Eu concentration. Under these alkaline conditions, solubility constraints should be controlling factors.

Finally, work began on the formulation of a glass source term by tests designed to quantify the properties of the glass in contact with clay materials (K_d, silica diffusion coefficient) and work continued on the development of the stochastic code. New results confirm the earlier conclusion of Si sorption : K_d is about 20 $ml.g^{-1}$ in unoxidized clay, and about 75 $ml.g^{-1}$ in oxidized clay.

C.1 Database Development

Work on designing and implementing the glass database was completed in 1996[1]. The *Versail* database will be used to compile pertinent information on R7T7 glass concerning its characteristics and physical properties, and notably its long-term behavior. The database was configured to allow multiple criteria queries. *Versail* was implemented under Sybase SQL Server System 10, a client-server package installed on a dedicated PC server running under Windows NT. The user interface uses *Object View 3.0* for display and updating of individual records, and *Business Objects 3.11* for query and search purposes.

The underlying logic of the *Versail* architecture is expressed in terms of tests, glass specimens and leaching experiments. A *test* covers information concerning the glass processing temperature and technique (platinum crucible, prototype, etc.). A *glass specimen* includes data on the glass fabrication date and characterization (density, homogeneity, chemical composition, etc.). A *leaching experiment* refers to data on a specific experiment, such as the operating conditions (temperature, SA/V ratio, duration, MCC-1 protocol, etc.), the leachate analysis results (element concentrations) and the alteration rates.

C.2 Experimental Work on Nuclear Glass Waste Dissolution Kinetics

C.2.1. pH-dependency of the saturation concentration of silica

In order to identify the nature of saturation effects in the glass dissolution mechanism, glass corrosion tests were performed in deionized water and in saline solutions at high sample surface to solution volume ratios (S/V=1000 m^{-1}). Experiments in granite water are still to be done. The pH-value was kept constant during the experiments at various preselected values. Experiments were run in duplicates at 80°C and 50°C. The glass composition (GP WAK1) closely resemble R7T7 type composition, however, the contents of SiO_2 and Al_2O_3 were 50 and 2.8 wt% instead of 45 and 5 wt% respectively and Uranium was absent in the glass. Solution aliquots were taken for solution analyses after 30 and 100 days. In few cases, samples were ultrafiltered to check for colloid generation.

Results of this study, obtained after 100 days of corrosion at 80°C are given in terms of normalized elemental mass losses (NL) in Figure 1 (50°C). Data points are individual values of each of the two duplicate tests. Using boron release data being indicative for glass matrix dissolution, three main groups of glass constituents were distinguished when comparing NL data (1) Silica, (2) elements which are leached with the same rate as the glass matrix is dissolved (Ca, Li, Mo under alkaline conditions), (3) elements which significantly retained (Ca under alkaline conditions, Al at all pH values except pH 1.

(1) Silica: At all pH values the measured solution concentration of dissolved silica represents truly dissolved material or extremely small polymeric species. No colloids larger than 18 Å were observed. For the pH range 1-9 it can be seen that the solution concentration of silica is almost independent of pH, thus confirming the usefulness of a simple silica saturation based first order dissolution rate law as approximate representation for saturation effects (Figure 2). This rate law appears to remain valid for pH values up to 11, though, the hydrolysis of orthosilicic acid needs to be taken into account. As shown in Figure 3, the experimental solubility data for silica can best be described mathematically by the solubility of an SiO_2 polymorph with a solubility about 30% less than that of amorphous silica (log $K_{50°C}$= -2,64, log $K_{80°C}$= -2,44). In very alkaline solutions, a mismatch is observed. In particular at pH 11 this may result from the fact that at this pH silica saturation was not yet reached.

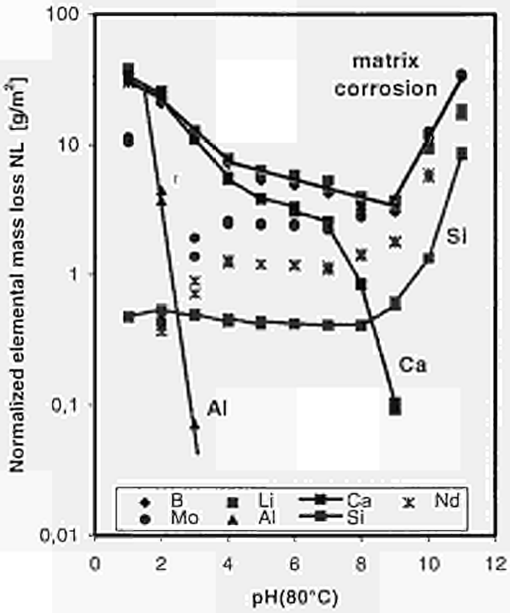

Figure 1: pH-dependency of the normalized release of consituents from GP WAK1 glass at S/V = 1000 m-1 and 80°C

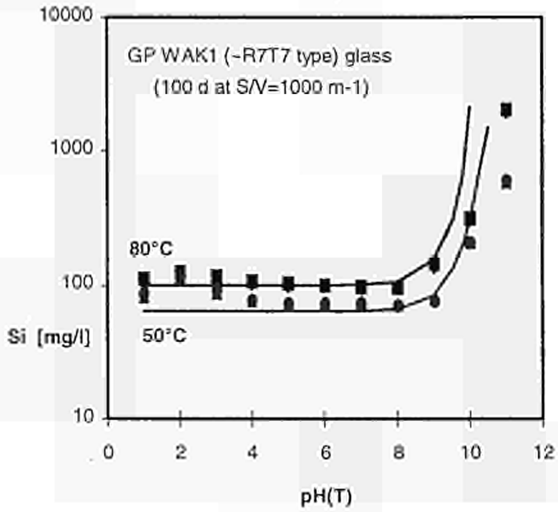

Figure 2. Comparison of calculated and measured saturation concentrations of silica.

(2) Matrix corrosion: The normalized elemental mass loss values for Boron and Lithium are very similar between pH 1 and 10, indicating glass matrix corrosion as common release mechanism. Ca is leached by the same mechanism at pH<4, at pH 1 this is true even for the release of Nd and Al, and at alkaline pH values for Mo. The pH dependency of glass matrix corrosion, shows the typical U-shape behavior as a function of pH. The corrosion rates of the glass matrix were small at all pH-values, indicating glass corrosion under saturated conditions. On the acid side, no correlation exists between the pH dependence of silica solubility and Boron (and Li, and Ca) release.

(3) Elements with a normalized mass loss smaller than Boron are retained to a significant extent in the surface layers of the glass. The data in Figure 1 show that mainly silica, Al, and

Nd belong to this group of glass constituents, however, at alkaline pH also Ca and at pH 11 even Li are retained.

C.2.2. Kinetics effects of the dissolved glass forming elements

Twenty leach tests with nonradioactive R7T7 glass powder specimens under flowing conditions at 90°C, either with initially pure water or with silica-enriched aqueous solutions, were achieved. The leachates are currently being analyzed and the results will be included in the next semiannual report. These tests are designed to identify the kinetically inhibiting role of silica.

C.2.3. Particular processes in the gel layer : Si migration experiment

At the end of '96 we sliced the clay core from one of the flow-through migration cells, to analyze the Si-32 migration profile. Previous measurements of the Si migration were carried out (see the program '91-'95 [1]) by analyzing the output solution. We also performed batch sorption tests on Si-32. The results confirm the earlier conclusion of Si sorption. K_d is about 20 $ml.g^{-1}$ (unoxidized clay), and about 75 $ml.g^{-1}$ (oxidized clay). The sorption process seems to be controlled by the first silicate anion ($SiO(OH)_3^-$).

C.3 Radionuclide Behavior during Glass Alteration

C.3.1. Determination of sorption isotherms

Experiments are started, using a similar experimental setup as described under C.2.1. Sorption studies were performed using the solid corrosion products of precorroded glass as substrate (precorrosion: initially deionized water, 30 days corrosion at 80°C at S/V=1000 m^{-1}). Sorption isotherms were determined at 25 and 80°C for Eu, U(VI) and Th. The behavior of Eu and Th represent the behavior of the chemically homologue tri- and tetravalent actinide elements. These three elements were added together to the solution resulting in initial concentration ranges of 10^{-4} to 10^{-6} m. The pH was kept constant at preselected values. The sorption process was followed as a function of time. After 10 days apparent equilibrium was reached. The quantity sorbed on the glass corrosion products and the calculated distribution coefficients R_D were deduced from the decrease of solution concentration after 10 days. Distinction was made between filtered and ultrafiltered solution samples to assess colloid generation. Though, R_D values have no physical bearing if colloids are present, the respective values are included to illustrate the colloid effect.

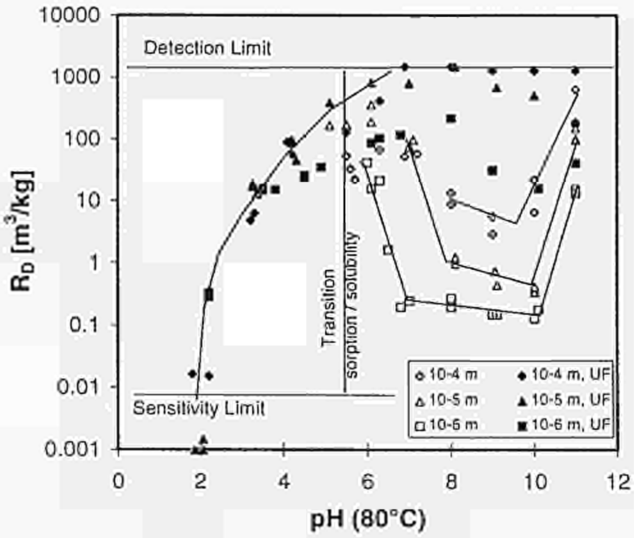

Figure 3. Sorption behavior of Eu on glass corrosion products as a function of pH.

As an example, R_D values for Eu sorption at 80°C are given in Figure 3. Solubility effects needs to be ruled out in order to unambiguously studying sorption behavior. R_D-values

independent on the initial Eu concentration were found in the pH range 2 to 6. clearly characterizing sorption behavior. In this pH-range, colloids were not observed. At higher pH-values, the R_D value increases with the initial Eu concentration. This may be an effect of solubility constraints with respect to phases such as $Eu(OH)_3$, $EuOHCO_3$, $Eu_2(CO_3)_2$. Indeed, in this range colloid formation (precipitation?) was observed.

C.3.2. α–doped glass alteration in silicate water with complexing anions

Leaching experiments were conducted with siliceous water (30 mg·l^{-1} Si) at 90°C in aerobic conditions with R7T7 glass specimens doped with natural uranium, ^{237}Np, ^{239}Pu or ^{99}Tc (the planned experiments with americium-doped glass have not yet been initiated). Additional experiments were performed in a reducing media (Eh/EnH = 200–250 mV) with glass specimens doped with U or Tc. The effects of anions (10^{-3} M of carbonates, phosphates and sulfates) were also investigated. Tests under aerobic conditions showed that the radionuclides are trapped in the alteration film in proportions exceeding 10% for Tc, 90% for U and Np, and 92% for Pu. The results currently available for the reducing medium indicate retention of over 10% for Tc and over 40% for U. The leachates were undersaturated with respect to solid phases such as $NaNpO_2CO_3$, $UO_2(OH)_2H_2O$, UO_2CO_3 and $Ca(UO_2)_2(PO_4)_2$. Conversely, the leachates were oversaturated with respect to $NpO_2(OH)$, $Ca(UO_2)_2(SiO_3OH)_2$, $Pu(OH)_{4(am)}$ and $PuO_{2(cr)}$. The potential role of chemisorption in controlling actinide retention phenomena therefore cannot be excluded by these geochemical arguments. Experiments specifically designed to identify this phenomenon were therefore undertaken.

R7T7 glass alteration gels doped with Np, Pu and Am were synthesized by static leaching of glass coupons at 300°C. SEM and X-ray diffraction examinations are scheduled.

C.3.3. Interaction of inactive and doped glasses with claywater

Preparation of corrosion tests on inactive glasses were started. A quality assurance (QA) system was also introduced, including validation of the results, calibration, etc. The experimental parameters were selected as to obtain the required parameters for the modeling, and to simulate the "worst case situation" for glass corrosion, i.e. interaction with Boom clay. The experimental parameters are as follows: (1) R7T7 glass SON68 and PAMELA glass SM539 will be used ; (2) SIC (synthetic interstitial clay-water) and WC (mixture of 2000 g/l Boom clay with SIC) media are retained., the volume of the interacting medium will be 7 ml or more ; (3) the temperatures 40°C and 90°C are fixed ; (4) Surface area to volume ratio (m^{-1}) is equal to, respectively,100 and 2500 (using powdered glass and plate) ; (5) the duration are 7, 28, 50, 90, 180, 270, 365, 540, 720, X days. After the tests some samples will be subjected to SIMS profiling analysis, to inform about the dissolution mechanism.

Preparation of corrosion tests on active glasses was also defined. Based on the results from the previous programme [1] we decided to focus on the backfill material (FoCa clay) as interacting material, because the data of radionuclide leaching in near field condition are most relevant. We also decided to focus on Np and Tc behaviour, because these are the most critical radionuclides in the long-term. The experimental parameters are as follows: (1) Glasses SON68 and SM539, doped with Np and Tc (1 condition for SON68, 2 conditions for SM539: one high Np/ low Tc, one low Np/ high Tc concentration in the glass) ; (2) real interstitial claywater (RIC) mixed with FoCa clay, and also RIC mixed with various solids (FoCa clay, Boom clay, sand, graphite, corrosion products) ; (3) two temperature, respectively 40°C and 90°C ; (4) the durations are equal to 28, 90, 180, 360, 540. Days ; (5) Surface area to volume ratio (in m^{-1}) of 100 (the volume of the medium will be about 15 ml).

C.4 Modeling: Interpretation of Experimental Results, Scaling Up, Extrapolation and Repository Performance Assessment

C.4.1. Geochemical Modeling of the interaction between glass, clay water and moist clay

Experimental validation of the GLADYS code began with the specification of a leaching protocol using R7T7 glass cylinders 2 cm in diameter and 3.5 cm high in contact with moist clay (Smectite 4a or Boom clay). A 2 year period is scheduled for the experiments. The test configuration was optimized on the basis of preliminary calculations using GLADYS (these calculations will be detailed in the annual report). Figure 4 shows the normalized mass losses calculated for various K_d values. Saturation conditions are reached earlier if K_d values are small.

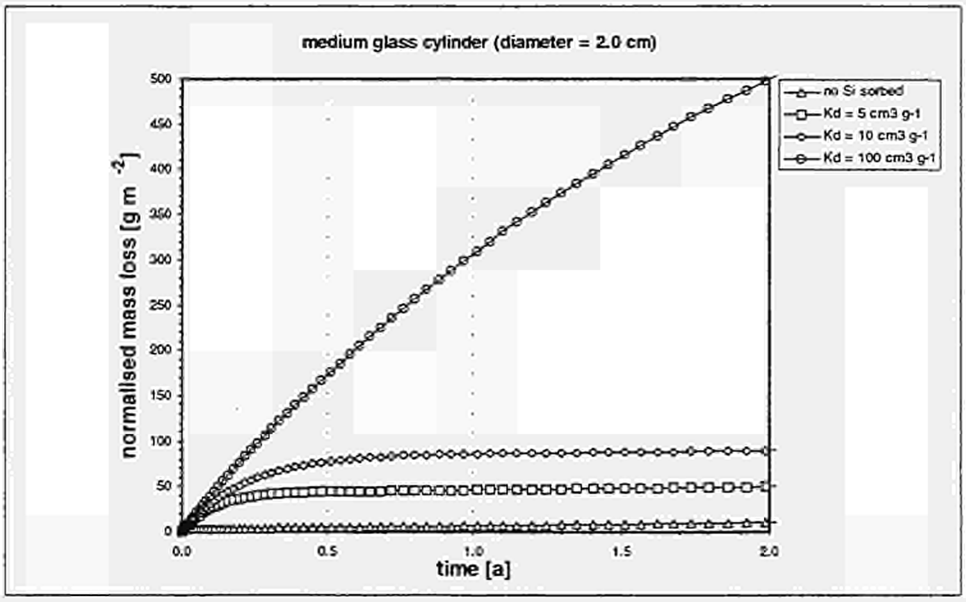

Figure 4. Normalized mass losses calculated with GLADIS for 4 silica distribution coefficients.

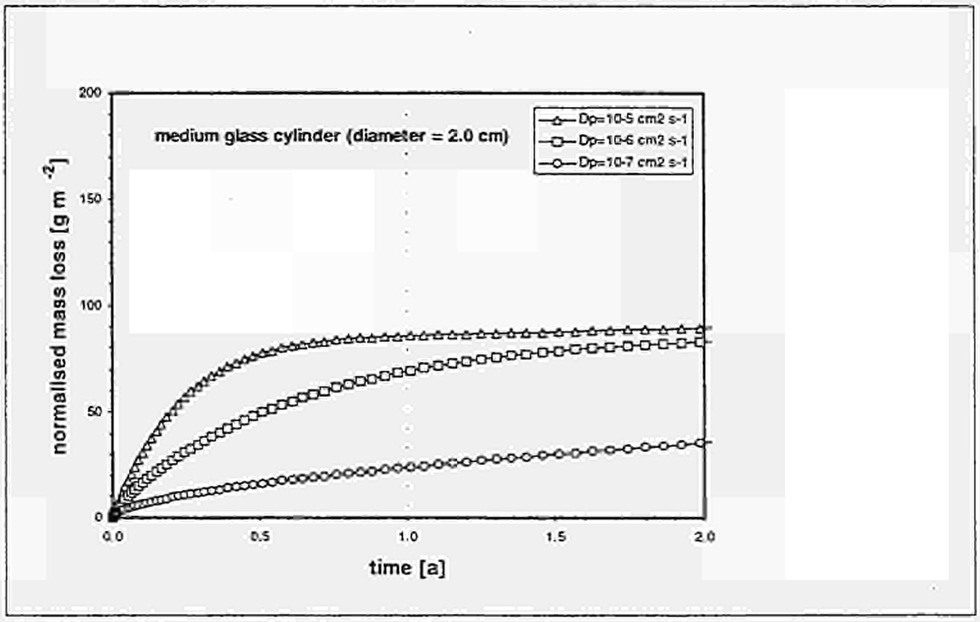

Figure 5. Effect of the pore diffusion coefficient on the normalized mass losses calculated with CLADYS. The actual value of D_p for silica diffusion through uncompacted water-saturated clay will most probably lie between 10^{-5} and 10^{-6} $cm^2.s^{-1}$ (D_p=5.10^{-5} $cm^2.s^{-1}$ for compacted Bentonite).

For a Kd value of 100 $cm^3.g^{-1}$, saturation is not yet reached after 2 years, but this condition is nevertheless being approached. Figure 5 shows that a decrease in the diffusion coefficient mainly changes the slope of the curves at early times. Different Kd values mainly affect the absolute normalized mass losses, while uncertainties in the diffusion coefficient have only an influence on the form of the curve. At present, the glass has been fabricated by melting oxides and carbonates at 1150°C, and the ratio of clay mass to leachant volume has been determined. The stainless steel leaching vessels (HP40 pots) are now being manufactured, and testing is scheduled to begin in February.

C.4.2. Mathematical modeling of glass/clay materials

We used the PHREEQE geochemical code to determine the mineral phases formed during the interaction of the glasses with pure solution (pure water or claywater), and to calculate pH. The predicted minerals include silica and zeolites for both glasses, and they could control the Si concentration in solution at high reaction progress. We started modelling the clay/water system with the PHREEQE code, considering the clay as a assembly of pure minerals. In a first approach we will use well characterized clay minerals such as smectite instead of Boom clay.

The mathematical modelling efforts were continuing the first approach [1], which was based on the Pescatore model [2]. This model combines diffusion of silica in the pore water of the clay with the glass dissolution rate law proposed by Grambow. We extended the model with a moving boundary. The model was used to fit the data of glass dissolution in clay slurries and pure clay. The model managed to fit the data (see Fig.6) though the model parameters could not yet be determined in detail. We also need to incorporate the ion exchange leaching process into the model.

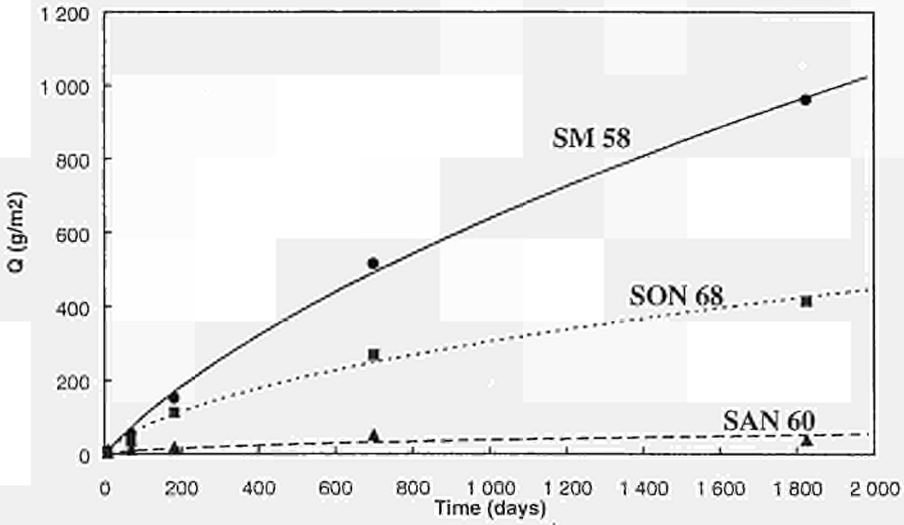

Figure 6. Experimental data (mass loss) and theoretical fitting for different glasses upon corrosion in pure Boom clay at 90°C.

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C.3.2-2 Corrosion evaluation of metallic materials for long-lived HLW and spent fuel disposal containers

Contract No: FI4W-CT95-0002	**Duration:** 1 Jan. 1996 - 31 Dec. 1998
Coordinator: **E. SMAILOS, FZK Karlsruhe/DE**	
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Partners: **ENRESA Madrid/ES, SCK-CEN Mol/BE, Univ. FUB Berlin/DE**	

A. OBJECTIVES AND SCOPE

In previous corrosion studies, two approaches were identified for the manufacture of long-lived HLW/Spent Fuel disposal containers that could act as a radionuclide barrier in repositories in rock salt, granite and clay. These are the corrosion allowance concept and the corrosion-resistant concept. For corrosion-allowance, carbon steels are the most promising material for all three rock formations. For corrosion-resistant, the strongest candidates are the alloy Ti99.8-Pd (Ti/0.2 Pd, TiGr-7) for rock salt, and stainless steels for granite and clay.

In the present research programme, further in-depth corrosion studies will be performed on the abovementioned materials in rock salt, granite and clay environments. These include: long-term immersion tests, electrochemical/radiochemical studies and slow strain rate tests. The objectives of the studies are:

- to investigate the potential effect of essential parameters on corrosion.
- to gain a better understanding of corrosion mechanisms.
- to provide more accurate data for a materials degradation model that can be used to predict the lifetime of such containers.

B. WORK PROGRAMME

B.1 Corrosion studies on candidate container materials in rock salt environments

1.1 Long-term immersion tests are performed on the TStE355 carbon steel (0.17 wt.% C) and Ti99.8-Pd in salt brines aimed at evaluating the effect of essential parameters on their corrosion behaviour. Such parameters are: the pH of the brines, the content of salt impurities, gamma-radiation, and welding (FZK).

1.2 Combined electrochemical and radiochemical studies are performed on Ti99.8-Pd in salt brines in order to get a detailed insight into corrosion kinetics and especially into the potential influence of the radiolytic product H_2O_2 on corrosion. Both unwelded and welded specimens are examined (FUB).

1.3 The resistance of Ti99.8-Pd (TiGr-7) and TStE355 carbon steel (0.17 wt.% C) to stress corrosion cracking (SCC) is investigated in a NaCl-rich brine (25.9 wt.% NaCl) at 170°C and strain rates of 10^{-4}-$10^{-7}s^{-1}$ by means of the slow strain rate technique (SSRT). For comparison, additional investigations are conducted in argon as inert reference medium (ENRESA).

B.2 Stress corrosion cracking studies in granite environments

The resistance of the AISI 316L stainless steel and TStE355 carbon steel to stress corrosion cracking (SCC) in a bentonite buffered granitic groundwater is investigated at 90°C at various rates (ENRESA).

B.3 Corrosion studies in clay/bentonite environments

Electrochemical corrosion studies are performed on candidate container materials aimed at investigating the influence of important environmental parameters on corrosion. Such parameters are: the temperature (16°C and 90°C) and the content of O_2, Cl^- and $S_2O_3^{2-}$ of the corrosion medium (SCK/CEN).

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The potential influence of important parameters on the corrosion behaviour of preselected container materials (carbon steels, Hastelloy C4 and Ti99.8-Pd) was investigated in salt and clay environments. Parameters examined were: Temperature (16°C-170°C), pH (3-7), H_2O_2 ($6x10^{-4}$-$1.7x10^{-1}$ mol/l), Cl^- (16-10000 ppm), gamma radiation (10 Gy/h) and strain rate (10^{-4}-$10^{-7}s^{-1}$). The investigations included long-term immersion tests, electrochemical/ radiochemical studies and slow strain rate tests.

Present results obtained in salt environments (salt brines) indicate that gamma radiation (10 Gy/h) and H_2O_2-concentrations smaller than 10^{-4} mol/l do not influence the corrosion behaviour of Ti99.8-Pd, and that this alloy is resistant to stress corrosion cracking at strain rates up to $10^{-7}s^{-1}$. pH values of the test brine between 3 and 7 do not enhance the corrosion rate of the carbon steel, but at the very slow strain rate of $10^{-7}s^{-1}$ the material shows a slight sensitivity to stress corrosion cracking. First results in clay water show that high alloyed stainless steels, Hastelloy C4 and Ti99.8-Pd are highly resistant to corrosion up to 30°C. However, the stainless steel AISI 316L is susceptible to pitting and crevice corrosion at high Cl^- concentrations in clay water. Finally, the carbon steel undergoes significant general corrosion in the oxydizing clay water. Further investigations in salt and clay environments are in progress. In general it can be stated that the project has proceeded in line with the work plan.

Progress and results

C.1 Corrosion studies on candidate container materials in rock salt environments (B.1)

C.1.1 Long-term corrosion tests (B.1.1) (F)

Influence of pH on corrosion of the TStE355 carbon steel in Q-brine

The influence of pH values between 3 and 7 on the corrosion of the TStE355 carbon steel was investigated in the $MgCl_2$-rich 'Q-brine' (26.8 wt.% $MgCl_2$) at 170°C. The results indicate that the steel is resistant to pitting corrosion. The general corrosion rates of the steels at the various pH values (177 µm/a - 209 µm/a) differ among one another only about 18% at the maximum (Figure 1). As such differences are within the statistical variations of the measured values, it can be concluded that initial pH values of the brine between 3 and 7 have no significant influence on the steel corrosion. Further investigations at lower pH values of 1 and 2 are in progress.

Effect of gamma radiation on the corrosion of Ti99.8-Pd in Q-brine

Irradiation corrosion studies up to 1 year were performed on Ti99.8-Pd in the Q-brine at 150°C and a gamma dose rate of 10 Gy/h. The experimental setup is described in a previous work [1]. Under the test conditions applied, the material is resistant to pitting corrosion both with and without irradiation. Furthermore, its general corrosion is, as expected, negligible low (< 1 µm/a), and a gamma radiation field of 10 Gy/h does not increase the corrosion rate of the material. Further investigations on welded specimens are in progress.

C.1.2 Electrochemical and radiochemical studies (FU)

The influence of the radiolytic product H_2O_2 on corrosion of Ti99.8-Pd was investigated in saturated NaCl brine at 25°C. Experiments were performed both at rest potential and at various anodic potentials of 0 V, 0.5 V and 1 V (passive range). The H_2O_2 concentrations in the brine varied between $6x10^{-4}$ and $1.7x10^{-1}$ mol/l. At the rest potentials (350 - 450 mV), the corrosion rate of Ti99.8-Pd is proportional to the H_2O_2 concentration in the brine (Figure 2). For the H_2O_2 concentration of $6x10^{-4}$ mol/l, which is realistic for disposal conditions, the corrosion rate is 0.5 µm/a which is very close to the value in the H_2O_2 free brine (0.1 µm/a) obtained in previous work [2]. This indicates that H_2O_2 concentrations in the brine smaller than 10^{-4} mol/l do not influence the corrosion rate of Ti99.8-Pd.

The potentiostatic measurements on Ti99.8-Pd were performed within the potential range from 0 V to +1 V (passive range) at 25°C and a high H_2O_2 concentration in the brine of $6x10^{-2}$ mol/l. Under these conditions, corrosion rates of 6 - 16 µm/a were measured which are clearly higher than the value measured in the H_2O_2-free brine (1 µm/a).

C.1.3 Stress corrosion cracking studies in salt and granitic environments (B.1.3 + B.2) (E)

The resistance of three candidate container materials (carbon steel TStE355, stainless steel AISI 316L and Ti99.8-Pd) to stress corrosion cracking (SCC) is investigated by means of the slow strain rate technique (SSRT). In the reporting time, following work has been done:

- Chemical, mechanical and metallographic characterization of the materials.
- Performance of slow strain rate (SSR) tests on the TStE355 steel and Ti99.8-Pd in NaCl-rich brine (25.9 wt.% NaCl) at 170°C and strain rates of 10^{-4}-$10^{-7}s^{-1}$.

The results of the SSR tests on unwelded and Flux cored arc (FCA) welded steel specimens show that the elongation, reduction of area, energy and true stress at fracture are clearly lower in the brine than in argon. In the metallographic examinations shallow secondary cracks (less than 12 µm/a) were observed for the unwelded and welded specimens after testing in the brine at the slowest strain rate of $10^{-7}s^{-1}$. This indicates a very slight sensitivity of the steel to SCC under the severe conditions of the SSR test. Scanning electron microscopic (SEM) examinations show a change from a fully ductile fracture surface (dimples) for specimens tested in argon, to a more brittle one when tests are performed in the brine.

The results of the SSR tests on Ti99.8-Pd (Table I) do not show any loss of ductility for unwelded electron beam (EB) and plasma arc (PA) welded specimens under the test conditions applied in brine and argon. Furthermore, no secondary cracks were observed in the metallographic studies. The fracture surface is fully ductile both in brine and argon, which indicates the resistance of this Ti-alloy to SCC in test brine.Further stress corrosion cracking studies are in progress.

C.2 Electrochemical corrosion studies in clay/bentonite environments (S)

The influence of temperature (16°C and 30°C) and Cl^- concentration (16, 1000 and 10000 ppm) on the corrosion behaviour of steels, Hastelloy C4 and Ti99.8-Pd was investigated in synthetic interstitial clay water. Emphasis is laid on the study of carbon steel and stainless steels. The results obtained so far can be summarized as follows:

- At all test temperatures and Cl^- concentrations, the high alloyed stainless steels (UHB 904L, Cronifer 1925hMo), the nickel alloy Hastelloy C4 and the Ti alloy Ti99.8-Pd are resistant to general, pitting and crevice corrosion
- The stainless steel AISI 316L is also resistant to general and local corrosion at all test temperatures and a Cl^- concentration of 16 ppm. However, at higher Cl^- concentrations (1000 and 10000 ppm) the steel exhibits crevice and/or pitting corrosion. For this steel, the breakdown potential decreases with increasing Cl^- content, which indicates an enhanced susceptibility to local corrosion.
- The carbon steel TStE355 undergoes significant general corrosion, mainly at the higher temperature (30°C) and the higher Cl^- concentrations in clay water.

Further electrochemical studies are in progress.

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GRAMBOW, B., SMAILOS, E., GECKEIS, H., MÜLLER, R., HENTSCHEL, H., „Sorption and Reduction of Uranium (VI) on Iron Corrosion Products under Reducing Saline Conditions“, Radiochimica Acta 74, 149-154 (1996).

Table I: SSR test results for welded and unwelded Ti 99.8-Pd (TiGr7) specimens in argon and NaCl-brine (170°C, 10^{-7} s^{-1})

	TiGr7		TiGr7 EBW		TiGr7 PAW	
	Argon	NaCl	Argon	NaCl	Argon	NaCl
Elongation (%)	30.0	31.3	23.6	26.4	19.1	18.2
Reduction in area (%)	71.9	68.7	68.7	69.2	69.4	70.8
Yield Strength (MPa)	178	181	187	185	194	199
True Stress Fract.(MPa)	538	540	581	573	592	577
Maximum Load (MPa)	228	243	246	244	248	239
Secondary cracks	None	None	None	None	None	None
Failure mode	Dimples	Dimples	Dimples	Dimples	Dimples	Dimples

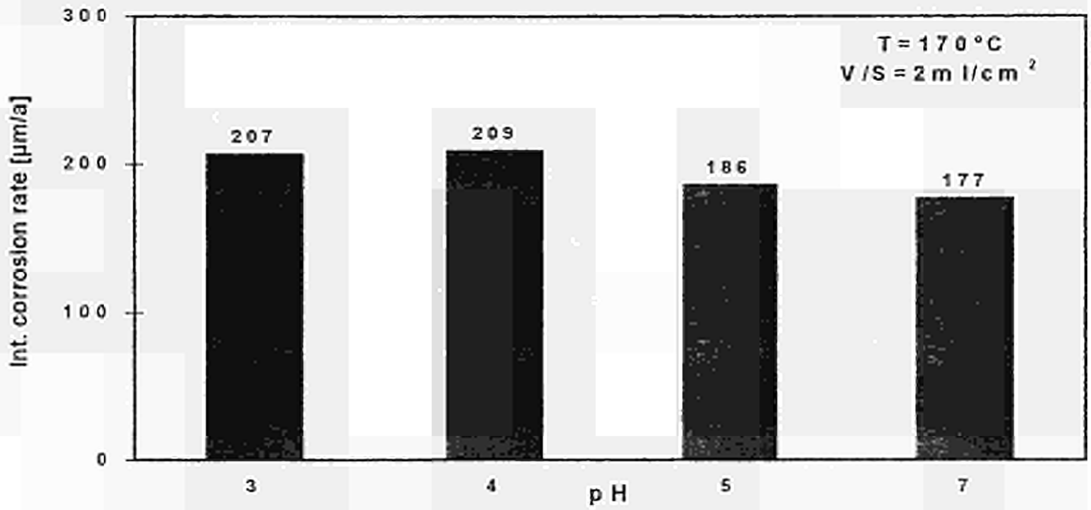

Figure 1: Corrosion rates of TStE 355 steel as a function of pH in Q-brine

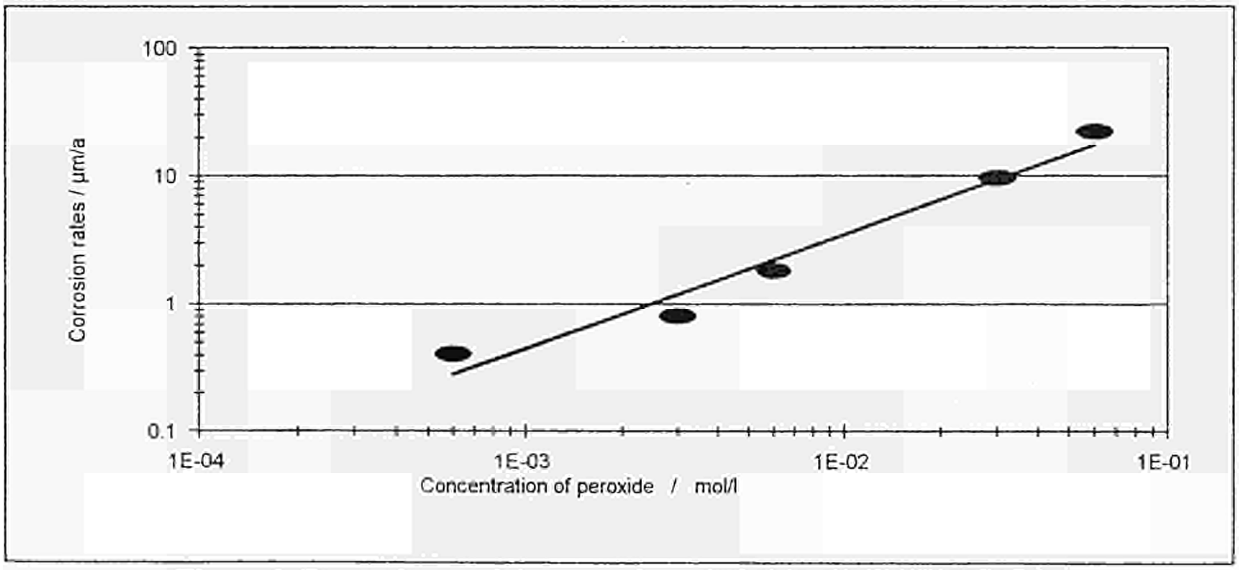

Figure 2: Dependence of the corrosion of Ti 99.8-Pd on the H_2O_2 concentration at rest potential (saturated NaCl-brine, 25°C)

C.3.2-3 Source term for performance assessment of spent fuel as a waste form

Contract No:	FI4W-CT95-0004	**Duration:**	1 Jan. 1996 - 31 Dec. 1998
Coordinator:	**B. GRAMBOW, FZK Karlsruhe/DE** Tel: +49 7247/825494, Fax: +49 7247/823927		
Partners:	**ENRESA Madrid/ES, CEA Valrhô/FR, Univ. Berlin/DE, JRC-ITU Karlsruhe/EC, SCK-CEN Mol/BE, STUDMAT Nyköping/SE, VTT Espoo/FI**		

A. OBJECTIVES AND SCOPE

The study aims at the understanding and quantification of processes controlling long-term dissolution/alteration of the fuel and associated radionuclide release/retention/reprecipitation in groundwaters or saline brines of underground repositories in granite, salt and clay formations. Five types of experiments are to be performed: dissolution tests, co-dissolution tests, precipitation tests and co-precipitation tests and electrochemical tests. Dissolution and co-dissolution tests will provide rate data and may allow the system to approach quasi-equilibrium conditions (saturation, steady state, etc.) from initially undersaturated conditions while precipitation and co-precipitation tests should facilitate an approach towards quasi-equilibrium from oversaturation. Fuel reaction rates and radionuclide retention mechanisms are to be determined as a function of groundwater composition, radiation field, the presence of canister corrosion products, backfill and repository rock. The objective is to develop models for assessing radionuclide release from spent UO_2 and MOX fuels for relevant long time spans. The model for radionuclide release will combine geochemical reaction paths, dissolution precipitation and mass transfer kinetics and radiolysis effects and be applied to source term quantification for each geological medium.

B. WORK PROGRAMME

B.1 Basic understanding of long-term spent fuel dissolution process

Basic understanding of spent fuel dissolution kinetics is provided by dissolution tests and by electrochemical tests. *Dissolution tests* of Spent (UO_2 and MOX) Fuel, SIMFUEL and UO_2 in various groundwaters intend to provide data on reaction rates, rate controlling processes and in some cases the results may give upper limits of release of significant radionuclides including colloidal contributions. Spent fuel matrix dissolution rates are to be assessed mainly from release data of Sr90. Colloid formation is analysed by ultrafiltration techniques. In the activities 1.1, 1.3, 1.4 and 1.5 the parameters pH, partial pressure of CO_2 and, in some cases, temperature, redox conditions granitic groundwater composition, S/V and humic acid contents are varied.

1.1 Spent fuel static dissolution test (FZK).
1.2 Surface area measurement of fuel powder (JRC-ITU).
1.3 Dissolution of Spent MOX and UO_2 fuel rod segments with preset defects (JRC-ITU).
1.4 Static dissolution of UO_2 and SIMFUEL (ENRESA, VTT, SCK/CEN).
1.5 Dynamic dissolution tests of UO_2 (ENRESA).
1.6 Electrochemical investigations on UO_2 and SIMFUEL electrodes (FUB).
1.7 Electrochemical investigations of spent UO_2 and MOX fuel electrodes (JRC-ITU).

B.2 Radionuclide retention during spent fuel dissolution

Due to solubility and sorption processes solution concentrations of many long-lived radionuclides are significantly lower than expected from congruent fuel matrix dissolution. This retention behaviour will be quite different in the various geological media due to the pH/Eh and pCO_2 dependency of sorption/(co)precipitation equilibria and due to differences in colloid stability.

Environmental parameters such as pH and pCO_2 will be varied in order to provide a database allowing a thermodynamic rationalisation of individual radionuclide behaviour.

The combination of these tests with the (co)precipitation tests helps to identify upper limits of solution concentrations (equilibrium, apparent solubility, etc.) and stable phases of significant radionuclides in the vicinity of potentially dissolving spent fuel.

2.1 Spent fuel, SIMFUEL and UO_2 precipitation tests (FZK, ENRESA, VTT).

2.2 Coprecipitation tests with Pu (ENRESA).

2.3 Determination of oxidation states during spent fuel dissolution (FZK).

B.3 Effect of near field materials

The presence of near field materials such as metallic canister materials, their corrosion products, clay backfill materials (bentonite) or the host rock minerals will strongly alter the dissolution behaviour of the fuels. The aim of the present study is to assess the effect of certain key materials: iron, iron corrosion products, bentonite and granite on spent fuel dissolution.

3.1 Co-dissolution of spent fuel, UO_2 and SIMFUEL with metallic iron (FZK, VTT, ENRESA).

3.2 Co-dissolution tests of UO_2 with iron (III) corrosion products (ENRESA).

3.3 Integral co-dissolution tests of spent fuel and SIMFUEL with rocks and/or backfilling materials (CEA).

B.4 Modelling

Model and source term development will be performed in three steps: (1) Process models, based on fundamental physical and chemical principles, shall accurately describe the experimental observations. (2) Process models will then be scaled up to relevant disposal configurations and decay times and temperature regimes (3) Finally, the obtained model shall contribute to source term quantification.

4.1 Geochemical reaction path and actinide chemistry (FZK, VTT, ENRESA).

4.2 Modelling of solid solution formation (FZK, ENRESA).

4.3 Modelling of radiolysis product formation (STUDMAT, FZK).

4.4 Source term quantification (FZK, CEA).

C Progress of work and results obtained

Summary and main issues: Procedures have been drafted for all experiments, including sample and leachant preparation, hot cell and glove box lay outs. Most experiments have been started and preliminary results are obtained. Dissolution rates of spent fuel appear similar in granite and salt environment both in presence and absence of iron (container material) being present. Similarly, dissolution rates of unirradiated UO_2 appear to be similar in air saturated saline (contractor V) and nonsaline (contractor E) granite water. Better understanding of redox effects on fuel corrosion were obtained: In granite groundwater, iron and reducing groundwater species controlled of U concentrations in solution $<10^{-7}$ M). Furthermore, a new method is developed to simulate the effect of oxidants in increasing UO_2 dissolution rates and corrosion potentials by applying external potentials on UO_2-electrodes. Spent MOX and UO_2 fuel electrodes were produced, the former showing lower corrosion potentials than the latter. With respect to radionuclide behavior upon spent fuel dissolution, similar solution concentrations of actinides were observed in dissolution and precipitation tests indicating coprecipitation as an important retention mechanism.

C.1 Basic understanding of long-term spent fuel dissolution process

C.1.1: Spent fuel static dissolution tests (F) Dissolution experiments of spent fuel powder in granite water have been conducted for about 200 days, including a wash cycle of 50 days to remove gap inventories and oxidized surface layers. Preliminary results indicate that the extent of dissolution of fuel powder in granite water is similar to NaCl-media (ca. 0.1% fractional release of Sr90).

C.1.2: Surface Area Measurement of Fuel Powder (IT) A Hg pycnometer for porosity determination of irradiated fuels used in the accompanying studies has been adapted and installed in a hot cells. Calibration and testing of samples will be carried out in the second half of this year, when burst test studies in this cell are completed.

C.1.3: Dissolution of spent MOX and UO_2 fuel rod segments with preset defects (IT): 4 UO_2 (burn-ups 20 - 55 GWd/tU) and 2 MOX fuel (burn-ups 23 - 55 GWd/tU) rods irradiated in BR-3 (Mol, Belgium) have been selected for experiments simulating fuel rod failure conditions. Approximately 6 cm long rodlets have been cut, stainless steel end caps screwed on and then defects cut in the cladding (a set of 3 x 1 mm ∅ holes at mid-height except for one with 2 sets of 3 x 1 mm ∅ holes at each end). The initial micro-structure of these fuels has been characterised by optical and scanning electron microscopy. For UO_2 fuels grain size is approx. 15 µm with large intragranular and intergranular pores. For MOX fuel the small-grained porous Pu-rich agglomerates are clearly visible in the UO_2 matrix, particularly at crack surfaces, and the fuel outer edge. Fuel rodlets were placed in fully-filled autoclaves for leach testing in demineralized water at 100° C. One autoclave containing the rodlet with the double set of defects is half-filled with demineralized water at 100° C so that one set of defects are immersed and the second set is only exposed to water vapor.

C.1.4: Static Dissolution of UO_2 and SIMFUEL (E, V, S) *UO_2 in 5 m NaCl and Granite-Bentonite Synthetic (GBW) water (E):* Static dissolution experiments were performed with UO_2(s) powder (particle size 50-100 µm, S/V of ≈1000 m^{-1}, 25°C). The evolution of uranium concentrations (see first part of Figure 3) under anoxic conditions in 5 m NaCl solution is similar to that under reducing conditions (H_2) reported in the previous EU project. This indicates that anoxic conditions are sufficient to maintain low uranium concentrations in solution. The decrease of uranium concentration with time confirms that UO_2 acts as a redox buffer: the redox potential remained constant at ≈ -300 mV_{SHE}. Static experiments in GBW-water were startet.

SIMFUEL in 5 m NaCl and GBW water (E): Uncorroded SIMFUEL powder of the selected particle size (50-100 µm) was characterized by SEM and TEM . The surface area was determined by BET method (0.043 m^2/g). The experimental procedure is described in detail in [2]. SIMFUEL leaching experiments were started both in 5 m NaCl and GBW media, both in absence and in presence of container material (Fe). Measured uranium concentrations showed strong discrepancies. Samples will be analysed again.

Dissolution Experiments of UO_2 pellets in saline granite groundwater (V): Dissolution experiments (25°C) with unirradiated UO_2 pellets in synthetic saline granite groundwater [2] were initiated both under oxic (air-saturated) and anoxic (anaerobic glove box, 99.999% N_2 atmosphere) conditions at 3 S/V values between 0.66 and 19.8 m^{-1}. The redox state (Eh) of the experiments under anoxic conditions was 0 - 130 mV_{SHE}. Under these conditions U has been analyzed to be in the hexavalent state. Lower Eh values (-255 to -170 mV_{SHE}) were obtained the addition of natural redox species of groundwater (0.01 -1 ppm Fe(II) and 1-5 ppm S(II)). Metallic iron was added in two tests. Under these conditions it is expected that dissolved uranium is predominantly in the tetravalent state.

Under *anoxic* conditions (see Figure 2) the solution concentration of uranium was constant after few days ($5 \cdot 10^{-8}$ M at S/V=19.8 m^{-1}, $1\text{-}2 \cdot 10^{-9}$ M at S/V=0.66 m^{-1}). The addition of reducing species under nitrogen atmosphere lowers the uranium concentrations in synthetic saline granite groundwater by a factor of 2-10. Also the addition of metallic Fe to the experimental system seems to lower the uranium concentrations in solution. Under *air-saturated* conditions, the rate of accumulation of Uranium in solution is constant and is proportional to surface area. The dissolution rate is $0.5\text{-}1 \cdot 10^{-3}$ g m^{-2} d^{-1}.

Static Dissolution of UO_2 in Interstitial Boom Clay Water. (S): Preliminary calculations performed at SCK•CEN predict that at a pH of 8.2 to 8.5 (likely the pH in the undisturbed Boom Clay host rock), uranyl-carbonate complexes will be the dominant solution complexes whereas at a pH lower than 6.5, humic acid complexes would be the predominant. Depleted UO_2 powder will be immersed at 25°C in real ("RIC") and synthetic ("SIC") interstitial clay water for various test durations using a target SA/V of 1000 m-1. An argon- and nitrogen purged glove box have been installed. Due to security regulations, the original glove-boxes had to be adapted. A third glove-box, purged with an argon/CO_2 mixture, will be installed in March 1997. The start-up of the experiments is foreseen in the first semester of 1997.

C.1.5: <u>Dynamic Dissolution Experiments</u> (E): Tests were performed with ≈ 0.1 g of UO_2(s) powder (particle size 100-300 µm) enclosed in a continuos thin-layer flow through reactor. The reaction rate in air saturated *Granite/Bentonite/Synthetic (GBW) water* was determined as $3.3 \cdot 10^{-4}$ g/m^2d. *The effect of radiolysis products* (0.1 m NaClO) was studied in 5m NaCl solution under anoxic conditions at pH 11.3 and 25°C. Final dissolution rates were ≈ 10^{-3} g m^{-2} d^{-1}, this value is lower than the one obtained in batch experiments in the previous EU project. Changes in the oxidized solid surface could be the responsible of these differences.

C.1.6: Electrochemical investigations on UO_2 and SIMFUEL electrodes (FU): A procedure was developed to simulate the effect of oxidizing agents on UO_2-dissolution by the use of electrochemical methods: First the dependence between potentials applied on the UO_2 electrode and radiochemically determined corrosion rates has measured for a given groundwater composition. Then the applied potentials were corrected for the potential depending resistance of the UO_2 electrode, using impedance measurements in the same groundwater. The resulting dependence between corrected applied potentials and UO_2-dissolution rates agrees well with a corresponding relation between these rates and the rest potential (open circuit potential) in presence of air and additional oxidizing agents ($KMnO_4$ or $K_2Cr_2O_7$) (see Figure 1).

C.1.7: Electrochemical investigation of spent fuel and MOX electrodes (IT) Electrodes have been prepared from three irradiated UO_2 fuels (30 - 54 GWd/tU) and a MOX fuel (20 GWd/tU), including polishing both sides to ensure good electrical contact between fuel and metallic support. The long term potential monitoring (up to 1000 h) in 95 % saturation NaCl brine at 25° C at neutral pH has shown that spent MOX has a lower rest potential E_{corr} = -270 mV_{SHE} and a different evolution in its E_{corr} value than the spent UO_2 fuels. For UO_2 fuels the highest burn-up fuel shows the highest (most anodic) E_{corr} value of - 96 mV_{SHE}. Optical microscopy of the high burn-up UO_2 fuel after 1000 h in 95 % NaCl shows considerable localised attack of the fuel surface. Often areas around larger pits or cracks are only slightly attacked. This may be due to the anodic pit making the surrounding area cathodic and hence protecting it from attack. Furthermore the SACV technique was modified and allows now very low scan rates down to 0.1 µV/S which correspond to a frequency of ~2.5 µHz in terms of impedance measurements. Both techniques combined can give more significant values for the polarisation resistance, which are expected in the range of some 10^6 ohms.

C.2. Radionuclide retention during spent fuel dissolution

C.2.1: Spent fuel, SIMLUEL and UO_2 precipitation tests: (F, E) *Spent fuel (F,E)*: Precipitation tests were performed inside a hot cell facility under remote control. Aqueous starting solutions of aqua regia dissolved spent fuel (burnup of 50 MWd/kg U) in 5 m NaCl solutions were titrated under anoxic conditions with NaOH to the selected pH values. This resulted in the instantaneous formation of yellow or orange precipitates (Na-polyuranates). Elements such as Sr or Cs remained in solution whereas the rare earth and actinide elements precipitated together with Uranium. The apparent equilibrium concentrations of Am, Cm, Pu and Np in solution were similar to those observed in spent fuel leaching test of the EU-project (1990-94). This indicates that solution concentrations of actinides encountered previously represent maximum values for the given chemical environment. As an example, in Figure 2, a comparison of results from precipitation and dissolution tests is shown for Np273. Coprecipitation of Np and Na-polyuranates is the likely cause for the agreement in the data.

SIMFUEL (E): For precipitation studies, a SIMFUEL pellet was dissolved in strong acid under oxidizing conditions. Aliquots of the SIMFUEL solution were added either to carbonate free 5 m NaCl solution or to air saturated granite-bentonite water. The pH of this solution was adjusted to the selected pH value by addition of NaOH. Preliminary results are available for NaCl-solutions in the pH-range 7 to 9.3. Due to precipitation, yellow to orange phases were formed. In parallel tests, at a given pH, apparent equilibrium concentrations of Uranium in solution (0.22 µm filtrate) varied from test to test by up to a factor of 50. This may be correlated with the particle size of the precipitates formed. Moreover, comparison of filtered and ultrafiltered solution samples indicated colloid formation. Minor elements concentration are not affected by the variability in the uranium concentrations. (see Table I). Sr did not precipitate at any pH value tested. La, Ce, Nd, Rh, Pd and Y concentrations in solution decrease with pH. At pH value 9.3 the concentration in solution decreased one order of magnitude with respect to the initial concentration in solution.

C.2.2: Coprecipitation tests with Pu (E): Glove box conditioning and isotopic analyses of the initial Pu solution were performed. Separation Pu-Am is going to be performed before starting the experiments. Experiments will start in the first half of 1997.

C.2.3: Determination of oxidation states of U and Pu in spent fuel leachates (F): Work will start early 1997.

C.3 Effect of near field materials

C.3.1: Co-dissolution of spent fuel, SIMFUEL and UO_2 with metallic iron (F, E, V):

Spent fuel (F): Static Co-dissolution tests were started with powdered high burnup spent fuel in the presence of iron powder and granite water (25°C, S/V = 3000 m^{-1}). Preliminary results indicate that dissolution rates are about a factor of 3 lower than the corresponding rates in absence of iron. The effect of metallic iron powder appears to be similar to that found for fuel pellets in 5 m NaCl-solutions (EU-project 1990-1994).

SIMFUEL (E): experiments were started

UO_2 (E): Static co-dissolution tests with unirradiated UO_2(s) powder in anoxic (N_2) 5 m NaCl solution (25°C, S/V ratio of 1000 m^{-1}) were performed in three steps: (1) Determination of U concentration as a function of time. (2) After reaching constant U concentrations, Fe powder was introduced. (3) After equilibration, a 10^{-6} m U(VI) concentration was introduced into the vessel. The results are shown in Figure 3. In the absence of Fe, U concentration decreased with time probably due to the redox buffer capacity of the UO_2(s) (see C.1.4). In the presence of metallic iron, the concentration decreases further because of the Fe reducing capacity. The U(VI) added at the third step is rapidly reduced and precipitates as UO_2.The final U concentrations were similar to those obtained in the EU project (1990-1994) with spent fuel in the presence of Fe. In a parallel test UO_2 particles were observed (by EDX) on the iron.

C.3.2: Co-dissolution of UO_2 with Fe(III) corrosion products (E): Experiments have not been started

C.3.3: Integral Co-dissolution tests of SIMFUEL and spent fuel with rock and/or backfill material (C): Equipment was set up in the *Clovis* hot cell complex; two process lines were prepared and installed in the cells and two identical process lines were set up for workbench use in a restricted access zone; acceptance and qualification tests were carried out; preparations were completed for start-up of the leaching tests and the first tests were initiated. A specific process was developed, set up and tested to allow simultaneous investigation of spent fuel and SIMFUEL leaching behaviour under identical experimental conditions in the presence of near field materials. The basic concept and its hot cell implementation are given in [2]. Two non-radioactive lines (four autoclaves) and two radioactive ones were started.

Experimental: Fuel powder and SIMFUEL powder samples were prepared with a specific area ranging from 0.01 to 0.02 $m^2 \cdot g^{-1}$. Reducing leaching conditions were adjusted with a Eh of –90 to –20 mV_{SHE}. An overall activity balance is attempted by measuring the radionuclides in the leachates and rinsing solutions by radiochemical analysis, $\alpha\gamma$ spectrometry and ICP-MS. A protocol for recovery and determination of the radioactivity sorbed on the environmental materials was specified [3]. The tests will be terminated at the beginning of March 1997.

C.4: Modeling

C.4.1: Geochemical reaction path and actinide chemistry (F, V, E) V: Preliminary modeling of uranium equilibrium solubilities in saline reference groundwater will be done using the geochemical code EQ3/6 (version 7.1 R121). The thermodynamic databases are the NEA and COM (composite) data bases.

C.4.2: Modeling of solid solution formation (F, E): Ideal solid solution models are currently tested to describe the behavior of trivalent elements during coprecipitation in the experiments of C.2.1.

C.4.3: Modeling of radiolysis product formation (ST) Modeling files were established for oxidation and dissolution of UO_2 by the products of water radiolysis and for radiolysis of aqueous chloride solutions. The file for radiolysis of aqueous of chloride solutions has been evaluated by FZK. Calculation of radiolysis effects from the previous EU-project on spent fuel were started. In these experiments, gas production and spent fuel dissolution rates were measured simultaneously. Distinction is made between three spatial zones in the aqueous medium: 1. A thin 30-50 µm layer of water on the surface of an irradiated UO_2 pellet is irradiated with α,β and γ radiation, total dose rate of 9520 Gy/h. 2. Irradiation of further 0.3 cm with β and γ at 2320 Gy/h and 3. γ irradiation of the rest of the solution with a mean dose rate of 80 Gy/h. Using only data from zone 1, corrosion rates of 0.02 $g.m^{-2}d^{-1}$ were predicted.

C.4.4: Source term quantification (F, C): First contacts were made with the SPA-project of the ongoing EC 1994-1998 research program. The fundamental assumptions inherent in the suggested source terms of the participants of SPA-group were critically evaluated, considering the uncertainties in the current knowledge of rate limiting mechanism in spent fuel long-term performance and conservative approaches are suggested. Certain open questions can be clarified within our ongoing project: the contribution of the cladding to radionuclide release, the effect of U(IV) sorption on metallic iron, the effect self-oxidation by radiolysis and the relation between semiconductor properties of UO_2 and dissolution mechanism and rates.

References

[1] K. Olilla, Mat. Res. Soc. Symp. Ser. 1997 in press
[2] Source term for performance assessment of spent fuel as a waste form, semiannual Report 01.01-30.06 1996 (July 1996)
[3] K. Le Lous, CEA Procedure SDMC/PR/79 issue A (August 1996)

Figures

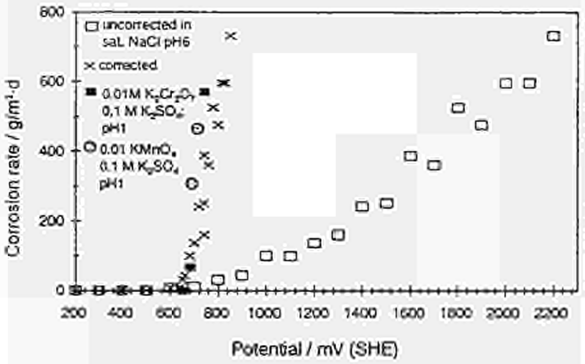

Figure 1: Dependence of UO_2 corrosion rates on applied potentials before and after correction - Comparison with corrosion rates/rest potentials in presence of oxidizing agents

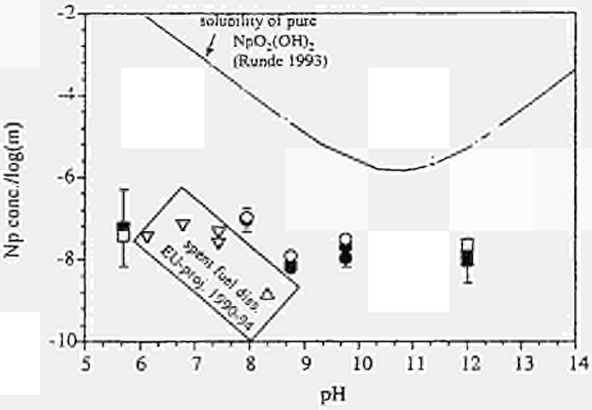

Figure 2: Comparison of Np data resulting from spent fuel precipitation with spent fuel dissolution data (EU-project 1990-1994) and with $NpO_2(OH)_2$ solubility

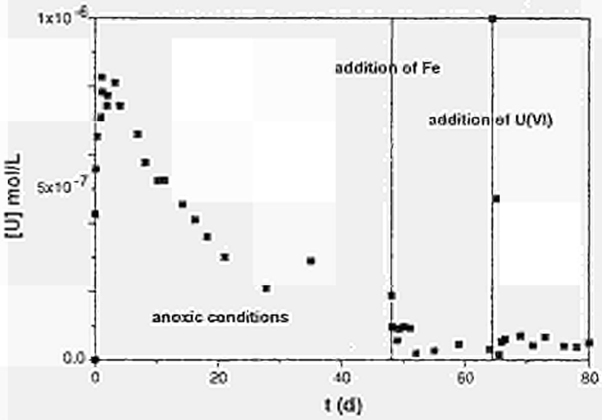

Figure 3: Dissolution of UO_2 in anoxic 5m NaCl solution. Temporal evolution of U concentration as a function addition of Fe. (C.3.1).

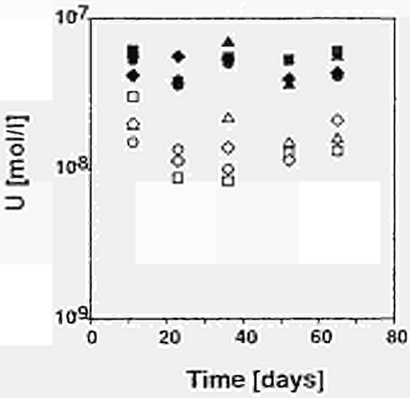

Figure 4: U concentrations in synthetic saline granite groundwater (anoxic, S/V=19.8 m^{-1}) filled symbols: no redox species, open symbols: 1 ppm S(II) and 0.01 ppm Fe(II)

Table I: Ratio of initial to final concentration in SIMFUEL precipitation tests (C.2.1)

pH	*Ba*	*Ce*	*La*	*Mo*	*Nd*	*Pd*	*Rh*	*Ru*	*Sr*	*Y*	*Zr*
7.4	2.1	6.6	4.9	57.9	8.7	25.7	126	5.6	1	8.1	--*
9.3	4.3	(*)	---	2.8	---	39.8	---	---	1.7	---	2956
9.3	3,7	(*)	---	2.5	---	---	---	---	1.3	---	--*
9.3	2.9	211	29	2.7	---	17.9	---	21*	1.3	23.7	--*

C.3.3-1 Round Robin test for Non Destructive Assay of 220 litre radioactive waste packages

Contract No:	FI4W-CT95-0007 **Duration:** 1 Feb. 1996 - 31 Jan. 1999
Coordinator:	**L.P.M. van Velzen, KEMA, Arnhem/NL** Tel: +31 26/3562593, Fax: +31 26/4433635
Partners:	**CEA Cadarache/FR, ENRESA Madrid/ES, ENEA Casaccia/IT, KFA Jülich/DE, Univ. TUM München/DE, SCK-CEN Mol/BE, Taywood Southall/UK, BELGOP Dessel /BE, JRC-ISIS Ispra/EC**

A. OBJECTIVES AND SCOPE

In most national quality checking laboratories of the European Union, equipment and procedures have been developed to measure waste forms for interim storage or disposal.

These equipment and procedures are based on various hardware and software techniques. An intercomparison (proficiency) test between the different laboratories and all their NDA techniques is a proven method of validating the results of these systems.

The main objective of this project is to obtain comparable representative data with various non-destructive examinations to improve the precision of the present assaying techniques for radioactive waste packages.

The intercomparison will be performed for fissile and non-fissile nuclides in 220 litre drums filled with representative matrix material. The specimens will be submitted to a wide range of NDT, e.g. gamma spectroscopy and scanning, ECT and TCT tomography, radiography, dose rate measurements, active and passive neutron measurement, etc.

B. WORK PROGRAMME

B.1 Selection of 220 litre packages

A total of 16 drums with non-fissile nuclides will be selected and prepared by the participants. Participants will seal and measure their respective specimen. A maximum of four drums containing fissile material will be provided by ENEA, CEA, KFA and SCK-CEN.

B.2 Equipment and procedures

All participants will review their equipment and procedures to assure satisfactory operation for the test; where necessary, modifications should be carried out to accommodate the standard 200 litre drum. A description of all equipment and procedures should be prepared.

B.3 Round Robin Tests

First, all participants will define the package characterisation parameters and data libraries. The waste packages will be transported to the participants in a sequence which will permit testing different packages simultaneously at a laboratory. The packages with fissile material will be submitted to passive and active neutron counting, the other packages will be assayed by gamma spectrometry.

B.4 Analysing test results

The analysis of the results will be reported by all institutes according to a predefined charactertisation list. A statistical evaluation procedure agreed by all partners will be applied to all results to determine the performances.

B.5 EC guideline or standard

After analysis of the results, a classification of the various techniques and procedures for assaying waste packages will be drawn up and published as part of a guideline.

C. Progress of work and results obtained

Summary of main issues

This European "Round Robin Test for non destructive assays of 200 litres radioactive waste packages" will be performed on 220 liters packages with waste from the nuclear industry. In this first project year effort has put in defining the detailed actions of each milestone, finishing milestones 1 & 2 and making preparations for the transport of the packages.

All 200 litres packages for the non-fissile as well as for the fissile Round Robin Test are carefully selected by the parcipants (milestone 1). These packages are selected out off a large number of packages from the different member states and will be provided to the project by the different partners. The detailed specification of each package fulfils the requirements of all participating partners. In total 18 packages are selected, 14 non-fissile and 4 fissile packages.

For an optimum result of this intercomparison existing equipment and procedures, which are tuned for national 200 litres packages, have to be modified (milestone 2). Most of the necessarily modifications are performed or are at a finishing stage.

The transport of the radioactive 220 litres packages is a difficult logistic problem, due to the nature of the transport, but also due to the limitations (total radioactivity, maximum number of packages, loading/unloading times of the truck, etc.) set by the different partners. At this moment a complete transport scheme is completed for the 14 non-fissile and 4 fissile packages, where the individual requirements of each partner is taken into account.

C.1 Selection of 200 litres packages

In order to focus the Round Robin Test on realistic conditions the form of the selected sources is restricted to 200/220 litres conditioned waste for non-fissile packages and artificial waste for fissile packages. The classifications of the non-fissile drums, which can be allowed in this Round Robin Test, are LSA I, LSA II or LSA III. These classifications are defined and described in the ADR (Accord Européen relatif au transport international des marchandises dangereuses par route Trb. 1959, 81).

The classification of the fissile drums is according 'page 12' of the ADR. The 200 litres sources with fissile material will be made out of existing reference standards which are present at the moment at some of the participating institutes. The packages will cover the range of matrices with a light density from 400 kg/m^3 up till bitumen with a density of 1300 kg/m^3. The amount of fissile material will not exceed the amount of 3 gram ^{239}Pu or ^{235}U per 100 kg. Due to the restricted amounts of non-fissile and fissile material the transport of these packages in the different Round Robin Tests can be combined.

The specified drums, which are selected by the partners for the Round Robin Test, have to fulfil to certain specifications. Therefore a special parameter list has been developed. This list is a compilation of all items, which have to be specified in the participating countries. Specified limitations are set by the ADR, handling licenses of partners and equipment restrictions.

The parameter list (updated till 16 December 1996) is given in appendix A. In this parameter list the detailed specifications of the 200 litres non-fissile reference drum (KEMA Test Vessel) is shown.

In table I an overview is given of the final selection of the 200 litres non-fissile drums and in table II an overview of the selection of the fissile drums, which is made by the project steering committee.

Table I Overview of the selection of 200 litres fissile sources.

No.	Partner	Type	Matrix	Nuclides
1	ENEA	MOX source	light Z	^{239}Pu, ^{240}Pu
2	ENEA	MOX source	high Z	^{239}Pu, ^{240}Pu
3	KFA	FUEL source	concrete	^{235}U, enriched
4	TUM/RCM	REPROCES-SING source	iron scrap	^{239}Pu, ^{240}Pu, Cm and fission pro-ducts

The radioactive content of the selected drums has been analysed by non destructive measurements. Some partners have also performed destructive sampling and analysis. All necessary data (according the parameter list in appendix A) of the drums have been transmitted to the coordinator of the project.

Table I Overview of the final selection of 200 litres non fissile drums (drum no.1 is completely homogeneous checked by KFA and CEA).

No.	Partner	Type	Nuclides	Matrix	Shielding	Density [kg/m^3]
1	KEMA	reference	natural	zirkonium ore with concrete	-	3000
2	KEMA	standard	artificial	super compacted	concrete (5 cm)	2200
3	KEMA	standard	artificial	super compacted	concrete (5 cm)	2200
4	ENRESA	standard	artificial	resins	-	1800
5	ENRESA	standard	artificial	filter cartridge	concrete (18 cm)	1860
6	WQCL	standard	artificial	raw waste	-	80
7	TUM/RCM	standard	artificial	bitumen	-	970
8	TUM/RCM	standard	artificial	super compacted	-	1800
9	CEA	standard	artificial	technical raw waste	-	170
10	ENEA	standard	artificial	concrete + BWR resins or + incin-erator waste	-	1900
11	ENEA	standard	artificial	pre compacted from decommis-sioning or a light metallic matrix	-	360
12	Belgopro-cess	standard	artificial	incinerator ashes	-	400
13	KFA	standard	artificial	concrete	-	1720
14	KFA	standard	artificial	concrete	-	1740

C.2 Equipment and procedures

In this Round Robin Test waste drums will be transported to all partners. For an optimum result of the intercom parison existing equipment and/or procedures at different laboratories have to be modified. The modifications are necessary due to the special 200 litres packages, which is the most common wast package in the participating countries, while most techniques and procedures are tuned only to national waste packages.

The changes and improvements introduced in the various non-destructive gamma assay systems affect the mechanical, the detection and control systems. The modifications are briefly described in "The Annual Progress Report 1996" [1].

C.3 Round Robin Test

An important item of the transport of the drums is the logistic problem of the transport itself. At this moment a transport schedule is setup according to the following requirements:

- each participant receives all non fissile packages
- each participant to the fissile Round Robin Test receives every fissile package
- each participant may use the measuring time for each package, which he wasable to define
- the loading, unloading times per partcipant
- a maximum of four non-fissile drums will be delivered at one time
- a maximum of one fissile drum will be delivered at one time
- limited possibilities for measuring drums a second time.

At this moment there is no data available of the selected drums, which are measured by the different partners.

C.4 Analysing test results

A first setup of the statistical data evaluation has been made by CEA. This setup has at this phase of the project only the aim to specify in general terms the objectives of the statistical evaluation of the measurements, which can be taken at least into account. The more detailed setup of the evaluation will be performed in the next period in cooperation with all partners.

The following objectives are of interest for the statistical evaluation:

- study of the repeatability in each laboratory.
- this repeatability of measurements can be characterized for each laboratory and for each package. The estimation of this repeatability will be more accurate by an increasing number of measurements
- the estimated repeatability can be compared with the calculated theoretical values for the different type of packages and also the coherence between the different measuring methods and the nature of the matrices of the packages
- comparison between laboratories. Statistical tests can be used to check if differences between results of different laboratories can or cannot be explained by the estimated repeatability per laboratory
- accuracy of fissile data. The fissile packages are made by adding certified calibration sources in artificial waste packages. This means, that mass and nuclide composition of the fissile packages are known. It will be then possible

to verify if differences between data of the certified sources (mass and nuclide composition) and experimental results can be explained by the estimated repeatability

- accuracy of the non fissile data
- the probable value of the activity per nuclide per package. This value can be estimated on the data of all measurements
- elimination of results. Elimination of results can be justified on statistical reasoning, in such cases the discrepancies and their causes will be investigated.
- calculation of a weighted mean
- estimation of the uncertainty of the associated mean values.

Next stages for preparing a final setup for the statistical treatment of the data of this Round Robin Test will be performed and tested in 1997.

C.5 EC-guideline or standard

The first set-up of the guideline will be prepared in 1997.

References

[1] Velzen, L.P.M. van; Bloem, J.; Delepine, J.; Chabalier, B.; Brunel, G.; Morales, A.; Piña, G.; Bardone, G.; Pedersen, B.; Sanden, H.J.; Filss, P.; Odoj, R.; Bücherl, Th.; Lierse, Ch.; Iseghem, P. van; Baeaten, P.; Bruggeman, M.; Lewis, A.; Daish, S.; Hendrickx, J.P.; Botte, J.; Round Robin Test for Non Destructive Assays of 200 litres Radioactive Waste Packages, Annual Progress Report 1996; KEMA Nederland B.V. report 41281-NUC 97-5096

Appendix A

PARAMETER LIST KEMA TEST VESSEL DETAILS (updated 16 December 1996)

-	Owner identification:	KEMA Nederland B.V.
-	Package identification:	KEMA TEST VESSEL
-	Data of empty drum:	
	* external dimensions (h*d)	860 * 570 mm
	* internal dimensions (h*d)	840 * 560 mm
	* material	steel
	* thickness	1 mm
	* weight (including lid and bolds)	16 kg
-	Source information	
	* fabrication date	1993-06-01
	* source description	natural zirconium ore/ENCI blast furnace mixture
	* gross weight (incl. drum)	684 kg (limit 800 kg per source, this is equivalent with 3600 kg/m^3)
	net weight (excl. drum)	668 kg
	density	3050 kg/m^3
	* shielding (not radioactive)	
	thickness side	none mm
	bottom	none mm
	top	none mm
	* chemical composition	natural zirconium ore/ENCI blast furnace mixture
-	Radioactivity content (based on net weight)	
	* total β/γ radioactivity:	115 MBq date: 930601 (limit 4 GBq)
	* total α radioactivity:	(*) MBq date: 930601 (limit 40 MBq)
	* fissile material	none MBq date: .... (limit 3g/100kg)
-	Method of determining the specific activity	
	* total β/γ radioactivity:	by measurements
	* total α radioactivity:	-
	* fissile material:	-
-	Dose Rates	
	* gamma dose rate at surface:	<5 μ Sv/h (limit 1.3 mSv/h)
	at 1 m :	<1 μ Sv/h
	* neutron doserate at surface:	0 Sv/h
	at 1 m :	0 Sv/h
-	Surface contamination	
	* alpha	none Bq/cm^2 (limit 0.037 Bq/cm^2)
	* beta	none Bq/cm^2 (limit 0.37 Bq/cm^2)
-	Toxic materials/dangerous materials:	none

PARAMETER LIST KEMA TEST VESSEL DETAILS (updated 16 December 1996)

- * Radionuclides concentration [Bq/g] date: 930601

^{226}Ra chain:	87	± 3
^{232}Th chain:	18	± 1.3
^{235}U chain:	5.9	± 0.6
^{238}U chain:	61	± 5

(*) This zirconium ore has from nature a high level of natural activity. Therefore it may be and is classified according to the ADR (line number 2700 point 2) as alpha emitters with low toxicity. In such a case (low toxicity) A1 and A2 values are unlimited for uranium and thorium chains (see line number 3700) for natural ore's (zirconium). The reason for using this special zirconium ore as additive in a cement matrix instead of ordinary sand is only done for archieving a homogeneous 200 litres source, which emits gamma's from 143 keV (^{235}U) till 1764 keV (^{214}Bi daughter of ^{226}Ra) and which can be detected easily by gamma assay systems. This Test Vessel will be transported during the Round Robin Test conform the ADR class 7 page 1.

C.3.3-2 Improvement of passive and active neutron assay techniques for the characterisation of radioactive waste packages

Contract No:	FI4W-CT95-0011	**Duration:**	1 Feb. 1996 - 31 Jan. 1999
Coordinator:	**T. BÜCHERL, TUM/DE**		
	Tel: +49 89/28914328, Fax: +49 89/3261115		
Partners:	**ANPA Roma/IT, SCK-CEN Mol/BE, KFA Jülich/DE, PTB Berlin/DE, CEA Cadarache/FR**		

A. OBJECTIVES AND SCOPE

The characterisation of neutron emitting nuclides and fissile material can be performed best with passive and active neutron assay techniques. While these methods have been used in safeguards successfully, precise measurements in waste monitoring are difficult due to the usually unknown and inhomogeneous matrices and the random localisation of the emitters in the bulk of these matrices. These problems are still present, although passive neutron counting and active neutron interrogation devices are commercially available.

The main objective of the project is the improvement of localisation and quantification of neutron emitters due to spontaneous fission (sf), (a, n)-reactions and induced fission within typical waste packages. These comprise drums ranging from 200l up to 500l filled with different (inhomogeneous) matrices with densities ranging from ca. 0.5g/cm^3 up to ca. 3.0g/cm^3.

B. WORK PROGRAMME

B.1 Passive Neutron Counting

1.1 The objective of this task is the development of the basics (i.e. a model) for improved localisation and quantification of neutron emitters based on well-defined matrices. After upgrading, the correct working of all components of the devices is tested. The subsequent measurements on phantom waste packages, which have well-defined matrices and geometry similar to typical waste, result in the data needed for the development of an interpretation model for localisation and quantification of neutron emitters. The sf- and (a, n)-emitters used for these measurements can be placed at different well-defined positions. Calculations with Monte Carlo methods accompany the measurements (TUM, ANPA, SCK/CEN).

1.2 To investigate time correlation methods, measurements on phantom waste drums with different well-defined matrices and neutron emitters are performed. The data of these measurements is the basis for the evaluation of the two topics:

i. Whether and how far TCM can be applied to real waste drums successfully;

ii. Validation of different TCM systems in use (TUM, ANPA).

1.3 The mechanics for the handling of an external moveable neutron source (e.g. ^{244}Cm) is set up. Measurements on phantom waste packages are performed after a short test phase to ensure the correct working of the device. Using this data and the results of corresponding Monte Carlo simulations, an interpretation model for the matrix correction is derived, resulting in a general algorithm that is transferred into a computer code (TUM, SCK/CEN).

1.4 The algorithms derived within tasks B1.1 - B1.3 are combined. Measurements on phantom drums and real waste packages verify the results of the data evaluation with the combined interpretation model (i.e. the developed computer code). The integration of the additional information resulting from TCM measurements in the combined interpretation model is investigated (TUM, ANPA, SCK/CEN).

B.2 Active Neutron Counting

2.1 The characterisation of two different existing sources, one based on the $^7Li(p,n)$ reaction, the other based on the (g, n) reaction either inside the matrix or on an external target will be performed by experimental studies and calculations. The parameters of these sources will be varied - as far as possible - to obtain optimum beam characteristics for the final application in an active neutron interrogation device (CEA, KFA, PTB).

2.2 The design of an appropriate detector system based on 3He and 4He detectors will be determined. This includes the theoretical and experimental determination of the detector characteristics as well as the reflection of their geometrical arrangement on the results of the measurements and the use of coincidence time correlation methods (CEA, KFA, PTB).

2.3 After the set-up and testing of the experimental equipment, experiments with random waste packages containing well-known matrices are performed taking into account the neutron source properties, the matrix composition and the detector characteristics. This will be followed by measurements on real waste packages and the test of the active neutron interrogation device in a simulative routine operational mode (CEA, KFA, PTB, TUM).

B.3 Recommendation of a State-of-the-Art and Practice Device

In this work package, the results obtained in the previous work packages are summarised and recommendations will be performed concerning State-of-the-art and practices for passive and active neutron counting devices, as well as recommendations for upgrading of existing systems. Furthermore, recommendations for the applicability of mobile devices will be given, derived from the results of the investigated stationary and mobile systems and from the information about commercially available portable neutron sources (TUM, ANPA, SCK-CEN, CEA, KFA, PTB).

C. Progress of work and results obtained

Summary of main issues

In passive neutron assay the main investigations were focused on the development of appropriate interpretation models for source localisation and quantification. Using simulated and measured data different models have been investigated. In parallel the studies of the applicability of different correlation techniques started both by experiment and by simulation.

In active neutron assay three external sources for neutron interrogation were investigated in detail. Based on these results different optimisation tasks took place considering the targets and the collimators.

Progress and results

1. Passive Neutron Counting

The investigations for setting up an interpretation model for source localisation and quantification and the testing of the performance of different correlation techniques started with measurements on artificial waste packages with homogeneous matrices consisting of concrete (density about 2 g/cm^3) and bitumen (density about 1 g/cm^3) followed by measurements of inhomogeneous matrices composed of supercompacted scrap of different densities. To perform the modelling by Monte Carlo calculations input files for the passive neutron assays of ANPA, SCK/CEN and TUM/RCM were set up and exchanged by ANPA and SCK/CEN.

At TUM/RCM measurements on test drums with different matrices were performed using a ^{244}Cm source (10 mg, $1.1 \cdot 10^5$ n/s) determining the gross counting efficiencies as a function of the positions of the neutron point source, the corresponding angular dependent count rate distributions and the correlation between the count rate distribution and the height positions of the point source and the detector respectively (Figure 1). Based on these data a preliminary simple model for source localisation was established, assuming that the detection efficiency η_i of the i-th detector depends on its shortest distance x_i to the source, on the matrix attenuation coefficient μ and on a polynomial A(z) which depends on the height z of the source position.

$$\eta_i = A(z) \cdot \exp(-\mu \cdot x_i)$$

First fitting procedures on measured data showed uncertainties of about 15 % at maximum indicating that an improvement of this simple model is necessary as it was expected.

The accuracy of a similar model based on a simple exponential neutron attenuation, as sometimes used in radiation-protection problems, and on a geometrical effect described by a r^2-law was investigated by SCK/CEN. The fact that the neutron detectors cannot be described as point detectors was taken into account by integrating the point detector response over the length of the detector. The response $R(r_i)$ of an elementary length element of detector i is then described by

$$R(r_i) = k \cdot \frac{\exp(-\mu \cdot r_i)}{(r_i + D_i)^2}$$

with r_i being the distance the neutron travels in the matrix from the source to the elementary length element, D_i being the distance the neutron travels outside the drum before entering the detector and μ being the attenuation coefficient characterising the matrix interaction with neutrons at a certain neutron energy. The factor k is a normalisation constant including the intrinsic detection efficiency. The response $R(i)$ for detector tube i is obtained by integrating over the active length of the detector. Application of a non-linear fitting routine on experimental or simulated data (Figure 2) as a function of the parameters (e.g. the distance of the source from the drum centre) showed, that the value of k is dependent on the source position due to moderation effects within the matrix. Furthermore an exponential attenuation law physically describes an uncollided intensity of a parallel neutron beam traversing a distance of material. Neutrons absorbed or scattered out of the beam are treated as being lost, but still have a chance to be detected. These neutrons will contribute to the overall response of the detector and are probably more susceptible to neutron absorption. In order to refine the model, Monte Carlo simulations of the detector response (^{3}He-detector tube/polyethylene) as a function of the neutron energy were performed (Figure 3). The results show that the energy dependent problem of neutron transport can be described qualitatively with only three energy groups, referring to high, intermediate and low energies respectively. These three energy groups will be considered in a refined model.

Simulation studies on the performance of different time correlation techniques and of neutron transport characteristics (e.g. neutron decay in space and time, die-away time, detection efficiency), when applied to large detection heads for cemented radioactive waste packages and point sources, are performed by ANPA and the University of Rome using Monte Carlo calculations. Special routines have been developed to simulate different time correlation methods (e.g. coincidence counting, multiplicity counting, time interval analysis) for different experimental set-ups. In order to validate the accuracy of these simulations, a preliminary series of experiments was performed, using a "well counter" JOMAR SYSTEMS JCC-15 and a scaled test drum (30-L, cemented, ^{252}Cf source at different positions). 30 measurements of the coincidence count rates (Real + Accidental, Accidental) and the total counting rate, each at 10 different matrix/source locations and with 3 different gate times (32, 64, 128 μs) were performed. The evaluation of the neutron source mass has been obtained from [(R+A)-A] data on the base of standard formulas and was compared with the corresponding Monte Carlo calculations (Table I). Typical delay distributions of detected neutrons for different source positions are shown in Figure 4.

2. Active Neutron Counting

The investigations were focused on the development of optimised external sources for neutron interrogation. While CEA considers a mobile (γ,n) source, PTB is working on an optimised target for a $^{7}Li(p,n)^{7}Be$-source. Due to the unexpected reactivation of their Sb-Be photo neutron source, KFA started their work with a comparison of the Sb-Be-source and the $^{7}Li(p,n)^{7}Be$-source.

At CEA experiments were performed to determine the main characteristics of the transportable linear accelerator (Varian Mini-Linatron). The maximum gamma dose rates as a function of the electron energy on the beam axis at 1 meter distance from the Bremsstrahlung target were measured using an air filled ionisation chamber with 7 aluminium slabs (five inner slabs of 0.25 mm thickness, 2.16 mm thickness at the entrance side and 1.65 mm thickness at the exit one) (Table II). The spatial distribution of the Bremsstrahlung photons was measured using a disk of natural ^{238}U at 1 m distance from the target moving it perpendicular to the beam axis. The (γ,n) and (γ,f) signatures were measured by fast neutron ^{3}He proportional counters. The variation of the neutron counts versus the position of the uranium disk (Figure 5) is representative for the photon dispersion. Due to the low neutron energy thresholds of (γ,n)-reactions for beryllium and deuterium which are equal to 1.9 MeV and 2.2 MeV respectively, the Bremsstrahlung photons with an end point energy of 9 MeV produce photo-neutrons when they strike the Be or the BeD_2 target. Different kinds of stochastic simulations using the MCNP 4A code were performed to determine the angular and spectral distributions of Bremsstrahlung radiation inside the tungsten converter (target). An analytical calculation program was developed resulting in the spectral distribution of the photo neutrons inside each slide of the (γ,n) target. In combination with the results of the Monte Carlo calculations of the photon and neutron transport, the (γ,n) production, transport and characterisation at 1 m distance from the photo neutron target was determined. The most interesting and intense (γ,n)-source corresponds to a 10 cm thick Be target. The (γ,n) production versus the angular position of the detector spot is given in Table III. The maximum neutron production corresponds to the angular position which is perpendicular to the beam axis.

An optimised neutron source for active neutron interrogation of 200-L (500-L) waste packages will be provided by PTB. This source has to fulfil several requirements: The energy of the neutrons emitted must be below 0.2 MeV to be separated from the induced fission neutrons, but must have an energy which is high enough for deep penetration (at least half the diameter of the drum investigated). In addition high neutron intensities and, optionally, a collimated neutron beam for scanning of the waste packages should be available. These requirements can be fulfilled using an external neutron source based on the $^{7}Li(p,n)^{7}Be$-reaction with $E_P \leq 1.95$ MeV. The basis for this source is an appropriate Li-target: A metallic Li-target has the highest yield but the current of the incoming photons is limited due to the low melting point of lithium (T_{melt}=180° Celsius). A further disadvantage is its oxidation instability. Another choice for the neutron producing target could be a LiF target which can withstand higher currents. But due to the $^{19}F(p,\alpha\gamma)^{16}O$ reaction high-energy photons (6-7 MeV) are emitted and could be a source of photon-induced fission neutrons. However, the cross-section for photo-fission is much smaller than for neutron induced fission. Owing to this fact it was finally decided to use a LiF-target. The influence of these high-energy photons, i.e. those from the $^{19}F(p,\alpha\gamma)^{16}O$ reaction, can be investigated by a further measurement with a CaF_2 target. Using this target only high-energy photons are produced. Monte-Carlo simulations, investigating the angular dependence of the spectral neutron fluence for both target assemblies (metallic Li and LiF) and for the design of an optimised collimator are performed.

A comparison of the characteristics of a $^{7}Li(p,n)^{7}Be$ source with a Sb-Be source was realised by KFA. Results of measurements of different natural uranium samples are shown in Table IV. For both sources the count rates increase linearly with the uranium mass with an error of about 20 %. The output is about three times higher for the $^{7}Li(p,n)^{7}Be$ source mainly due to its higher source strength resulting in an improved detection limit compared to the Sb-Be system. In Table V the fission count rates for measurements with both systems on samples of natural uranium (100 g and 400 g respectively) placed at different positions in a rotating drum filled with concrete are shown. For the Sb-Be system the count rates decrease from the inner to the outer position and a variation of 20 % of the net count rate per gram ^{235}U can be observed, i.e. the variations due to different mass content and different radial position are of the same order! The results using a $^{7}Li(p,n)^{7}Be$ system and a 100 g natural uranium show an increase of the count rates in the outer regions of the drum. This is due to a change of the geometrical arrangement: The detector was placed at 90° with respect to the accelerator beam axis. In combination with the results of further investigations performed, the measurements confirm the assumption that the best results for active neutron interrogation will be obtained by use of a $^{7}Li(p,n)^{7}Be$ neutron source.

3. Recommendation of a State-of-the-Art and Practice Device

The opportunity to perform an outdoor measuring campaign at a national intermediate disposal facility using the mobile passive neutron assay SANDRA of TUM/RCM was used to gather information and experiences on that field of operation. The results will help to define (recommend) an appropriate assay for mobile investigations.

Table I: Relevant matrix characteristics of neutron transport simulations

matrix (source location)	detection efficiency e (%) simulated	experimental	detection die-away time t_{det} (ms)
air	5.60	6.15	86
cement (1-B)	3.91	3.76	157
cement (1-C)	4.86	4.73	159
cement (1-A)	3.83	3.77	157
cement (2-B)	4.98	5.10	136
cement (2-C)	6.23	6.32	139
cement (2-A)	4.97	4.91	136
cement (3-B)	6.56	6.47	117
cement (3-C)	8.04	8.24	119
cement (3-A)	6.44	6.60	117

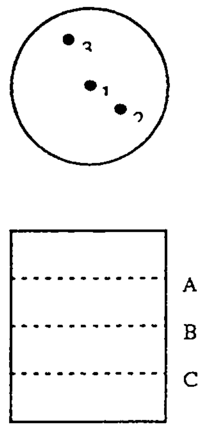

Table II: Maximum gamma dose rate

electron energy in MeV	6	9	11
max. γ-dose rate in Gy/s	0.33	0.42	0.2

Table III: (γ,n) production of a Be-target (10 cm thick, r=0.4 cm, E_c=9 MeV) versus the angular position of the detector spot.

Cartesian co-ordinates in cm (z = 0.00 cm) x in cm	y in cm	angular position in degree	neutron flux* in $n \cdot cm^{-2}$	neutron energy in MeV	neutron dose* ($\cdot 10^{-2}$ Gy)
106.30	0.00	0.0	$4.47 \cdot 10^{-11}$	2.081	$1.32 \cdot 10^{-19}$
102.89	25.88	14	$1.44 \cdot 10^{-10}$	2.297	$4.51 \cdot 10^{-19}$
92.90	50.00	28	$1.68 \cdot 10^{-10}$	2.275	$5.24 \cdot 10^{-19}$
77.01	70.71	43	$1.78 \cdot 10^{-10}$	2.262	$5.52 \cdot 10^{-19}$
56.30	86.60	57	$1.82 \cdot 10^{-10}$	2.257	$5.66 \cdot 10^{-19}$
6.30	100.00	86	$1.85 \cdot 10^{-10}$	2.304	$5.80 \cdot 10^{-19}$

(*) per electron

Table IV: Fission neutron count rates from samples of different uranium content. Measurements performed with the Sb-Be and $^7Li(p,n)^7Be$ source. Samples in the central position of a concrete filled 200 l waste drum.

	Sb-Be source (26 keV)			Li(p,n)Be source (163 keV)		
Natural Uranium content in g	Fission count rate in s^{-1}	Net fission count rate in s^{-1}	Count rate per mass ^{235}U in s^{-1}/g	Fission count rate in s^{-1}	Net fission count rate in s^{-1}	Count rate per mass ^{235}U in s^{-1}/g
	0.083			0.258		
20	0.112	0.029	0.207	0.316	0.058	0.414
50	0.140	0.057	0.163	0.479	0.221	0.631
100	0.190	0.107	0.153	0.696	0.438	0.626
190	0.267	0.184	0.138	1.040	0.782	0.588
400	0.430	0.347	0.124	1.485	1.227	0.438
	Average:		0.157			0.540
	Error:		0.032			0.105
	Relative error		20%			19%
	Detection limit in mg ^{235}U:		174			89

Table V: Fission neutron count rates for natural uranium samples in different position of a concrete filled 200 l waste drum. Interrogation by a Sb-Be and $^7Li(p,n)^7Be$ neutrons respectively.

	Sb-Be source and 400 g nat. uranium			$^7Li(p,n)^7Be$ source and 100 g nat. uranium		
Radius in mm	Fission count rate in s^{-1} (Background: 0.083 cps)	Net fission count rate in s^{-1}	Count rate per mass ^{235}U in s^{-1}/g	Fission count rate in s^{-1} (Background: 0.302 cps)	Net fission count rate in s^{-1}	Count rate per mass ^{235}U in s^{-1}/g
0	0.430	0.347	0.124	0.720	0.418	0.597
75	0.337	0.254	0.091	0.802	0.500	0.714
175	0.309	0.226	0.081	0.917	0.615	0.879
220	0.313	0.230	0.082	0.823	0.521	0.744
Average:			0.094			0.734
Error:			0.020			0.116
Relative error			21%			16%
Detection limit in mg ^{235}U:			290			71

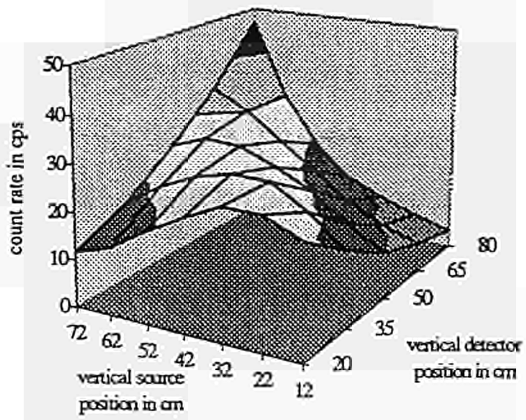

Figure 1: Count rate distribution for horizontal detector tubes as function of the axial source height and the detector position.

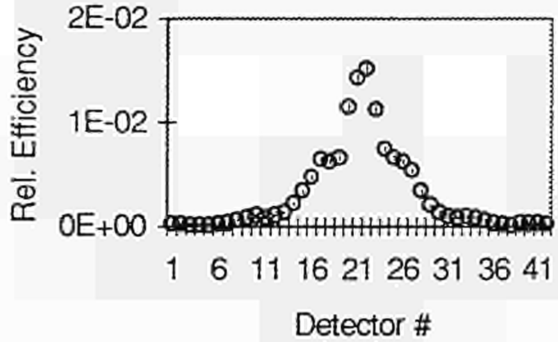

Figure 2: Simulated detector response distribution for a point source positioned off-centre (SCK/CEN system).

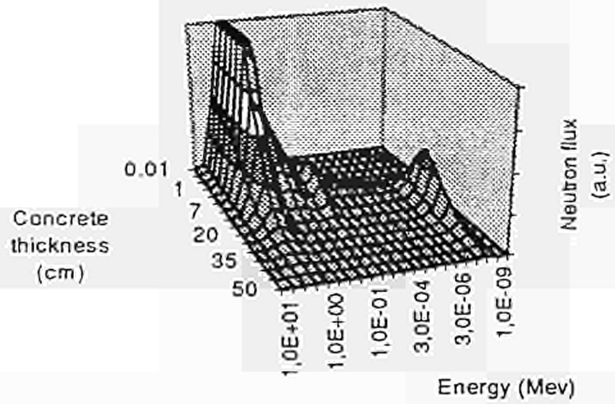

Figure 3: Neutron flux as function of layer thickness (data at the upper left corner cut).

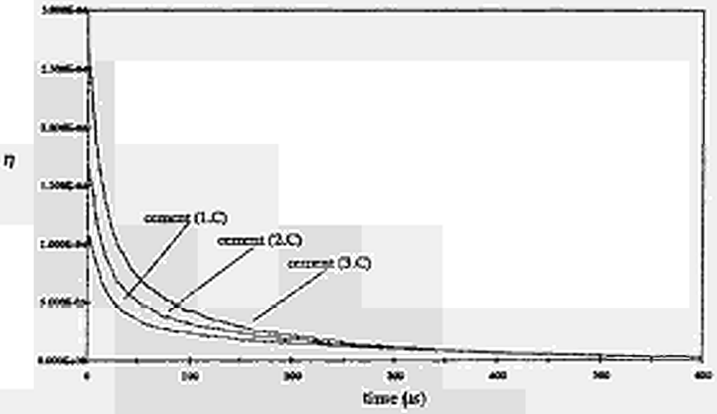

Figure 4: Delay distribution of detected neutrons for different source positions.

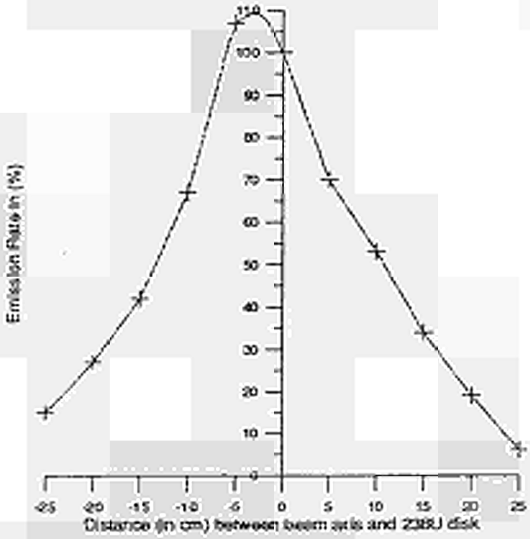

Figure 5: Spatial distribution of the Bremsstrahlung photons of the Mini-Linatron.

C.3.3-3 Evaluation and standardisation of fast analytical techniques for destructive radwaste control

Contract No:	FI4W-CT96-0019 **Duration:** 1 July 1996 - 30 June 1999
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A. OBJECTIVES AND SCOPE

Present destructive measurements of radioactive waste are based on traditional analytical methods including many and often rather complicated radiochemical and radioanalytical procedures. According to the increasing analytical tasks, routine controls on radioactive waste demand new fast methods which allow the processing of many samples per week with high accuracy. In order to minimise time, manpower and other costs, samples should be separated and measured in automated systems with as few steps as possible.

The research programme aims at investigating the application of fast wet chemical laboratory techniques as well as solid state mass spectrometry of untreated samples for routine analysis in comparative tests on standards and genuine radioactive waste (LLW) of various compositions. The project comprises evaluation of fast pretreatment techniques of samples and fast separation of radionuclides by High Performance Liquid Chromatography as well as the detection of radionuclides either in solutions or solids. On the basis of the results achieved in the project standardised or harmonised methods for destructive quality control on radioactive waste in the European Union will be suggested to the European Commission and documented in a manual (final report). Furthermore, as a result of the project, suggested reference standards are to be generated for later introduction and checking of destructive laboratory routines throughout the testing facilities of the European Network for Quality Assurance/Quality Control.

B. WORK PROGRAMME

B.1 Development of fast chemical laboratory routines

Preparation techniques for specimens to be investigated are fast dissolution methods of various kinds of radioactive waste and pre-chromatographic operations for removal of interfering inactive nuclides. The chromatographic investigations shall include (a) High Performance Ion Chromatography (HPIC) of radioactive cations or anions, (b) High Performance (Pressure) Reversed Phase Chromatography (HP-RPC) of radioactive nuclide complexes after precolumn derivitisaiton with an organic extractant and Low pressure Ion Extraction Chromatography (IEC) or Solid Phase Extraction (SPE). Radioactive nuclides and/or inactive carrier elements will be investigated. Examples of waste types are cemented waste or evaporator concentrates. (FZA, ENEA, Univ. Innsbruck).

B.2 Coupling of chromatographic methods and fast nuclide detection techniques

As sensitive methods for nuclide detection of chromatographic fractions Liquid Scintillation Counting (LSC) and High Resolution Inductively Coupled Plasma Mass Spectrometry (HR-ICP-MS) have been chosen. The feasibility of new scintillation cocktails with improved solubility in water for LSC measurements of nuclides in different chromatographic solutions as well as the application of HR-ICP-MS on various solutions has to be investigated in detail. The best combination of chromatographic separation methods and HR-ICP-MS for various analytical tasks will be investigated. (KFA, ENEA, Univ.Innsbruck).

B.3 Development of solid state mass spectrometrical techniques

Mass spectrometrical techniques for the analysis of special radwaste types are to be evaluated. In addition, the detection limits of solid state mass spectrometry for various nuclides with different half lives will be determined (KFA, Univ. Antwerpen).

B.4 Validation tests

Validation tests shall include analyses of either simulated waste solutions with given matrix composition comprising various amounts of relevant nuclides or real waste solutions (KFA, ENEA, Univ. Innsbruck, Univ. Antwerpen).

B.5 Use in routine mode and definition of fast laboratory routines

According to the results obtained from work under B.4, the techniques developed and validated will be adopted (on a routine basis) for a characterisation campaign on real radioactive waste. The experiences acquired in routine mode will enable the definition of a more reliable and complete analysis protocol to be adopted (All partners).

B.6 Comparison, validation and recommendations

Analytical results obtained from work under B1-B5 will be discussed, and the remaining problems will be defined. Fast laboratory techniques for routine analysis in European quality control will be suggested with respect to cost (equipment and manpower), capability to operate within defined detection limits (All partners).

C. Progress of work and results obtained

Summary of main issues

The results so far obtained show that HPIC can be applied to the separation of a wide range of radioactive anions or cations. Nevertheless due to problems in mixing aqueous buffers with organic or partially organic LSC cocktails the coupling of ion chromatographic methods with LSC is not always favorable. Often the more organic and ion free or ion poor solvents used in RP-HPLC offer better options. Therefore the evaluation of RP-HPLC separations will be extended. Currently the design of new complexing ligands for the different separation problems including actinides is intensively investigated. Research activities on IEC materials focus on highly stable column materials consisting of extractants which are chemically bound to polymers. A material for the solid phase extraction of lanthanides was already synthesized and tested.

The investigations on solid state mass spectrometry included electrically conducting as well as non conducting samples. GDMS of conducting samples was tested on Zircaloy and the applicability of different sample pretreatment techniques was studied. LA-ICP-MS was evaluated using concrete samples doped with some radionuclides. The results obtained so far are promising in view of low detection limits for long-lived radionuclides in concrete or a metallic matrix. Future research activities on GDMS will also include non-conducting (isolating) materials like glasses. Soil samples containing low concentrations of U and Th will be mixed with copper and will be analysed with GDMS. The experimental parameters for the analysis of radioactive concrete samples by LA-ICP-MS will be optimized.

C.1 Development of fast chemical laboratory routines

Research activities on HPIC (work package 1) in the first six months of the project included up-grading of instrumentations for low active samples as well as for the separation of medium and highly active samples. A chromatographic system for the separation of radioactive transition metals and Group II radionuclides as well as minor actinides was developed. The system consists of a polymeric cation exchange column and tartaric acid buffers as eluents. Colorizing agents are used for post column detection of uranium and thorium, transition metals as well as magnesium, calcium and strontium. The amount of radioactivity in the single elemental fractions is measured by off-line techniques. The influence of important chromatographic parameters on the separation was intensively studied. Preliminary results will be published on the conference RADWAP, 23-27 June, in Würzburg, Germany. Separation of Tc-99 as TcO_4^- from other anions was succesfully performed on a polymeric anion exchange column with mobile phases containing $Na_2CO_3/NaHCO_3$ and 4-cyanophenol.

The approach for work package 2 (covers simultaneously tasks of the work packages 1 (IEC), and 3 (sample pretreatment and nuclide/matrix separations). This approach consisted in the design and synthesis of efficient complexing ligands which may be used to form neutral, organic metal complexes for RPC. The thermodynamical and kinetic stability of the corresponding metal complexes as well as the selectivity of these ligands versus a large variety of metal ions may be investigated sufficiently by using these ligands in a polymer bound form in sample clean-up. Concerning IEC or SPE, a series of new polymer based stationary phases has been synthesized. Stationary phase-1 (INSP-1) consists of a high capacity, cis-1,4-dicarboxylate derivatized polymer which may be regarded as a polymer bound succinic acid (Figure 1). Due to the great chelating potential of the cis-1,4-dicarboxyl group the resins can be prepared in a wide range of capacities.

Using resins of the above type the separation of lanthanides at pH = 5.2-5.6 from other metal ions such as Al(III), Fe(III), Mg(II), Ca(II), Sr(II), Ba(II), Co(II) and Ni (II) employing masking and demasking techniques in order to ensure selectivity was succesful. So far the new resins may be considered as highly efficient and selective stationary phases for the quantitative extraction or preconcentration of lanthanides from radwaste. The use of further masking reagents and mixtures thereof, respectively, for enlarging the range of applicability is currently under investigation. Additionally the design of other new stationary phases especially those containing dipyridyl-functionalities is investigated intensively.

Furthermore sodium diethyl dithiocarbamate (NaDDTC) was selected and tested as a complexing ligand for the separation of radioactive metals by RP-HPLC. DDTC complexes of Ni, Co, Zn, Cr, Cu, Te, Hg, Mn, Se and Mo were synthesized in aqueous solutions and could be extracted by chloroform. After evaporation to dryness the extracted complexes were redissolved in methanol and injected into different chromatographic systems. RP-HPLC using stationary phases on the basis of C-18 modified silica and aqueous mixtures of methanol and acetonitril as eluents showed promising results. Weighable amounts of the complexes could easily be detected via typical absorptions in the ultraviolett range of the optical spectrum. Additionally an approach to form the DDTC complexes by pumping the aqueos sample through a NaDDTC loaded guard column was successful. The new complexation and enrichment technique offers the possibility of a fully automized chromatography without handling of open radioactive samples and chloroform. First results will be published at the analytical conference ANACON, 6-8 April 1997 in Konstanz, Germany.

C.2 <u>Coupling of chromatographic methods and fast nuclide detection techniques</u>

Concerning work package <u>4</u> (optimization of on-line/off-line LSC) beta-spectra of different radionuclides were measured by off-line LSC and have been collected in a manual. Using these spectra in the logarithmic energy scale, radioactive isotopes may be identified in separated single elemental fractions as well as not too complex mixtures of different radioactive elements.

The possibilities of on-line LSC were studied by means of a detector with a novel background reduction technique. Tests on efficiency and detection limits especially included Pb-210 and the radioactive transition metals Fe-55, Co-60, and Ni-63. For practical reasons and due to the rapid progress in work package <u>4</u>, investigations foreseen in work package <u>5</u> (coupling of liquid chromatography and LSC) for the second year of the project were already integrated in these works. Different possiblities of coupling on-line liquid scintillation counting and a HPIC system for the separation of radioactive transition metals were proven. In this connexion the applicability of LSC cocktails developed for aqueous or highly ionic aqueous mixtures was also evaluated. Figure 2 comprises the separation of Pb-210 from its daughter nuclide Bi-210, as well as from Ni-63 and Fe-55 using on-line LSC as the detection method. Currently on-line LSC is investigated in connexion with RP-HPLC.

Preliminary work on coupling of liquid chromatography and HR-ICP-MS (work package 6) included investigations on concentration and isotopic composition of uranium in a nitric acid leachate of reactor graphite by ICP-MS. In comparison with LSC or alpha spectrometry ICP-MS showed very low detection limits for long-lived actinides and Tc-99. In addition it is possible to determine Se-79 in liquid samples by ICP-MS. Further investigations will occur in the second year of the project as foreseen in the work programme.

C.3 Development of solid state mass spectrometrical techniques

For the first year of the project the evaluation of LA-HR-ICP-MS and HR-GDMS of solid radioactive samples is foreseen in work package 7.

The results so far obtained on GDMS with conducting samples show that measurement of trace elements using the VG9000 glow discharge mass spectrometer is possible. The technique combines the necessary sensitivity with a high mass resolution to obtain sub-ppm concentrations. Conductors such as metals, as well as non-conductors can be analysed using this technique.

Previous investigations on pretreatment techniques for GDMS of conducting samples focused on zircalloy presumably cut using spark erosion with a brass wire. Several samples have been prepared in three different ways to check for possible surface contamination and to evaluate the efficiency of the cleaning process.

Summarizing it seems that spark erosion of the zircalloy samples causes relatively high surface enrichment of Cu, and Zn, as well as significant surface enrichment of B, Ba, and Mg. The spark erosion process therefor not only removes material from the sample but also transport of material from wire to sample occurs. This exchange process can result in more than a superficial contamination of the sample and will be difficult to remove. Further testing and analyses will be performed to characterise this contamination in order to find an appropriate cleaning process or to select a 'cleaner' way for cutting the samples. No particular difficulty is experienced for the analysis of the radionuclides ^{232}Th, ^{235}U and ^{238}U which were present in the samples at a level of circa 20 ppb.wt (Th) and ca 450 ppb.wt (U). Other trace elements can directly be determined at levels from 20 ppb.wt to 6000 ppm.wt.

Ongoing activities on GDMS of conducting samples include depth profiling on the zirconium samples performed by secondary ion mass spectrometry. Furthermore a mapping of a cross section through the sample will be made using a scanning electron microscope and if technically possible SIMS in order to get a good view on the effects of cutting the sample using spark erosion.

Investigations on non conducting samples focused on LA-ICP-MS. Experiments were performed using a Nd:YAG laser equipped laser ablation system developed for the analysis of ceramic material. The laser ablation chamber is coupled either with a quadrupole-based or with a double-focussing sector field ICP-mass spectrometer. Using simulated samples doped with Tc-99, U-233 and Np-237 an analytical method for the determination of long-lived radionuclides in cemented radioactive waste was developed and tested. Figure 1 comprises mass spectra of a concrete standard doped with actinides and a concrete blank sample. Preliminary results show that the detection limits should be lower if the double focusing ICP-mass spectrometer is used.

Results obtained in this project were published in a report of the Research Centre Jülich as well as in lectures and posters on the 3. Symposium Massenspektrometrische Verfahren der Elementspurenanalyse, 1996 in Jülich, Germany.

Figure 1: Backbone of INSP-1 used for the solid phase extraction of lanthanides

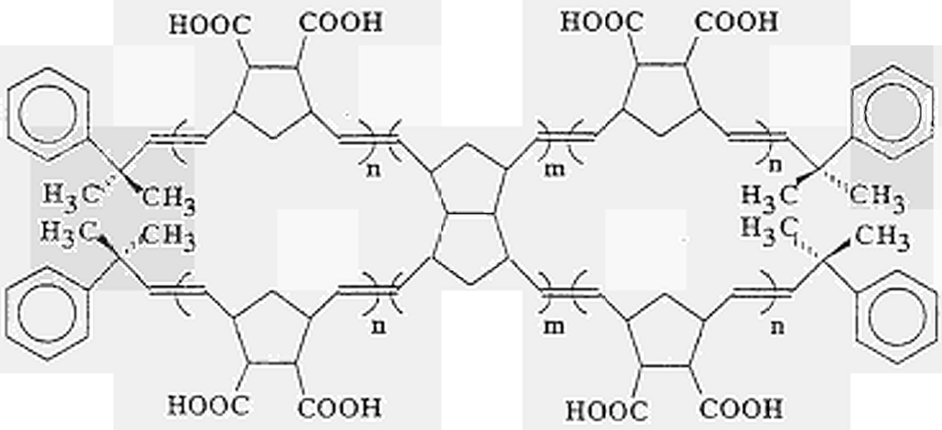


Figure 2: HPIC separation of some radionuclides using peak detection by on-line LSC

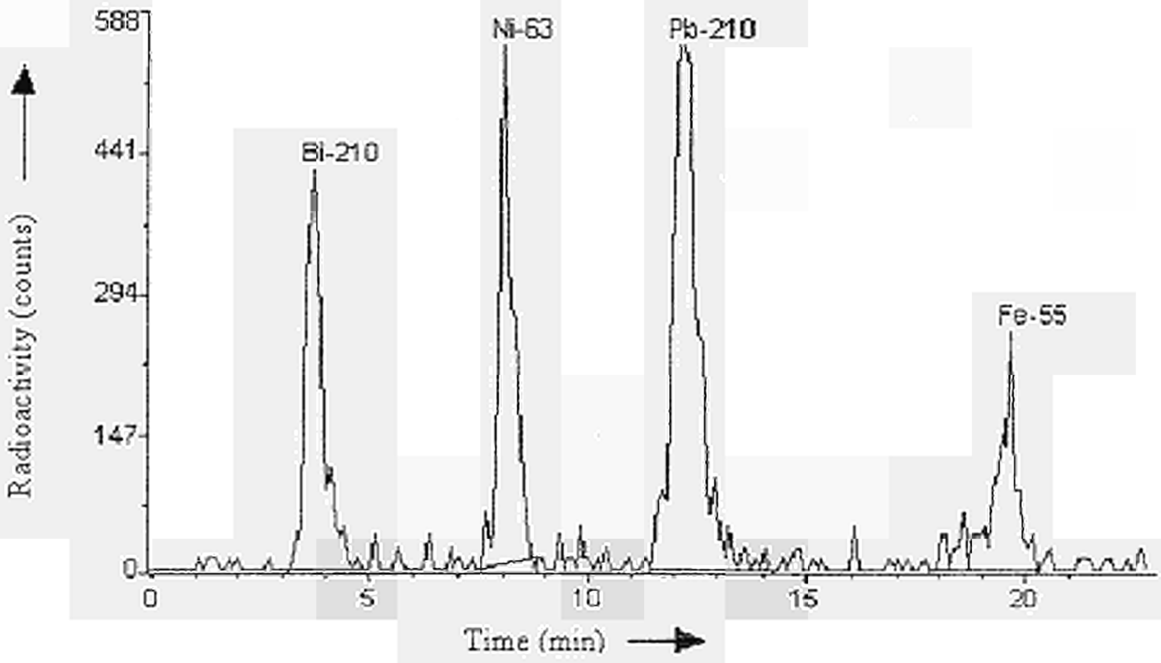
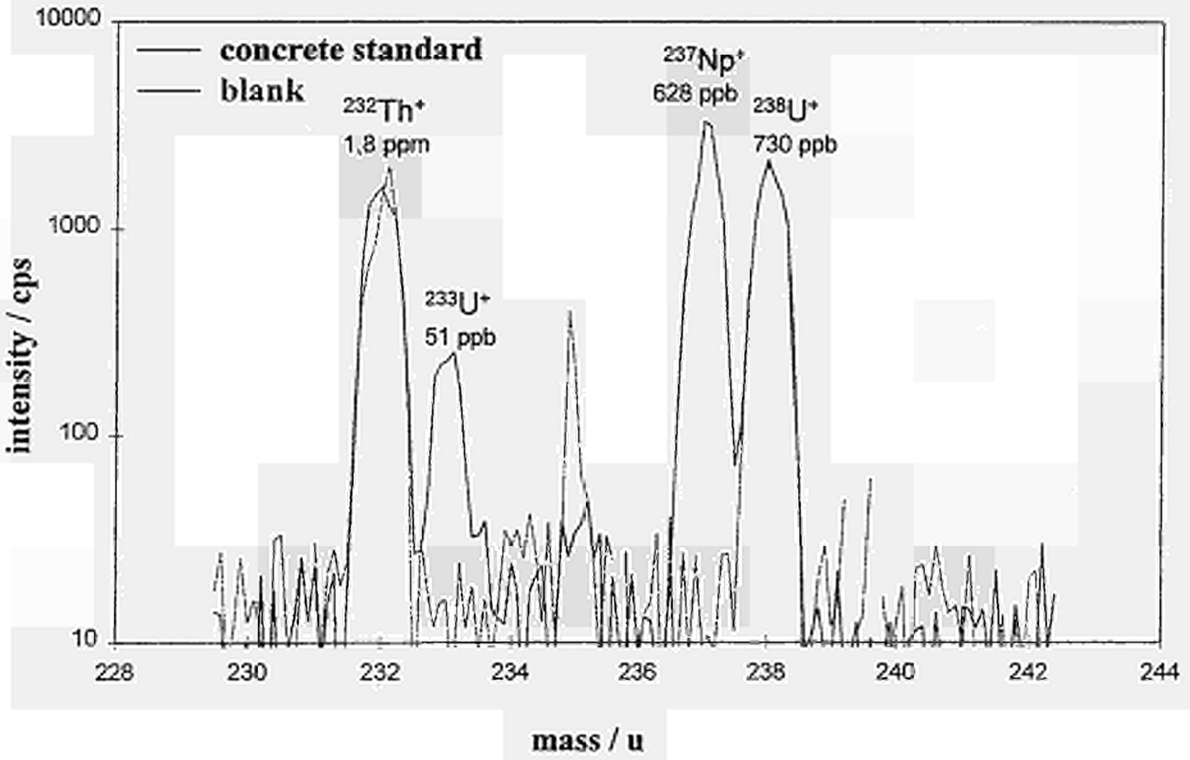


Figure 3: Mass spectra of a concrete laboratory standard and a concrete blank sample obtained by LA-ICP-MS



C.3.3-4 Characterisation of accessible surface area of HLW glass monoliths by high energy accelerator tomography and comparison with conventional techniques

Contract No: FI4W-CT96-0023	**Duration:** 1 May 1996 - 30 April 1999
Coordinator: **M. SENÉ, AEA Technology plc, Harwell/GB**	
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Partners: **BAM Berlin/DE, CEA Valrhô/FR**	

A. OBJECTIVES AND SCOPE

The total accessible surface area of HLW glass blocks is an important parameter in all safety calculations for a final repository because the leaching ratio of radioactive nuclides is proportional to the total accessible surface of the waste-loaded glass blocks. Similarly the extent of phase separation within glass blocks is crucial in determining their long-term behaviour in the repository environment. Reliable characterisation of these aspects of HLW (and also MLW) glass blocks is clearly important.

The primary objective of this proposal is to quantitatively characterise the accessible surface area and the extent of phase separation in a number of actual inactive glass monoliths produced by CEA. Other objectives are the i) demonstration of a practical, non-destructive and quantitative technique for characterising these parameters of HLW and MLW glass blocks and ii) comparison of this non-destructive technique with existing characterisation techniques.

B. WORK PROGRAMME

B.1 Development of beam hardening model and procedure for quantitative computer tomography (CT) measurements

The intensity required for optimum resolution in the CT images will be established by using an electron linac source. Work will be carried out to provide stable and reliable operation at high intensity. The beam hardening effects on the resulting CT images will be minimised by use of Cerenkov detectors. Corrections for beam hardening effects will be included, if necessary, by theoretical modelling of the CT scanner system (AEA Techn., BAM).

B.2 CT measurements on samples and development of image processing system

Tomographic measurements will be made of glass blocks supplied by CEA using the procedure defined in B.1. Comparative measurements will also be made using ^{60}Co as the g-ray source for the CT scanner. The beam hardening model will be integrated into the image processing system and used to evaluate the total accessible surface area. During this period CEA will supply a second block which will also be subjected to the same measurements (AEA Techn., BAM, CEA).

B.3 Leach tests on samples and analysis of CT data

The CT data on the glass blocks will be processed to give quantitative information on the properties of the blocks. At the same time the glass blocks will be returned to CEA who will carry out soxhlet leach tests for comparison with the results from the non-destructive test method (AEA Techn., BAM, CEA).

B.4 Assessment and comparison of characterisation techniques

An assessment will be made of the results of the non-destructive and destructive analyses of the glass blocks. The limitations and range of applicability of the non-destructive and destructive techniques will be considered and their implications for the establishment of QA and QC procedures will be assessed (AEA Techn., BAM, CEA).

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The project officially started on 1st May 1996. However, due to some administrative delays and previous commitments, it was not possible to start work on this project until July 1996. Consequently completion of the first work package (B.1.) is estimated to be delayed by 1 month but the overall timescale of the project is not affected.

The project commenced with a meeting at BAM in July between AEA Technology and BAM. Following this meeting and during the period of this report work has concentrated on the following three aspects of work package B.1.

i) The bremsstrahlung γ-ray intensity required for optimum resolution in the CT images is being established using the electron linac source. Work has been carried out to provide stable and reliable linac operation at high intensity. Further work is planned to complete this over the next few months.

ii) A model is being developed to a) correct, if necessary, for beam hardening effects in resulting CT images, b) detect the presence of cracks in the resulting images and c) determine the total accessible surface area from the resulting processed images by a statistical technique. Work on all areas (a, b & c) of the model has started.

iii) The use in this model of CT images that have obtained with Cerenkov detectors (which minimize beam hardening effects) is being compared with the use of CT images obtained using conventional scintillation detectors.

As a result of this work, at the end of work package B.1. one of three possible procedures will have been identified for making quantitative measurements of glass blocks.

i) Use of standard scintillation detectors in CT scanner. Correction of the resulting images using theoretical model for beam hardening. Computer processing of the final image.

ii) Use of scintillation and Cerenkov detectors in the same detector array to provide optimum resolution and information on beam hardening. Correction of resultant images using theoretical model for beam hardening together with extra information obtained from two detector systems. Computer processing of the final image

iii) Use of Cerenkov detector systems to minimize beam hardening effects. Computer processing of the final image.

Over the next 6 months the first test glass block will be fabricated by CEA. This will be an inactive sample of actual HLW glass blocks produced at the Marcoule plant. The detailed specification of these glass blocks is at present under discussion. A meeting between AEA, BAM and CEA at Marcoule is planned in February 1997 to finalise this.

C.1. Development of beam hardening model and procedure for quantitative computer tomography (CT) measurements

LINAC During the period of this report a number of significant improvements have been made to the BAM LINAC. As a result of this work dose rates of about 55-60 Gy/min have been sustained for periods of more than 24 hours. An example of this performance is shown in Figure 1. In addition to the sustained output, the number of arcs in the linac, which cause missing output pulses, and hence missed projections in tomograms, have been reduced almost to zero. In addition to the modifications already carried out a new control system has been ordered which will detect LINAC trips and restart the LINAC automatically up to ~10 times. This is useful if long measurements are to be made.

MODELLING Work has started on all three components of the image processing model. With regard to the beam hardening calculations during the period of this report work has concentrated on calculating the bremsstrahlung γ-ray spectrum from the linac and the

γ-ray responses for the Cerenkov and scintillation detectors. This has been carried out using the Monte Carlo based electron/γ-ray transport code EGS4 (Ref. [1]). Figures 2-4 show the examples of these calculations. Figure 2 shows the bremsstrahlung γ-ray energy spectrum for incident electron energies of 10 and 12 MeV. The differences in the spectra are significant, hence it is important to be sure of the electron energy from the linac. Figures 3 & 4 show the response of the scintillation and Cerenkov detectors to mono-energetic γ-rays.

Once an image, corrected for beam hardening, has been generated the next step in the process of determining the accessible surface area is to use image processing techniques to determine the presence and magnitude of cracks and voids. The measurement of cracks is a particularly challenging problem. Various approaches to this problem have been described in the literature and have been reviewed. At present the most promising approach is being developed in collaboration between BAM and 'Bayerisches Forschungszentrum für wissens-basierte Systeme' at Passau, Bavaria. They have proposed a method based for the extraction of crack structures from grey level images (Ref. [2]). Work on evaluating this technique is still continuing.

The images generated using the CT scanner only represent a sample of the whole volume of the glass block. To provide an accurate estimate of the accessible surface area for the whole block from this data statistical techniques will be employed (Ref. [3]). The applicability of this approach was tested in a previous contract. Unfortunately the poor resolution of the images and artefacts (particularly due to beam hardening) made reliable crack detection impossible. The improved tomographic images that this project aims to obtain will enable this model to be fully validated.

CT IMAGES Following the improvements to the LINAC described above work has started on obtaining images of existing test glass samples with both scintillator and Cerenkov detectors to enable a detailed comparison to be made of the validity of the model for the two types of detector.

GLASS FABRICATION The first of the two full scale R7T7 inactive reference glass block samples to be supplied by CEA will be fabricated during the next 6 months. At present discussions are under way between AEA, BAM and CEA concerning the details of the steel canister in which the glass will be poured. In addition to there is also discussion over transport to, and particularly, from BAM as there is some evidence from previous work that the transportation of the glass block can itself cause damage and hence invalidate the comparison between the CT and leach testing results.

References

[1] NELSON, W.R., HIRAYAMA, H. and ROGERS, D.W.O. Stanford Linac Centre Report SLAC 265, UC-32 (1985).

[2] DONNER, K. Proc. Applications of Artificial Intelligence (AI 1991)213.

[3] GOEBBELS, J., ILLERHAUS, B. and REIMERS, P. Proc. 5th Int. Conf. on Radioactive Waste Management and Environment Remediation, Berlin September 3-7 1995. (ASME 1995)

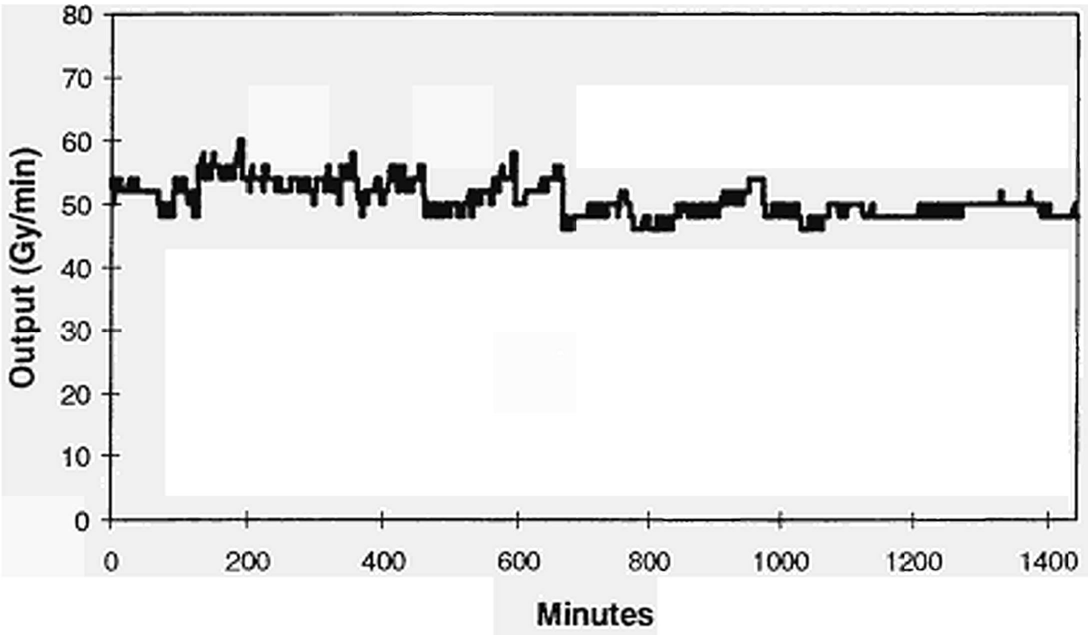

Figure 1: LINAC output over a period of 24 hours, as monitored by reference detector.

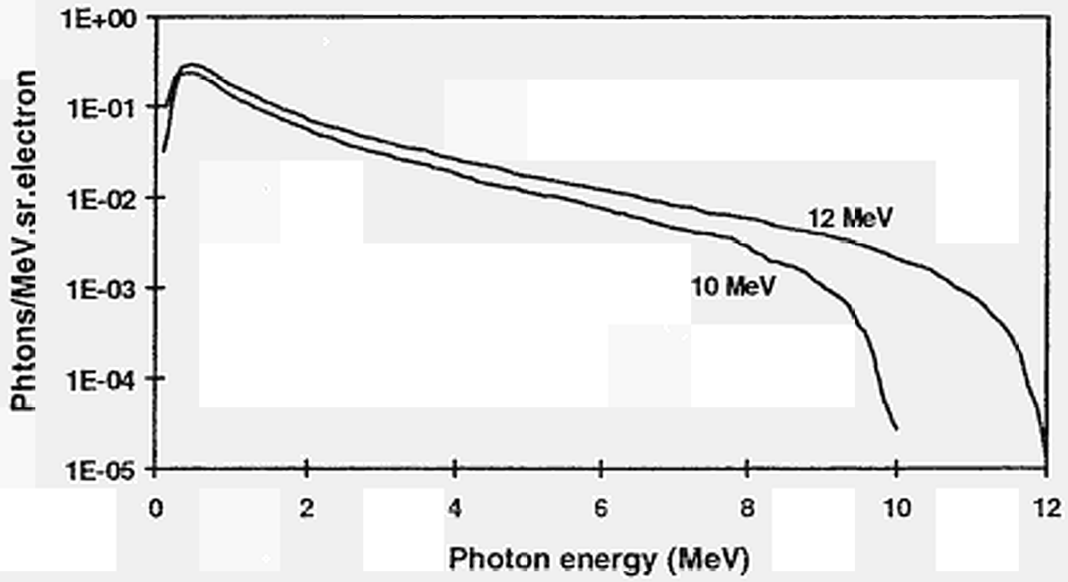

Figure 2: Bremsstrahlung γ-ray spectra for 10 and 12MeV electrons for the BAM linac.

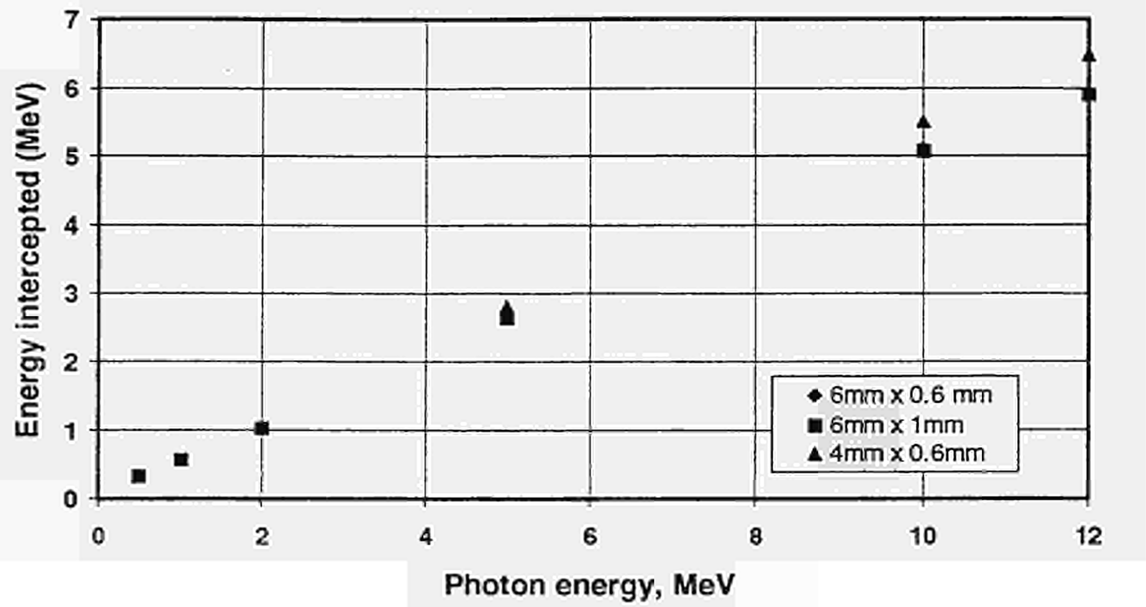

Figure 3: Response of scintillation detector (total energy deposited in detector) as a function of γ-ray energy for different collimator openings.

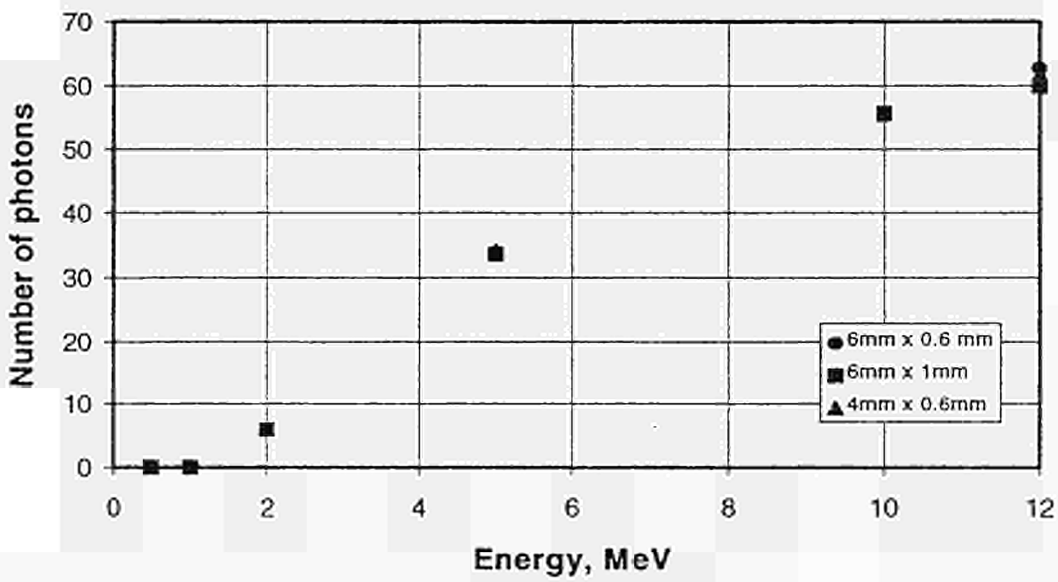

Figure 4: Response of Cerenkov detector (number of Cerenkov photons per incident γ-ray) as a function of γ-ray energy for different collimator openings.

C.3.4-1 CATSIUS-CLAY : Calculation and testing of behaviour of unsaturated clay

Contract No: FI4W-CT95-0003 **Duration:** 1 Jan. 1996 - 31 Dec. 1998
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Tel. +34/3.401.68.66 Fax: +34/3.401.65.04 e-mail: alonso@etseccpb.upc.es
Partners: ANDRA Chatenay-Malabry/FR, Clay Technology Lund/SE, ISMES Bergamo/IT, SCK/CEN Mol/BE, Univ. UPC Barcelona/ES, Univ. Liège/BE, Univ. UWCC Cardiff/GB

A. OBJECTIVES AND SCOPE

Compacted expansive clays are adopted as engineered barriers around waste canisters in reference concepts for geological repositories. After emplacement in saturated formations these barriers experience a transient wetting and swelling phase governed by the rate of absorption of natural water and a transient temperature regime controlled by the decaying heat power input induced by the canister.

In recent years a number of research groups in several European countries have developed models for the behaviour of unsaturated expansive materials. From the experimental point of view the results of fairly comprehensive suction controlled tests on expansive clays are available. In addition full scale hydration experiments are in progress. Therefore a benchmark exercise is being implemented with the objectives :

- to assess the accuracy and reliability of the numerical predictions.
- to establish the useability and capability of codes to model the thermohydromechanical behaviour of unsaturated clay barriers.
- to provide an evaluation of existing laboratory testing methods in view of the data and parameter requirements of THM models.

B. WORK PROGRAMME

The work programme consists of three stages which will be performed by all partners.

B1. Verification exercises

Its main purpose is to check that the codes involved are correctly programmed to solve the field equations. Two cases are treated :
- Two-phase infiltration in a finite column of unsaturated soil.
- Thermal convection in saturated low porosity rigid medium.

B2. Validation exercises at laboratory scale

Two laboratory tests will be modelled :
- Oedometer suction controlled tests on samples of compacted Boom clay pellets.
- Small scale wetting-heating test of compacted bentonite.

B3. Validation exercises at a large "in-situ" scale

Two large hydration tests will be modelled :
- The BACCHUS 2 in-situ hydration experiment on Boom clay pallets performed in the HADES underground laboratory at Mol.
- The FEBEX heating and wetting experiment on compacted bentonite.

C. Progress of work and results obtained

Summary of main issues

In accordance with the Project objectives and schedule, work during its first year (1996) included (s. Table I for acronyms used to designate each partner):

STAGE 1. Work Package 1. (Benchmark 1.1: Infiltration in a Finite Column of Unsaturated Rigid Porous Medium and Benchmark 1.2: Thermal Convection in a Saturated Rigid Porous Medium). 1st Project Meeting held in Barcelona (Spain) the 31st of May of 1996. The results of stage 1 have been reported in a 1st Topical Report which will eventually become a report within the EUR series.

BM 1.1 was solved correctly by all partners. BM 1.2 was divided in two parts (Part A: low porosity and Part B: high porosity). Partners solved the BM using different strategies and assumptions (SCK, due to problems with their code, will not provide predictions for this BM). For part A, predictions were satisfactory. For part B, predictions were discrepant, specially when pressure calculations are compared. These difficulties have been traced to simplifying assumptions used by some of the codes used.

STAGE 2. Work Package 2. (Benchmark 2.1 part A: Oedometer Suction Controlled Tests on Samples of Compacted Boom Clay). 2nd Project Meeting held in Córdoba (Spain) the 19th of November of 1996.

For BM 2.1 part A, predictions by several partners (AND, UPC, UOL, UWC) approached experimental results with an acceptable accuracy. SCK, due to problems with their code, will not provide predictions for this BM. BM 2.1 part B: Blind Suction Controlled Swelling Pressure Test on a Sample of Compacted Boom Clay will be reported in 1997.

C1. Verification exercises

a) Benchmark 1.1: Infiltration in a Finite Column of Unsaturated Rigid Porous Medium. The onedimensional flow of a fluid inside a rigid homogeneus isotropic unsaturated medium due to a prescribed flow was considered. Two situations were envisaged: i) vertical flow and ii) horizontal flow. Assuming a constant permeability and a linear variation of degree of saturation with suction, an analytical solution was found. It was subsequently used as the reference solution. Partners were asked to submit predictions using 1D, 2D and 3D elements and meshes of varying refinement. Discrepancies were found to be small in all cases. As an illustration, Figure 1 shows the distribution of pressures along the soil column for one of the cases specified (horizontal flow, 1D refined mesh, $t = 10^7$ seconds). Table II shows the code used, method of solution and cases actually solved by each partner.

b) Benchmark 1.2: Thermal Convection in a Saturated Rigid Porous Medium. The case refers to a rigid homogeneous isotropic and saturated porous medium of infinite extent with a uniform spherical source of heat. The benchmark requires the calculation of temperature and pressure distributions and evolutions inside and outside the spherical source of heat. Various approaches were followed by partners to simplify the problem proposed in this BM: some (AND, UOW) simplified the problem adopting Hodgkinson's approach [1], others (ISM, UOL) simplified the problem only uncoupling the heat balance from the fluid flow and others (CLA, UPC) used no simplifications at all. Table III shows the code used, method of solution and cases actually solved by each partner.

Part A: Low porosity. According to Hodgkinson [1], if the porosity of the medium is low ($n = 10^{-4}$ was specified in the BM) and the permeability of the medium is low, some hypothesis can be made to simplify the problem. The hypotheses used by Hodgkinson essentially neglect the heat transfer by convection and the thermal expansion of the fluid and lead, in fact, to uncouple the heat balance from the fluid motion, being buoyancy the driving force of the fluid. Temperature predictions among partners reproduced Hodgkinson's theoretical values (identified as CIM in the comparison figures) with small relative discrepancies (Figure 2). Pressure predictions among partners also reproduced Hodgkinson's theoretical values (except ISM). Differences among partners may be attributed to: i) the effect of the simplifications made to solve the problem (thermal expansion of the fluid, some convective terms) and ii) numerical issues (mesh used, time integration step).

Part B: High porosity. If the porosity of the medium is high ($n = 10^{-1}$ was specified in the BM), Hodgkinson's hypothesis can no longer be accepted. No analytical solution is available in this case. Temperature predictions among the partners show similar trends. Regardless of uncoupling or not the heat balance from fluid motion, pressure predictions showed two different trends: i) predictions by partners using Hodgkinson's approach (AND, UOW) and ii) predictions by partners not using this approach (CLA, ISM, UPC, UOL) with the exception of CLA. Considering the center of the spherical source of heat, Hodgkinson's approach predicts a zero pressure increase, whereas the flow of the fluid due only to thermal expansion produces an important pressure increase, as a simplified computation shows. These two mechanisms agree with the two trends observed in the predictions provided by partners, and would explain the differences found among them. Therefore, differences among partners may again be attributed to: i) the effect of the simplifications made to solve the problem and ii) numerical issues. Figure 3, which shows the pressure distribution for t = 50 years, provides an illustration of preceeding comments.

Summarizing, we may consider two main (simplified) limiting cases: i) fluid only driven by buoyancy (Hodgkinson's approach) and ii) fluid only driven by thermal expansion. Part A would then be close to the first limiting case, whereas part B would be close to the second limiting case. In both part A and part B, the uncoupling of the heat balance from the fluid motion would not be important.

C2. Validation exercises

Benchmark 2.1 part A: Oedometer Suction Controlled Tests on Samples of Compacted Boom Clay. A series of oedometer tests using ideal samples were performed (reported in [2]) to gain insight into the behaviour of unsaturated expansive clays. The soil samples were prepared using highly compacted Boom clay which was crushed and sieved to get isolate aggregates of an average size of 2 mm (pellets). These pellets were compacted to get 5 samples with an initial dry density of 14 kN/m^3. Each sample was tested at a constant vertical stress by varying the suction. Using the experimental curves suction vs. vertical strain for the 5 samples, partners were asked to calibrate their models. The comparison of the experimental results against data provided by some partners showed good coincidence (AND, UPC, UWC) and by some others only a coincidence in the main trends was observed (UOL). CLA provided some partial results, while ISM delayed their results to a future time. Figure 4 shows a comparison between predictions by partners and experimental results for a net vertical stress of 392.4 kPa.

References

[1] HODGKINSON, D P, Annals of Nuclear Energy Vol 7, pp. 313-334 (1979)

[2] ALONSO, E E, LLORET, A, GENS, A, YANG, D Q, 1st International Conference on Unsaturated Soils UNSAT'95, Paris, 6-8 September 1995. Proceedings pp. 11-16.

Table I: Acronyms used in CATSIUS CLAY to designate coordinator and partners

CIM	CIMNE
AND	ANDRA
CLA	Clay Technology
ISM	ISMES
SCK	SCK-CEN
UPC	Technical University of Catalonia
UOL	University of Liège
UWC	University of Wales-Cardiff

Table II: Summary for Benchmark 1.1

	AND	CLA	ISM	SCK	UPC	UOL	UWC
Code used	CLEO	ABAQUS	ABAQUS	PORFLOW	C BRIGHT	LAGAMINE	COMPASS
method of solution	FEM	FEM	FEM	NPI	FEM	FEM	FEM
Dimensions used in the analysis							
1D	•			•	•	•	
2D	•	•	•	•	•	•	•
3D		•	•	•	•	•	

Table III: Summary for Benchmark 1.2

	AND		CLA		ISM		SCK	UPC		UOL		UWC	
Code used	CLEO		ABAQUS		ABAQUS			C BRIGHT		LAGAMINE		COMPASS	
method of solution	FEM		FEM		FEM			FEM		FEM		FEM	
Coupling energy balance - fluid motion (CASE)	A	B	A	B	A	B		A	B	A	B	A	B
energy balance ⇔ fluid motion			•	•				•	•				
energy balance ⇒ fluid motion	•	•		•	•	•			•	•	•	•	•
Type of equations solved (CASE)	A	B	A	B	A	B		A	B	A	B	A	B
general equations			•	•				•	•				
simplified equations	•	•			•	•				•	•	•	•
Domain where the solution is sought (CASE)	A	B	A	B	A	B		A	B	A	B	A	B
half plane $(0,+\infty)\times(-\infty,+\infty)$			•	•	•	•		•	•	•	•	•	•
quarter of plane $(0,+\infty)\times(0,+\infty)$	•	•											

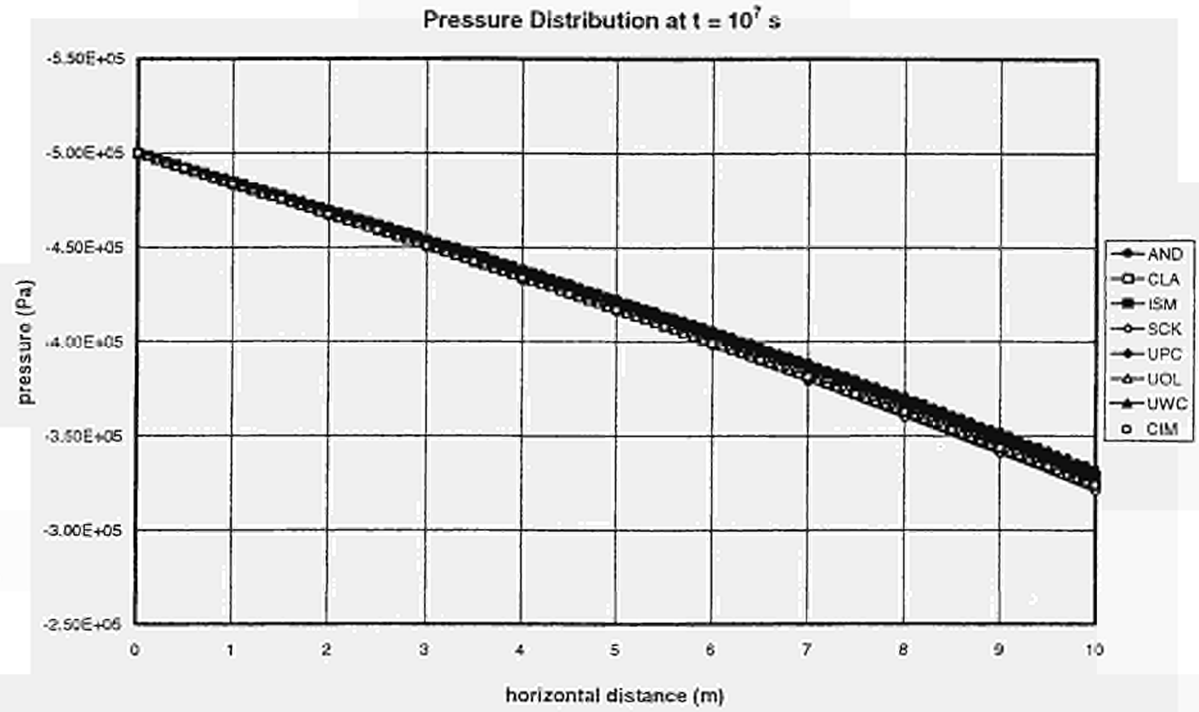

Figure 1: Benchmark 1.1: Infiltration in a Finite Column of Unsaturated Rigid Porous Medium. Case D: Horizontal Flow using 1D Refined Mesh.

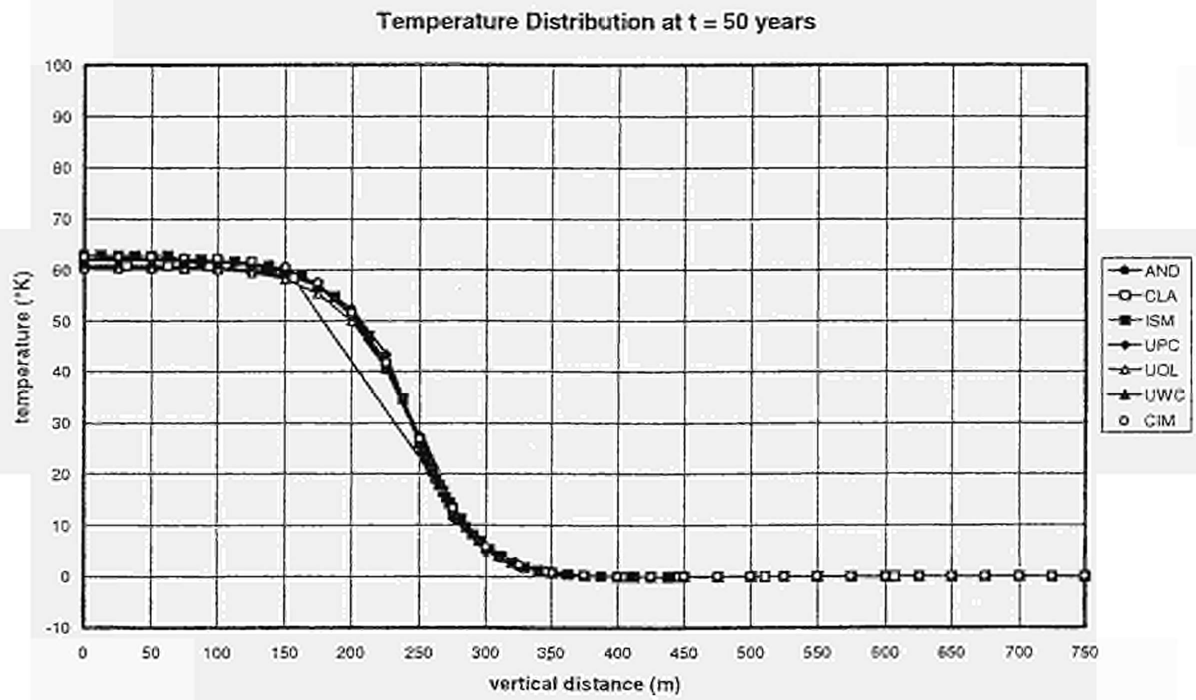

Figure 2: Benchmark 1.2: Thermal Convection in a Saturated Rigid Porous Medium. Case A: Low Porosity.

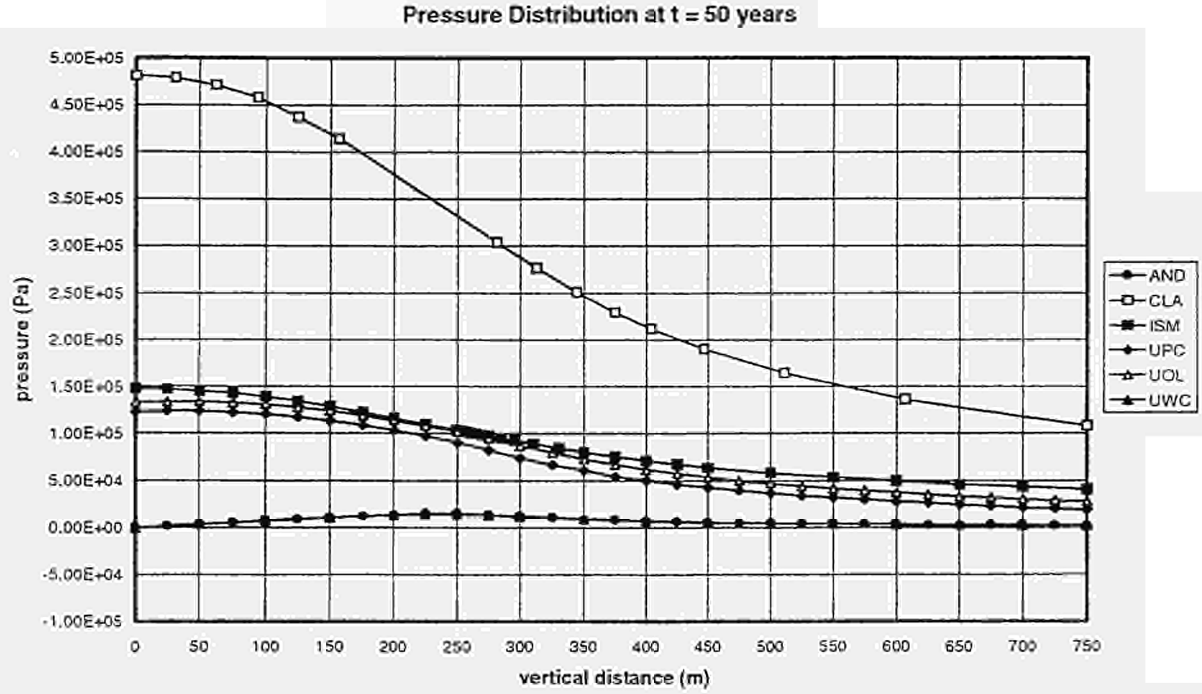

Figure 3: Benchmark 1.2: Thermal Convection in a Saturated Rigid Porous Medium. Case B: High Porosity.

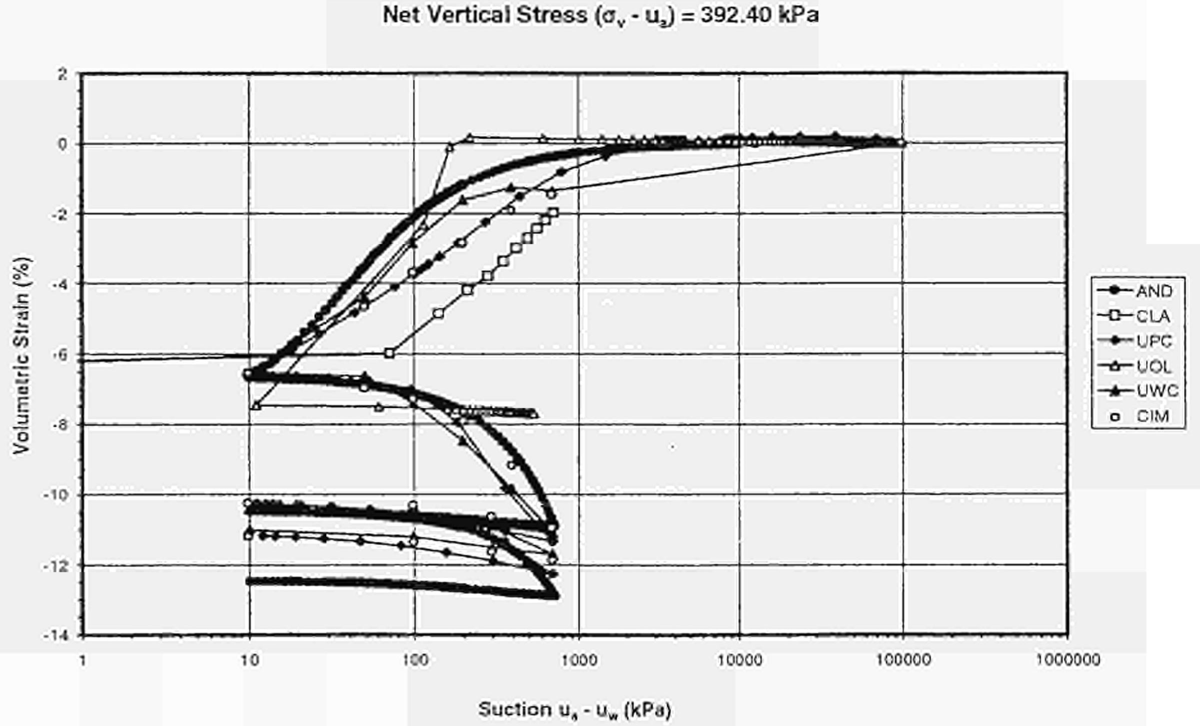

Figure 4: Benchmark 2.1 part A: Oedometer Suction Controlled Tests on Samples of Compacted Boom Clay.

C.3.4-2 Microstructural and chemical parameters of bentonite determinants of waste isolation efficiency

Contract No: FI4W-CT95-0012 **Duration:** 1 Jan 1996 - 28 Feb 1999
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Partners: VTT Espoo/FI, Univ. Hannover/DE, Univ. KTH Stockholm/SE

A. OBJECTIVES AND SCOPE

Engineered clay barriers are being proposed in the conceptual design of radioactive waste repositories since most clay types (especially smectites) have a very low hydraulic conductivity and exhibit excellent retardation capabilities for cationic radionuclides. The project aims at improving the understanding of sorption and diffusion mechanisms in smectite clays by working out a complete microstructural model and correlate it with the sorption and diffusion of cations and anions for ordinary commercial bentonites and for organophilic clays.
The work will comprise both theoretical and experimental studies and the models developed for predicting radionuclide retention and transport through saturated clay barriers will contribute to the evaluation of the performance of clay barriers.

B. WORK PROGRAMME

B1. Development of general microstructural model for bentonite (CLAY TECH.)

- Numerical modelling
- Laboratory experiments

B2. Model for diffusion and pore water chemistry (VTT)

- Adoption to microstructural model
- Experimental studies on pore water chemistry

B3. Organo-bentonite (Univ. Hannover)

- Preparation of organophilic bentonite (smectite)
- Investigations of surface chemistry of organo-bentonites
- Sorption/desorption experiments
- Determination of radionuclide diffusivities in organophilic bentonite
- Incorporation of data into the diffusion model
- Stability testing of the organo-bentonite

B4. Cationic and anionic diffusion mechanisms in bentonite/organo-bentonite (KTH)

- Input to the diffusion transport and surface chemistry models
- Experiments

C. Progress of work and results obtained

Summary of main issues

Experimental work within all four packages have contributed to yield a solid basis for developing a microstructural model of bentonite that takes density and porewater chemistry into consideration. The relevance of initially proposed clay embedding methods for electron microscopy has been verified and the technique is now available for preparing samples that have been used in laboratory experiments performed within the project. Numerical modeling for determining microstructural changes by simulated compression has been started in 2D and yielded reasonable results.

Numerical modeling of the microstructure for simulating ion migration is under way using a probabilistic model of ions occupying a certain specific volume in the electrolyte, paying special attention to the Stern layer. Experimental studies on the porewater chemistry have been started.

Experiments have been made with bentonite treated with alkylammonium ions for determining the anion and cation sorption capacities. Evaluation has been obtained through XRD and surface charge measurements. A major outcome of the study is that the sorption capability of bentonite for iodide ions can be substantially improved.

The mode of diffusion of Sr^{2+}, Cs^{+} and I^{-} has been investigated in experiments with compacted bentonite and the apparent diffusivity and transport K_d have been obtained by computer simulation of the experimentally measured flux and concentration profiles in the clay. For Cs^{+} and Sr^{2+} the ionic strength dependence of the distribution coefficients clearly points at ion exchange as the main sorption mechanism.

C1. Development of general microstructural model for bentonite (CT)

- Numerical modeling

A suitable grain size distribution for numerical modeling has been derived and a geometrical 2D model specified. This model has been applied for calculation of compression-induced strain using finite and boundary element methods. Adequate strain parameters have been derived and numerical computation made of the deformed microstructural arrangement resulting from compression under 10, 50 and 100 MPa.

- Laboratory experiments

MX-80 powder has been compressed to a few representative densities and successfully percolated by a non-polar liquid to check the pore geometry of non-hydrated clay.

Preparation of clay by use of methyl/butyl methacrylate for subsequent microstructural analysis using XRD and electron microscopy turns out to give at least approximately the same microstructural pattern as assumed for hydrated clay (Figure 1). This preparation technique will therefore be used throughout the study.

A technique for visual simulation through automatic picture analysis of electron micrographs will be used in the microstructural modeling.

C.2 Model for diffusion and porewater chemistry (VTT)

- Adaption to microstructural model

The pore geometry of clay specimens has been modelled as a bundle of non-interconnected, tortuous negatively charged capillaries. For these pores, the equations governing the local chemical equilibrium have been numerically integrated on a grid which dynamically adjusts according to the prevailing physico-chemical conditions. The discretization of the governing equations and the coding of the first version of the space charge model are practically finished.

- Experimental studies on porewater chemistry

An apparatus for porewater extraction by squeezing has been developed and experiments are under way (Figure 2). In a first series the time for chemical equilibrium in bentonite/water systems has been investigated and it has been demonstrated that considerable time is required (Figure 3).

C.3 Organo-bentonites (ZSR)

- Preparation of organophilic bentonite (smectite)

A suitable technique for treating bentonites with alkylammonium ions has been developed, tested and applied. Three parameter variations were employed: a) alkylammonium ion incorporation into bentonite charged with different exchangeable cations (eg. Na^{+}, Ca^{2+}, Mg^{2+}), b) sorption effects after incorporation of alkylammonium ions differing in organic carbon chain length and aliphatic or aromatic structure, and c) sorption effects after different levels of alkylammonium precharging. The investigations were accompanied by determinations of the basal spacings from XRD-measurements and by evaluation of the surface charge of the original and of differently saturated organo-mineral materials.

- Investigations of surface chemistry of organo-bentonites

The particle surface charge of the original and different $HDPy^{+}$-treated MX-80-bentonites have been determined using polyelectrolyte-titration. The results of the measurements at different pH of the dispersion (adjusted with HCL or NaOH) are shown in Figure 4. They indicate that the mass specific charge of the bentonite shifts from negative to positive values as a consequence of the $HDPy^{+}$ treatment. It is remarkable that the symbol of the charge start to change when $HDPy^{+}$ is applied in amount of about 70% CEC (55% exchange of interlayer cations), considering the initiation of iodide sorption at this level of $HDPy^{+}$ saturation. The pH-effect on surface charge appears not to be substantial.

- Sorption/desportion experiments, determination of radionuclide diffusivity, testing of diffusion model and stability testing of organo-bentonites are being planned.

C.4 Cationic and anionic diffusion mechanisms in bentonite/organobentonite (KTH)

Input to the diffusion transport and surface chemistry models

Diffusion experiments have been interpreted by applying a diffusion model that considers filter porosity and thickness, referring to the break-through curve. The ionic strength dependence of the distribution coefficients as evaluated by applying computer simulation clearly points at ion exchange as the main sorption mechanism. An example of a test with recorded and calculated break-through curves is shown in Figure 5.

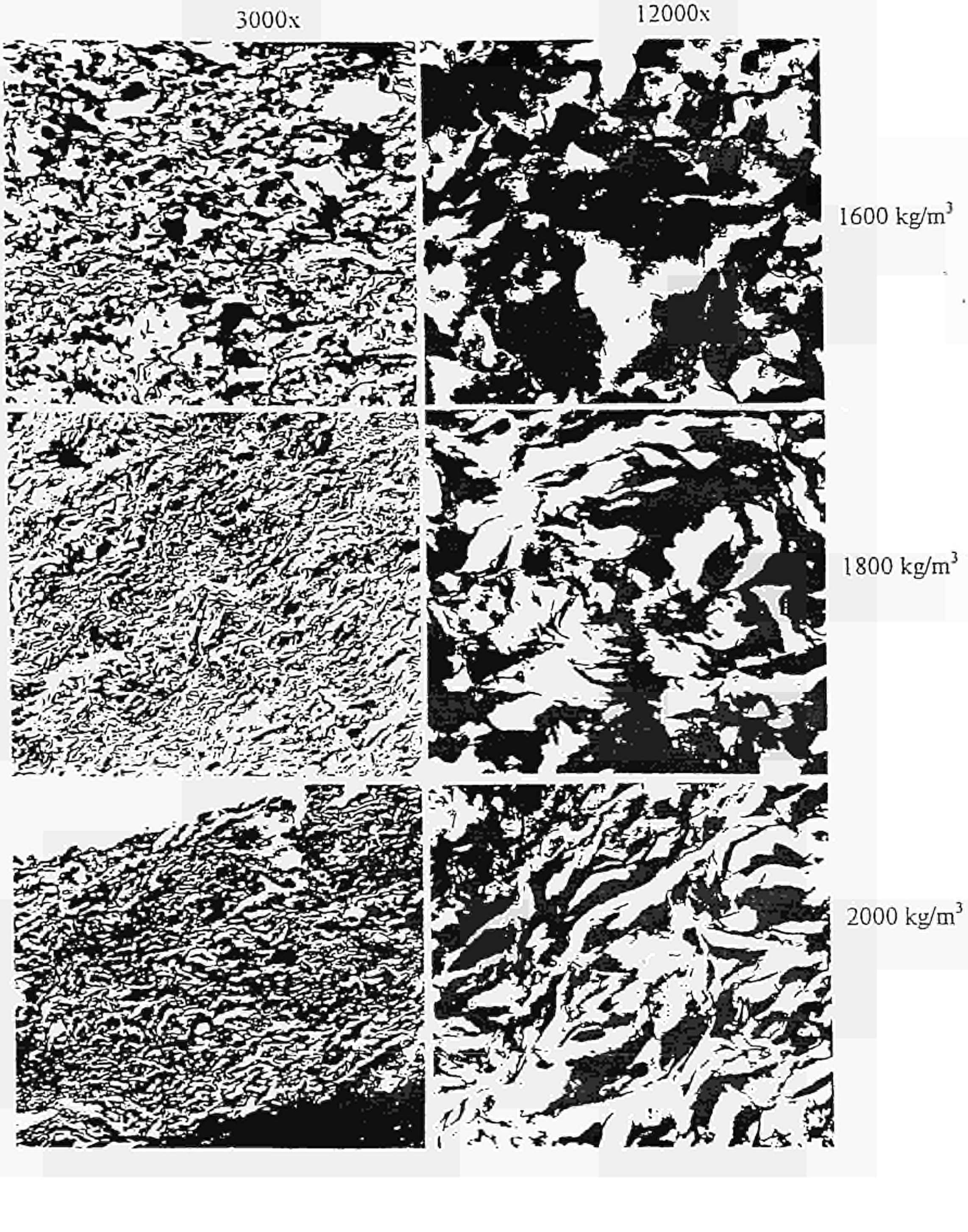
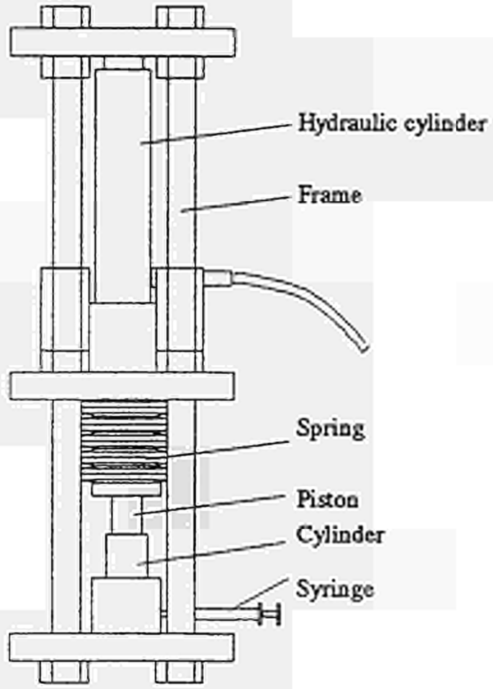


Figure 1. TEM micrographs of MX-80 at different densities.



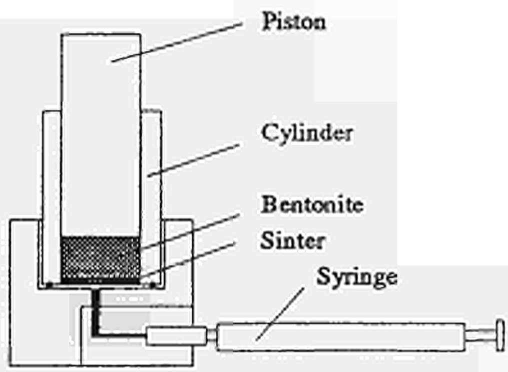


Figure 2: Upper: Pressing apparatus (left) and compaction cell (right)

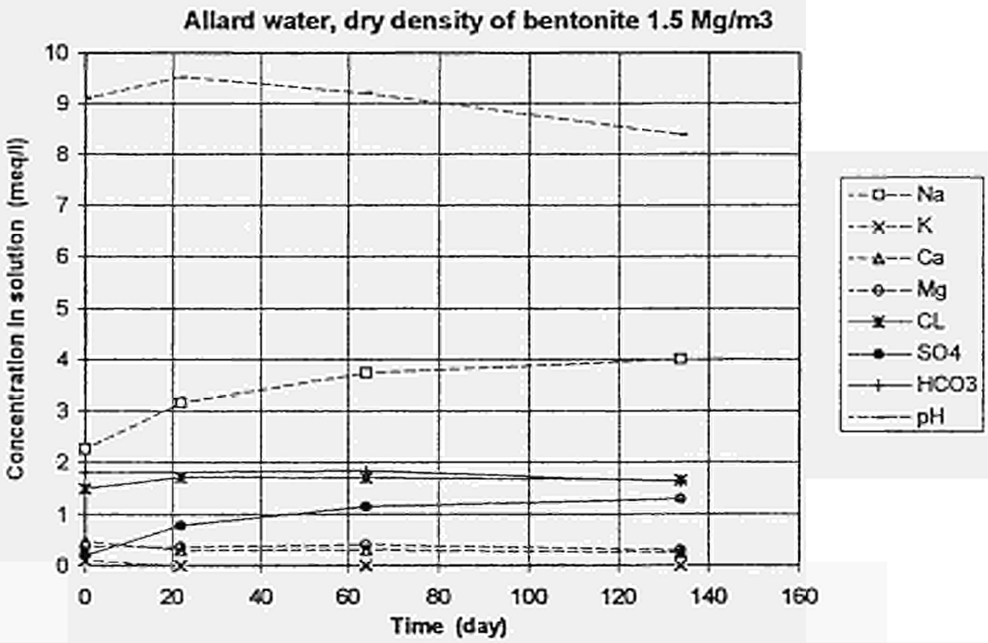


Figure 3. The concentrations of the experimental solution as a function of time. MX-80 bentonite with fresh water.

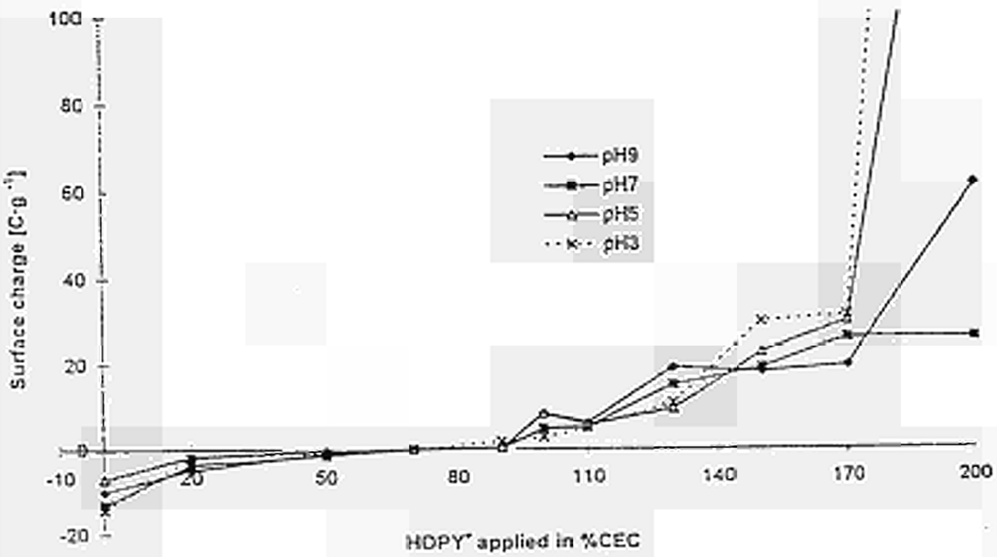


Figure 4. Surface particle charge of MX-80 bentonite measured at different pH in the original and in the different $HDPy^+$-treated forms.

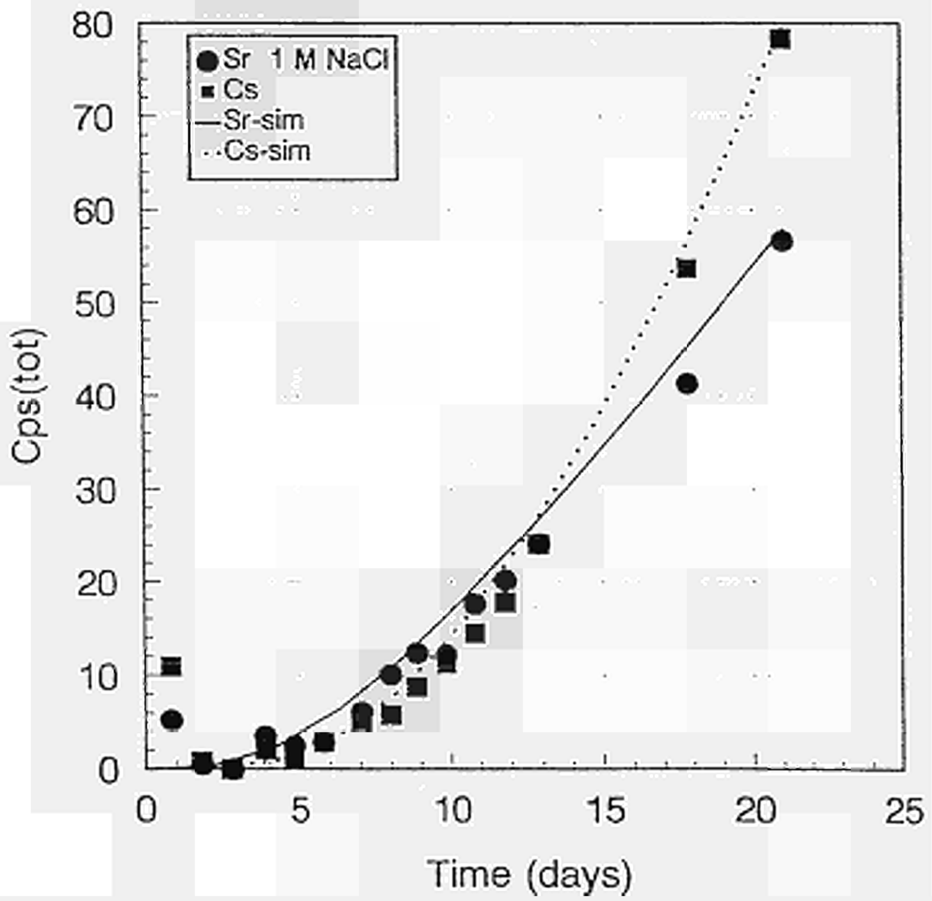


Figure 5. Experimental and calculated break through curves. Dry density 1.8 g cm^{-3}, 1 M NaCl solution.

C.3.5-1 PROGRESS : Project of research into gas generation and migration in radioactive waste repository systems

Contract No:	FI4W-CT96-0024	**Duration:**	1 May 1996 - 30 April 1999
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Partners:	**TVO Olkiluoto/FI, ISTec Garching/DE, SCK/CEN Mol/BE, QuantiSci Henley-on-Thames/GB, Univ. Birmingham/GB, Univ. "La Sapienza" Roma/IT, Univ. Exeter/GB, BGS Keyworth/GB, ISMES Bergamo/IT, Univ. UPC Barcelona/ES**		

A. OBJECTIVES AND SCOPE

There is currently a good understanding of the principle gas generation mechanisms within a repository for low- and intermediate level waste and understanding of gas migration in porous media is reasonably advanced. However there remain uncertainties about the kinetics of gas generation processes, in particular microbial degradation of organic wastes, and there is a further need for a clearer understanding of the way in which gas migrates in different host rocks under consideration for deep geological repository systems.

The main objectives of the project are therefore :

- to supplement experimental measurements both in laboratory and in-situ
- to interpret and analyze the experimental data in order to further develop mathematical tools
- to derive design requirements for waste repositories
- to provide necessary input for realistic calculations of the consequences of gas generation on performance assessment of waste repositories.

B. WORK PROGRAMME

B1. Subproject : GASGEN : Gas generation in radioactive waste repositories

B 1.1 Large scale gas generation experiment (TVO)

A large scale gas generation experiment will be carried out in an underground tunnel located in the VLJ repository at Olkiluoto. A metal vessel will be filled with 16 perforated steel drums, containing typical low-level operating waste from BWR'S together with specimen of concrete, and flooded with water. In addition laboratory experiments will be performed and predictive gas generation modelling undertaken using the GAMMON computer programme.

B 1.2. Development of correlation between experimental gas generation measurements and characteristic waste parameters. (ISTec)

Statistical evaluations of existing data from global gas generation experiments using real waste packages will be carried out and additional carefully designed experiments performed focussed on measurements of key parameters influencing gas generation within waste packages. Gas generation models will be further developed enabling correlations to be identified between characteristic waste parameters and gas generating processes.

B 1.3. Gas generation in repository in clay (SCK/CEN)

Experiments will be performed to measure gas generation by anaerobic metal corrosion. Batch experiments will be carried out with a mixture of clay, water and metal powder.

B2. Subproject : GAMERS : Gas Migration in European Repository Systems

B2.1. **Gas migration in low permeability fractured rock**

B2.1.1. Review and modelling of natural gas migration (QuantiSci)

A literature review on previous studies and available gas exploration data will be undertaken and further calculations carried out with two-phase flow codes.

B2.1.2. PET experiments on water-saturated single fractures (AEA - Univ Birmingham)

Positron Emission Tomography (PET) techniques will be applied to image gas flow through fractures in typical natural rock samples. The observed gas migration will be modelled by the development of a programme to describe gas migration through single water-saturated rough fractures.

B2.1.3. Laboratory and field experiments on microbubbles (Univ. Roma/AEA)

Columnar experimental devices will be used to study the kinetics of microbubble flow through porous media and the transport of tracers linked to air bubbles through saturated porous and fractured media. A supporting field programme will be undertaken to study the natural evidence of material transport by gas flow from underground.

B2.1.4. Gas injection experiment at a fractured rock site (AEA Technology, Univ Exeter)

A gas injection experiment will be performed into a shallow inclined borehole at Sellafield to study the localization of gas upon release at the land surface.

B2.2. **Gas migration in salt** (ISTec)

B2.2.1. Investigation of design requirements

Two-phase flow of gas and brine will be evaluated for establishing design requirements for a repository in salt as a function of various relevant parameters.

B2.2.2. Brine migration along grain boundaries

Physical models will be devised that apply to brine migration along grain boundaries and a study undertaken on its possible influence on brine supply.

B2.2.3. Provision of necessary input data

Experiments will be carried out to measure two-phase flow parameters (relative permeabilities, capillary pressure as a function of saturation) in porous salt media.

B2.2.4. Host rock convergence

The TOUGH2 code will be further developed by including the impact of geomechanical effects such as convergence in the modelling of two-phase flow.

B2.3. **Gas migration in clay**

B2.3.1. Laboratory experiments on gas migration under isotropic stress conditions

BGS and SCK/CEN will perform gas flow experiments on clay cores. Special attention will be paid to the detection of preferential pathways.

B2.3.2. Physical soil model tests (ISMES)

Centrifuge tests will be performed on reconstituted saturated clay specimens.

B2.3.3. In-situ experiments (SCK/CEN)

In-situ gas injection experiments will be carried out in the HADES facility at Mol as a continuation of the experiments carried out under the MEGAS 1 project.

B2.3.4. Modelling (QuantiSci - UPC)

The MEGAS 1 experimental results will be further modelled from the perspective of non-Darcy phenomena. A common framework for model and numerical code development will be established to ensure compatibility between numerical models.

B2.3.5. Development of a geomechanical gas migration code (QuantiSci - UPC)

Coupling of a geomechanical and preferential pathways gas migration model and testing against experimental results.

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The PROGRESS project contract started on May 1, 1996. A launch meeting was held in the UK from 21-22 May, 1996, to cement the interactions between partners and promulgate the detailed scope and objectives of the whole programme amongst them. The first six-monthly progress report was delivered to the European Commission in November 1996, in accordance with the contract requirements. At that time the estimated EC contribution to the costs incurred was about 208 kECU, or about 10% of the project budget of 2139 kECU.

As it is early in the project, work has not progressed far enough to have reached many of the technical milestones. The only technical deliverable so far due is a report from topic C2.1.1 below. This was delivered in draft on schedule, but it is being revised to incorporate new data.

C1. **Subproject : GASGEN** : Gas generation in radioactive waste repositories

C1.1. Large scale gas generation experiment (TVO)

A laboratory-scale degradation test of waste materials was started in anaerobic conditions, with the aim of determining the gas production capacity of the materials. A second set of specimens is being studied because of segregation of material during grinding of the first set.

In support of the planned large-scale experiment in the Construction Tunnel of the VLJ Repository at Olkiluoto, computer modelling was carried out using the GAMMON code to predict gas generation rates. The GAMMON runs suggest that hydrogen will be the dominant gas evolved in the experiment over at least the first two years.

Detailed planning of the large-scale experiment was carried out. Low-level reactor operating waste, compacted in 16 steel drums, is waiting to be emplaced into a concrete box and enveloped by a steel tank.

C1.2. Development of correlation between experimental gas generation measurements and characteristic waste parameters (ISTec))

As this activity is not being funded be the European Commission, no progress report on the item has been provided. A report of the work will be provided to the annual review meeting.

C1.3. Gas generation in repository in clay (SCK/CEN)

A new batch reactor has been designed to perform the gas generation experiments. Acquisition of the construction materials leading to building of the reactor is in progress. Two kinds of metallic powders have been selected for the anaerobic corrosion experiments, namely an iron powder and a stainless steel 316L powder. They represent two different options for canister materials.

C2. **Subproject : GAMERS** : Gas Migration in European Repository Systems

C2.1. **Gas migration in low permeability fractured rock**

C2.1.1. Review and modelling of natural gas migration (QuantiSci)

A literature review has been carried out covering hydrocarbon industry experience and information on the source and generation of petroleum, the mechanisms for the migration of petroleum (liquid and gaseous) from source rocks through carrier rocks to reservoir traps and then leakage from there to surface seeps, the use of seeps in identifying gas sources, relevant case histories, and modelling techniques used within the industry for gas migration.

The industry consensus was found to be that migration of gas to the reservoir occurs in a separate phase from the water and is driven by buoyancy forces resulting from density differences. The dominant mechanism for leakage from traps through overlying cap rocks is disputed, but buoyancy provides the driving force. Capillary leakage through pore spaces is thought to be more significant than flow through fractures except within a few hundred metres of the surface or at high pore-fluid pressures. Methane seeps are widespread and associated with subsurface gas accumulations, but are not necessarily from a petroleum source. A draft report on the literature review was completed in November 1996, but is being revised to include new data that has been received as a result of the initial requests for information.

Only a small amount of quantitative data on natural analogue gas migration was revealed by the literature survey. This has limited the scope for the application of modelling tools to simulate the data, but a programme has been devised to undertake such modelling studies as are appropriate.

C2.1.2. PET experiments on water-saturated single fractures (AEA Technology, Univ Birmingham)

Work has progressed on acquiring and mounting two samples of fractured rock for the experimental part of this work programme. The first sample is a piece of Triassic sandstone core approximately 35 cm long with a single fracture running its entire length. The second sample is a small block of Borrowdale volcanic rock from the Lake District National Park, Cumbria. The block overall is about 30 cm x 40 cm and contains several distinct fracture features. As a result of difficulties in preparing the samples, no PET imaging studies have so far been undertaken. Some developments to the PET data processing methodology was required to allow work on irregular shaped blocks to be carried out. This has been accomplished.

The modelling work undertaken has been on the development of a computer model of gas migration through rough water-saturated fractures that will be used to model the results of the PET experiments. Testing of an implementation of a published algorithm revealed some difficulties with that particular approach. The most significant of these was that, in calculations using relatively large numbers of grid blocks in the discretisation of the fracture roughness, the algorithm led to the selection of short timesteps and hence excessive computation times. In an attempt to overcome these difficulties, a new algorithm, which is conceptually simpler than that published has been devised and implemented. The new approach allows longer time steps to be enforced than in the earlier model. Testing of the new algorithm appears to indicate that it is working satisfactorily.

C2.1.3. Laboratory and field experiments on microbubbles (Univ Roma, AEA Technology)

An experimental structure, designed to simulate the pressure / temperature conditions occurring at depth, was constructed for the laboratory study. The system consists of a columnar device hosting an initially saturated porous medium that can be heated up to a temperature of 70°C and pressurised up to 5 bars. A series of tubes and manual or solenoid valves make it possible to generate micro-bubbles and to control working parameters. Numerous experiments on bubble transport will be conducted using this apparatus, primarily in regard to the influence of the pressure-temperature regime and the importance of the properties of the geological medium (i.e. porosity, permeability, etc.). If bubble transport is detected, studies will also be undertaken on the role of different carrier gases on transport and on the way in which micro-particles are linked to the advecting micro-bubbles (e.g. surface tension or as an aerosol).

The first stage of the field investigations carried out in the Tolfetano-Cerite volcanic district (Latium) have been focused on the definition of areas in which gas-phase transport could be detected. To this aim, data has been collected and correlated from soil-gas and geo-electrical surveys as well as from geological mapping. From this data it has been possible to select some sectors near the village of Sasso di Furbara which are characterised by the coincident presence of enhanced endogenic gas leakage, electrical resistivity lows and diffuse sulphide mineralisation. These will be the areas subjected to further work focused on the study of the capacity for solid-particle transport by endogenic gas fluxes.

C2.1.4. Gas injection experiment at a fractured rock site (AEA Technology, Univ Exeter)

Work on this activity is scheduled to start during 1997.

C2.2. **Gas migration in salt** (ISTec)

C2.2.1. Investigation of design requirements

In order to investigate preliminary design requirements for a repository in rock salt, a scenario analysis is underway. This analysis has the objective to identify the potential evolution with time of porosity and permeability characteristics of the crushed salt backfilling. Based on this analysis the orders of magnitude for the porosity and permeability development used for preliminary calculations will be defined and the initial conditions will be set.

C2.2.2. Brine migration along grain boundaries

Current knowledge about principle brine sources and brine migration processes in rock salt has been reviewed and included in a status report.

C2.2.3. Provision of necessary input data

Work has been carried out to identify a suitable equation-of-state (EOS) model to describe equilibria of brine-gas mixtures. Existing measured equilibrium data for brine-gas mixtures in the literature have been collected. For the acquisition of accurate vapour pressures and salt solubility at different temperatures and brine compositions the EQ3/6 EOS model of the Lawrence Livermore National laboratory will be used. The EQ3/6 algorithm has been computationally implemented. Further verification calculations are underway to explore the existing Pitzer data files for salinity conditions.

The significance of effects like precipitation of salt for permeability and porosity variation has been analysed.

The EOS7 EOS model of the Lawrence Berkeley Laboratory has been implemented and verified. This module for the TOUGH2 code is currently being analysed for the possibility of including additional salt specific properties, solubility, and eventually a numerical coupling with EQ3/6.

Other Darcy two-phase flow codes have been analysed with respect to the possibility of including salt specific modifications and extensions.

Additional data needs are for two-phase flow parameters for potential backfilling materials in saline repositories. Different organisations have been invited to submit proposals for the measurement of these parameters. The proposals are being assessed.

C2.2.4. Host rock convergence

Current knowledge about salt creep has been reviewed and included in a status report. Different salt creep laws have been compared and the law of Zhang and coworkers has been identified as appropriate for the planned numerical analysis.

C2.3. Gas migration in clay

C2.3.1. Laboratory experiments on gas migration under isotropic stress conditions (BGS, SCK.CEN)

Work has been carried out to derive relationships between total stress, gas entry pressure at breakthrough, and gas permeability for existing experimental data on Boom clay and bentonite. A framework of linear elastic fracture mechanics is being used to derive these relationships in terms of the initiation and propagation of fractures formed by the applied gas pressure.

During the previous MEGAS experiments on gas migration through Boom clay, "intermittent" or "burst-type" gas flow was observed. In order to examine the details of these rapidly varying flows, an acoustic emission (AE) sensor has been purchased and linked via a fast data acquisition card to a 32-bit microcomputer. A computer program has been written to process the signals. The sensor has been mounted on a waveguide which is fixed to the base of BGS's new axisymmetric gas injection apparatus. The aim of the experiments will be to monitor acoustic emissions generated by microcracking. It has been observed that the rapid expulsion of gas from a compact clay results in fairly conspicuous "crackling" sounds (in the audible range) which may be indicative of the rupture of interparticle water films.

For the planned work by SCK.CEN on gas migration in Boom clay, the construction has been ordered of three new isostatic cells following the plans for similar cells used previously in phase 2 of the MEGAS project. Some slight modifications have been introduced, namely in the height of the cell (16 cm instead of 30 cm) and in the membrane surrounding the clay sample. This membrane will be a copper sheath of 0.1 mm thick, which will be laser-welded onto both end caps supporting the clay sample. This will avoid any leak of helium into the confining fluid during gas injection, as was observed in experiments conducted in phase 2 of MEGAS.

C2.3.2. Physical soil model tests (ISMES)

The work of this activity involves soil centrifuge testing. Preparations have been made for this by carrying out checks on all equipment necessary for the planned test. This includes a preliminary check of both the apparatus and the procedure for the tests.

C2.3.3. In-situ experiments (SCK.CEN)

For the planned in-situ gas injection experiments a choice has been made concerning the emplacement of the gas injection point in the MEGAS E5 set-up. Gas will be injected in filter 13 located on multipiezometer C (in the vertical plane of the central multipiezometer A). The reasons for choosing this site are the following: there is confidence that the vicinity of multipiezometer C has not been perturbed during previous gas injection undertaken during the first phase of MEGAS; filter 13 occupies a central position in the 3D set-up; its neighbouring filters are close and located at two different distances (25 and 50 cm). A pressure controller has been purchased to maintain a constant gas injection pressure during the whole duration of the gas flow. Prior to the gas injection, a single point water permeability measurement around filter 13 is being made.

C2.3.4. Modelling (QuantiSci, UPC)

The development of a generic model for gas migration via a single propagating gas pathway through an argillaceous medium has been completed. Initial work making the generic model more specific to Boom clay has been undertaken by incorporating a microstructure model for Boom clay. A key feature of the Boom clay model is that the information and propagation of the gas pathway is controlled by the stress field.

Sub-models have been formulated to assist in quantitative characterisation of the behaviour of two different postulated methods by which a gas pathway might propagate through an argillaceous medium: drainage of pre-existing water filled capillaries; dynamic creation of pathways. The two sub-models have been used to review the experimental results from MEGAS 1. Comparison with the experimental results has highlighted how the single gas pathway models are able to provide an insight into the model of gas migration in the experiments: it is found that much of the behaviour in the MEGAS experiments can be explained using the single gas pathway model with a dynamically created gas pathway.

The UPC computer program CODE BRIGHT is being used to simulate the results of clay core gas injection laboratory tests and in situ experiments performed in the MEGAS 1 project. This will allow cross verification and calibration/validation of models and the study of sensitivity to parameters.

A series of improvements have been made to the computer programs developed by UPC to model coupled thermo-hydro-mechanical problems in porous media.

C2.3.5. Development of a geomechanical gas migration code (QuantiSci, UPC)

The generic model developed as described under C2.3.4 above explicitly includes the standard stress tensor. An idealised microstructure model for Boom Clay has been used to develop simple expressions for the components of the stress tensor. With more detailed thought about the physical basis of the way by which the stress fields are represented, it is planned that this should lead to the implementation of a geomechanical gas migration code for predicting the formation of preferential pathways. Since the focus is on gas pathway propagation, it is no longer planned to consider a modification of Darcy's Law, taking into account the effect on permeability and porosity of effective stress.

C.3.6-1 GESAMAC : Conceptual and computational tools to tackle long-term risk from nuclear waste disposal in the geosphere

Contract No: FI4W-CT95-0017	**Duration:**	1 Jan 1996 - 31 Dec 1998	
Coordinator: P. Prado, CIEMAT Madrid/ES			
Tel. +34.1.34.66.528	Fax: +34.1.34.66.269	E-mail : prado @ ciemat.es	
Partners: JRC-EI Ispra/IT, Univ. Bath Bath/GB, Univ. Stockholm Stockholm/SE			

A. OBJECTIVES AND SCOPE

The overall objective of the project GESAMAC (GEosphere modelling - geosphere Sensitivity Analysis - Model uncertainty in geosphere modelling - Advanced Computing in stochastic geosphere simulation) is to tackle areas of uncertainty and develop some conceptual, methodological and computational tools which can be used in actual safety analysis case studies. The project focuses on issues such as (i) models of radionuclide migration, (ii) sensitivity analysis in the field of modelling radionuclide migration (iii) rigorous treatment of different sources of uncertainty and (iv) the realization of a computational framework in which all the various stages of a safety study can be tested and applied.

B. WORK PROGRAMME

B1. Geosphere Modelling (CIEMAT)

Study the effects of parameter and model assumptions on the predictions of the model related to the physico-chemical reactions between the liquid and solid phases to describe the transport of two species.

B2. Sensitivity Analysis (JRC-EI)

Investigation of Monte Carlo based variance measures (also known as Sobol' sensitivity indices), quasi random numbers and usual regression/correlation methods.

B3. Uncertainty Analysis (Univ. Bath)

Quantitative synthesis of all sources of uncertainties (scenario, structural, parametric, predictive) combined in a Bayesian framework of model uncertainty to the specifics of groundwater transport of radionuclides.

B4. Parallel MC Driver (Univ. Stockholm)

Design and testing of a parallel Monte-Carlo driver for geological disposal safety calculations.

C. Progress of work and results obtained

Summary of main issues

The milestones and deliverables programmed for the first year of the project and recorded in the technical annex of the contract [1], have been successfully completed for each work package [2-5]. During 1996 the project GESAMAC was mostly concentrated on: 1) the development of a multi-layer mono-dimensional groundwater transport code taking into account a selected number of physico-chemical reactions between the solid and liquid phases (see B.1). The new version released was presented at the second meeting of the project in Brussels; 2) the investigation of the more recent variance based test for sensitivity analysis of model output (see B.2.); the FAST (Fourier Amplitude Sensitivity Test) and Sobol' indices were compared by means of a computational experiment, showing the advantages and disadvantages of both methods. 3) the exploration of Markov chain Monte Carlo (MCMC) and Laplace approximation approaches in GESAMAC uncertainty framework (see B.3.); it has been concluded that MCMC will not be needed in early phases of the project, and may not be necessary even when all forms of uncertainty are considered; also, has been developed a general scheme of the four different sources of uncertainties in GESAMAC; 4) the development of a conceptual design and the technical specifications of a parallel Monte Carlo driver in the existing platforms (see B.4.); a conceptual design of the parallel PREP (PRE-Processor) code has been developed and tested with a simple model. Additionally a test case study, in order to test the methods and tools developed for each teams, has been proposed for 1997.

C.1. Geosphere Modelling (CIEMAT)

During 1996 this part of the GESAMAC work package was mostly concentrated on the development of one dimensional groundwater transport code that considers different physico chemical reactions between the liquid and stationary phases involved. In particular the activities in this work package started by analysing the last version of GTM (Geosphere Transport Model) series developed at the JRC of Ispra and named GTMCHEM (GTM with CHEMical reactions). GTMCHEM retains most of the original GTM1 (GTM release One) structure and capabilities and incorporates new chemical reactions. GTMCHEM was recorded in a more modular and user friendly version that permits Monte Carlo simulations without changes to the source program for each simulation. In particular the different chemical reactions managed by the code were separately programmed, letting the code its automatic management to customise the source/sink terms for each chemical species involved at each simulation. The information related to the chemical reaction implicated are transferred to the code through a "chemical matrix" (see Table I) included in the input file. The new version has been tested (see Figure 1) and documented [6]. Current version solves the one dimensional advective/dispersive transport equation with the following possible physico chemical reactions between solid and liquid phases: adsorption by linear isotherm in equilibrium (k_d concept), adsorption by slow reversible reaction (Langmuir type), homogeneous first order chemical kinetics reaction, irreversible reactions (filtration, biodegradation, etc.) and radioactive decay.

C.2. Sensitivity Analysis (JRC-IT)

The objective of this part of the GESAMAC project is to develop better sensitivity analysis methods for use in the probabilistic assessment. During 1996 the activities in this work package were concentrated on theoretical progresses in the sensitivity analysis methods which resulted in papers in the open literature [7,8].

The main conclusions of the sensitivity analysis studies conducted through this period were: a) the theoretical foundation of the ANOVA-like decomposition of variance in experimental design and that of FAST, Sobol' indices is the same; b) Bootstrap can be used to quantify the error in the numerical estimate of the sensitivity indices; c) FAST and Sobol' indices of the first order yields the same number, i.e. they are identical in their predictions; d) FAST is more computationally efficient that Sobol' indices; e) FAST seems to be biased compared to the Sobol' indices and to the expected (analytical) values; and e) the Sobol' indices offer a very useful measure of sensitivity, linked to the total effect (main plus interactions) of a parameter (see Figure 2). At present FAST does not offer such a possibility. A new sensitivity analysis estimator is already under study, and is based on an attempt to make FAST unbiased and capable of computing the higher order terms (as Sobol' indices) using a lower sample size. Another approach which will be taken is that of reducing the sample size needed for Sobol' indices by innovative approaches to sample selection.

C.3. Uncertainty Analysis (Univ. Bath)

The aim of this work package refers to a more rigorous treatment of model uncertainty combining all sources of uncertainty (scenario, structural, parametric and predictive). The objective for the first year of the project was to explore Markov chain Monte Carlo (MCMC) and Laplace approximations to assessment of posterior model probabilities, with a limited subset of relevant scenario and structural choices. The role of Markov Chain Monte Carlo in GESAMAC simulations have been considered; it has been concluded that MCMC will not be needed in early phases of the project, and may not be necessary even when all forms of uncertainty are considered. The framework of application of a full predictive uncertainty assessment in GESAMAC (see Figure 3) and how to propagate all sources of uncertainty has been stabilised (liaising with B.1. and B.2). Initial calculations have been performed using the first version of GTM (see C.1) ant the PSACOIN Level E Intercomparison specifications (PSAG, OECD/NEA). Initial discussions have been maintained on developing a Bayesian layer to select structure and scenario at random in PREP (see C.4.).

C.4. Parallel MC Driver (Univ. Stockholm)

The aim of this work package within GESAMAC is to develop a parallel Monte Carlo driver. The driver is intended to exploit the potentialities of high-performance computing environments (workstations working in parallel or parallel supercomputers). The driver will have the same functionality of the serial PREP code, but include also the inclusion of a Baysean layer for probabilistic assessments (see C.3.).

During the first year the conceptual design of the Monte Carlo parallel drive has been developed [5]. This work has included a desktop study and the development of a code prototype. In short the desktop study addressed a) how to implement a parallel code using MPI (Message Passing Interface) to monitor the input of submodels that make radionuclide transport calculations, b) how to sample the input parameters and call these submodels and c) how to prepare the output for statistical post-processing, uncertainty and sensitivity analyses (see Figure 4). It was necessary to discuss with the other work packages to define how to implement a Baysean layer (see C.3.), to study the GTM code in order to analyse how to design an interface to this submodel (see C.1.) and to understand the ongoing work on Sensitivity Analysis methods (see C.2.). A prototype code has been developed to be able to address in a concrete way some of the aspects of the conceptual design. This first prototype implemented on a parallel supercomputer [9] has been tested as a probabilistic monitor of a transport code for fractured media. The resulting work has been accepted and will appear in the proceedings of the WM'97 conference in Tucson, Arizona in March 97 [10]. Other work using the parallel Monte Carlo driver prototype which has been submitted to publication are [11,12].

References

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[11] ANDERSSON, M., MENDES, B., PEREIRA, A., Introducing Heterogeneity of Fractured Media in a Monte Carlo Model for Use in Risk Assessments of High-Level Radioactive Waste. Submitted to publication. (1996)

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Table I. Example of the 'chemical matrix' in the input file for GTMCHEM

```
.......
MATRIZ-Q:    N°REA TIPO NCHA NELM N°ORD CTE-R(LY=1) CTE-R(LY=2) CTE-R(LY=3)
               1    1    1    1    1     4.40E+9     4.40E+9     4.40E+9
               1    1    1    2    2     4.40E+9     4.40E+9     4.40E+9
               1    1    1    3    3     5.51E+1     5.51E+1     5.51E+1
               2    2    1    1    1     4.00E-1     0.00E+0     0.00E+0
               2    2    1    4    2     1.00E-2     0.00E+0     0.00E+0
               0    0    0    0    0     0.00E+0     0.00E+0     0.00E+0
.......
```

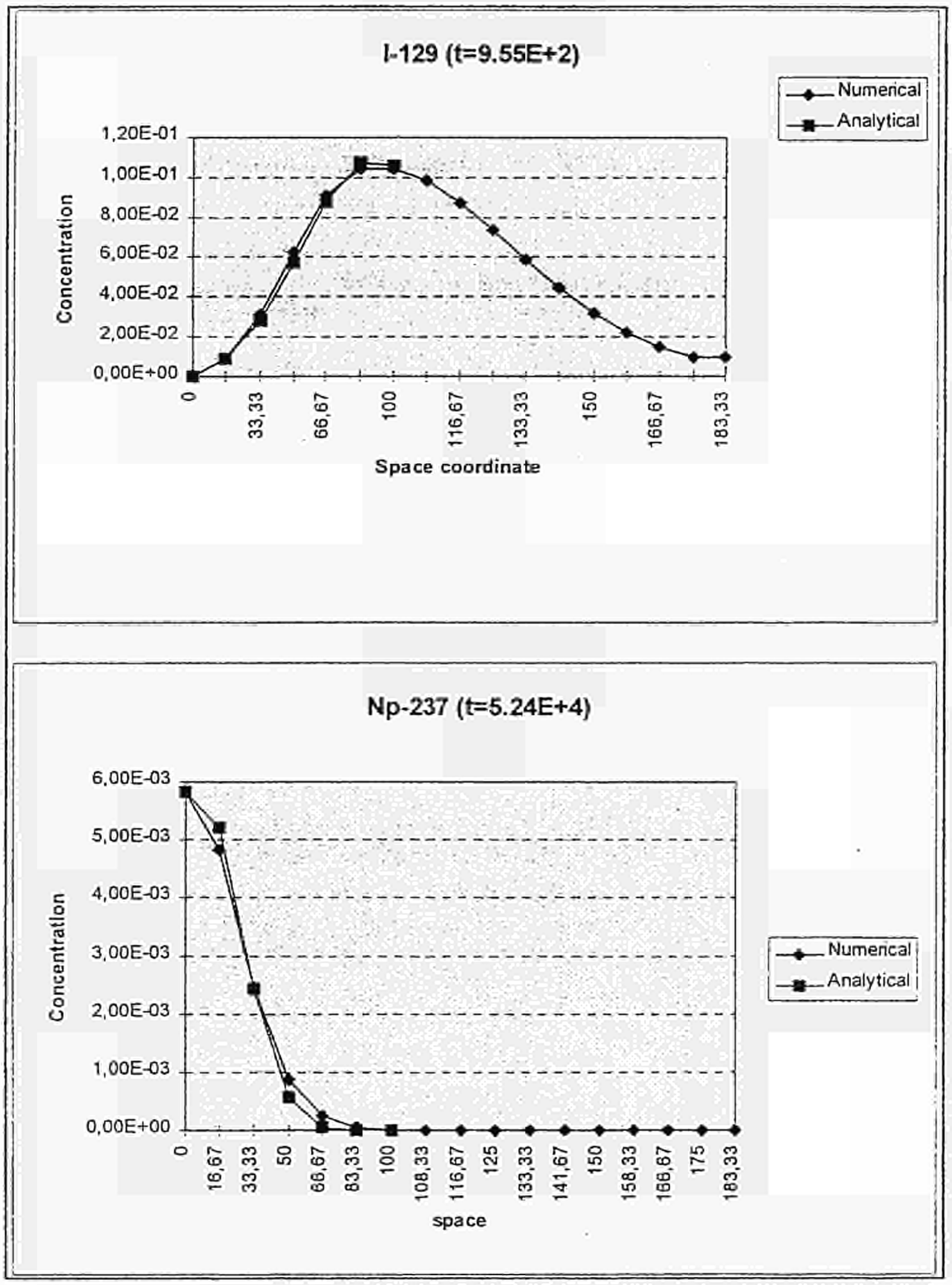

Figure 1: Outputs from GTCHEM (PSACOIN Level E)

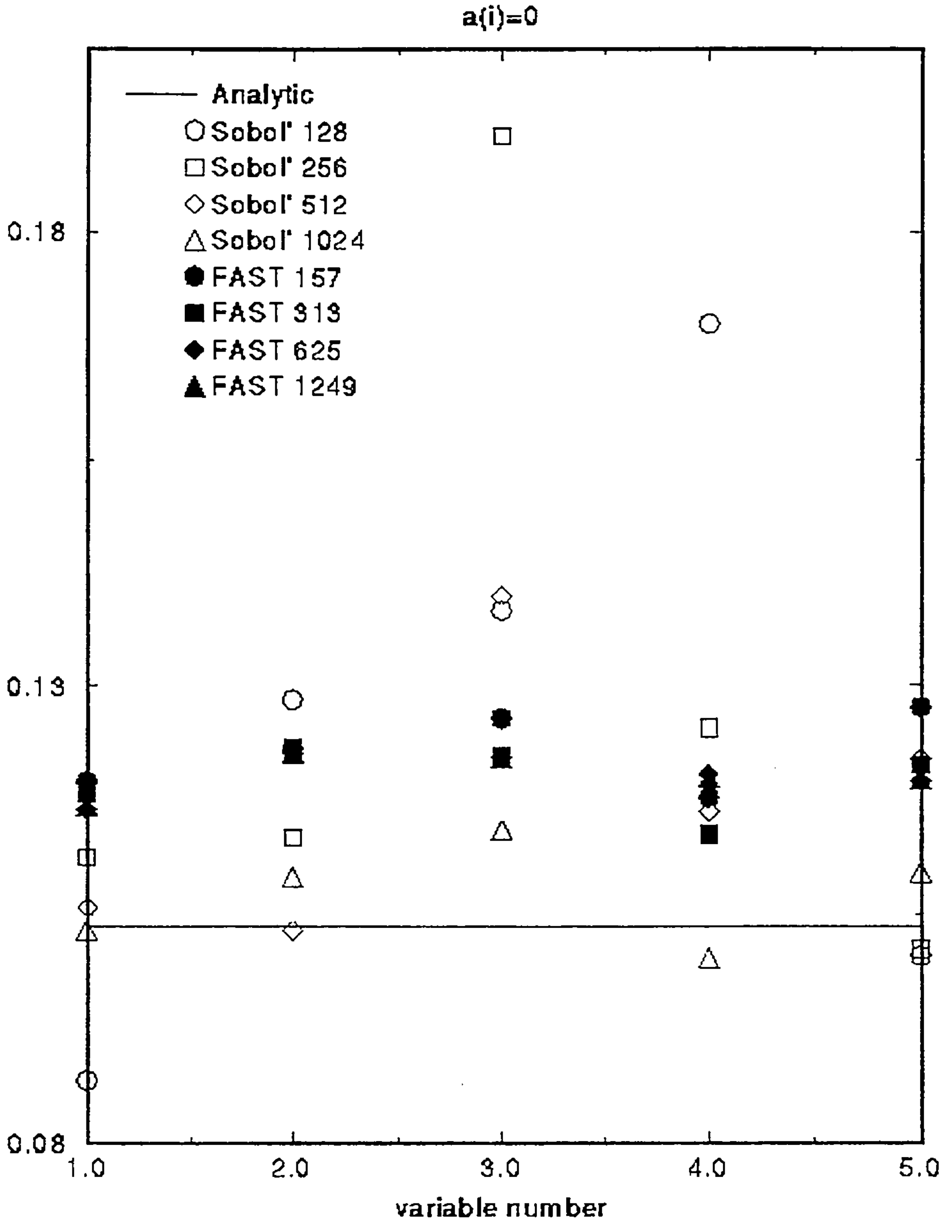

Figure 2: Comparison of FAST and Sobol' indices. The abscissa is time in year, and the ordinate is the value of the main effect for the input variables, as computed using FAST (full symbol) and Sobol' indices (empty symbol).

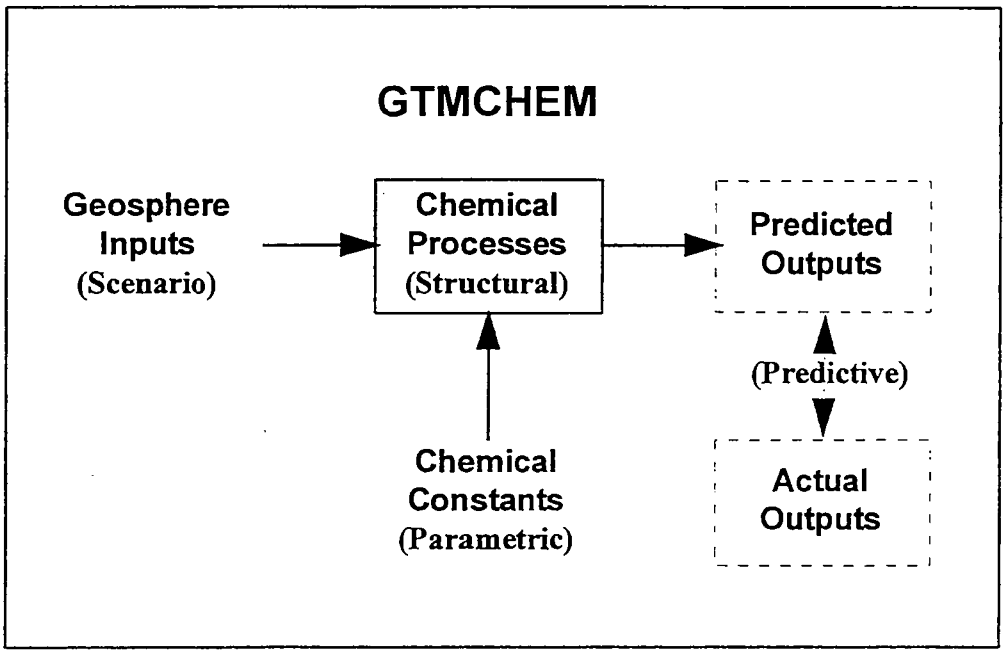

Figure 3: Schematic illustration of the four sources of uncertainty in GESAMAC

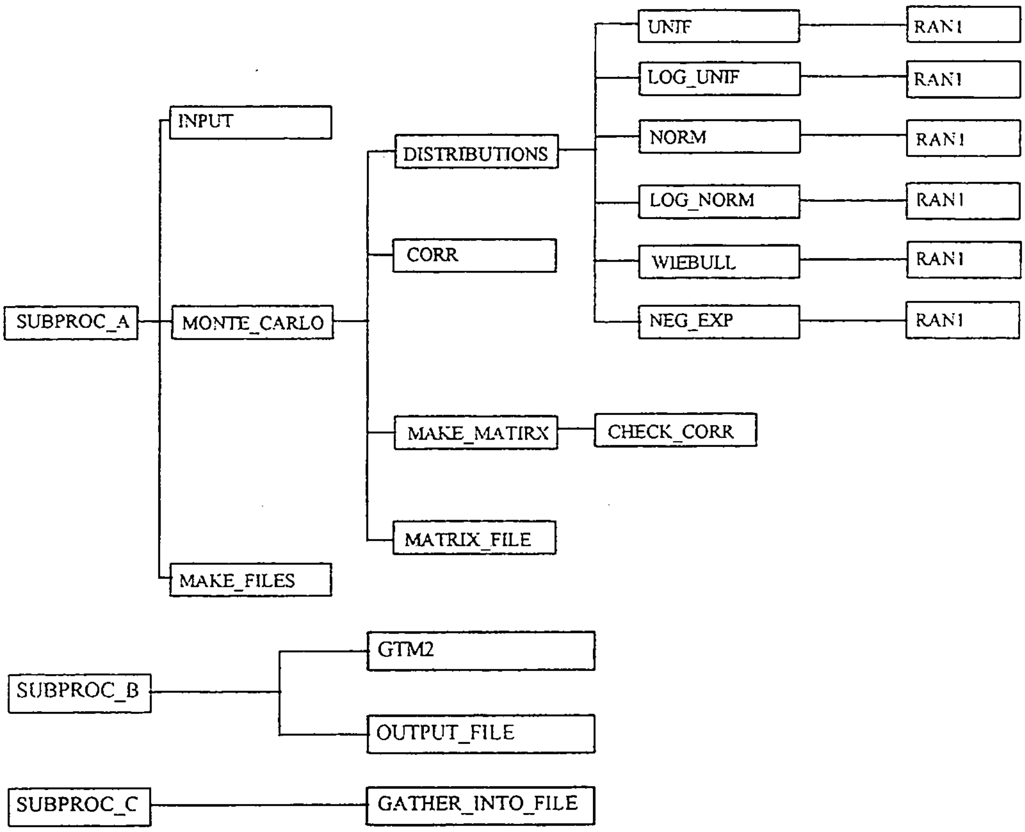

Figure 4: Conceptual model in different subprocesses A, B and C.

C.3.6-2 CARESS : The role of colloids in the transport of radionuclides from a radioactive waste repository : impact of colloid-borne radionuclide transport on safety assessment calculations

Contract No: FI4W-CT96-0021 **Duration:** 1 May 1996 - 30 April 1999
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Partners: **ARMINES Fontainebleau/FR, CEA/DCC Fontenay-aux-Roses/FR, CIEMAT Madrid/ES, CNRS/CSNSM Orsay/FR, Univ. Nantes/FR, GSF/IFH Neuherberg/DE, INFM Legnaro/IT, Univ. Loughborough/GB, QuantiSci Henley-on-Thames/GB, RMCE Abingdon/GB**

A. OBJECTIVES AND SCOPE

The project contributes to improve the knowledge and understanding whether colloids have critical impact upon the transport and retention of radionuclides in the geosphere and if they do, how the radionuclides associated with mobile colloids should be treated in safety assessment calculations.

The various objectives of the project are :

- to identify mobile phases which could be associated with radionuclides in the far field of a radioactive waste repository and to establish the conditions under which the colloidal transport is likely to be of importance. This is not limited to ambient colloidal distributions but also considers the near field as a potential source of colloids;
- to characterize colloid transport and radionuclide association with the mobile colloid phase using a number of established coupled processes models both on laboratory and field scales, and to assess how robustly these models can represent the physico-chemical processes involved;
- to develop effective guidelines for recognizing and accounting for the significance of colloids in radionuclide transport;
- to assess the implications of colloid-borne transport of radionuclides upon performance assessment calculations.

B. WORK PROGRAMME

B1. Laboratory experiments (CEA-DCC, AEA, CIEMAT, GSF, CNRS, Univ. Nantes, Univ. Loughborough, INFM)

Including colloid characterization and generation, batch monolith, model fracture and coloumn experiments.

B2. Model development and implementation (QuantiSci, ARMINES, RMCE)

This work package will focus on design of laboratory and field experiments, process modelling, application of 3 D particle tracking to colloid transport and surface interaction as well as colloid transport/migration modelling by consideration of upscaling and implications for PA.

B3. Field scale colloid migration experiments (AEA, CEA-DCC, CIEMAT)

Provide colloid transport data for testing various models developed under WP2 for scaling up conditions.

C. Progress of work and results obtained

Summary of main issues

The main issues within this reporting period are concerned with the design and commencement of batch and column experiments, a review of the current state of modelling with regard to colloids (transport and surface interactions) and the selection of a site for the field colloid tracer experiment in a porous medium.

Progress of work and results obtained

Monodisperse colloids have been synthesised and distributed (B.1.) for use in batch and column experiments. In dynamic retention experiments (B.1.) designed in collaboration with the modelling task (B.2.), transport of (unreactive) gold colloids has been demonstrated in a novel synthetic fracture. The particle tracking code is being adapted and the experimental system modified in order to successfully model transport of reactive colloids (B.2.). Batch experiments are underway to study the generation of bentonite colloids as examples of near-field colloids. Sand column experiments are in progress to study the transport of silica colloids as a function of associated radioelements or organic coatings and of the system chemistry and hydrodynamics (B.1.). A review of the present state of modelling of colloid transport and surface interactions is underway (B.2.) Atomic Force Microscopy (AFM) has been used to successfully image silica colloids on a mica surface (B.1.). Two possible sites have been identified for the field colloid tracer experiment in a porous medium at Dukovany in the Czech Republic and at the Winfrith site in Dorset, UK (B.3.).

C.1 Laboratory experiments (CEA-DCC, AEA, CIEMAT, GSF, CNRS, GERMETRAD, LUT, INFM)

As a common source of monodisperse colloids for laboratory experiments, synthetic silica, hematite and ceria colloids have been characterised and distributed to the partners. The use of Particle Induced X-ray Emission (PIXE) has been successfully tested for quantification of europium retained on silica monoliths (cm-size polished sections). These experiments to study the reversibility of metal retention are being extended to the use of Eu, Th and U in the presence or absence of colloids. Sorption kinetics are studied in synthetic fracture experiments designed in collaboration with the modelling task (B.2.) to ensure successful modelling. The transport of unreactive gold colloids has been successfully demonstrated through a uniform polycarbonate 'fracture' (Figure 1). AFM has been used to image the number density and size of colloids on a flat mica surface (Figure 2) prior to such measurements on the synthetic fractures. Silica colloids were found to facilitate europium transport through sand columns and the percentage mobilisation increased with decreasing europium concentration. The presence of humic acid (HA) led to a dominant HA facilitated transport. The breakthrough of silica colloids in vertical columns of packed sand was found to occur before that of the conservative tracer (^{36}Cl) for up-down flow. In studies of the generation of colloids under near-field conditions, the flow of synthetic granitic water through a column containing a mixture of crushed granite and bentonite resulted in the formation of subspherical particles of silica, iron and aluminium oxyhydroxides and mineral fragments.

C.2 Model development and implementation (QuantiSci, ARMINES, RMC-E)

A significant modelling effort has involved taking an active part in designing laboratory and field experiments to ensure that all necessary data are collected and that uncertainties and ambiguities in the database are minimised. Design input to the synthetic fracture studies (B.1.) is ongoing following the successful demonstration of the transport of unreactive gold colloids through a uniform fracture. An iterative adaptation of the experimental equipment has been started. This should help further reduce the dimension of the system to be solved, greatly aiding the successful modelling of colloid transport, allowing for the simple determination of surface sorption terms in the case of reactive colloids. The prototype tracking code has been adapted in order to broaden the possible inlet conditions allowed, thus reducing the discrepancies due to model and experimental limitations. A modelling review is underway to focus on the present state of modelling with regard to the transport and surface interactions of colloids.

C.3 Field scale colloid migration experiments (AEA, CEA-DCC, CIEMAT)

Field colloid tracer experiments provide data for testing the models developed in B.2 for scaled-up conditions. It is necessary to identify suitable sites, characterise the colloids and groundwater at such sites and design the experiments with the modellers playing an active part, using an up-scale of the experimental systems from the laboratory scale. There are currently two sites available to the project under consideration for the field experiment in a porous medium, at Dukovany in the Czech Republic and at Winfrith in Dorset, UK. The Dukovany tracer test area covers about 100m × 100m with three wells and nine piezometers drilled to a depth of 15m in the Tertiary sands. Tracer tests have previously been carried out under dynamic and natural flow conditions using dye-tracers and in a single well experiment using radioactive tracers. In principle, permission has been given to perform radioactive tracer experiments at a site within the Atomic Energy Establishment at Winfrith. The site is underlain by Tertiary strata consisting of Bagshot sands in beds several metres thick and a few hundred metres in lateral extent underlain by London Clay and the Reading Beds. The site has been extensively characterised in terms of geology, mineralogy and hydrology and contains many boreholes available for use.

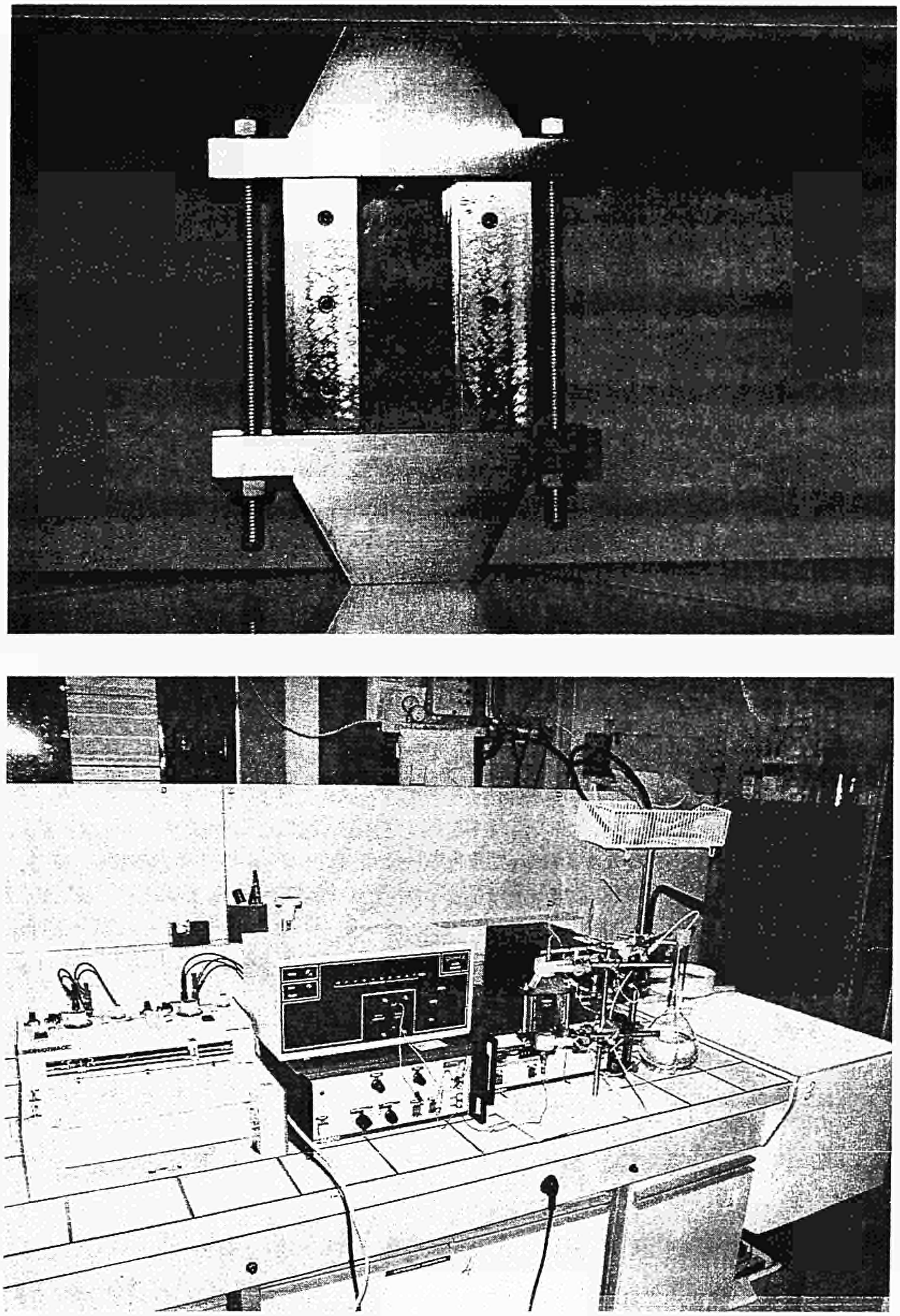

Figure 1: Two views of synthetic polycarbonate fracture (7.5 × 5.5cm, 100µl volume) used to study transport (vertical) of gold colloids.

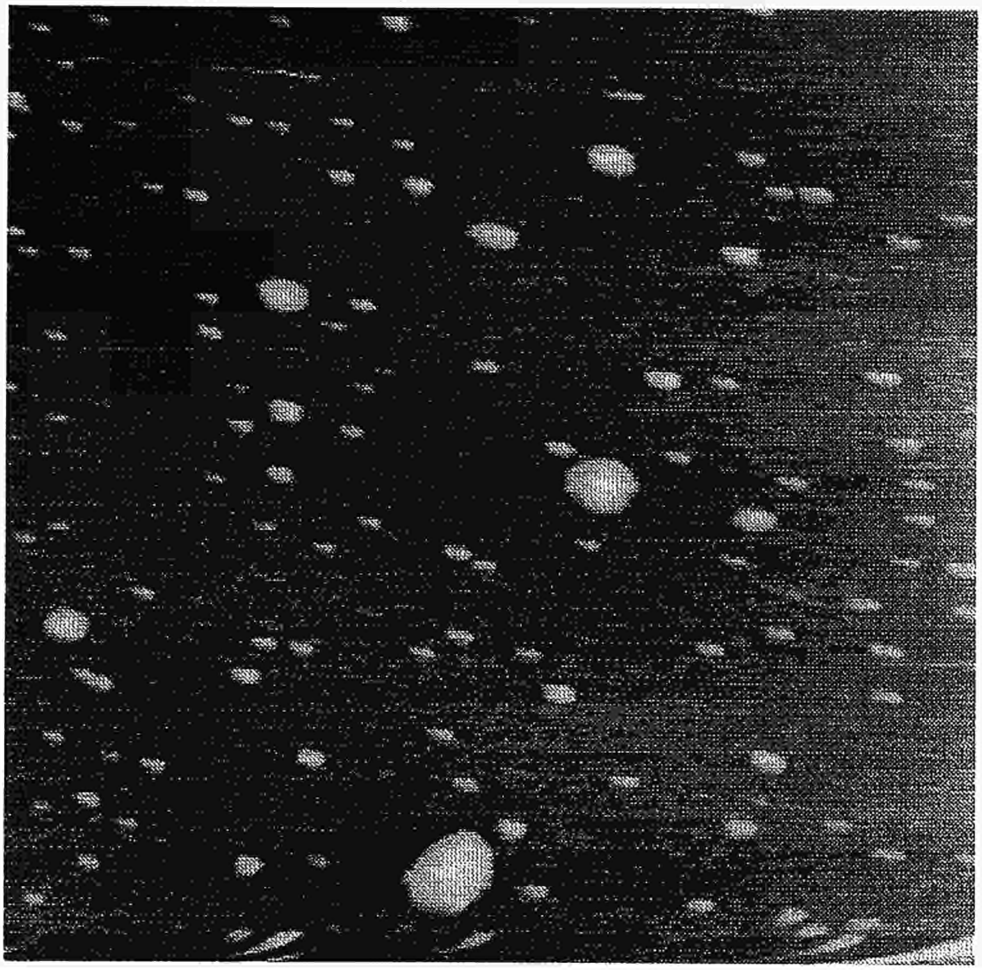

Figure 2: An Atomic Force Microscopy (AFM) scan (5×5 μm) of 14nm silica colloids adsorbed onto mica. The brightest images represent a height of about 20nm above mica surface.

C.3.7-1 Transport of radionuclides in a natural flow system at Palmottu

Contract No:	FI4W-CT95-0010 **Duration:**	1 Jan 1996 - 30 June 1999	
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A. OBJECTIVES AND SCOPE

The Palmottu U-Th ore deposit, located in a granitic host rock in southern Finland, provides an excellent location for analogue studies to perform an integrated study :

* to assess the radionuclide transport from the U-Th ore deposit along well defined groundwater pathways in the fractured crystalline rock and
* to address in Performance Assessment (PA) terms near-field spent fuel solubility/dissolution and far-field radionuclide mobilization/transportation/retardation aspects.

The project comprises two phases (phase 1 : Jan 96 to May 97 and phase 2 : June 97 to June 99) and five main work packages. Phase 1 includes the assessment of natural flow paths and PA exercise with respect to evaluate and systematise into databases existing mineralogical, hydrogeochemical and geochemical data. Phase 2 will investigate a selected flow paths with respect to water-rock interaction, to redox processes, to migration processes and testing of models and PA concepts.

B. WORK PROGRAMME

B1. Work packages phase 1

WP 1 : Understanding the natural flow system
WP 5 : PA-activities

B2. Work packages phase 2

WP 2 : Geochemical evolution of the water-rock system
WP 3 : Redox processes affecting RN mobilization
WP 4 : Migration of radionuclides
WP 5 : Conclusions relevant to repository PA (Scenarios, Near Field/Far Field)

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

Phase I of the Palmottu Natural Analogue Project commenced on January 1st, 1996, and it is scheduled to be completed by May 31, 1997. With respect to the volume of performed activities in 1996, the majority of activities belong to WP 1: Understanding the natural flow system, and a minor part to WP 5: Performance assessment exercise of Phase I. WP 1: Understanding the natural flow system comprises five major tasks which are:

Task 1.1: Up-dating the structural model of the site
Task 1.2: Hydraulic testing and interpretation
Task 1.3: Hydrochemistry as an indicator of groundwater flow
Task 1.4: Flow modelling and integrated evaluation of hydraulic and hydrogeochemical results
Task 1.5: Low-contamination drilling

The initial meeting of the project was held at Saariselkä, Finland on March 11-14 with the objectives (1) to present all available data to the consortium partners, (2) to review and up-date the Phase I plans, and (3) to produce detailed working plans and schedules for the main tasks and activities of Phase I. The internal hydrogeological assessment of the project results was performed as a series of four planning and follow-up meetings dedicated to hydrogeology, hydrogeochemistry and the integration of both of these issues. In planning and performance it was ensured that the needs of the PA issues of the project were continuously taken in account.

The studies of Phase I have progressed according to the plans and the results with respect to flow conditions at Palmottu will be published as a document for the European Commission at the end of Phase I in 1997.

Progress and results

C1. Task 1.1: Up-dating the structural model of the site

1. Objectives and main activities

The study of this task focused around the following main issues in order to produce and document a reliable hydro-structural model of the study site:

* Precise levelling data, image processing of relief maps, interpretation of structural features;
* Fracture/fracture zone identification and orientation;
* Conceptualization of the structural data;
* Conclusions and reporting: Potential fractures/fracture zones for hydraulic testing.

2. The conceptual hydro-structural model

The site-scale conceptual hydro-structural model was preliminary updated to incorporate six vertical structures (V1 to V6) and one subhorizontal structure (H1). These structures were bordered by regional-scale class III and IV structures. Based on feed-back from hydraulic tests, the following planar structures of the model were abandoned and replaced by 3-dimensional geological units: V1 and V2 were replaced by the rather extensive Palmottu brooklet fracture zone, incorporating the major uranium horizons and located between the Western and Eastern Granites. The structures V4 and V5, located at the contacts of the Western Granite, were replaced by the tabular Western Granite body. Based on cross-hole measurements and hydraulic head data, the structure V6 of the Eastern Granite was modified to have a branch (E1) striking westward toward the South-East corner of Lake Palmottu. The subhorizontal structure H1 was modified and presented as a zone with a larger width of several tens of metres, alternatively H1 could represent the bedrock from the previous H1 up to ground surface.

C2 Task 1.2: Hydraulic testing and interpretation

1. Objectives and main activities

The hydraulic testing programme targeted the identifcation of the most prominent groundwater flow paths of the study site and at characterising their hydraulic properties. The study focused on the following activities:

* Flow meter measurements from boreholes in the central part of the study site and in the new deep borehole;
* Pressure field monitoring from packed-off boreholes;
* Cross-hole interference tests with recovery monitoring;
* The application of the inverse modelling approach to pressure data and hydraulic conductivity distribution.

This task was the most extensive study programme within WP 1, mainly due to the large amount of field work and the extensive data sets produced which were to be refined by different modelling approaches. For example, the cross-hole testing programme incorporated 23 boreholes, each divided into 2 to 4 borehole sections. Consequently, up to 70 separate observation intervals were in use during the cross-hole testing campaign. In total 17 short cross-hole test were conducted.

2. Conclusive results

The main conclusions from the hydrogeological characterisation of the Palmottu site can be summarized as follows (December 1996):

1. The fluctuation of hydraulic head in the packed-off borehole sections is strongly dependent on precipitation. Generally the hydraulic head in the central area reflects the local topography, however, a few exceptions have also been seen. *E.g.* high head values are found in boreholes R302, R318, R332 and R384, cross-cutting the eastern granite. These results together with results from cross-hole tests give support for the proposed new structure E1.

2. Two main groundwater flow directions can be delineated; one northwestward towards Lake Palmottu and the other towards the southeast. Seasonal changes in precipitation do not seem to have any major influence on the persistence of this pattern.

3. The main groundwater flow occurs in the uppermost 100 m section of the bedrock where predominantly subhorizontal flow can be inferred. The cross-hole tests may indicate an apparent subhorizontal zone of greater thickness than assumed in the previous structural model but its upper and lower boundaries are uncertain. Possibly, the upper boundary of the zone extends up to the surface, which implies that the uppermost, approximately 100 m of the bedrock may be treated as a separate hydrogeological unit.

4. The assumed structures V4 and V5 in the Western Granite are likely to be hydraulically interconnected forming one hydrogeological unit. The calculated transmissivity of this unit is only slightly higher than that of the surrounding mica gneiss.

5. The structure V6 in Eastern Granite constitutes a rather isolated hydrogeological unit. This unit has lower transmissivity than the other structures and thus may act as

a hydraulic barrier to groundwater flow across the granite. Based on cross-hole tests, the presence of a cross-cutting structure E1 is possible, but the transmissivity values are quite low.

6. The results of the cross-hole tests in the assumed structure V1 were inconclusive (mixed responses) and therefore, both the geometrical interpretation of this structure and its hydraulic properties are uncertain, and, thus, the hydraulic importance of the structure is open. Interestingly, however, the propagation of pressure during pumping has an elongated response within the structure V1, indicating preferential subvertical hydraulic connections.

C3. Task 1.3: Hydrochemistry as an indicator of groundwater flow

1. Objectives and main activities

The objectives of this task was to acquire representative hydrochemical data from the site in order to support and further refine the hydro-structural model of the site. The study focused on the following activities:

* Groundwater sampling for major and trace element analyses, relevant isotopes and microbes;
* Systematisation and quality assurance of the hydrogeochemical data;
* Testing interpretations for improving conceptual hydrogeochemical models;
* Conclusions: Implications of hydrogeochemistry for groundwater flow.

2. Groundwater sampling activities

The main groundwater sampling campaign, from packed-off sections of the old exploration boreholes, took place between April and July 1996. To-date, 18 packed-off sections from 10 boreholes have been sampled and analyzed. Samples of meteoric water, surface water and groundwater from the overburden were also taken during the 1996 field season. Of the new deep borehole, samples were taken during percussion drilling (SDD) from the upper part of R385, and chemical characterisation (CCC) with the double packer system continued when drilling was finished. Groundwater samples from deeper drilled parts of R385 and R386 were taken using the SKB mobile field laboratory after the drilling campaign had been completed between July and early December 1996. In total, representative groundwater samples were received from 12 water-conducting fracture zones of the new boreholes R385 and R386.

3. Characterisation of the new deep borehole

The upper part of R385 (24-117.9 m) is characterised by dilute Ca-HCO_3 or Na-Ca-HCO_3 type waters with TDS-values below 0.2 g/l, deeper (217-222.9 m) a Na-HCO_3-Cl type was encountered with a TDS slightly exceeding 0.2 g/l. The deepest sampled section, 403-408.9 m, is characterised by Na-Cl water with TDS over 1.5 g/l, indicative of less dynamic hydrological conditions.

Based on a first interpretation of the Eh-data from boreholes R385 and R386 the following statements can be made: Both downhole and surface electrodes yield rather constant Eh-values within 5 to 10 days after the initial equilibration. All electrodes gave relatively close and strongly negative Eh values for the various zones except for the most surface-close section 94-99.9 m, where a rather extensive scattering of Eh-values could be seen. Within the lowest zone (403-408.9 m) a high sulphide concentration of 3.7 mg/l was measured.

4. Site hydrogeochemistry

According to the main chemical components three different water types could be distinguished: near-surface dilute bicarbonate groundwater and deep saline Na-SO_4

and Na-Cl-type groundwaters. Environmental isotopes (^3H, $\delta^{18}O$, $\delta^{13}C$ and ^{14}C) reveal that at least two different sources for the water itself: a glacial melt component and groundwater recharged during the 1960's. The former occurs as a considerable component in both saline water types and the latter is mainly associated with the bicarbonate water. The high SO_4/Cl and Br/Cl ratios and results of principal component analysis indicate that seawater is not a likely component of the saline waters. The data suggest that the most extreme saline Na-SO_4 and Na-Cl groundwater samples contain up to 40 % glacial meltwater.

The main mass transfer in water-rock interaction is likely connected to the carbonate system. Organic derived initial carbonate is mixed with dissolved fracture carbonate (calcite) along flowpaths of the bicarbonate waters. This is indicated by the carbon isotope data of dissolved carbonate and by thermodynamic calculations. During this progressive evolution, secondary carbonates start to control the bicarbonate content of the groundwater. Saline Na-Cl water shows methane derived ^{13}C signatures for the dissolved carbonate implying sulphate reduction which is consistent with the high measured sulphide concentrations in the deeper fracture zones. The behaviour follows the evolutionary pathway of bicarbonate except in the most saline Na-Cl samples, demonstrating the importance of solid carbonates to the Ca balance. Minor hydrolysis of rock-forming silicates and ion-exchange reactions are considered likely in order to produce cations, also supported by Sr isotope signatures of dilute groundwaters.

C4. Task 1.4: Flow modelling and integrated evaluation of hydraulic and hydrogeochemical results

1. Objectives and main activities

The target was to define one or several natural flow paths which will satisfy the needs of the forthcoming transport analogue study (Phase II). This task comprises:
* Flow modelling performed at regional, site and detailed scales;
* Integration and combined evaluation of all hydrogeological and hydrochemical information with respect to a transport analogue study.

2. Regional flow

Based on a three-dimensional modelling approach and the finite element method a regional flow modelling was performed indicating that the predominant regional groundwater flow is toward the southeastern corner of the Palmottu regional area, *i.e.* off from Lake Palmottu toward the extensive valley some 1.5 km south from it. A comparison of cut planes representing different depths shows that the regional groundwater flow is predominantly horizontal in character. The volumentary fluxes through the horizontal cut planes decrease strongly so that infiltration below 150 m is only 14 % of the infiltration through the ground surface.

3. Site-scale flow

The site-scale flow model comprises a bedrock volume of 0.2 km^2 * 0.4 km in the close vicinity of Lake Palmottu. The most dominant site-scale structures were incorporated into the model: V1 - V6 and H1. The main inlet of regional groundwater flow is from the western part of the area, predominantly along regional East-West striking structures. The Palmutunoja Brooklet fracture zone is the main outlet channel from the site-scale area, and it is characterised by horizontal flow. Calculations indicate that flow flux along this structure is only 1 % of the flux of surface water flow along the Palmottu Brooklet.

The infiltration rate along the fracture zones V4 and V5, both connected to the tabular Western Granite, is quite high. The flow decreases steeply with depth, and at 150 m it is only 3% of the flow at the 30 m level. Approximately one third of the flow is trending toward the drainage area (Lake Palmottu), whereas two thirds are trending toward the regional flow field. The flow toward the drainage area is mainly vertical with an upward trend below Lake Palmottu, whereas the flow in the south-western part of the zone is predominantly horizontal after an initial infiltration in the bedrock.

A hydraulic head maximum is located at the Palmottu Brooklet fracture zone around drilling profile 336. Th head maxima acts as a local water divide separating flow toward the northeast (Lake Palmottu) and toward the southwest to the topographically lower valley region.

C5. Task 1.5: Low-contamination drilling

1. Objectives and main activities

The objectives were to improve and test drilling techniques which would allow a sampling of representative formation groundwaters before serious contamination by flushing (or open-hole effects) would have taken place. The following main activities were involved:

* Evaluation of available low-contamination drilling techniques with respect to representative hydrochemical sampling and eventual modification of techniques for the present purpose;
* Sampling interlinked with drilling of the deep borehole;
* Petrography and fracture characterisation during and subsequent to drilling.

2. Fracture characterisation based on the deep borehole

In order to characterise fracture zones suitable for hydrogeochemical sampling, TV imaging and flow meter measurements of the deep borehole were conducted. The total number of fractures observed by TV-imaging was 527, *i.e.* 48 % of those fractures observed manually during drill-core logging (approximately 1100). Based on manual logging, the number of open fractures was 83, and the majority of these fractures seem to be subhorizontal or gently dipping. When presented statistically on a stereographic projection, the maxima of open fracture planes focus around 330°/10°, 190°/23°, 80°/34° and 220°/82°. Three of these figures indicate subhorizontal planes and one a subvertical plane, which coincides with the average local schistosity. Only a few open fracture zones with good hydraulic yield were observed during the flow meter measurements: 90.94, 96.32, 115.72, 222.80, 406.30 and 407.28 m in borehole R385, and 30.20, 35.00 and 80.00 m in borehole R386.

The flow meter measurements of the deep borehole support the view of a relatively permeable upper part of the bedrock down to approximately 150 m, whereas in deeper parts water-conducting fractures are quite sparse.

C6. Workpackage 5: Performance assessment exercise of Phase I

An important part of the Palmottu project is the close integration of the natural analogue study with performance assessment (PA) of repositories for radioactive waste. To ensure that this integration takes place, the project includes a special part in Phase I to deal with PA. The objectives for Phase I activities consist of the following three parts:

* Incorporation of PA expertise;

* Establishing and evaluating of mineralogical, geochemical and hydrogeochemical data sets based on PA needs;
* Planning of Phase II.

The main part of the work in WP 5 is planned to be performed in Phase II. The planning of Phase II activities requires a reasonable understanding of the hydrological, geological and chemical system at the site. In order to acquire sufficient understanding and to ensure close integration between PA and the geoscientific evaluation, PA-experts have contributed throughout the planning and reviewing activities of Phase I.

Based on preliminary information of the natural flow system at Palmottu the following main PA issues will be of relevance for WP 5 in Phase II:

1. Conclusions regarding the mobilisation and transport of uranium in the "Eastern Granite";
2. Conclusions regarding mechanisms for dissolution and alteration of uraninite in the mineralisation zone in the central part of the site;
3. Conclusions regarding different mechanisms for retardation of dissolved uranium in the mineralisation zone in the central part of the site. The mechanisms could include for example sorption, matrix diffusion, precipitation and mineralisation;
4. Conclusions relevant to the development of geochemical evolution scenarios, in particular scenarios related to glaciation.

The detailed planning of Phase II will be completed in early 1997.

C.3.7-2 Natural analogue of the thermo-hydro-chemical and thermo-hydro-mechanical response of clay barriers

Contract No: FI4W-CT95-0014 **Duration:** 1 Jan 1996 - 30 June 1998
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A. OBJECTIVES AND SCOPE

The overall objective of the project is to investigate one or more natural analogues of the THM and THCM changes undergone by heated clay formations and to determine how applicable the field data are for studying PA issues of clay barriers. The focus of the project is to investigate in detail a suitable natural analogue of the behaviour of clay under increased thermal gradients from the viewpoint of geomechanical and chemico-mineralogical alterations with the aim of understanding the chemical, hydraulic and mechanical changes related to the rise in temperature, the coupling of the changes and their time scales.

The work will address the current state of conceptual model by considering the clay zone where the temperatures have been in the range actually expected in a repository and in the surrounding host rock. Attention will be given to three potential analogue sites : Orciatico (IT), Island of Skye (UK) and Col de Perthus (FR).

B. WORK PROGRAMME

B1. Preliminary identification of PA issues (All partners)

This work package will attempt to identify issues of concern to PA caused by thermal alteration of clays and provide a base line and structure of the study.

B2. Review of literature of data and models related to the issue (All partners)

The review will critically examine the various changes due to the rise in temperature and split them into four areas : (i) THM-changes (ISMES), (ii) chemical changes (BGS), (iii) mineralogical changes (IPSN) and (iv) gas permeability changes (Univ. Rome).

B3. Consideration of the required characteristics of a natural analogue (All partners)

This work page will identify features or characteristics which are needed in an analogue in order to unambiguously investigate particular processes or issues identified in WP 1 and 2.

B4. Assessment of potential analogue sites (All partners)

Available information will be reviewed to assess the features of potential analogue sites in France, Italy and UK.

B5. Site investigation and evaluations (All partners)

This task will be planned and performed in close contact with the modelers and should maximize the chance that the obtained data are relevant to PA issues. It will be tried to establish links between the thermal, hydraulic and mechanical properties of the clay. The work package is subdivided in several tasks.

C. Progress of work and results obtained

Summary of main issues

The project inception meeting was held on January 26, 1996. Based on decisions taken at the meeting, the partners proceeded to work on a number of preliminary issues. In particular ISMES was assigned the task of collecting and reviewing existing knowledge about the Orciatico intrusion and of identifying suitable locations for the subsequent sampling and detailed investigation of clay properties and thermal alterations (activities within work packages 4 & 5). Somewhat similar tasks were assigned also to BGS for the dykes on the Isle of Skye and to IPSN for the dykes that intersect Toarcian sediments in the region of the Tournemire tunnel (w.ps. 4 & 5). Rome University (RU) was expected to review the literature about soil gas data in thermally altered clays and in particular to investigate the potential of soil gas surveying to clarify some relevant features of the Orciatico analogue (w.ps. 4 & 5). Regarding the theoretical aspects of the project, ISMES was expected to address the status of modelling thermo-hydro-chemo-mechanical processes in clays (w.ps. 1 & 2), BGS to assess the implications of natural analogue studies for performance assessment (PA) (w.ps. 1 & 2) and IPSN to performe some parametric thermal calculations of the temperature distribution in clays intruded by magmatic materials (w.p. 4).

During 1996 progress was made on all issues. In addition RU has performed a geoelectrical survey of the Orciatico intrusion to clarify the underground shape of the intrusive body and the contact with the surrounding clay (w.p. 5). At the time of the geophysical survey, ISMES has carried out a preliminary sampling, taking shallow clay samples along an alignment at 90° with the clay-intrusion contact. The purpose of the preliminary sampling is to obtain empirical inputs to be used in reconstructing the paleotemperatures experienced by the clays (w.p. 5).

C.1 Preliminary identification of PA issues

A review of how natural analogues can be used in performance assessment (first draft started, completion by end of February 1997) is being performed by BGS.

A database of literature relevant to the project is being compiled (a large amount of references have now been obtained, and a first release of an EndNote database is expected in mid-1997, though this will grow as the project proceeds).

C.2 Review of literature on data and models related to the issue

A good understanding of clay behaviour is important in defining the best testing programme for clay samples, in interpreting the results of both laboratory determinations and field observations and in developing adequate conceptual models of clay response to heating.

A state-of-the-art report on the mechanical behaviour of clays and on its coupling to hydraulic, thermal and chemical phenomena and on their mathematical modelling has been compiled by ISMES. Experimental findings regarding thermo-mechanical behaviour under representative repository conditions are briefly discussed, together with a critical review of models of thermo-plasticity of clays and of applicability of Darcy law to water flow in saturated clays. Relevance of natural analogue studies to long-term performance of repositories for heat-generating waste is also addressed.

In many repository designs, long-term waste isolation depends on the longevity of clay-rich barriers, either as engineered elements of the repository or as part of the geosphere. Heating by radioactive decay in the waste may be the most important factor with the capability of causing adverse and irreversible alterations of the clays.

Most experience on thermal alterations of clays is derived from laboratory experiments and regards short-term effects. The thermal increase of pore water pressure may cause a significant enhancement of water flow away from the heat sources. This is expected to happen relatively soon after repository closure and, therefore, should have no direct impact on radionuclide migration, unless waste containers were to fail very early. In addition, increase of pore water pressure may cause a drastic decrease of effective stress, causing mechanical failure of the clay, particularly during the cooling phase. A secondary effect of mechanical failure may be an increased hydraulic conductivity.

As far as long-term effects are concerned, these are linked to irreversible changes of clay structure and mineralogy and to modifications of the transport properties. Both temperature rise and high pore water pressure may irreversibly change hydraulic conductivity and diffusivity either through fracturing (e.g., caused by pore pressure rise) or through illitization of smectite and other physico-chemical changes.

Observations and analyses at natural analogue sites offer a number of potential advantages, such as: duration of heating comparable with a repository, representative dimensions of heated clay (for sizeable magmatic intrusions), realistic conditions from the viewpoints of boundary conditions and formation heterogeneity. There are also problems associated with the interpretation of natural analogue data, for example regarding: the temperature of intrusion, that is way too high and requires careful assessment of the zone where temperatures have been analogous to a waste repository; the mechanical effects of intrusion and the resulting strain; the effects of the post-intrusion geological history, such as hydrothermal phenomena, weathering and unloading by erosion of the overburden.

Keeping in mind the preceding considerations, modelling capabilities and experimentally determined thermo-hydro-mechanical properties of clays have been reviewed, paying particular attention to their relevance to the present project.

At least one fully operational computer code, based on a basic thermo-plastic Cam-clay model (CCTeru), is available for immediate use and could serve for the simulation of short-term effects. This model cannot handle long-term variations.

There are also a few models for variable permeability or sediment dewatering which could be used as a guide to develop an acceptable model or to expand the thermo-plastic model mentioned above.

An important step of analogue modelling would be the coupling of hydro-mechanical models with models of chemical reactions and physico-chemical processes, such as dehydration, precipitation of silica or carbonates and metamorphic alterations. Resulting models should include the constitutive relationships of coupling between the chemical reactions and their effects, and the mechanical properties and permeability.

In conclusion, existing models permit to simulate many short-term phenomena and the long-term process of fracturing, due to cooling. Several long-term phenomena can be modelled by modifying the coupled thermo-plastic model or by using a few different and somewhat odd models. Initially, simplified simulations could be performed to assist, hopefully, in sampling and analyzing the site. The potential exists for the successful development of a coupled model which includes the main long-term phenomena.

A draft report addressing this work package has been completed during 1996; editing will be finalized by March 1997. The report includes a comprehensive list of references that will be provided to BGS for merging with the database mentioned under C.1.

C.3 Consideration of the required characteristics of a natural analogue

Desirable characteristics of a thermo mechanical natural analogue have been discussed and identified as follows:

- relatively recent event to limit subsequent alterations that might confuse interpretation;
- system initially at low temperature and pressure to avoid interpretation difficulties due to ambiguity of alteration causing events;
- single intrusive event;
- the presence of smectite minerals would allow to obtain information relevant to assess the response of buffer/backfill materials;
- content of sufficient amounts of organic materials and suitable minerals for indipendent determination of paleotemperatures;
- no disturbance of the analogue zone (paleotemperatures between 100°C and 150°C) by magmatic fluids.

Eventually agreement was reached on the fact that no candidate analogue met all requirements and that the Orciatico intrusion appeared to be the most favourable one.

C.4 Assessment of potential analogue sites

During 1996 three sites have been reviewed: Isle of Skye in UK, Col de Perthus in France and Orciatico in Italy. Project participants recognized that all three sites presented interesting aspects and they all deserved some effort.

As a preliminary activity to any site evaluation, IPSN and ERM performed some simplified, parametric thermal calculations of the effects of magmatic intrusions in clays. With the simple assumptions that have been used, such as intrusion in the shape of a cylinder and conduction as the only heat transfer mechanism, the extension of the heated zone depends linearly on the diameter of the intrusion. In particular the temperature of 100°C would be reached in the clay at a distance from the contact roughly equal to the diameter of the cylinder, as shown in the table below.

Actual site investigations are described in next section.

Table 1 - Main results of the IPSN/ERM thermal calculations

Diameter of the dyke	Approximate distance where 100°C peak has been calculated	Corresponding time to reach 100°C
1 m	1 m (possibly ~3 m)	<1 year (possibly ~1 year)
10 m	10 m (possibly ~35 m)	1÷2 yrs (possibly ~10÷20 yrs)
100 m	100 m (possibly ~300 m)	100 yrs (possibly ~ ≤1000 yrs)
1000 m	1000 m (possibly ~2000 m)	10,000 yrs (possibly ~ ≤100,000 yrs)

C.5 Site investigations and evaluations

Isle of Skye

The British Geological Survey has been investigating thermally-altered mudrocks on the Isle of Skye, Scotland. Jurassic sediments outcrop over various parts of Skye and surrounding islands, though are best exposed along the coast in northern Skye in a region known as the "Trotternish Peninsular". The Jurassic rocks have been intruded by Tertiary dykes and are overlain by Tertiary lavas.

Two field visits were organised to study the Jurassic rocks of northern Skye. The first visit in March 1996 was a preliminary one, to investigate sites previously identified by literature review, and to identify new sites. Five potential sites were eventually identified and samples taken to constrain their suitability for more detailed study. The sites were:

- Potential analogue site 1 - L\'98 n Ostatoin (NG 4076 7268).
- Potential analogue site 2 - L\'9d b Score, below Score SW (NG 3994 7256).
- Potential analogue site 3 - L\'9d b Score, below Score NE (NG 4002 7265).
- Potential analogue site 4 - L\'9d b Score, base of cliff below Bealach Iochdarach (NG 4084 7345).
- Potential analogue site 5 - L\'9d b Score, below Duntulm Castle (NG 4105 7401).

A total of 31 rock samples were taken from the first field visit, and the following analytical techniques were applied:

- Bulk X-ray diffraction (completed).
- Clay X-ray diffraction (completed).
- Surface area measurement (completed).
- Backscattered SEM on impregnated samples (nearly completed).

The X-ray diffraction work was completed quickly in order to provide information for the second field visit. The above work will be written up in two BGS reports, one concerning the background to the visit and the sampling itself (first draft already completed), and the other concerning the mineralogical observations (first draft ~80% completed). It is hoped to have both of these reports completed by the end of March 1997.

The second field visit in July 1996 was combined with the second progress meeting of the project. This gave other project participants an opportunity to visit the exposures on the Isle of Skye and discuss the preliminary results from the initial sampling. In summary, the following was decided about the five potential analogue sites on the Isle of Skye:

- Potential analogue site 1 - Analysis of samples collected during the preliminary visit indicated that some of the sediments had a smectite content of up to 45%. However, XRD analysis also revealed the presence of significant grossular garnet. This could only have formed at temperatures well in excess of 200 °C (maximum limit of interest to this study). Our conclusions were that this site was not suitable as an analogue due to its complexity and thermal history.
- Potential analogue site 2 - Analysis of samples collected during the initial visit indicated that the sediments contained between 13% and 22% smectite. There appeared to be no igneous bodies in the immediate vicinity. Our conclusions were that this site was not suitable as an analogue because no heat source was present.
- Potential analogue site 3 - Analysis of samples collected during the preliminary visit indicated that the sediments contained between 12% and 30% smectite. A "knife-edge" contact was observed between the sediments and a sill, but this was parallel to bedding. Our conclusions were that this site was not suitable as an analogue because of its small size and because the contact with the heat source was parallel to the bedding.
- Potential analogue site 4 - This medium sized cliff section showed a relatively simple relationship between a near vertical dyke about 3 m wide and horizontal Jurassic sediments. Most of the sediments appeared to be of shelly limestones and sandstones, however there were several mudstones within the sequence. Analysis of samples collected during the preliminary visit indicated that these mudstones contained between 14% and 26% smectite. Field observations revealed subtle differences in colour and physical properties of one particular mudstone away from the contact with the dyke. These differences were over a distance scale similar to that predicted by thermal modelling. Our conclusions were that this

site was the most suitable of those visited as an analogue, mainly because of its geological simplicity and apparent thermal aureole.

- Potential analogue site 5 - Sediments adjacent to a large sill were highly baked and converted to hornfels. Analysis of samples collected during the preliminary visit indicated that the sediments contained about 28% smectite only 10 m away from the contact, but that this was virtually all destroyed in the hornfels zone. Our conclusions were that this site was not suitable as an analogue because of the possible complex shape of the sill, and because the site could only be accessed during low tide.

As a result of the above observations several samples were taken (about 30), but were concentrated on site 4. Samples of mudstone were carefully taken at increasing distances from the contact with the dyke and was repeated on both sides of the dyke. Samples of adjacent shelly limestone were also taken to provide material for vitrinite reflectance studies.

The following analytical techniques have been or are being applied:

- Bulk X-ray fluorescence (completed).
- Clay X-ray diffraction (completion expected mid-1997).
- Surface area measurement (completed).
- Backscattered SEM on impregnated samples (completion expected mid-1997).
- Hardness testing (will be investigated by mid-1997, but may require measurements to be performed on *in situ* samples.
- Vitrinite reflectance (completion expected mid-1997).
- Crystallinity studies (completion expected mid-1997).

Although Site 4 appeared the most suitable, preliminary results appear to indicate that significant "back reaction" has occurred (i.e. smectite has formed after the intrusive event) and that smectite content actually increases at the mudstone/dyke contact. The results from the other techniques will help constrain this further.

Six samples from the Orciatico analogue site were received in December 1996. These have now been digested to reveal their solid organic material, and are undergoing preparation for vitrinite reflectance studies. The results of this work is expected by mid-February 1997.

Col de Perthus

In the past years IPSN has studied a number of dykes that intrude Toarcian sediments in the area near the Tournemire tunnel (Aveyron, south of Massif Central, France). Existing data provide a comprehensive picture of the chemical and mineralogical properties of both intrusive body and thermally altered argillaceous sediments. The literature produced by the previous studies has been collected and reviewed.

The structural situation of these dykes is similar to the one observed for the dykes on the Isle of Skye, with the possible significant difference that the French dykes are even thinner than the Scottish ones. This implies that the heated zone is quite thin and that the heating transient has lasted a short time. As a consequence it might not be feasible to identify reliably the analogue zone, that is the section of mudrock where paleotemperatures have reached between 100 and 150°C. Nevertheless the possibility of obtaining some data, for a significantly different argillaceous material, that could be compared with the Scottish and Italian ones, has been judged of sufficient interest to justify an additional effor on one of the French dykes. In conclusion the dyke of Col de Perthus, which is the thickest one, has been selected for further study.

Orciatico

The Orciatico intrusion has been studied in the 80s by ENEA and the University of Pisa (Ref. [1]). The emphasis of those investigations has been on the chemical and mineralogical alterations of the heated clays. During the previous study 12 shallow boreholes were drilled (maximum depth about 20 m) crossing the contact zone in only four cases. The study of the previous reports and site visits have permitted to determine that the contact is characterized by significant variability in respect to both orientation and thickness of the thermally metamorphosized contact zone. This situation has been interpreted as an indication that heat transfer from the magma to the surrounding clay has not taken place isotropically. This would appear to imply that conduction has not been the only heat transfer process at work. The variation of thickness of the layer of thermally altered clay (thermantite), that usually separates the magmatic rock from the apparently unaltered clay, seems to support a non-homogeneous heat transfer.

If we assume that, at Orciatico, at least locally, heat transfer may have been disturbed by fluid flow and/or degassing near the boundary of the intrusion (a logical hypothesis), it becomes very difficult to choose the proper sampling location, considering also that results of thermal calculations, based on a somewhat idealised model, place the 100^{O}C isotherm, for an intrusive body 1000 m across, at a significant distance from the intrusion boundary. Quite different results could be obtained assuming a more realistic model of heat transfer and using different shape and boundary conditions for the magmatic body.

As a matter of fact some geoelectrical soundings, performed on behalf of the University of Rome during December 1996, have confirmed that the Orciatico intrusion consists of a laccolith, as already stated in the literature (Ref. [2]).

Figure 1 shows a cross section of the intrusion and surrounding sediments along an alignment roughly orthogonal to the surface contact between intrusion and clays, near the south-eastern end of the outcrop (Figure 2). Along the same alignment (cross section D-D') ISMES has taken six shallow clay samples. The samples, taken in double, have been sent to BGS and IPSN, where they will be analysed with the main purpose of deriving the paleotemperatures experienced by the materials.

Empirical determinations of paleotemperatures are believed to be essential for the determination of the location where clays have been heated to temperatures in the 100°C to 150°C range. Actual data are hoped to enable to overcome the great uncertainty associated with modelling results in such complex contact geometry.

References

[1] ENEA,1983. Final Report of Contract 152-80-7 WASI: Studi sulle modificazioni delle proprietà subite dall'argilla per l'azione termometamorfosante provocata dall'intrusione di un ammasso subvulcanico (Orciatico).

[2] L. Trevisan & G. Giglia, 1978. Introduzione alla geologia. Pacini editore.

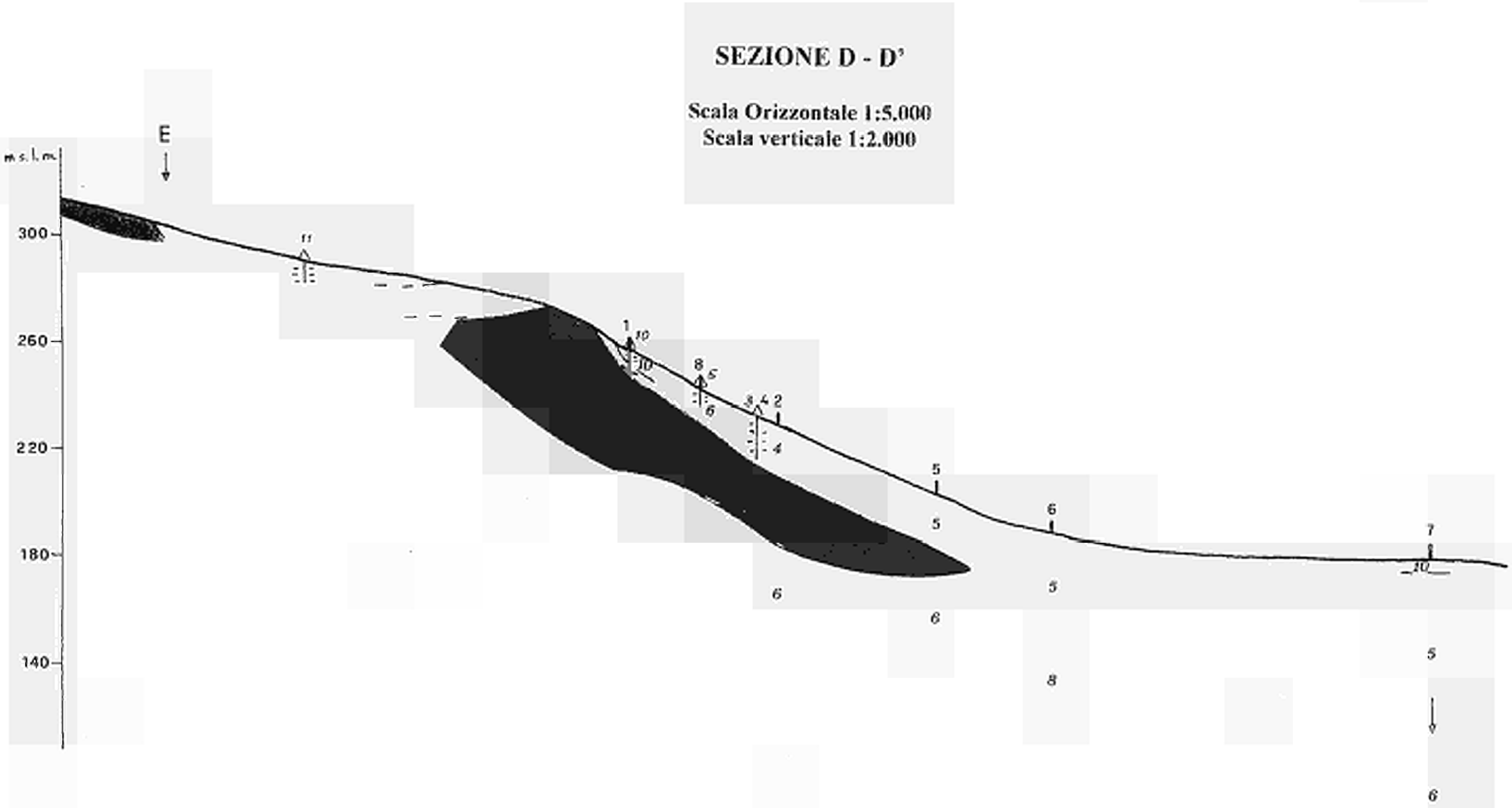

Figure 1: Cross section D-D' on which six shallow clay samples have been taken at points 1, 2, 5, 6, 7 and 8. Yellow indicates clay, red indicates high resistivity material, presumibly intrusive rock.

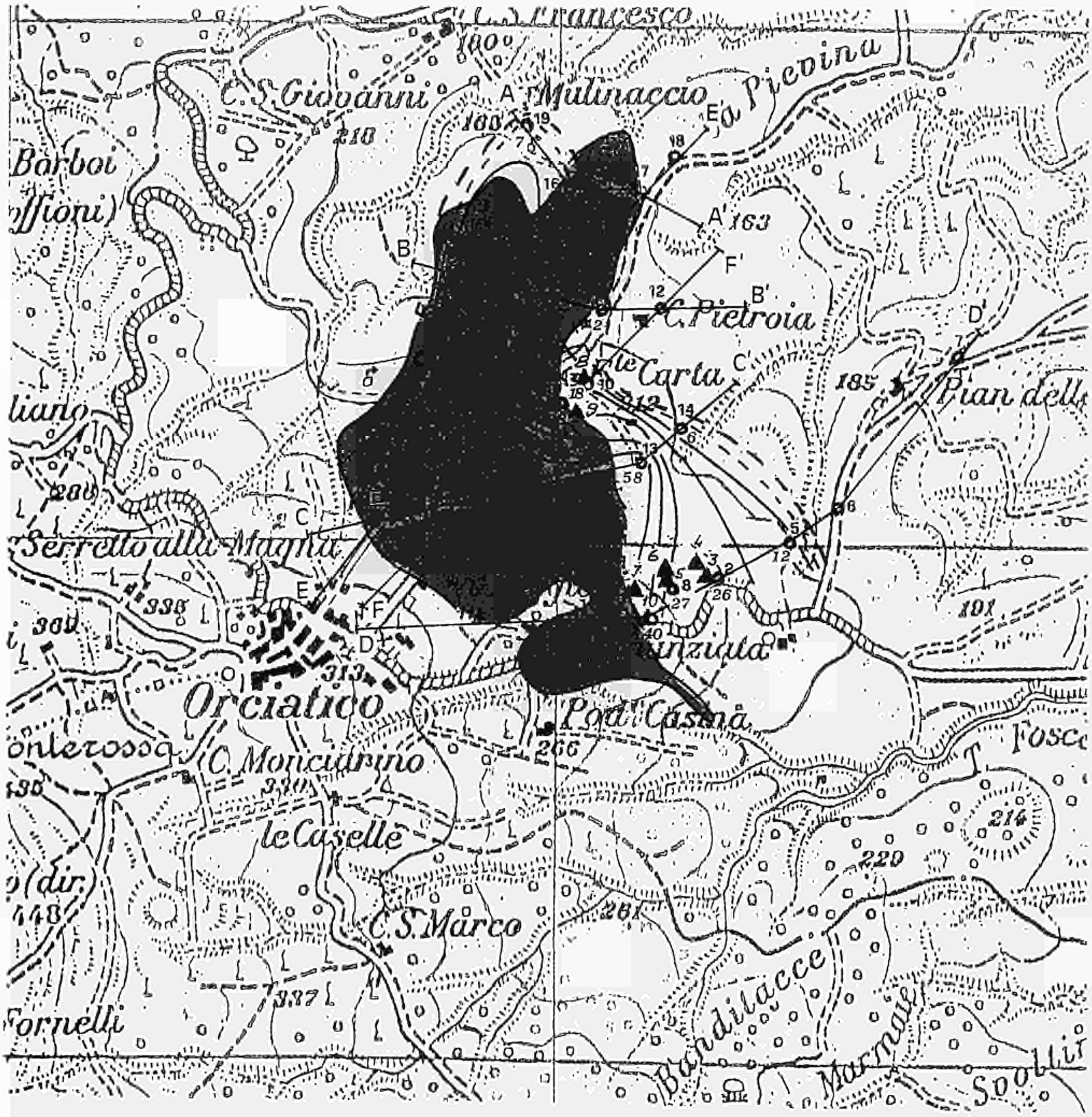

Figure 2: Map of thickness of high resistivity layer. It shows also location of cross sections, including section D-D', shown on Figure 1.

Scala 1 :10.000

Legenda

7 Sondaggio elettrico e suo numero

5 Spessore in m del complesso resistente

30 Curva di isospessore e suo valore in m

Limite del complesso resistente

2 Sondaggio geognostico e suo numero

Area di affioramento delle selagiti e termantiti

C.3.7-3 OKLO Natural Analogue phase II : Behaviour of nuclear reaction products in a natural environment

Contract No: FI4W-CT96-0020 **Duration:** 1 June 1996 - 31 May 1999
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A. OBJECTIVES AND SCOPE

The OKLO Natural Analogue project will :

- focus on a quantitative assessment of processes of radionuclide migration/retention within the Oklo basin and relate these processes to analogous parts of a deep repository system for high level radioactive waste;
- provide suitable data for repository PA-models by considering near-field (source term), far-field (geosphere) and overall PA aspects;
- test models of radionuclide transport in the geosphere against data from a well characterized natural system;
- improve knowledge on the confinement capacity and mechanisms of geological material.

The scientific work will be developed on two different locations of the OKLO basin (SE Gabon) : (i) a deep sited reaction zone (Oklo/Okélobondo) and (ii) one shallow reaction zone (Bangombé).

B. WORK PROGRAMME

B1. Geological History (CEA,SKB,CNRS, ANDRA, ENRESA)
Task 1 : Site characterization
Task 2 : Reaction zone characterization (Bagombé)
Task 3 : Fractures

B2. Location of Fission Products (SKB, ENRESA, CEA, ANDRA, CNRS, Univ. Oviedo, CNRS)
Task 4 : Source term
Task 5 : Interface near-fied to far-field
Task 6 : Far-field

B3. Radionuclide Transport-Hydrogeology (Univ. Oviedo,SKB/Conterra, CEA, ENRESA, UPC)
Task 7 : Hydrology
Task 8 : Hydrochemistry
Task 9 : Flow and transport properties

B4. Modelling of Radionuclide Transport (CEA, ARMINES, SKB, ENRESA, CNRS, UPC, QuantiSci)
Task 10: Geochemical processes
Task 11: Hydrodynamics and solute transport

C. Progress of work and results obtained

. Summary of main issues

The first half year of the project was mainly devoted to the field missions to Gabon. The missions were organized from August to November 1996 to collect water and rock samples at Oklo and Bagombé sites and to complete the drilling programme at Bagombé (Figure 1).

The necessary water and rock sampling to start the project scientific activities has been completed and samples have been distributed to each participant.

. Progress and results

1. Field missions

A preparatory mission went to Gabon in June to coordinate the project field activities with the COMUF activities (COMUF: mining company in charge of Oklo and Bagombé U ore exploitation). A problematic issue was the Bagombé field work. The COMUF plans for Bagombé were to mine out the entire ore before end of 1996 with a preparatory uncapping starting in July. These plans would have severely affected the implementation of field experiments planned at Bagombé [1]. On CEA request, the COMUF work was been post poned until the end of 1996. this problem of the site preservation has led to the decision to complete the sampling and drilling programme as soon as possible before the end of 1996.

B3, all tasks: a groundwater sampling campaign has been organized at the Bagombé site before the drilling campaign, from 27 August to 14 September 1996 [2, 3]. the same mission has sampled boreholes and drips down the Oklo-Okélobondo mine. 25 water samples have been collected for chemical and isotopic analyses (Table I).

B1 and B2, all tasks: The drilling programme at Bagombé initially planned five new boreholes of twenty meters each. The preliminary geological study on site of the first set of cores led to an extention of the drilling programme. In total eight boreholes were drilled during the period from 29 August to 14 October 1996 [4]. The total lenght of core recovered is around 120 meters. The cores have been produced with a triple correr which has enable the preservation of the fractures infills minerals. This drilling programme has already permitted to better localize the Bagombé reactor zone (Figure 2). All cores have been shipped to Strasbourg and were strored within CNRS premises for logging before distribution. Several rock sampling have been undertaken during field tour either at Bagombé or at the Oklo open pit as well as down the Oklo-Okélobondo mine and at the COMUF cores-room in Mounana. Samples were selected for fracture minerals studies [5, 6].

2.Other activities

B1 and B2 : A library of the Oklo samples collected since the discovery of the phenomenon has been set up at the CEA Nuclear Center of Cadarache. The total amount of samples now available is around five tons of rock material from different reaction zones and different geological horizons. A document listing for each sample its reference and origin, the sampling date, the type and the amount of material available and the total radioactivity, has been produced by the CEA/DCC. 7033 samples have been recorded and included in this document. The sample distribution from the Cadarache Library is organised by the project coordinator and following the safety rules for the transportation of natural radioactive material when necessary.

B1, Task 1: The synthesis on existing data for the geology of the Franceville basin is progress.

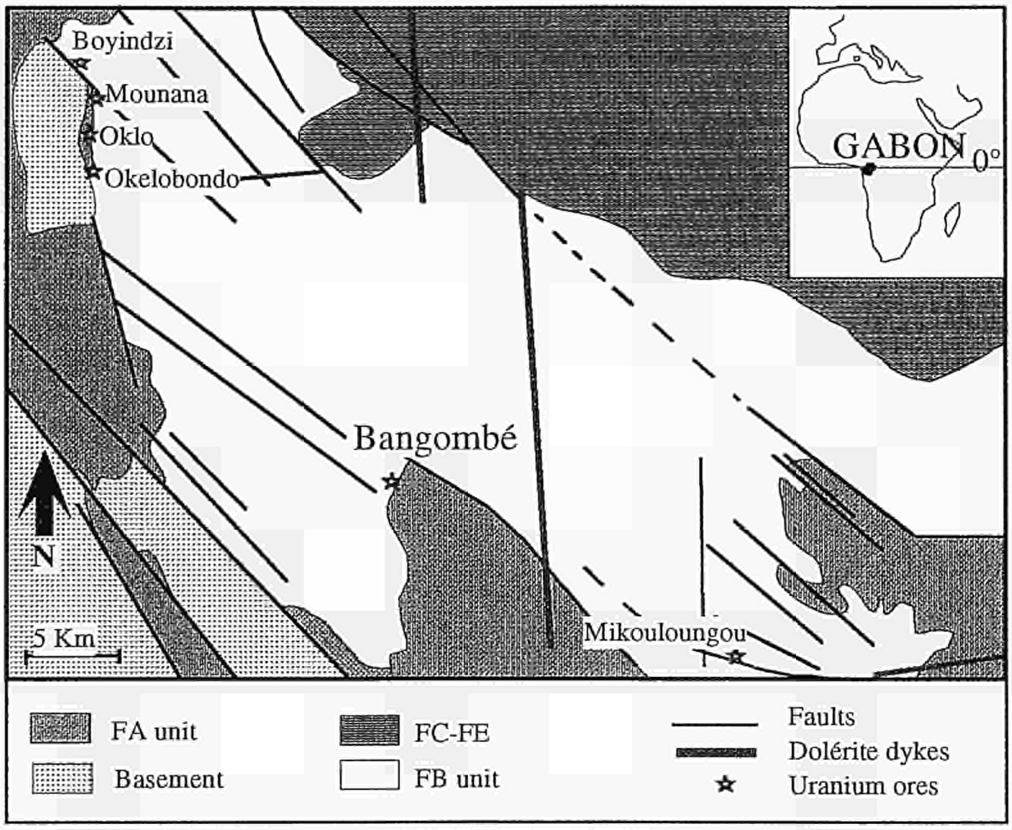

Figure 1 : Geological map of the Franceville basin showing the location of Oklo and Bagombé[4]

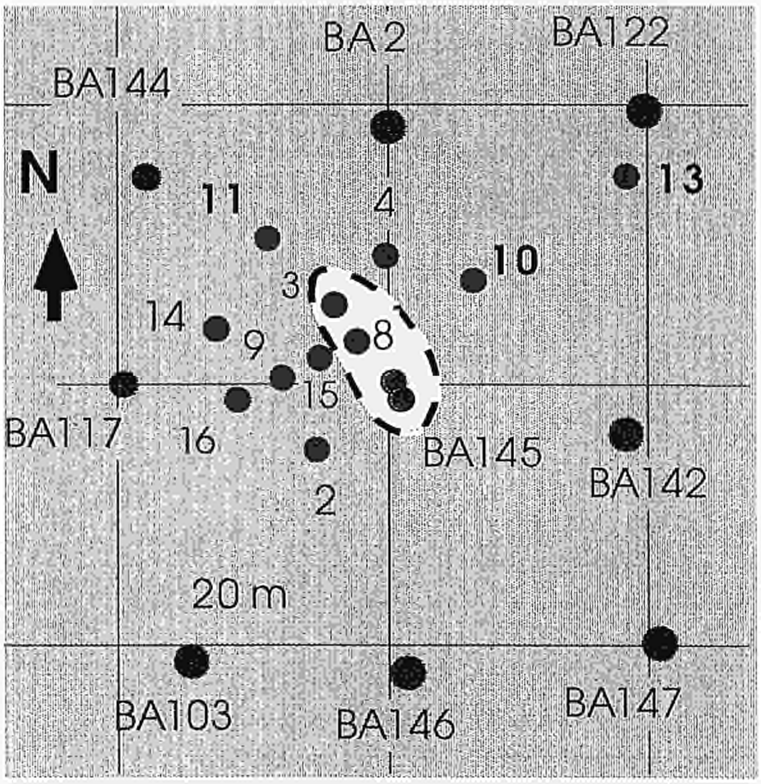

Figure 2 : Location of COMUF boreholes (blue dots) and " Oklo-Natural Analogue" BAX boreholes (red dots); possible shape and contours of the reactor (yellow; dashed)

References

[1] LOUVAT, D, CEA Travel Report SESD 96-522 (1996). Rapport de mission au Gabon, juin1996.

[2] LOUVAT, D, BELTRITTI, A and CASTELLIER E, Travel Report SESD 96-580 (1996). Rapport de mission au Gabon, août1996.

[3] LEDOUX, E, ENSPM-CIG Report (1997). Echantillonnage des eaux à Okélobondo et Bagombé. Rapport Oklo-Phase II, 8p.

[4] BROS, R and GAUTHIER-LAFAYE, F, CNRS Travel Report (1996). Réalisation des forages de Bagombé pour le programme Oklo-Analogue Naturel Phase II, 45p.

[5] MATHIEU, R and CUNNEY, M, CREGU Travel Report (1996). Rapport d'échantillonnage pour le programme Oklo-Analogue Naturel Phase II, 37p.

[6] RAIMBAULT, L, ENSPM Technical Report LHM/RD/96/57 (1996). Rapport d'échantillonnage pour le programme Oklo-Analogue Naturel Phase II, 26p.

TABLE I

sample reference	**sampling date**	**Chemist. majors & traces**	**water isotopes**	**dissol.U isotopes**	**^{14}C DIC & DOC**	**nobles gases**	**dissol. Rare Earth**	**dissol. ^{36}Cl & ^{129}I**	**dissol. F.P.**	**colloids & Org.Mat**
Bagombé										
BAX01	03-sep-96	X	X	X	X	X		X		X
BAX02	31-aoû-96	X	X	X		X	X	X	X	X
BAX03	31-aoû-96	X	X	X		X	X	X	X	X
BAX04	01-sep-96	X	X	X		X	X	X	X	X
BAX05	02-sep-96	X	X	X		X				X
BAX06	03-sep-96	X	X	X		X				X
BAX07	30-aoû-96	X	X	X		X	X	X	X	X
Okélo Surface										
OK158	05-sep-96	X	X	X	X	X		X		X
OK168	05-sep-96	X	X	X	X	X		X		X
OK199bis	05-sep-96	X	X	X	X	X		X		X
Okélo mine										
OK0	05-sep-96	X	X	X	X					
OK2	12-sep-96	X	X	X						
OKH2	09-sep-96	X	X	X	X			X		
OKH3	10-sep-96	X	X	X	X			X		
SA31	09-sep-96	X	X	X						
SA32	09-sep-96	X	X	X						
SA34	09-sep-96	X	X	X	X	X		X		
SA36	09-sep-96	X	X	X						
SA954	11-sep-96	X	X	X						
SA992	11-sep-96	X	X	X						
SA995	11-sep-96	X	X	X						
SA996	11-sep-96	X	X	X						
SA997	11-sep-96	X	X	X						

C.4.1-1 Dismantling of PWR, BWR and VVER reactor pressure vessels and internals

Contract No.:	FI4D-CT95-0001	**Duration:**	1 Jan. 1996 - 31 July 1999
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Partners:	**KRB-A Gundremmingen/DE, EWN Lubmin/DE**		

A. OBJECTIVES AND SCOPE

The main objective of this project is to demonstrate the feasibility of Reactor Pressure Vessel (RPV) dismantling at reasonable costs and dose commitments, the comparison of different dismantling techniques and the execution of this major operation on the BR3 Pressurized Water Reactor in Mol and the KRB-A Boiling Water Reactor in Gundremmingen, being the most commercialized reactor types in the EU Member States. Moreover, the project deals with the future dismantling of the EWN VVER reactors in Greifswald.

Specific data will be gained on real costs, dose commitments and waste generation. This data generated on actual components will then be made available for the European Data Bases EC-DB-TOOL and EC-DB-COST.

The project will be carried out by three organisations: KRB and EWN in Germany and SCK/CEN in Belgium. Nevertheless, like in former projects, other European contractors could be associated to the project.

B. WORK PROGRAMME

B.1. Project coordination
B.2. Selection and testing of techniques
B.2.1. Selection of techniques for cutting RPV walls in air and underwater (SCK/CEN, KRB, EWN)
B.2.2. Support, positioning and driving system (SCK/CEN, KRB, EWN)
B.2.3. System for the collection and filtration of swarfs and debris (SCK/CEN, KRB)
B.2.4. Full-scale testing of techniques (SCK/CEN, KRB, EWN)

B.3. Radiological survey and radioprotection optimization
B.3.1. Radiological characterization of RPV (KRB, SCK/CEN)
B.3.2. Radiological inventory of VVER reactors and internals (EWN)
B.3.3. Radioprotection optimization to cope with ALARA (SCK/CEN, KRB, EWN)

B.4. Dismantling by various techniques
B.4.1. Dismantling of the RPV at BR3 (SCK/CEN)
B.4.2. Dismantling of the KRB-A RPV (KRB)
B.4.3. Application to core components and structures of VVER-type reactors (EWN)

B.5. Collection of specific data (SCK/CEN, KRB, EWN)

C. Progress of work and results obtained

Summary of main issues

The contract starting at the beginning of 1996, this year was mostly devoted for the three decommissioned facilities to studies, licensing, engineering and some tests. Some actual preparatory work was also carried out in the three plants.

At KRB-A, the contents of the first period were mainly characterised by theoretical work and studies on dismantling of the reactor pressure vessel. Referring to the objectives and scope, calculations were done on the radiological survey (B3) and techniques for cutting in air and under water were evaluated in view of feasibility and spreading of aerosols and hydrosols (B2).

At EWN, the preparatory work consisted in the dismantling of equipments in the Steam Generator Room of Unit 5 as well as the one-piece removal of the six steam generators and main coolant pumps and their transportation to the Interim Storage Nord (ZLN) where they will be stored until later dismantlement. The preparation of the specification for the tendering documents for the dismantling equipment as well as the preparation of the licensing document for the dismantling of equipment in Unit 2 and for the model testing were carried out and filed.

At BR3, a comparison study on the Reactor Pressure Vessel (RPV) dismantling under water and in the air was performed. This was also required by the financing authorities in charge of the liabilities funding management. This study led to select the underwater option and to a formal agreement of these authorities to proceed with the D&D of the plant.
Studies were then started for the disassembly of the RPV from its primary loop and for the leaktightness of the pool when getting through the RPV. Engineering studies of two options of the RPV dismantling, i.e. in situ or as one piece removal and then dismantling in the pool, were then carried out. Finally, in parallel, a first radiological characterization of the RPV, based on dose rate measurement, was performed.

C.1. Project coordination (Task B.1.)

The first progress report was issued in common and a coordination meeting was organized at KRB-A. The coordination continues through frequent contacts between the different contractors. This is also guaranteed by monthly status reports of the participants.

C.2. Selection and testing of techniques (Task B.2.)

At KRB-A, two detailed proposals for the dismantling of the reactor pressure vessel (Fig.1) were offered by industrial partners. The studies were evaluated with regard to the feasibility. Both studies are accepted in principle, but a decision for the final strategy was not taken up to now.
One of the studies combines the use of thermal as well as mechanical cutting techniques. Both kind of tools are licensed for the dismantling in KRB-A. The other proposal is based on mechanical cutting techniques only.
The basic idea is to cut the vessel into ring segments and to transport these segments to the storage pool or a special cutting cabin for post-dismantling into smaller pieces for packaging. Thermal cutting can be used for the upper and lower part of the vessel, because the activity is rather low. The proposal in this case is to use oxygen cutting and to incline the torch by 30°, so that the process starts in air, facing the outside of the vessel, but the slag will be blown out of the kerf right below the water level or at least directly on the water surface. Due to this the

cutting thickness will rise from 124 mm to about 151 mm. A special draining device fixed to the outer side of the vessel will catch leaking water if necessary.
For the core section of the RPV remote-controlled mechanical cutting is foreseen in both studies.
The strategy is to use the water for shielding purposes as long as possible. This can be done when cutting from the inside of the pressure vessel only to a certain depth. The final cut will be done in air after draining the water to a lower level.
The disadvantage of using mechanical tools is, that these tools ask for heavy tool support, which must counteract the reaction forces.

At EWN, in the framework of a conceptual review with the aim to optimise the techniques and reduce the costs, the strategy was adapted, in such a way that reactor 5 will only be dismounted and transported to the interim storage. The same is valid for the core internals. The components will be subject to a decay storage of 40 - 70 years and afterwards a treatment and conditioning without remote techniques will be possible.
For the cutting of the activated components one cutting installation for two reactor units is foreseen (both located in one reactor hall). For this purpose the steam generator room of unit 2 and 4 will be utilized.
The cutting installation consists basically of a dry cutting caisson and a wet cutting caisson (i.e. under water cutting) where the higher activated components are cut in the wet caisson.
The two cutting caissons as well as the packing station are implanted below openings to the reactor hall, so that the loading and unloading of these areas can be performed without problems.

At BR3, the two optional strategies for cutting the RPV, in air or under water, were analysed and compared. An industrial partner made a proposal for dry cutting, using the one-piece removal of the RPV in the pool and then dismantling the RPV with the tools situated at the outside of the RPV.
Nevertheless, due to the high complexity of the needed equipment, to the radiations hazards implied by removing the RPV from its pit, and finally to the cost comparison of both options, the cutting under water was selected as reference option.

However, the idea of a one-piece removal was retained and analysed for the underwater cutting. It will allow much easier access to the external shroud of the RPV (see Fig.2) as well as to the thermal insulation (glass fibres) encapsulated between the shroud and the RPV.

Regarding the good experience gained by the internals dismantling, the mechanical cutting techniques, including a circular saw for making rings and a band saw for segmenting them, will be used. Both types of tools and equipment were foreseen to cut wall thicknesses like the RPV wall. Nevertheless, some cold tests will probably still be required for defining the right cutting parameters and for training the personnel.

C.2.2. Support, positioning and driving systems (Task B.2.2.)

At KRB-A, heavy supports will be necessary for mechanical cutting (Fig.3) or for moving the ring segments to the storage pool or a special cabin for post-dismantling. The existing crane in the reactor building can be used for driving and positioning these supports. In case of separating the transport of segments from the cutting device, an additional transport system must be used, and the cutting device must be able to fix itself to the pressure vessel.
Besides this, an additional working platform in the storage pool for the steam dryer system could be useful, to get closer to the working area.

A closed cabin must be constructed in case of post-dismantling the segments in air with thermal cutting techniques. For post-dismantling under water in the storage pool, an underwater band saw will be necessary. For cutting the upper and lower section of the vessel with thermal cutting technique, a special tent must be available to cover the working area and to contain the aerosols.

At EWN, two types of cutting are foreseen, depending on the activity of the workpieces :

Dry cutting caisson : The dry cutting station is divided into a pre- and postcutting area separated by a shielding wall. Through a vertical gate in the shielding wall it is possible to move a transport waggon between the cutting areas. The dry cutting caisson is equipped with an underpressure control system.

The component to be cut is placed in the precutting area on a turntable. The turntable is mounted on the transport waggon. The component is fixed with a tie rod. In the precutting area the horizontal cuts are performed. The cut piece is transported with the transport waggon to the postcutting area, where it is cut into pieces suitable for final storage. The normal cutting tools are bandsaws. In the framework of the testing programme autogen and plasma cutting will also be tested.

In the packing station which is directly connected to the caisson, the containers are loaded and covered. During filling the container is equipped with a protective sleeve in order to avoid contamination.

Wet cutting caisson : This caisson consists basically of a bottom structure with the cutting devices, different transport and handling devices, water purification system, emptying and filling connections and a ventilation system. The caisson is separately ventilated and kept at underpressure. Neighbouring rooms are not radiologically influenced.

The component to be cut is placed in the caisson on a turntable. The cutting is normally performed with bandsaws like in BR3, horizontally as well as vertically. Optionally also plasma-, as used in KRB-A, and CAMC- cutting devices are foreseen. For the handling two manipulators are foreseen. One is used for the handling of the cutting tool and the other one for the handling and transport of the pieces, which are placed in wiremesh baskets. The loaded baskets are transported to the packing station with an overhead crane and placed in a container. Subsequently the container is automatically closed and the documentation is put together.

At BR3, after selecting the underwater cutting option, the study mostly focuses on the clamping of the workpiece. Indeed, the whole pressure vessel (30 T) and then parts of it will have to be supported and clamped. The tool support and driving systems require to be stiff enough, for mechanical cutting, but the equipment for the internals dismantling showed to be alright and will probably be reused, with some adaptation.

C.2.3. System for the collection of swarfs and debris (Task B.2.3.)

At KRB-A in principle different cutting techniques can be used for the dismantling of the RPV. The experience gained from the cutting of the reactor head showed, that the segmenting in air with thermal cutting tools is possible with an effective suction and filtering device. Cutting under water would reduce the emission of aerosols to a minimum, but this of course is not possible, because dismantling must be done in situ and the water level has to be beneath the cutting position. The idea to cut the vessel from outside with thermal tools and to blow the slag on the water surface will reduce the emissions to a certain amount, but it must be taken into account, that oxygen cutting is working with a high flow of gas, which will curb the filtering effect of the water substantially.

Opposed to this, mechanical tools will have no spreading of aerosols at all, especially when cutting under water.

The investigation and experience in working with mechanical and thermal cutting techniques in air and under water has shown that the emissions of the tools can be handled with the existing filtering system. The capacity of filtering systems is strongly depending on the cutting technique and the cutting environment. Mobile filtering systems are very flexible and they can be renewed easily. Recleanable systems should be used for stationary systems in air and under water.

For the BR3, the former small underwater filter installation seems to be too limited in flow rate and gave also a lot of problems for its maintenance. Moreover, the consumption of filters and strainers was quite high. The related waste cost was also quite high.

Therefore, a search for other systems using as first filtration device the cyclone system, avoiding the use of strainers, has been carried out. Finally, an industrial equipment, based on this system was found and has been ordered. It delivers a flow rate of 20 m^3/h (i.e. more than 3 times higher than before) and the main particles and swarfs are collected in drums through a cyclone system. Cold tests of the system will be done during I/97.

C.3. Radiological survey and radioprotection optimization (Task B.3.)

C.3.1. Radiological characterization of RPV (Task B.3.1.)

The interesting topic of this work package will be the intercomparison of the 3 types of reactors (PWR, BWR and VVER).
Moreover, the radiological characterization of the RPV is important to manage the material flow and the waste category to which each part belongs.

For KRB, a theoretical calculation of the activity for the whole RPV has been performed in the year 1983 by an external company. Based on this study, Table I shows the total activity inventory for some RPV sections calculated for the year 1997. For the selection of appropriate RPV cutting techniques the amount of aerosols generated at the cutting kerf is one important parameter. In general, the specific activity of the RPV wall in the core section is higher by a factor of 1000 higher than for the other RPV sections. In order to optimise the material flow, further sampling and activity measurements must be carried out to determine the exact activity for such RPV material which can be given for controlled recycling by melting (< 200 Bq/g).

At BR3, a first characterization done by underwater contact dose rate measurements has been performed. The results are shown in table II. These results show where it is interesting to get samples for further detailed characterization in order to determine the material flow. For the Belgian waste package requirements, the solid waste has to be separated between HLW (i.e. contact dose rate higher than 2 Sv/h), MLW (contact dose rate between 2 mSv/h and 2 Sv/h), and LLW (contact dose rate lower than 2 mSv/h). The package requirements, handling and cost are different for these 3 types of waste.

Existing samples of the pressure vessel are presently analysed to build a complete radioactive inventory of the RPV and to be able to define precisely the cut levels in order to optimize the waste management.

C.3.2. Radiological inventory of EWN reactor and internals (Task B.3.2.)

Based on the different activity levels of the reactor components, major calculations were performed in context of the analysis of the container concept, in order to arrive to an optimal container selection. Taking into account the decay during interim storage, the loading of the containers will be performed in such a way that after the interim storage the containers can be transported without additional shielding on road and rail.

According to the new strategy for reactor 5, the necessity for additional shielding during interim storage and for the transport was calculated. As a result of these calculations it was shown that for the pressure vessel and the cavity bottom additional shielding is not needed. For the protecting tube unit, core basket and reactor cavity, an additional shielding is necessary. For the interim storage all components will be shielded in order to obtain a sufficiently low ambient radiation level in the storage hall.

A preliminary investigation on α-contamination in the primary loop and related components was performed. It can be concluded that α-contamination is present and more detailed investigations must performed.

Nuclide vector determination : In order to guarantee that limits on radioactivity content are not overpassed it is necessary to consider all nuclides present. In order to limit the analytical effort only a limited number of complete analyses, including pure β- and α-emitters, are performed on representative samples. In the practical work, only easy-to-measure γ-nuclides are measured and other nuclides can be calculated. In total 60 samples were used for this purpose and they were analysed by an external laboratory. Based on these measurements it was possible to define only 3 major nuclide vectors covering basically all equipment.

All analysis and the nuclide vectors have been controlled by the authorized expert and finally licensed by the authority and are shown in table III.

C.3.3. Radioprotection optimization to cope with ALARA (Task B.3.3.)

In parallel with the radiological survey of the RPV and internals, the optimization of the radioprotection has also been started in the different projects.

At KRB, the programme to get a complete dose rate profile for the RPV has been started up with radiological measurements nearby the bioshield area. The first results are listed in table IV and will be used for the pre-planning and calculation of the collective dose for the RPV dismantling. This values will be checked again after completing the removal of all RPV internals.

After draining water from contaminated components, problems during dismantling could occur by the normal drying procedure in the atmosphere which could result in a substantial release of aerosols into the working area. Therefore the inner surface of the RPV vessel will be cleaned before cutting, with a high pressure water jet to remove the contamination layer as far as possible.

Because of the dismantling work, which will be done done from the outside of the RPV, it is necessary to clean the areas around the vessel above and below the bioshield from a wide-spread dust layer (B.3.3.). By this, the hazard of aerosol incorporation for the dismantling staff is substantially reduced. Figure 4 gives a picture with a view to the area around the RPV. The space between the RPV and the bioshield is still filled with aluminium insulation material.

At BR3 (the dismantling being carried out under water and regarding the experience gained during the preceding phase of the project) it is mainly the operation time in the controlled area which has to be minimized as well as the maintenance of the tools and equipments.

Nevertheless, the preparation of the RPV cutting requires a radioprotection optimization study, as it involves the dry separation of the RPV from its support (24 bolts), the dry cutting of the hot and cold legs, and the leaktight closure of the pool openings.

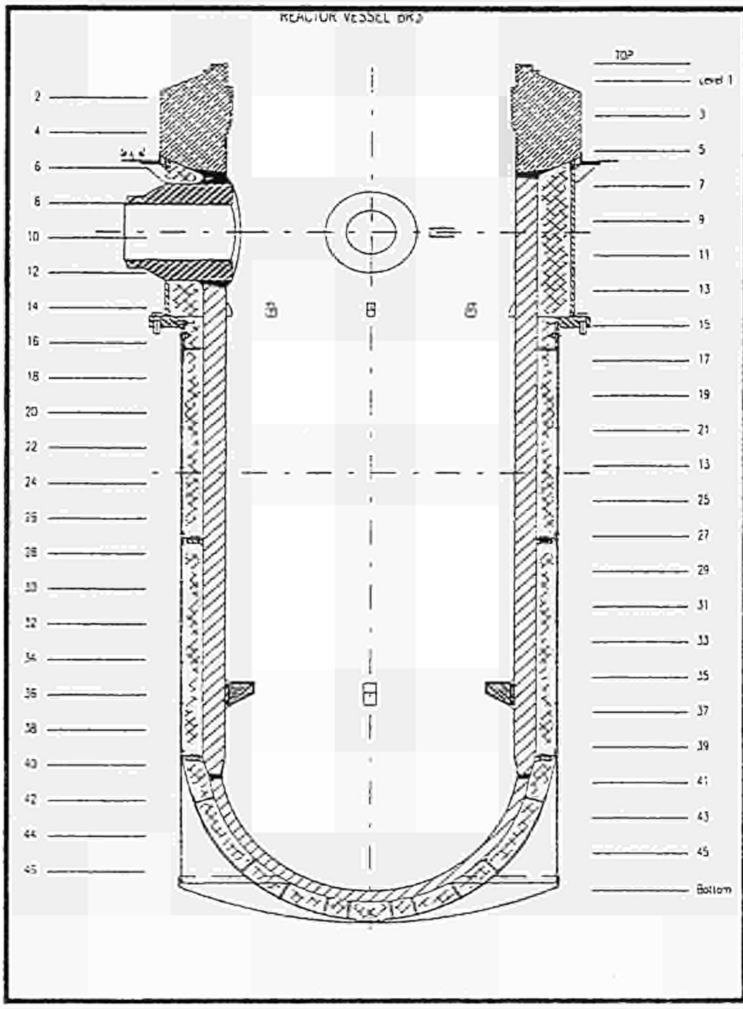
For this last operation, involving some dismantling of auxiliary loops in the space surrounding the primary loop, a computer-based dose uptake modelling system has been developed. It uses a 3D model of the environment and the measurement of radiation sources and ambient dose rate at some points and allows then to calculate the dose integrated by the operators and the effects of alternative operations (see fig. 5).

Table I : Calculated activity of the KRB-A RPV

RPV section at level	Mass (tons)	Activity (Bq)	
		in the ferritic material	in the austenitic cladding
Flange	58.1	2.4 E+03	6.2 E+02
Water separator	22.6	1.1 E+04	4.5 E+03
Feedwater inlet	21.6	4.3 E+07	1.8 E+07
Upper core	19.4	3.2 E+09	1.2 E+09
Central core	34.5	3.1 E+12	1.3 E+12
Lower core	22.0	3.7 E+09	8.4 E+08
Recirculation inlet	39.8	5.8 E+09	1.1 E+09
Buttom dome	42.0	-	-

Table II : Contact dose rate of the BR3 Pressure vessel

(Measurements done under water)



Level mm	Dose rate in mSv/h				
Distance from mid plane	Fuel Transfer Tank Side	Hot leg side	Cold leg 1 side	Cold leg 2 side	Mean value
2240	1,3				**1,3**
2040	1,3				**1,3**
1840	2,5				**2,5**
1640	3				**3**
1440	5				**5**
1240	15				**15**
1040	55				**55**
840	200	80	70	200	**137,5**
640	800	200	250	600	**462,5**
440	1500	800	800	1300	**1100**
240	2500	1800	1500	2500	**2075**
40	3500	2200	2200	2500	**2600**
-160	2500	2500	2500	2500	**2500**
-360	2000	2500	2500	2000	**2250**
-560	1000	1800	2500	1000	**1575**
-760	300	400	500	350	**387,5**
-960	70				**70**
-1160	35				**35**
-1360	9				**9**
-1560	8				**8**
-1760	15				**15**
-1960	70				**70**
-2160	400				**400**
-2360	800				**800**

Table III : Nuclide vector at EWN

Area	% of total activity			
	Co - 60	**Cs - 137**	**Fe - 55**	**Ni - 63**
CA unit 5	28	3	65	4
CA units 1 - 4	17	2	71	10
TH units 1 - 5[1]	18	2	60	20

(all nuclides > 1% listed)
CA = controlled area
TH = Turbine Hal

Table IV : Results of dose rate measurements at KRB-A

Place	Dose rate (μSv/h)	
	Contact to the RPV wall outside	**1 m distance to the RPV wall**
Above the bioshield +17.9 m	910	170
Core section +12.9 m	63,000	12,000*
Below the bioshield +8.9 m	370	70

* Calculated

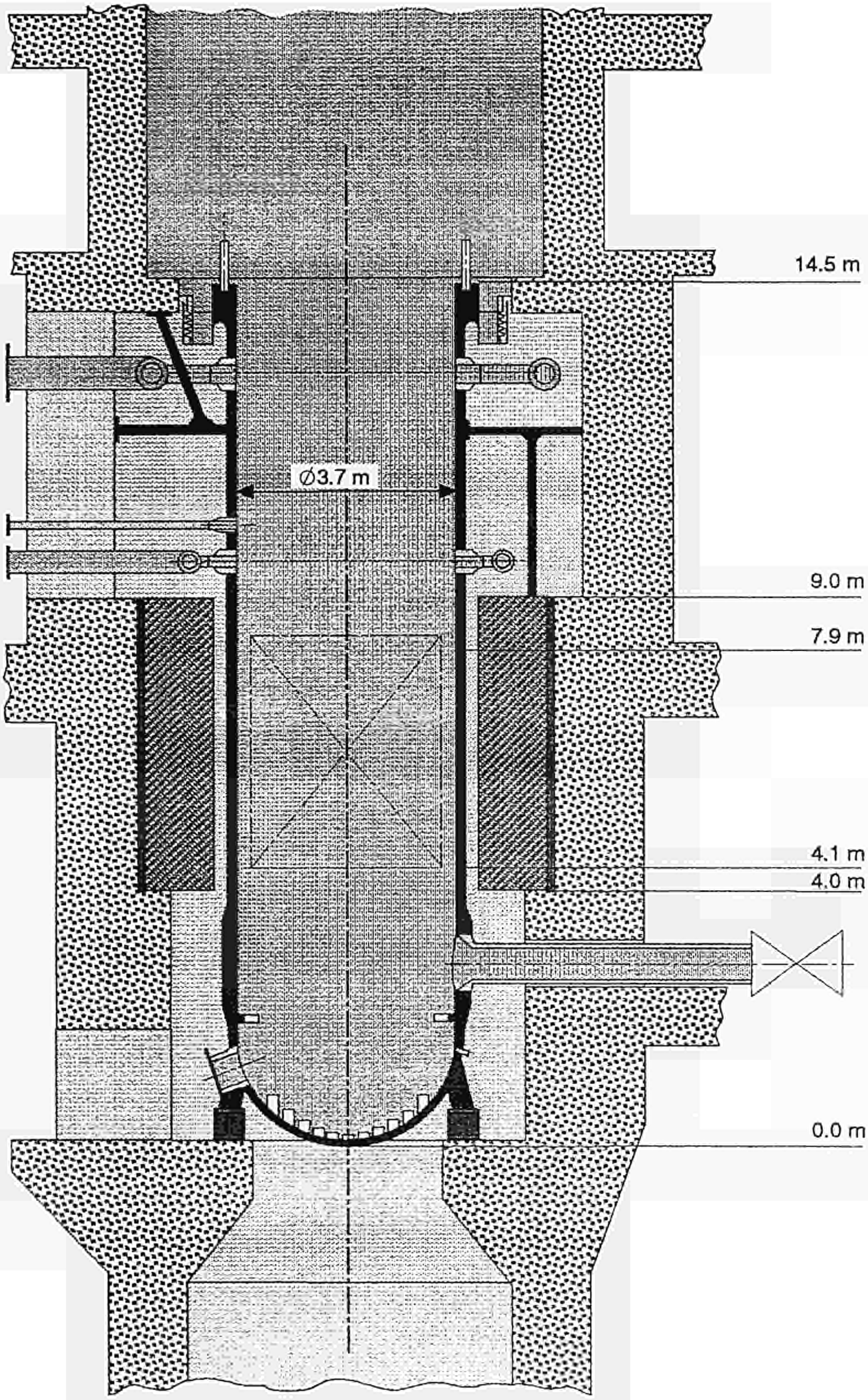


Figure 1

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KRB **Scheme of the RPV without internals**

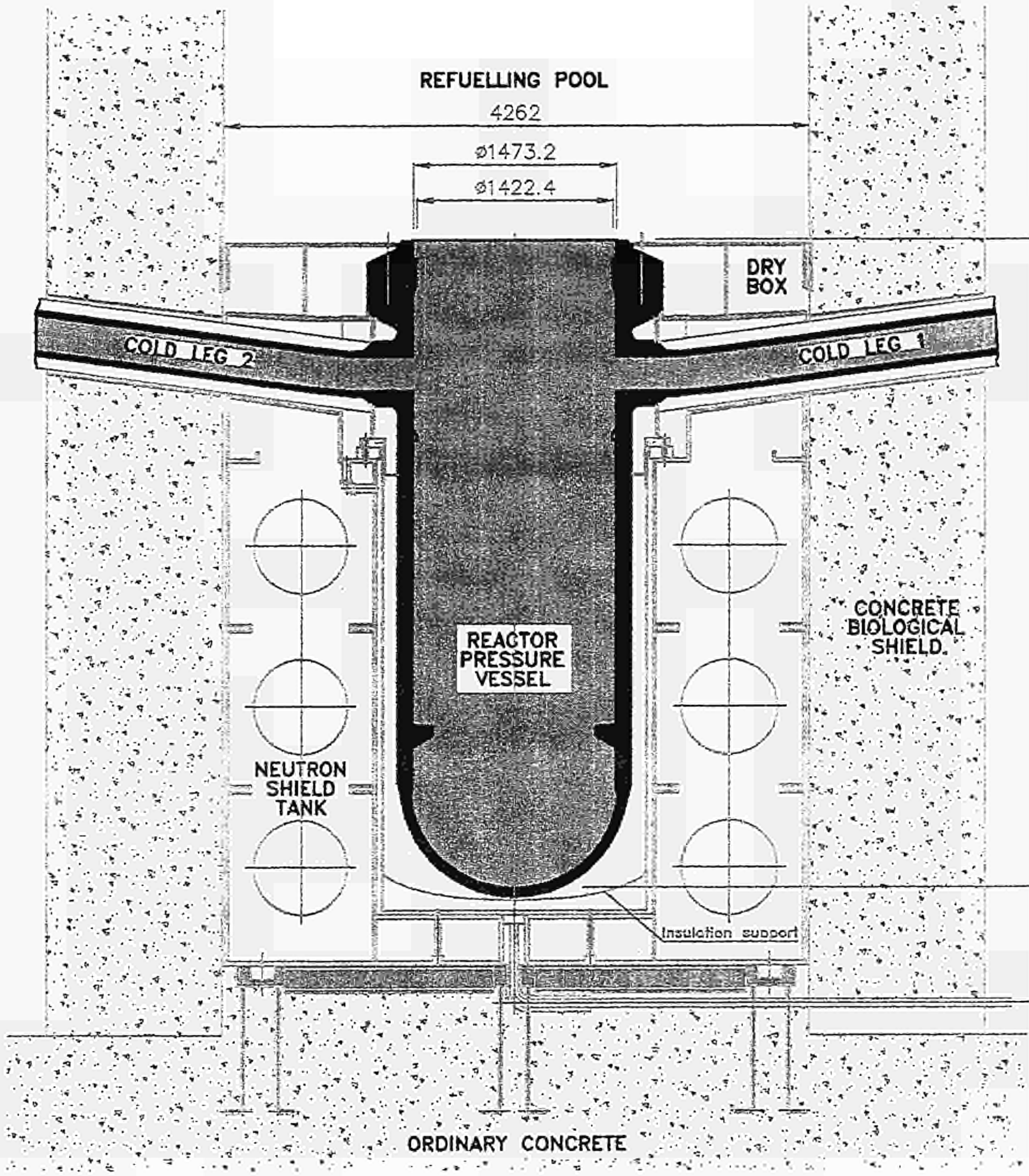


Figure 2: BR3 Reactor Pressure Vessel
Schematic view

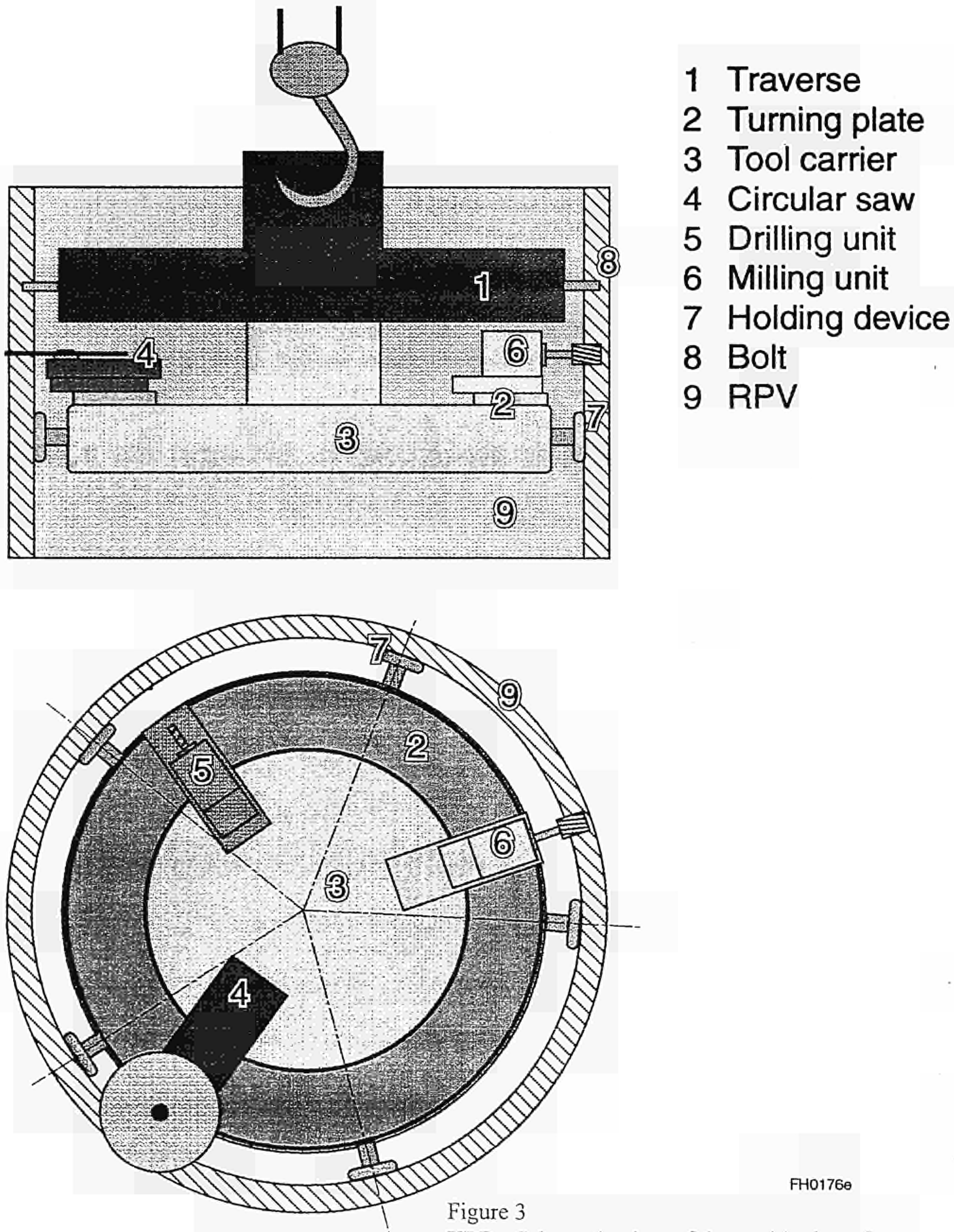


Figure 3
KRB - Schematic view of the positioning of mechanical tools and supports for dismantling the RPV

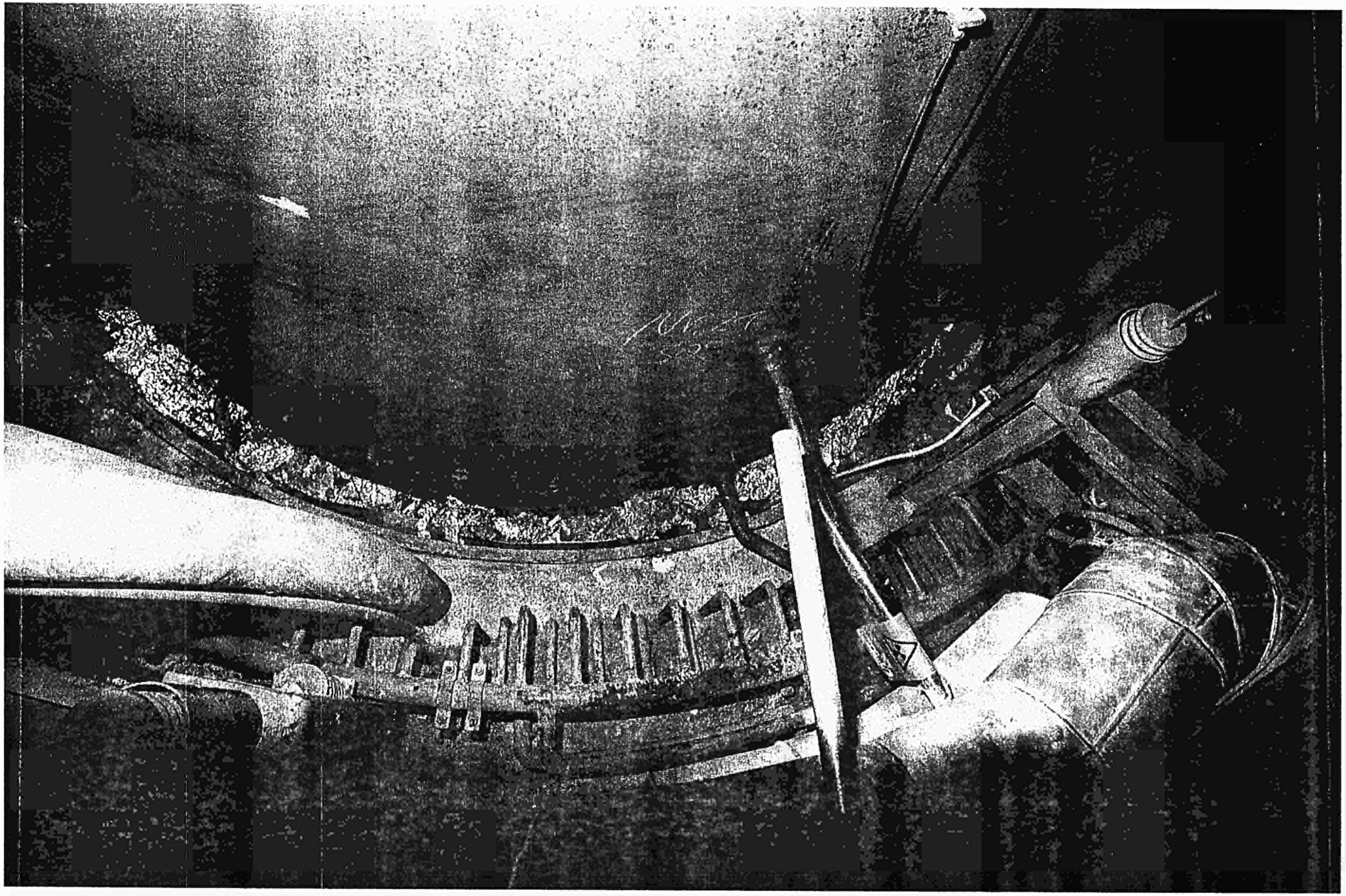
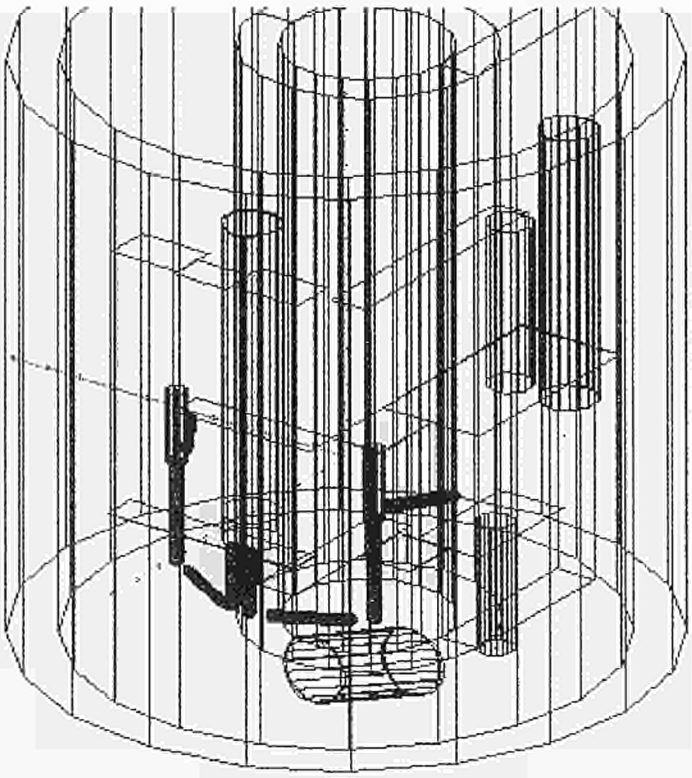
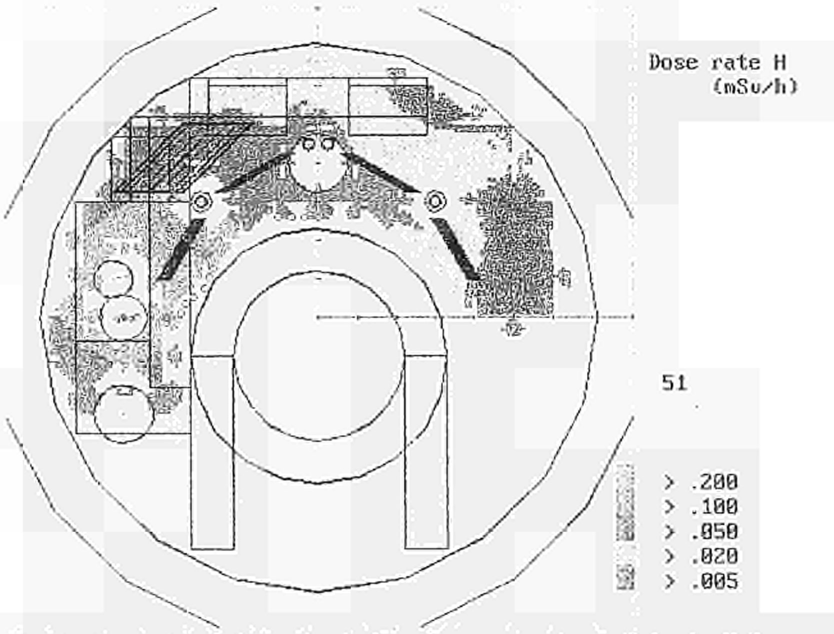


Figure 4 : Upper part of the KRB-A RPV above the bioshield. This area was cleaned to reduce the dose uptake during preparation of the dismantling work



Three dimensional model used, with the main equipments (radiation sources) included.
(View inside the plant container)



Typical result of the calculation (cross section of the building at one level).
The programme is then able to compute the cumulated dose uptake of the operators.

Fig.5 : 3D model for the radiation dose uptake calculation and optimisation of BR3.

C.4.1-2 ASCADIN - Coordinated development of a powerful thermal cutting tool (LSI)[1] with an innovative self-cleaning air filtration system

Contract No.: FI4D-CT95-0002	**Duration:** 1 April 1996 - 30 April 1999
Coordinator:	**J.P. Grandjean, CEA-CEN Valrhô, Bagnols-s-Cèze/FR** Tel. +33/4/66 79 63 04 Fax: +33/4/66 79 64 22
Partners:	**KEI Heidelberg/DE, Camfil SA, Pont Ste-Maxence/FR**

A. OBJECTIVES AND SCOPE

The main objective of this project is to develop a thermal cutting tool suitable for very thick steel plates (the LSI process) and a self-cleaning filtration system that will allow the tool to be used without significantly modifying the building ventilation system.

The development target is to cut 250 mm thick plates in air at a rate of about 100 mm-min^{-1} and to assess the mutual effects of workpiece thickness, cutting rate and secondary wastes. The cutting rates will be compared with those measured under contracts FI2D-0013 and -0019, and possibly with other relevant studies; aerosol generation will also be compared with data compiled for only smaller thicknesses (<150 mm) in the previous EC decommissioning programmes.

The acoustic self-cleaning electrostatic filter will be tested to evaluate its ability to trap and recover aerosols.

The cutting test results will be transferred to the European data base EC DB TOOL.

The outcome of the work should be the demonstration of the suitability of thermal cutting methods such as the LSI process (producing large amounts of aerosols) for 250mm thick plates in connection with a new CAMFIL self-cleaning filter.

B. WORK PROGRAMME

B.1 LSI Process Qualification
B.1.1 Fabrication of the LSI cutting tool (KEI)
B.1.2 Personnel Training (KEI, CEA)
B.1.3 Preparation of the test cell (CEA)
B.1.4 Cutting speed versus thickness (CEA)
B.1.5 Secondary waste production versus thickness (CEA)
B.1.6 Data for EC-DB-TOOL (CEA)

B.2 ASCADIN Qualification
B.2.1 Filter clogging measurements (CEA)
B.2.2 Filter efficiency measurements (CEA)

B.3 CAMFIL Filter Development
B.3.1 Specification (CAMFIL, CEA)
B.3.2 Fabrication of the CAMFIL aerosol filter (CAMFIL)
B.3.3 Filter efficiency measurements (CEA, CAMFIL)

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[1] LIS - Lichtbogen Sauerstoff Impulsschneiden

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues

The Tasks B.1.1, B.1.2, B.1.3, and B.3.1 were performed in 1997. The LSI cutting tool was manufactured, and tested in Saclay. The CEA personnel were trained and the cutting cell has been prepared.. Camfil designed a new filter, according to the project specification.

C.1. LSI process qualification (B.1)

Task B.1.1 : Fabrication of the LSI Cutting Tool. The LSI machine was manufactured by KEI and tested at Heidelberg in September. During October, the machine was improved to be able to cut plates up to 200 mm thick.

Task B.1.2 : Personnel Training. KEI had to wait four weeks because CEA needed some safety and security authorisations from the Saclay centre to be allowed to use the LSI cutting tool in the cutting cell : the main risks for the personnel came from the Oxygen flow rate needed by the LSI process in the cell, and the eyes irradiation by the powerful light of the cutting operations. When CEA was allowed to start the tests, in December,then the first tests of the LSI machine on the cutting table were carried out. The CEA personnel was trained for cutting, and repairing the machine.

Task B.1.3 : Preparation of the Test Cell. The cutting cell was mounted at Saclay and the complete ventilation loop was installed in June 1996. The cutting table was delivered in September and the Oxygen and electricity connected in October.

Task B.1.4 : Cutting speed versus Thickness. This task started at the end of December and should finish in March 97.

Task B.1.5 : Secondary Waste Production versus Thickness. This task started at the end of December and should finish in June 97.

Task B.1.6 : EC DB TOOL. This task will be performed in 1998.

Figures : Figure1 : The cutting table
Figure 2 : The LSI cutting process

C.2 ASCADIN Qualification (B.2)

This work package will be performed in 1997.

C.3 CAMFIL filter Development (B.3)

Task B.3.1 : Specification. The specification of the filter has been performed by the CEA and CAMFIL ; the type of filtering (cartridges) and declogging (air flow) processes have been selected.

Task B.3.2 and B.3.3 : Fabrication of the CAMFIL aerosol filter and Filter Efficiency Measurements. These tasks will be performed respectively in 1997 and 1998.

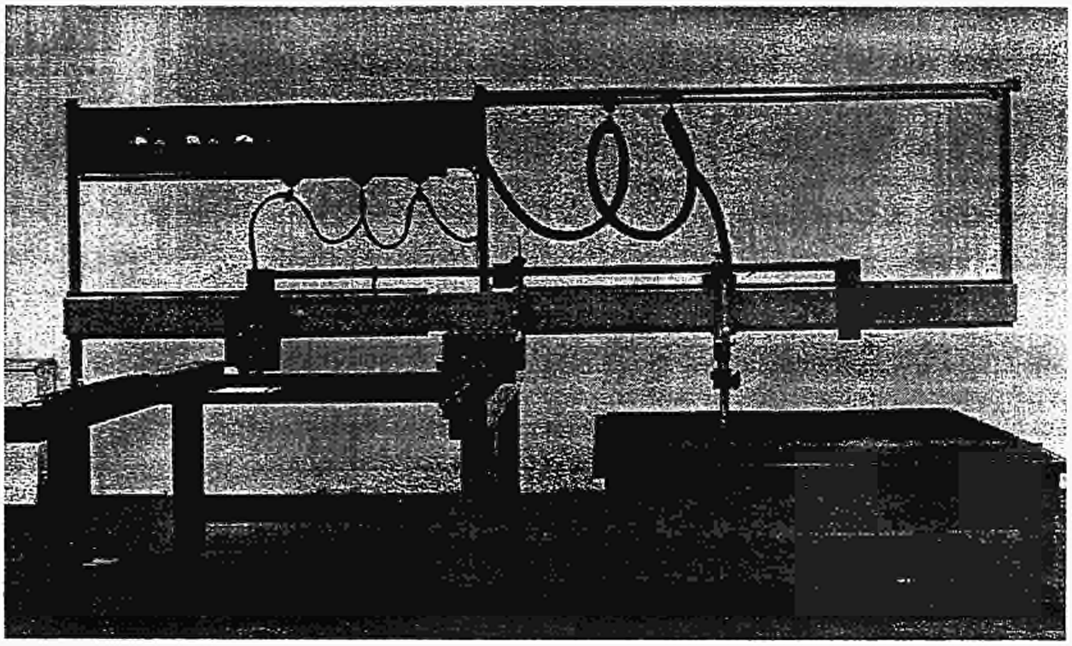


Figure 1 :The cutting table

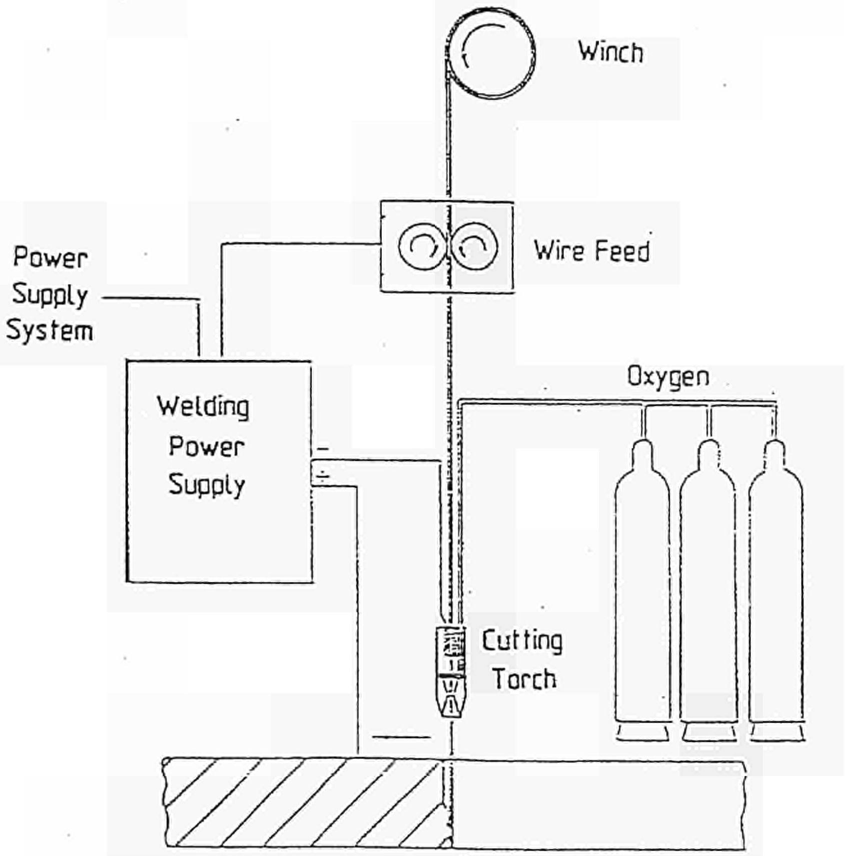


Figure 2 : The LSI cutting process

C.4.1-3 Remote cutting by a laser unit for dismantling the nuclear installations (CLAUDIN)

Contract No.: FI4D-CT95-0005	**Duration:** 1 Jan. 1996 - 31 December 1998
Coordinator: **J.P. Grandjean, CEA-CEN Valrhô, Bagnols-s-Cèze/FR**	
Tel. +33/4/66 79 63 04	Fax: +33/4/66 79 64 22
Partners: **RADIUS Gent/BE**	

A. OBJECTIVES AND SCOPE

The main objectives of the current project, are:

- To show the feasibility of remote cutting by Nd/YAG laser
  - using optical fibre transport
  - and maintaining a cutting head/workpiece distance of more than 200 mm (and up to 1 metre
  - using new spherical lenses in ZnS, with minimal absorption of the coatings.
- To establish the feasibility of remote cutting by Nd YAG laser without gas assistance.
- To determine the influence of focal length and other cutting parameters versus thickness.
- To assess the secondary wastes produced such as sediments, aerosols, etc., and to show how clean the process is.

Testing the laser cutting tool in the same cell as was used for contract FI2D-0013 will allow to make an objective assessment of the laser in conditions similar to those in real dismantling, and to compare it with the five other tools already tested in the above-mentioned contract under standard cutting and ventilation conditions.

B. WORK PROGRAMME

B.1 Manufacture of optical systems
B.1.1 Specification of the 200 mm focal length optical system (CEA, RADIUS)
B.1.2 Manufacture and testing of the 200 mm focal length optical system (RADIUS)
B.1.3 Specification of the 500 and 1000 mm focal length optical systems (CEA, RADIUS)
B.1.4 Manufacture and shop testing of the 500 and 1000 mm focal length optical systems (RADIUS)

B.2 Cutting tests
B.2.1 Parametric tests (CEA)
B.2.2 Speed versus thickness graph (CEA)

B.3 Quantification of secondary wastes
B.3.1 Influence of thickness on waste generation (CEA)
B.3.2 Influence of focal length on waste generation (CEA)
B.3.3 Data for EC-DB-TOOL (CEA)

C. PROGRESS OF WORK AND RESULTS OBTAINED

Summary of main issues :

Tasks B.1.1, and B1.2 were performed in 1996. The 500 mm focal length cutting head was manufactured by RADIUS Engineering and delivered to Saclay at the end of December.

C.1 Manufacture of optical systems (B.1)

Task B.1.1 : Specification of the 500 mm focal length optical system : CEA and RADIUS established a specification for the fabrication of the first cutting head, with a focal length of 500 mm.

Task B.1.2 : Manufacture and testing of the f = 500 mm optical system : RADIUS manufactured the cutting head which was delivered to Saclay in December, with a delay of about 3 months. See figure 1 in annex.

Figure 1 :The 500 mm focal length cutting head

C.2 Cutting trials

Task B.2.1 : Parametric tests : this task should start in October, but will be delayed since the beginning of 1997 because of the task B.1.2 delay.

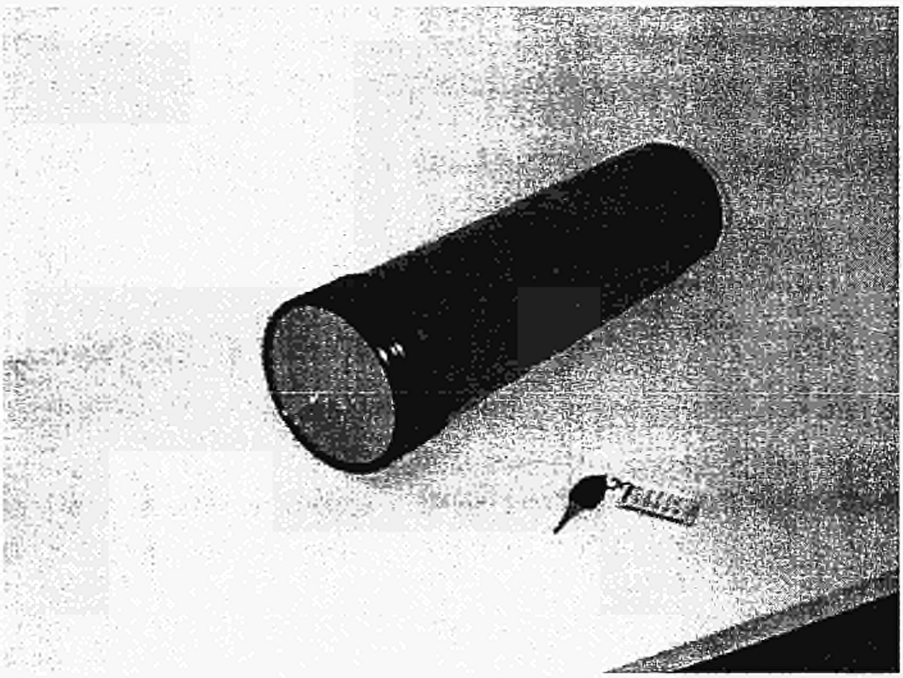


Figure 1 :The 500 mm focal length cutting head

C.4.1-4 Remote dismantling operations - WAGR decommissioning project

Contract No.: FI4D-CT95-0006 **Duration:** 1 March 1996 - 28 Feb. 1999
Coordinator: **B. Spruce, UKAEA, Risley/GB**
Tel. +44/1925/253 193 Fax: +44/1925/252 100
Partners: **CEA-UDIN Valrhô/FR**

A. OBJECTIVES AND SCOPE

The purpose of this project is to carry out the remote dismantling with low occupational doses of the WAGR reactor core and pressure vessel. The work will be managed on a commercial basis by a competitively appointed prime-contractor.

Operations to remove these elements of the reactor internals will be undertaken with the use of innovative dismantling techniques involving amongst others the following partly developed equipment:

- computer controlled Remote Dismantling Machine using stereoscopic television cameras to assist in the dismantling process;
- video-gamma-camera to identify and sort the most radioactive materials;
- operators training on virtual CAD scenes;
- ultra-violet laser to decontaminate vital parts of the machine before maintenance;
- acoustic-cleaning of electrostatic pre-filters.

Dismantling of the reactor core will be undertaken in discrete stages which are classified in a proposed project programme as individual tasks. Completion of each task is a measured objective prior to proceeding with the following task. A summary programme is submitted to illustrate the milestones.

This project will be undertaken jointly between UKAEA and the CEA to develop dismantling equipment to facilitate reactor core removal.

B. WORK PROGRAMME

B.1 Installation of Dismantling Equipment (CEA with UKAEA support)
B.1.1 Virtual CAD Modelling
B.1.2 Electrostatic filter cleaning equipment
B.1.3 Laser decontamination equipment
B.1.4 Gamma camera installation

B.2 Reactor Core Dismantling Tasks (UKAEA with CEA support)
B.2.1 Dismantling operations of the reactor core will begin with the removal of reactor operational waste.
B.2.2 This task will concentrate upon the hot box removal - a complex steel structure designed for reactor hot coolant collection and also to facilitate fuel loading operations;
B.2.3 Dismantling operations will continue with the loop tubes removal.
B.2.4 Dismantling of the reactor neutron shield, including inner and outer neutron shield;
B.2.5 Final task: the dismantling of the core and its restraint structure, i.e. removal of graphite core assemblies, the reflector and the core restraint bands.

B.3 Collection of specific data for the EC decommissioning data bases (UKAEA, CEA)
B.4 Radiological, technical and economical balance (UKAEA)

C. PROGRESS OF WORK AND RESULTS

Summary of Main Issues

Since award of the contract UKAEA and CEA have collaborated in a good manner and jointly agreed solutions to the technical problems arising in the definition of work against each Contract Task. These problems have occurred as a result of integrating the proposed innovative equipment into the WAGR Project without adversely affecting the WAGR work programme already in progress. This programme is controlled by several major commercial contracts which should not be disrupted.

Whilst the technical problems were being addressed, UKAEA and CEA negotiated a supplementary agreement which would facilitate direct communications for collaboration matters. The supplementary agreement was necessary to ensure minimum disruption to the commercial contracts already in effect at WAGR.

Considerable discussion between CEA and UKAEA was entered into to examine and agree the system to be adopted for the virtual CAD modelling. The finance available under the EC contract was not sufficient to underwrite the best technical solution for the model to meet the needs of training of operators in remote dismantling techniques. Similar discussions were also needed to define an access route for the gamma camera entry into the reactor bioshield. EC Contract funding was not sufficient to cover the costs involved.

However CEA and UKAEA have reached agreement and made proposals for additional funds from their respective organisations.

Progress and Results

C.1

(B1.1) Virtual CAD Modelling

The first step in this area involves comparison of those software solutions which can be envisaged for developing the reactor vessel Environment Simulator.

Two such CAD robotic systems for plant modelling are habitually used at the CEA:-

- ROBCAD from TECHNOMATIX in the field of feasibility studies. This is a very expensive system and is more suited to design departments. It offers considerable liberty in modelling and comparison of different scenarios for remote control applications.

- Act from ALEPH Technologies is more applicable to the field of operations work preparation. Being an on-the-job tool, this system has the advantage of being dedicated to a particular installation and is thus more accessible to the operator for any given environment.

In view of the UKAEA's requirements the former system should enable an easier evolution of the model over the period of dismantling. After examination of the two systems, Robcad was selected as the most appropriate application for use at WAGR.
The second step of the work involves collecting the plans needed for modelling the reactor vessel, the Remote Dismantling Machine (RDM) and the environment. Supply of plans from UKAEA but the following parts have already been modelled:-

- the RDM arm,
- the reactor vessel and the hot box.

An initial simulation of the hot box has now been done, as shown in the Figure 1.

(B1.2) Electrostatic Filter Cleaning Equipment

The WAGR is equipped with electrostatic filters which stop a large proportion of dust particles before they reach the VHE filters. This protects the VHE filters especially when dealing with smoke from the use of thermal cutting tools.

The electrostatic filter has numerous advantages over other types of prefilter, particularly its low loss of charge, reduced space requirement, low cost and satisfactory performance.

The greatest drawback of these filters is currently the problem of cleaning them when radioactive particles are present.

An electrostatic filter loses its efficiency progressively as the collection plates become clogged with dust. Frequent cleaning is needed when they are being used intensively. CEA have tested and measured the feasibility of a liquid-free cleaning method which can be applied in situ without demounting the cassettes and this is ACOUSTIC CLEANING.

Waves produced by the vibration of a membrane displace the air in which they travel: this is acoustic energy. This energy has two main parameters: the fundamental frequency measured in Hertz and its intensity measured in decibels (dB).

Compressed air is used to vibrate the membrane of our horn, producing sufficient acoustic energy to break the adhesive bonds between the particles and the collection surfaces and thus cleaning the electrostatic filters.

The characteristics of the horn used are as follows (manufacturers data):-

- Frequency : 250 Hz
- Acoustic pressure level (at 1 m) : 145 dB
- Air supply pressure during signal : 0.3 to 0.4 MPa
- Air consumption during signal : 20 to 30 l/s
- Mass : 20 kg

The electrostatic filter housing will have an orifice closed by a membrane. This membrane is currently made of a plastic material.

The horn will be fitted to the WAGR No. 2 electrostatic filter (ESP 2) which is connected directly to the inside of the reactor vessel and is thus likely to be the more contaminated of the two electrostatic filters in the installation.

The CEA is currently carrying out trials to optimise the choice of the separation membrane and of acoustic insulation. Additionally UKAEA is assessing the changes to the ESP installation.

(B1.3) Laser Cleaning Equipment

During the working time the tools, grabs etc, used during dismantling phases inside the core, will become contaminated with dust from graphite erosion or steel cutting. Decontamination methods to allow maintenance and refurbishment of the tools need to be established.

The CEA proposed concept for decontamination involves the direction of high energy (> 1 MW) ultraviolet laser pulses towards the tools to achieve excitation levels at which the electrons participating in molecular bonds at the tools surfaces are dissociated.

CEA found that this could be used to clean dusty equipment, especially graphite dust deposits, without damaging the tool surface. Each shot can treat 0.5 cm^2 at time. The advantage of this method is that it allows dry cleaning and the system can easily be computerised. The fine residual powder generated is transported by a ventilation system extract and trapped on an electrostatic filter.

A laser decontamination facility is to be built, as an extension to the proposed waste processing route containment, at ground floor level beneath Heat Exchanger D.

The proposed waste processing containment will be increased in size to accommodate the laser, control desk and grab handling 'A' frame and associated rotating unit for handling the contaminated tools during cleaning.

An air extraction ducting will be run from the containment and connected into the existing WAGR active extract ventilation system.

A 380 V3 phase electrical power supply, cooling water chiller unit and laser gas supplies will be needed outside of WAGR outer containment. These services will require a weatherproof and secure housing in addition to electrical supplies for the chiller unit. The chiller unit will stand within a bunded area. Penetrations will be cut through the WAGR outer containment to allow passage of these services into the laser decontamination facility.

The laser and the beam transport have all been purchased and are undergoing extensive tests in Marcoule.

The laser facility to be built inside the WAGR containment, is being assessed by UKAEA to ensure co-ordination with other dismantling preparatory works in progress.

(B1.4) Gamma Camera Installation

Over the last few years, CEA has developed a gamma camera prototype for fast localisation of irradiating zones or materials, which is called Aladin. This project has been substantially worked on using CEC financial support, outwith this Contract.

Since this gamma camera has been successfully operated at CEA or COGEMA plants, it was decided to introduce it as an innovative technique into the WAGR decommissioning project.

Work on this task can be divided in two parts:-

- development of a new gamma camera by CEA, more industrial and less expensive than the prototype system.

- preparation of its installation into the upper part of the reactor core.

1. Development of "Aladin 2"

The first prototype Aladin is steadily operated in France for CEA needs, therefore a new device has been developed, including some enhancements, that appeared to be necessary after the first operations with the prototype:-

- updating of the photonics;
- superimposition to a colour image, delivered by a separate CCD camera;
- cold lighting, the light being conducted by a fibre optics bundle;
- PC workstation software (whereas the first one was running on Unix workstation);
- Laser telemeter;
- gamma probe for background measurements.

The device Aladin 2 was firstly tested in October 1996. Trials are still underway in Marcoule.

2. Gamma Camera Installation

During our meetings, it has been progressively stated to introduce the camera inside the reactor vault, above the core. The main reason was that it would be able to monitor continuously the irradiating zone distribution of the working area and its evolution.
This requires some penetrations through the reactor biological shield to be cut. Two opposite locations were defined, so that a view of the whole of the reactor vessel could be seen. Figure 2 displays one of them.
The camera will be inserted into the vault and supported by a pole through the reactor biological shield, and will be operated from a control panel located on the second floor level. When needed, the device will be transferred from one location to the other.
Additionally, CEA are studying the possibility of sorting radioactive wastes when suspended by the waste route crane, before being transferred to the sentencing cell prior to loading into waste boxes.
General features of the pole have been outlined by CEA in November. Its fabrication will be started after Hazop analysis, and acceptance by UKAEA.
Preparation of Safety documentation for the preceding tasks has commenced.
Approval of the documentation will be sought from UKAEA's Safety Authority before any work within WAGR can proceed.

C.2 Milestone deliverables numbers 1-4 inclusive are scheduled for late 1997 and progress in the initial B1 group of tasks, led by CEA with UKAEA support, are on programme to meet the dates quoted in the contract.

C.3 Milestone deliverables 5-9 inclusive for the B2 and B3 tasks, are scheduled to commence early 1998. There is a delay due to problems relating to preparatory work to facilitate reactor core dismantling which may affect these deliverables. The recovery of delays now appearing in the WAGR Project programme are being assessed by the WAGR Project Managers and other contractors outside the EC Contract boundary. Upon receipt of this recovery assessment the Co-ordinator will review the Contract deliverables schedule.

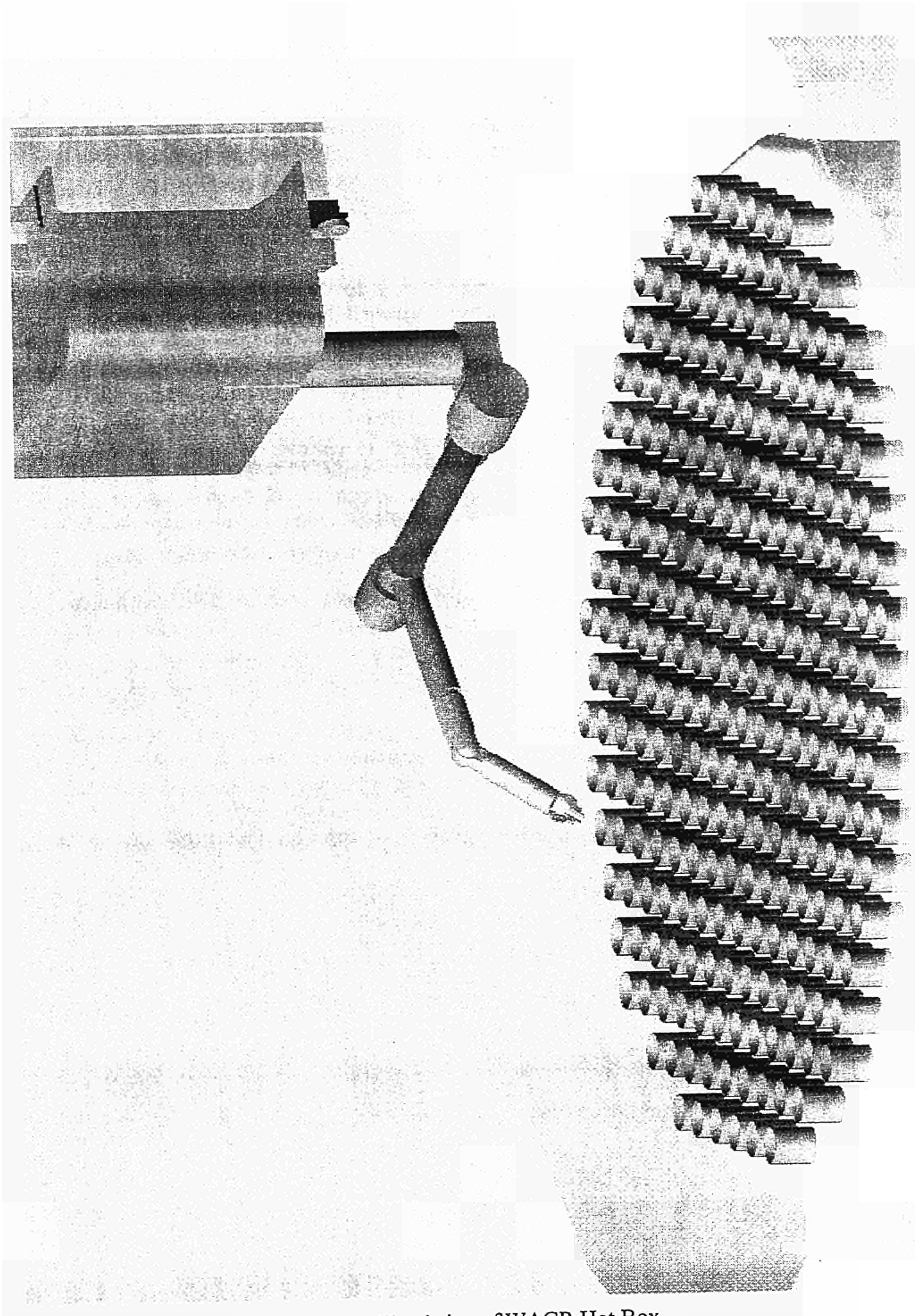


Figure 1: Simulation of WAGR Hot Box

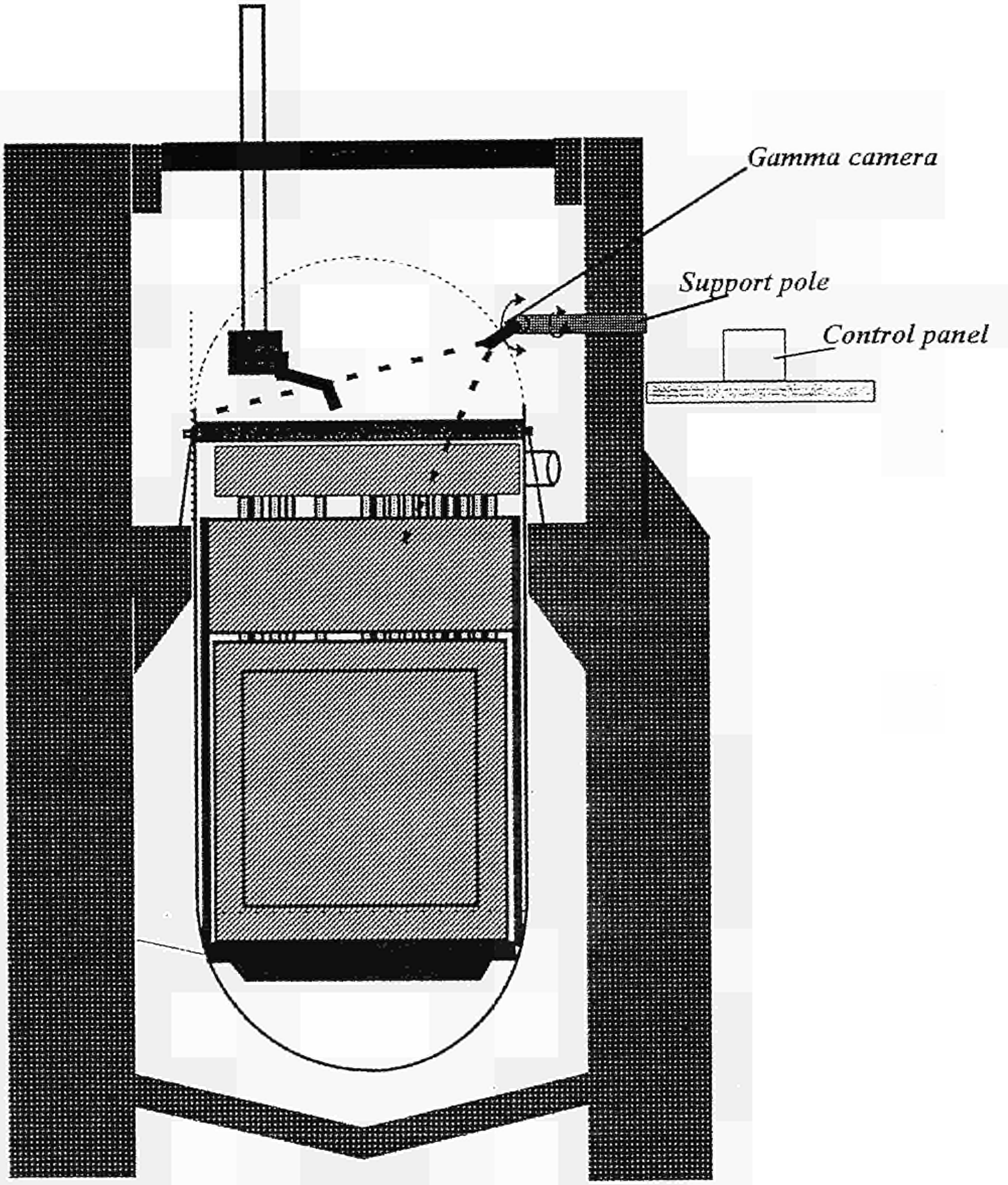


Figure 2: Introduction of the gamma camera into the reactor vessel

C.4.1-5 Remote-operated, manipulator-aided underwater dismantling of complex aluminium and steel structures by CAMC

Contract No.:	FI4D-CT95-0007	**Duration:**	1 Jan. 1996 - 31 Dec. 1998
Coordinator:	**F.W. Bach, Institut für Werkstoffkunde (IW), Universität Hannover/DE**		
	Tel. +49/511/762 43 16	Fax: +49/511/762 52 45	
Partners:	**CIEMAT Madrid/ES, VAMCE Linz/AT**		

A. OBJECTIVES AND SCOPE

Contact Arc Metal Cutting (CAMC) is the thermal sputtering of metallic materials by means of a discontinuous, recurrent high power short circuit arc ignited by contact between the non-consuming electrode and the workpiece.

One of the main objectives of this project is the investigation of different electrode materials, the optimization of the electrode cooling, the scavenging, and the process control in order to reduce electrode wear and secondary waste products.

Another main objective is the development of a remote-operated modular underwater manipulator handling system (MHS) of Voest-Alpine/MCE.

The main aim of this development is to prove the possibility to expand the flexibility of an installed handling system, e.g. the machine for fuel exchange, by simply adapting a remote-operated manipulator to obtain a tool guiding device with an enlarged freedom of movement for complex dismantling tasks.

B. WORK PROGRAMME

B.1 Development of the CAMC tool and technique (IW)

B.2 Inactive and active cutting tests
B.2.1 Selection and characterization of the components (CIEMAT)
B.2.2 Definition of the trajectories (CIEMAT)
B.2.3 Manufacture of a representative model for cold tests in JEN-1 (CIEMAT)
B.2.4 Manufacture of representative models for cutting tests at IW (IW)
B.2.5 Determination of cutting parameters (IW)
B.2.6 Estimation of waste product quantities (IW)
B.2.7 Training of CIEMAT staff at IW (IW/CIEMAT)
B.2.8 Delivery of a prototype CAMC tool to CIEMAT (IW)
B.2.9 Adaptation of the CAMC tool delivered by IW (CIEMAT)
B.2.10 Cold cutting tests in the JEN-1 facility (CIEMAT/IW)
B.2.11 Cutting of radioactive material in the JEN-1 facility (CIEMAT)
B.2.12 Measurement and analysis of cutting effluents (CIEMAT)

B.3. Adaptation and qualification of a modular M/S manipulator for underwater use (VA-MCE, IW)

B.4 Installation in a nuclear facility (option) (All partners)

B.5. Supply of relevant data to the European data base EC-DB-TOOL (IW)

C. Progress of work and results obtained

Summary of main issues

The CAMC process was investigated. In parameter studies the electrode material as well as the emissions initiated by the process were the focus of the investigations. The cutting parameters for the JEN-1 stainless steel components, which were initially selected by CIEMAT, have already been determined. A CAMC tool for adaption to both the master/slave manipulator of VAMCE and the guiding device which is present on site of CIEMAT and IW is under design. The selected components present a diversity of origin, shape, dimensions and composition in order to test the cutting tool in different working conditions. These components are both made of aluminium and stainless steel. To take the new working environment water into consideration several modifications had to be make in hardware and software of the M/S manipulator. They mainly extend to the slave mechanism and the electronical control.

C.1 Development of the CAMC tool and technique (IW)

C.1.1 Investigation of the electrode (IW)

The investigation of the electrode have been done within the parameter studies (see C.2.5). Electrodes made of pure graphit and carbon-reinforced electrodes were tested.
The test series showed that there are narrow limits for the use of electrodes made of pure graphit. They must not be stressed by mechanical forces or be stimulated to swing, otherwise they will break. To use the pure graphit electrodes the idling voltage should not exceed 50 V and the maximum cutting speed should be limited to 300 mm/min for an austhenetic stainless steel plate with a thickness of 20 mm. The carbon-reinforced graphit electrode have a distinct higher mechanical stability and so it can be used with an operating idling voltage of > 70 V. The maximum cutting speed is higher than 1000 mm/min for an austhenetic stainless steel plate with a thickness of 20 mm. To compare the electrode wear of both materials several cutting tests were done with parameters which took the limits of the pure graphit electrode into consideration. The result was, that the pure graphit electrodes fundamentally shew less wear than carbon-reinforced electrodes with the same process parameters (see Figure 1).

C.1.2 Design and manufacturing of tool (IW)

The main characteristics of the CAMC tool are light weight, compact geometry and an interface both for the master/slave manipulator of Voest-Alpine mce (the tool will be taken by the gripper) and the tool guiding device of CIEMAT (the tool will have the possiblity to be assambled to the device). The clamping of the electrode is realized by a remotely operated pneumatic cylinder. The final design will be finished in March 1997.

C.2 Inactive and active cutting tests

C.2.1 Selection and characterization of components (CIEMAT)

The components initially selected present a diversity of origin, shape, dimensions and composition in order to test the cutting tool in different working conditions. These components are both made of aluminium and stainless steel. The Core grid (Al-Mg3,frame with thickness up to 150 mm), grid support (Al-Mg3, essentially double-T profiles 120 mm x 100 mm, thickness 20 mm) and the fuel elements frame of the JEN-1 reactor (Al 99,5, complex structure) have been selected as aluminium components (see Figure 2). The radio-logic characterization of the core grid and the grid support has been done by α, β and γ spectrometry (see Table I). The other selected aluminium components are not yet completely characterized, nevertheless their dose rate is about 0.08 mSv/h. Fuel element racks coming from spent fuel pools of spanish nuclear power plants (AISI 304, see Figure 3) and several components of JEN-1 reactor (tubes, bars, etc.) made of stainless steel have also been selected. The radiologic charac-teristics of the fuel element racks are not yet completely defined. Concerning the JEN-1 stainless steel components, such as tubes and bars, their diameter respectively their thickness is about 80 mm and they present a dose rate from 0.01 to 4 mSv/h.

C.2.2 Definition of the trajectories (CIEMAT)

The definition of the trajectories will be completed at the time of training of the CIEMAT staff at IW Hannover (3rd quarter 1997). It is assumed that cutting trajectories do not present special problems associated to shape and characteristics of selected components, the main restrictions could be due to the moving possibilities of the underwater cutting device inside the limits of the cutting vessel of CIEMAT.

C.2.5 Determination of cutting parameters (IW)

The aim of extensive parameter studies was to achieve minimum electrode wear, minimum emissions and maximum cutting speed. The parameter which were variied are idling voltage, feed speed, water flow rate and the electrode material (see C.1.1). In addition to the results in C.1.1 it was detected that the water flow rate for the scavenging influences directly the electrode wear. An effective scavenging ensures that molten particles are flushed out of the kerf and won´t ignite any further arc between kerf wall and electrode. The parameter studies have been made with stainless steel plates so far, so the cutting parameters for cutting the stainless steel components selected by CIEMAT have already been determined. The boundary condition remote operated cutting, hand guided by master/slave manipulator, effects that mechanical forces onto the CAMC-electrode can´t be avoided, so the electrode will be made of carbon-reinforced graphit for all cuts. The idling voltage should be chosen between 45 V and 60 V to ensure a sufficient kerf width for flushing out the molten workpiece particle and on the other side to limit the kerf width in order to minimize the emissions. The water flow rate has to be as much as possible. Good cutting results with a low electrode wear were achieved with a flow rate of 70 l/min. The parameter studies with aluminium workpieces are still running.

C.2.6 Estimation of waste products (IW)

Within the parameter studies the emissions were also measured. The emissions caused by the CAMC process are 96% - 98% sedimentation, 1.5% - 3.5% hydrosols and 0.01% - 0.6% aerosols. The distribution depends on the trajectory direction, electrode material, idling voltage und cutting speed. Basic statements can be made as follows. The total emission depends on the kerf width, which is mainly dependent on the idling voltage. The aerosol emission decreases with rising idling voltage but increases with rising cutting speed. Boring / sinking holes initiates three times more total aerosol emission and twice more gas emission (important because of production of hydrogen) than cutting the same cross-section. That must directly influence the cutting strategy, boring with the electrode shall not be done to seperate components. For example cutting a 20 mm thick stainless steel with a carbon-reinforced electrode, an idling voltage of 71V and a cutting speed of 200 mm/min produces an aerosol emission of 62.4 mg per m cutting length. The average diameter of the particles is 0.35 μm.

C.3 Adaption and qualification of a modular M/S manipulator for underwater use (VAMCE, IW)

C.3.1 Adaption of MHS to underwater use (VAMCE)

To relieve the decontamination of the slave arm all aluminium parts of the outer body have been coated to achieve a smooth surface. Any other parts of the area of contact are made of austenitic stainless steel. Thus decontamination can be done easily. The slave arm is electrically operated by seven servomotors with planetary gearing. To prevent them from water breaking-in three different housings were necessary. These housings are equipped with watertight connector sockets for non-interchangeable plugs. All driving shafts were supplied with special seals that can support pressure from each side. These cases are connected with adjustable compressed-air supply as our tests have shown that additional pressure is necessary to activate the seals on the driving shafts. In addition superpressure helps to detect leaks in underwater application.

C.3.2 Adaption of a control (VAMCE)

The new working environment water changes the moving dynamics of the slave arm which is then even different to the dynamic behaviour of the master arm in atmosphere. The buoyancy has also to be embeded in the control software. In order to cope with the increased computational effort an upgrade of the main processor card was necessary. The new card is based on a Motorola 68040-33 Mhz CPU running under the operating system OS9 and its task is to achieve real-time control of the 14 axes robot system. To implement the new CPU in the system several changes in the operating system OS9 were necessary, mainly the rewriting of hardware drivers. Moreover new installation routines and adaptions of the existing control software for the new CPU were carried out and testet. At underwater service there are two main responsible aspects for a conspicious change of the robot system behaviour: the mechanical changes necessary to make the slave arm waterproof and the fact, that the whole slave arm moves in the new medium water. The mechanical changes result in a change of weight relations and in new friction conditions (for example new gaskets). The transfer of the slave arm under water surface effects new weight relations again according to the different extend of buoyancy of each component as well as a total change of system dynamics due to the increased resistance of the medium water. While the effects caused by the mechanical changes could be measured directly, the impact of underwater motion could - up to now - only be evaluated in a theoretical approach. Afterwards the parameters of the control algorithm were newly calculated and implemented. Moreover the software for electronic gravity compensation had to be adapted to the new situation. In the first configuration of the MHS underwater manipulator prototype the absolute position of every axe was measured by potentiometers. The absolute position is needed for the electronic gravity compensation as well as for the protection of the endpositions. Seeing the drawbacks of using potentiometers (vulnerable system, possible damage in the gearbox of potentiometer difficult to be repair) for absolute position measuring, a new, more simple and user & service friendly method has been implemented, mainly based on software changes. After manufacturing the robot, the axes can be put in a basic position that is obvious and easy to define. Via a comfortable user interface, the manufacturer resets the control, and the resolver counts are saved in a permanent memory. From then on, whenever the manipulator is moved, every millisecond the new position is updated in this permanent memory - when the manipulator is switched off, it cannot be moved (brakes). Due to the nature of the permanent memory even at power break down the absolute position is not lost. The permanent power is supplied by a new accu card in the VME system, guaranteeing no data loss for more than 10 years

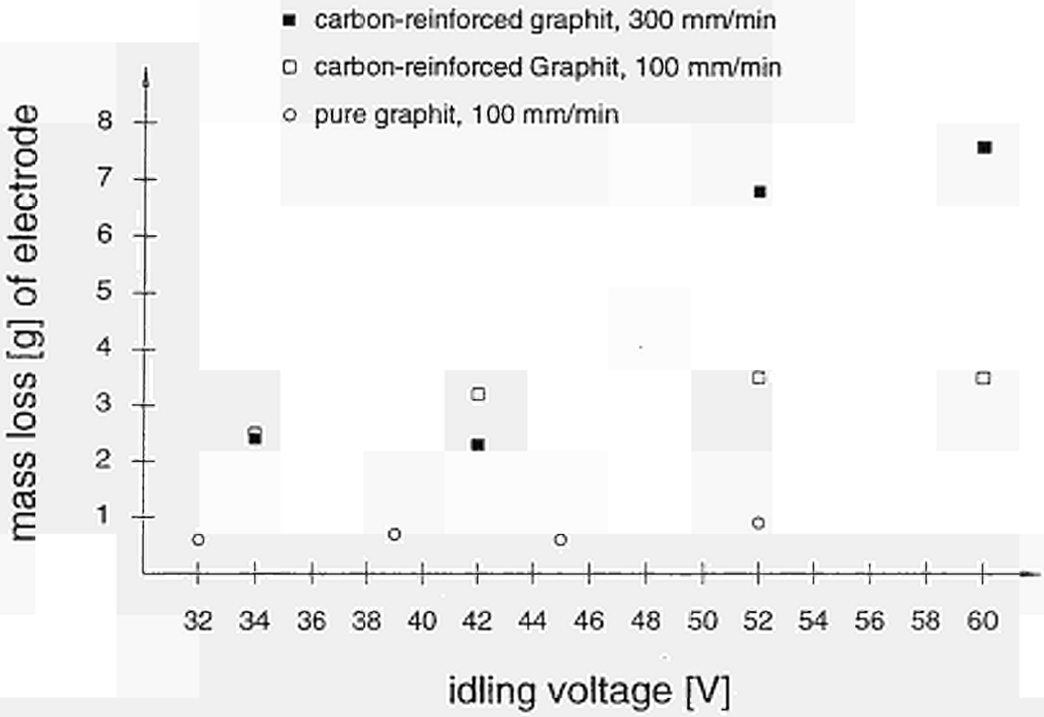


Figure 1: Comparison of the CAMC electrode mass loss between carbon-reinforced graphit and pure graphit; workpiece stainless steel, 20 mm thick

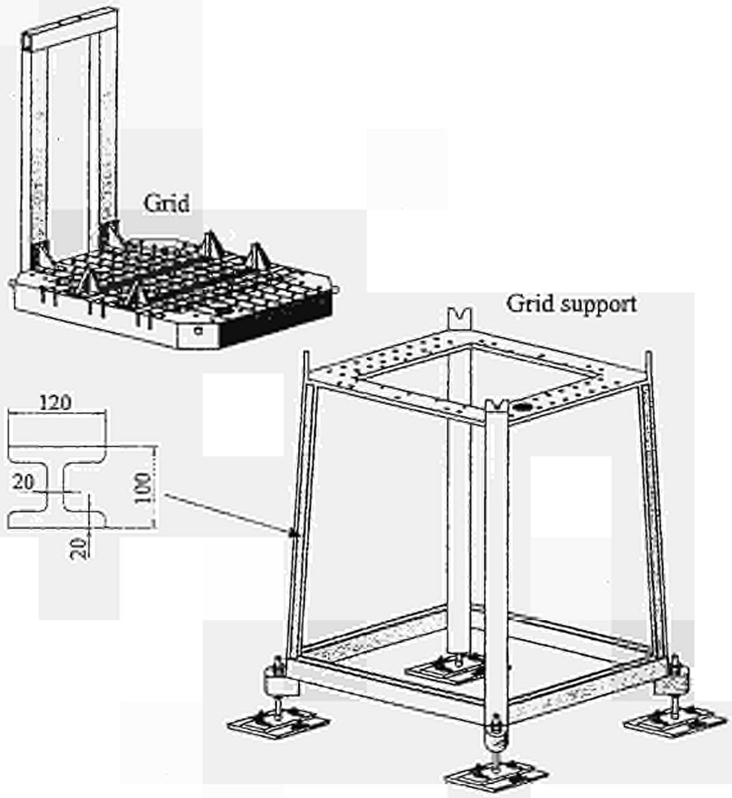


Figure 2: Selected aluminium components grid and grid support

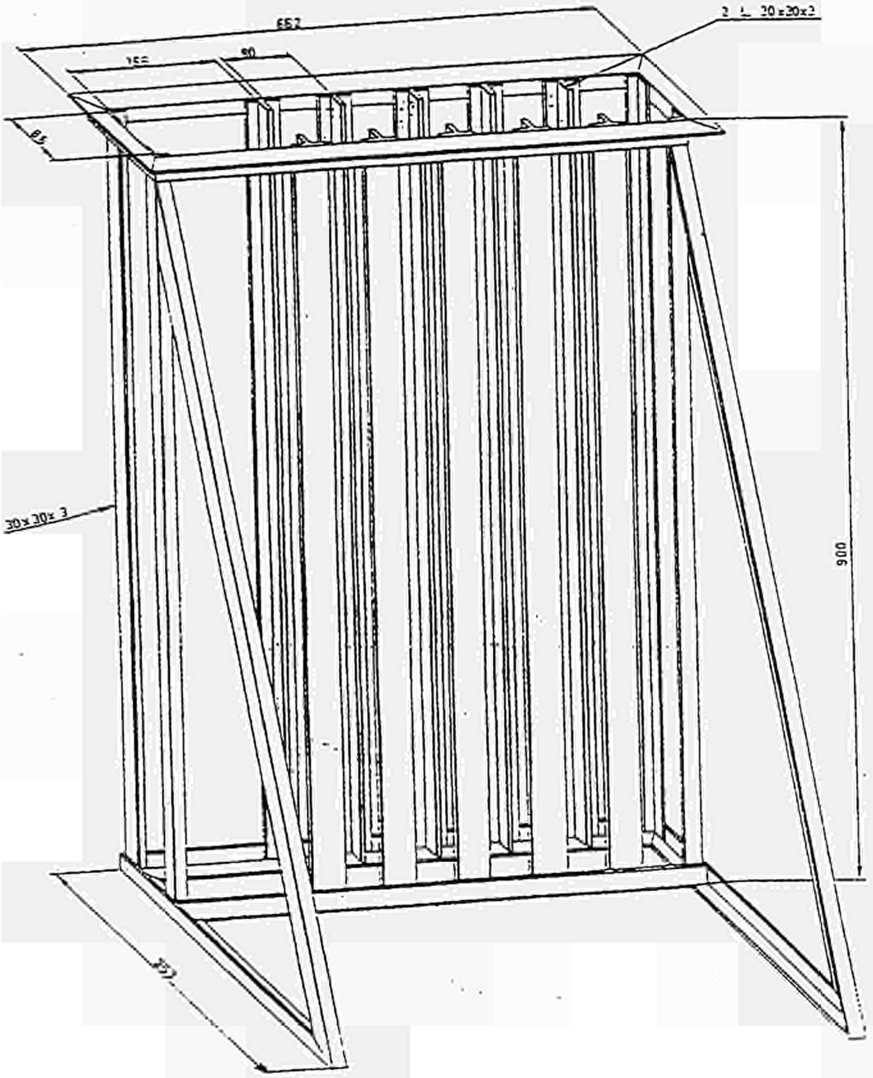


Figure 3: Fuel element rack

Table I: Specific activity of selected aluminium components core grid and grid support

Nuclide	Specific activity [Bq/g] Core grid	Specific activity [Bq/g] Grid support
Fe 55	2.9 E+02	4.3 E+03
Co 60	1.1 E+04	8.7 E+02
Ni 63	5.5 E+01	7.2 E+02
Nb 93	-	5.8 E+03
Cs 137	5.7 E+01	1.4 E+01
Sr 89	-	6.8 E+00
Sr 90	5.3 E+01	1.0 E+01
Pu 238	8.0 E-01	-
Pu 239	2.6 E+00	4.0 E-01
Am 241	4.0 E-01	-

C.4.2-1 Further development of the EC-DB-TOOL data base and collection of technological performance data from the decommissioning of nuclear installations

Contract No.: FI4D-CT95-0004	**Duration:** 1 Jan. 1996 - 31 Jan. 1999
Coordinator:	**F.W. Bach, Institut für Werkstoffkunde (IW), Universität Hannover/DE** Tel. +49/511/762 43 16 Fax: +49/511/762 52 45
Partners:	**CEA-UDIN Valrhô/FR, AEA Tech Harwell/GB, CIEMAT Madrid/ES, ANPA Roma/IT, EWN Lubmin/DE, FRAMATOME Paris La Défense/FR**

A. OBJECTIVES AND SCOPE

Data bases are suitable for storing and evaluating decommissioning experience.

The objective of this project is to continue, improve and extend the data base on cutting techniques and associated filter systems *EC-DB-TOOL* initiated in the previous programme (contract FI2D-0056) to decontamination and recycling techniques, as well as to VVER reactor data. To improve EU-wide use, the English users' vocabulary will be translated into French, German, Italian and Spanish.

The data base development will continue with the mouse-driven ORACLE 7 software at Universität Hannover. Other PC-friendly and possibly cheaper softwares will be assessed in parallel (e.g. with MS ACCESS).

B. WORK PROGRAMME

B.1. Further development of EC-DB-TOOL
B.1.1. Data base upgrade (Uni. Hannover)
B.1.2. Structure extension for decontamination and recycling techniques (Uni. Hannover)
B.1.3. Assessment of PC-friendly interface software (Uni. Hannover)
B.1.4. Translation of data base vocabulary (Uni. Hannover, CEA, ANPA, CIEMAT)
B.1.5. Connection of EC DB TOOL to EC DB COST (Uni. Hannover, NIS, EWN)

B.2. Data collection
B.2.1. Data forms for new techniques (Uni. Hannover, Framatome, CIEMAT)
B.2.2. Data collection (all)

B.3. Revising and updating data (all)

B.4. Initiation of the EU-wide use of the data base
B.4.1. EU wide access (Uni. Hannover)
B.4.2. Questionnaire for comments (Uni. Hannover)

B.5. Documentation (Uni. Hannover, CEA, UKAEA, CIEMAT, ANPA)

B.6. EU decommissioning network for systematic exchange of experience and data (EC, all)

C. Progress of work and results obtained

Summary of main issues

The database has been upgraded to the current ORACLE 7 software (B.1.1). The data forms need to be constantly improved (see also B.3.2).

A concept for database tables to extend EC DB TOOL (B.1.2) to recycling and decontamination techniques has been developed .

Other interface software to access the Oracle database has been considered and a Visual Basic user interface chosen for an evaluation. (B.1.4).

A concept for the connection of the EC DB COST and EC DB TOOL has been worked out and implemented (B.1.5).

The database vocabulary was translated into Spanish and inplemented in the database (B.1.6).

A central issue of the project is the data collection and input of data into the database (B.2.1). All partners responsible for the data collection have started to identify data sources and some of them have already provided data sheets.

Data collection sheets for melting (considered as a recycling technique) and decontamination techniques have been developed (B.2.2).

Possible solutions for the establishment of a EU-wide access to EC DB COST and EC DB TOOL for data input and retrieval were worked out and are now under evaluation (B.4.1). A special attention is given to data considered as confidential and to how institutions outside this project can be addressed to provide data.

Progress and results

1. Further development of EC DB TOOL (B.1)

Database upgrade (B.1.1)

The EC DB TOOL database has been upgraded to the current ORACLE 7 version V7.3.2. The existing ORACLE Forms 3.0 user interface forms have been migrated to ORACLE Forms 4.5. The new forms on Unix workstations like the SunSparcstation are now based on the Motif graphical user interface (Figure 1). By this, the user friendliness was efficiently improved. The development of EC DB TOOL version 2.1 was finished lately. The forms are now being tested. They are available on a PC with Windows 3.x, too (Figure 2). Both versions are compatible.

Structure extension for decontamination and recycling techniques (B.1.2)

Based on the data questionnaires developed by CIEMAT and FRAMATOME, a first concept for a table structure has been developed by Uni. Hannover.

EC DB TOOL with other interface software (B.1.4)

It was agreed by UKAEA, CEA and Uni. Hannover that Visual Basic forms (based on the concept shown in Figure 3) will be developed by UKAEA which would emulate the existing Oracle input forms. A prototype form was developed and sent to Uni. Hannover for trials. The Visual Basic solution also needs an Oracle driver on the client machine to connect to the central database. The data access is slowed down by the use of the Visual Basic solution.

Connection of EC DB TOOL and EC DB COST (B.1.5)

A concept for the connection of the EC DB COST and EC DB TOOL databases was worked out by NIS and Uni. Hannover as shown in Figure 4. It comprises a table that assigns a dismantling task identification number to a combination of a working step and working package identification number as well as a form to make the assignment. This concept was implemented at Uni. Hannover.

Translation (B.1.6)

The partners responsible for translating the vocabulary into Spanish, Italian and French have been provided either with an Oracle form (Figure 5) as well as a database export file or a list with relevant terms. CIEMAT carried out the translation from English into Spanish of a first set of about five hundred terms. This translation was sent to the Coordinator by electronic mail and imported into EC DB TOOL.

2. Data collection (B.2)

Continuation of data collection (B.2.1)

Uni. Hannover has reviewed and analysed data from the last technical reports from former EU R&D programmes, final reports and literature. The data has been included in the database.

At UKAEA a strategy for the collection of data was formulated and a programme of work agreed upon for the forthcoming period.

CIEMAT began this activity with the collection of decommissioning data of its own installations, since these are the most important ones within the installations in process of decommissioning in Spain.

The activity carried out by ANPA in this first year was mainly addressed to the collection of information on the relevant decommissiong data in Italy. The collected information is now being treated in order to be suitably put into the EC DB TOOL data sheets.

According to the contract the main task of the EWN in 1996 was the collection of data. Available technical information from the decommissioning of the Greifswald Unit 5 was collected and put into the data collection sheets.

CEA has scrutinized the documentation arising from the AT1 decommissioning project. This search provided the data to establish 18 sets of data collection sheets in French on the cutting tools used. These have not yet been translated into English.

Data sheets concerning SODP decontamination on Dampierre and Agesta Steam generators, and EMMAC decontamination on Gravelines 2 primary pipes were filled in by FRAMATOME. These sheets are being prepared and will be submitted in I/97.

Data questionnaires for new techniques (B.2.2)

CIEMAT has prepared a data questionnaire for melting (considered as a recycling technique). CIEMAT made the revision of the questionnaires, jointly with FRAMATOME and CEA. The questionnaires were sent to all partners for comments.

FRAMATOME had in the first half year established a preliminary questionnaire and collected information related to the decontamination processes for maintenance or dismantling operations. In the second half year, FRAMATOME prepared the data questionnaires on decontamination techniques. These were revised after some comments from CEA.

3. Revising and updating the data (B.3)

Use of new features on present data (B.3.2)

see database upgrade

4. Initiation of an EU-wide network for a systematic data exchange (B.4)

EU-wide access (B.4.1)

Possible solutions for the establishment of an EU-wide connection to both databases for data input and retrieval have been worked out by NIS and Uni. Hannover. The solutions which will be examined in the ongoing work programme are as follows:

- access via Internet (TCP/ IP protocol)

- access via Network programmes via ISDN

Heart of this is a client/server concept consisting of a central database server at Hannover, providing access to the most current data (Figure 6) and making the database administrable. Any partner of the users' group will be able to connect his client computer located anywhere in Europe via Internet or ISDN to the server to retrieve and add data.

CIEMAT initiated the on-line connection to EC DB TOOL at Hannover. There were several contacts and information exchange with the Coordinator concerning the connection.

EU-wide users' group (B.4.2)

The PR-paper was revised and adapted to the current EC DB TOOL version by Uni Hannover. Furthermore, Uni. Hannover participated in a poster session at the Annual Meeting on Nuclear Technology '96 in Mannheim and presented EC DB TOOL (Ref. [1]).

Security aspects to assure that data which is considered confidential can only be accessed by authorized users were studied and are now under evaluation; first steps were taken to constitute a EU Users' Group.

References

[1] HÜSKE, M., BACH, FR.-W., Annual Meeting on Nuclear Technology '96, Mannheim, 21-23 May 1996, Proceedings, pp. 587-590.

Figure 1: Current ORACLE Forms 4.5 „Dismantling-Task"-Form on Motif 1.3 - GUI

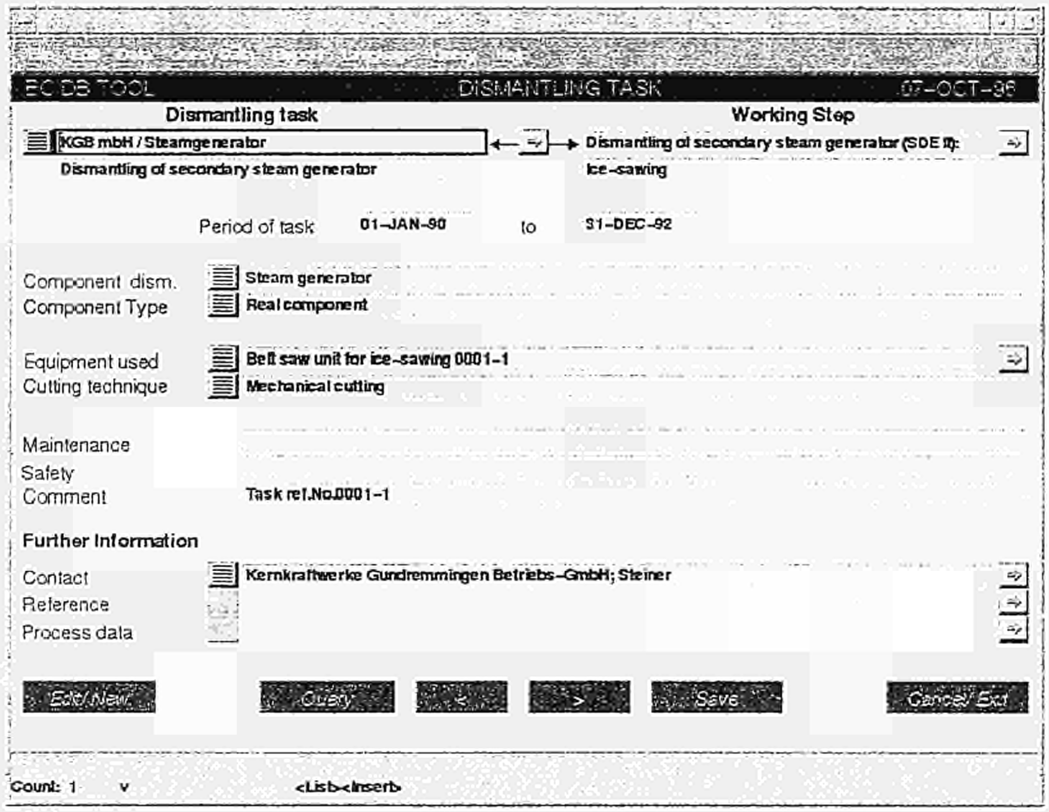


Figure 2: Current ORACLE Forms 4.5 „Dismantling Task"-Form on Windows 3.x

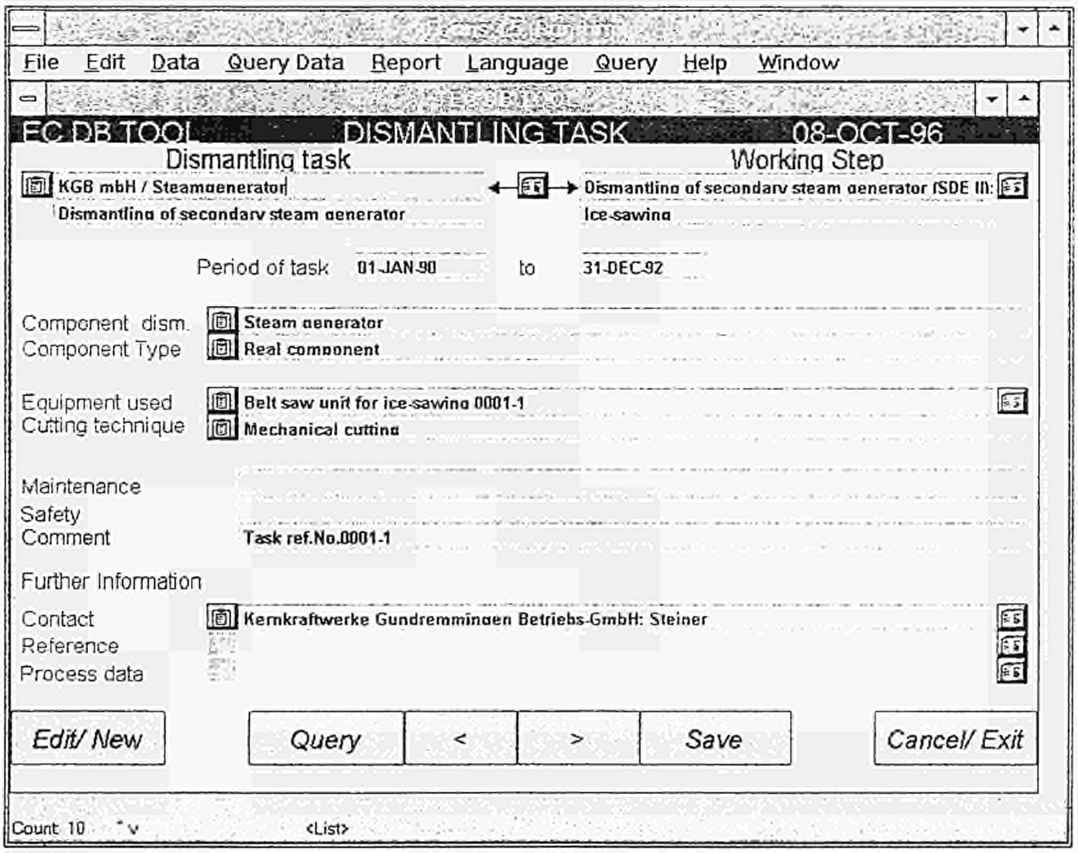


Figure 3: Implementation using Visual Basic

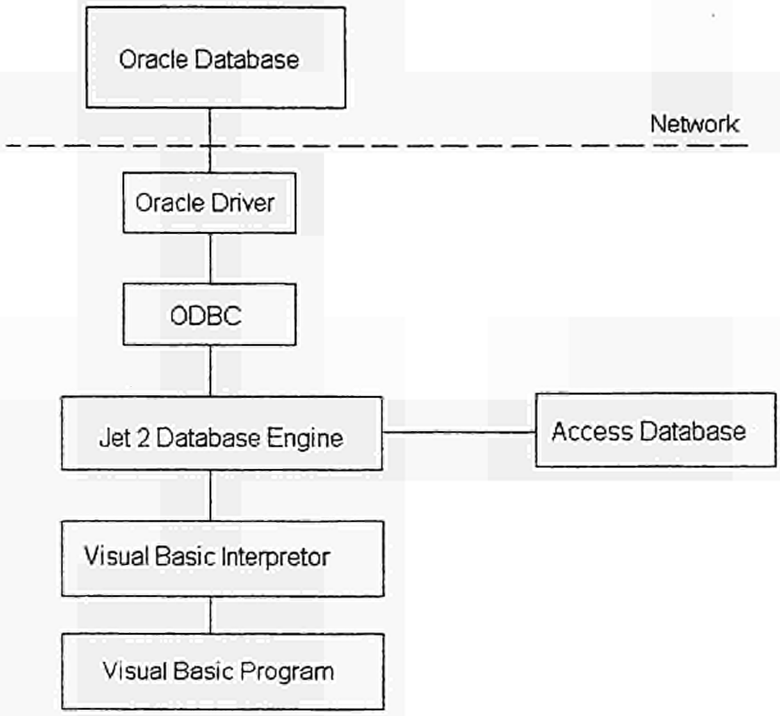


Figure 4: Concept for connecting EC DB COST and EC DB TOOL

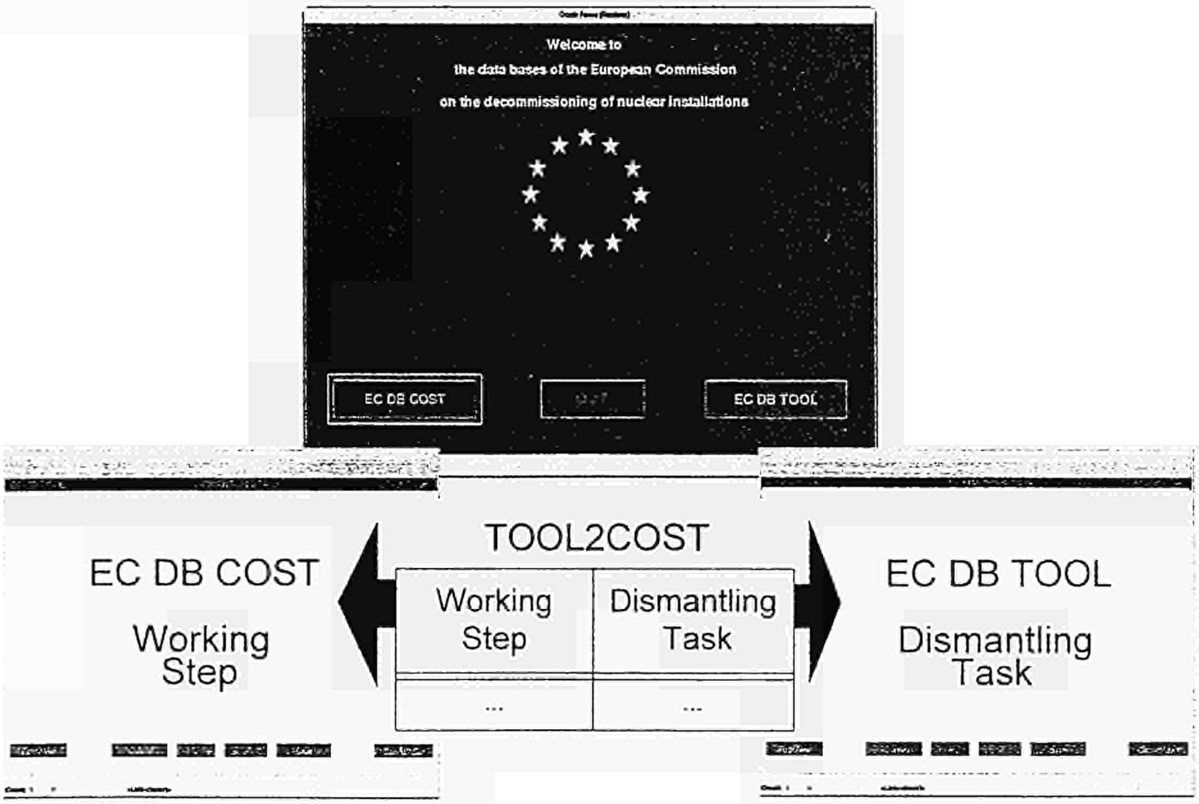
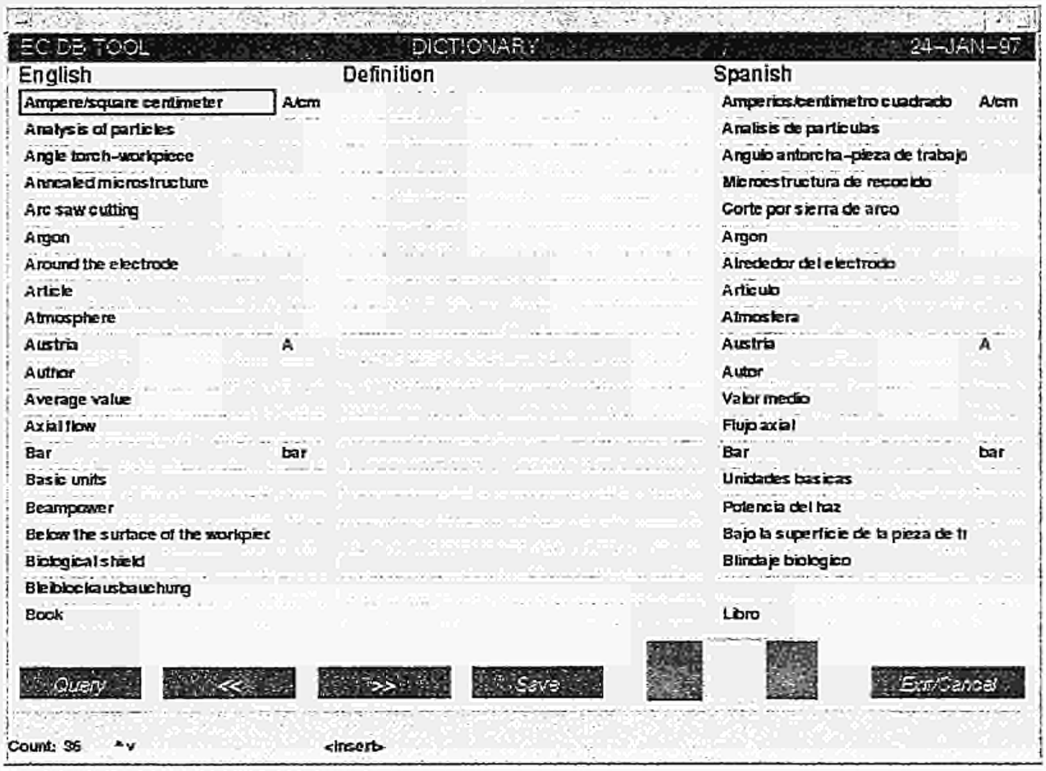


Figure 5: Form for the translation of the terms into Spanish



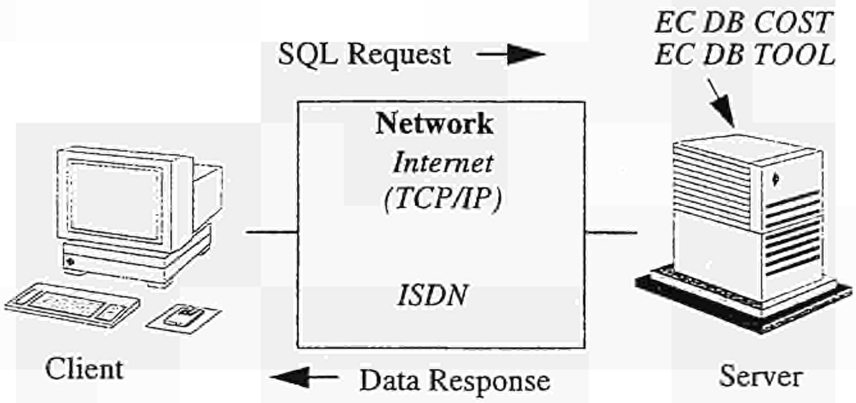
EC DB TOOL — DICTIONARY — 24-JAN-97

English		Definition	Spanish	
Ampere/square centimeter	A/cm		Amperios/centimetro cuadrado	A/cm
Analysis of particles			Analisis de particulas	
Angle torch-workpiece			Angulo antorcha-pieza de trabajo	
Annealed microstructure			Microestructura de recocido	
Arc saw cutting			Corte por sierra de arco	
Argon			Argon	
Around the electrode			Alrededor del electrodo	
Article			Articulo	
Atmosphere			Atmosfera	
Austria	A		Austria	A
Author			Autor	
Average value			Valor medio	
Axial flow			Flujo axial	
Bar	bar		Bar	bar
Basic units			Unidades basicas	
Beampower			Potencia del haz	
Below the surface of the workpiec			Bajo la superficie de la pieza de tr	
Biological shield			Blindaje biologico	
Bleiblockausbauchung				
Book			Libro	

Query | << | >> | Save | Exit/Cancel

Count: 36 <insert>

Figure 6: Possible network connections to EC DB COST and EC DB TOOL



C.4.3-1 Further development of the EC-DB-COST and collection of data on costs, radiation exposure and waste from the decommissioning of nuclear installations

Contract No.: FI4D-CT95-0003	**Duration:** 1 Jan. 1996 - 31 Dec. 1998
Coordinator:	**P. Petrasch, NIS GmbH Hanau/DE** Tel. +49/6181/18 51 44 Fax: +49/6181/12 00 33
Partners:	**CEA-UDIN Valrhô/FR, BNFL Risley/GB, ONDRAF/NIRAS Brussels/BE, CIEMAT Madrid/ES, EWN Lubmin/DE, FRAMATOME Paris La Défense/FR, ANPA Roma/IT**

A. OBJECTIVES AND SCOPE

Data bases are suitable for storing and evaluating decommissioning experience, and will facilitate cooperation and information exchange between organisations and persons involved in the decommissioning of nuclear installations.

The objective of this project is to continue, improve and extend the data base on cost and dose uptake *EC-DB-COST* initiated in the previous programme (contract FI2D-0059) to waste streams, recycling techniques and VVER reactor data. To improve EU-wide use, the English users' vocabulary will be translated into French, German, Italian and Spanish.

The data base will be upgraded to the mouse-driven ORACLE 7 software. Other PC-friendly and possibly cheaper data base software, eg MS-ACCESS, will be assessed in parallel.

B. WORK PROGRAMME

B.1 Further development of EC-DB-COST
B.1.1 Data base upgrade (NIS)
B.1.2 Structure extension to decommissioning masses (NIS)
B.1.3 EU-wide user connection concept (all)
B.1.4 Using EC DB COST with other interface software systems (NIS)
B.1.5 Connecting of EC-DB-COST to EC-DB-TOOL (NIS, EWN)

B.2 Continuation and improvement of data collection, input (all)

B.3 Revising and updating data (NIS, partners)

B.4 Evaluation of the data stored in EC-DB-COST
B.4.1 Principles of cost and radiation exposure estimates (NIS)
B.4.2 Minimization of radioactive waste (all)

B.5 Initiating a EU-wide users'group and testing of data base by selected users (all)

B.6 Documentation and users'guide
B.6.1 Preparation of the data base documentation (NIS, all)
B.6.2 Preparation of the users'guide (NIS, all)
B.6.3 Implementation of an on-line help system (NIS, all)
B.6.4 Testing the users'guide. (All)

B.7 Decommissioning network for systematic exchange of experience and data (EC, all)

C. PROGRESS OF WORK AND OBTAINED RESULTS

Summary of main issues

The activities carried out during the year 1996 mainly include the tasks B.1.1, B.1.3., B.2.1. and B.2.2. i.e.:

- establishing of a data collection strategy and continuous data collection from decommissioning projects
- improvement and acceleration of the data acquisition by revision and simplification of the data collection sheets
- creation of a concept to upgrade EC-DB-COST from ORACLE 6 to ORACLE 7.x version and start of programming
- adaptation of the data collection sheets with respect to the new structure
- creation and discussion of a concept about the integration of EC-DB-COST in a European network and specification of necessary equipment
- creation of an interface as a premise for a connection between the two data bases EC-DB-COST and EC-DB-TOOL to offer a complete system.

Progress and results

1. Data base upgrade (B.1.1.)

Activities have been started to implement the Relational Database Management System (RDBMS) ORACLE 7.x. This system is more user-friendly and powerful than ORACLE 6 version and the implementation is the first step for the EU-wide connection to the data base.

A new data base structure has been built up as the first step for an upgrade of the data base. The data collection sheets were revised and adapted with respect to the new design.

A new entity relationship model was created showing the elements of information and the way they are logically related. Also a new function hierarchy was created to enable the design of the application.

The implementation of the ORACLE 7.x RDBMS will be continued and should be finished during the course of the next year.

2. EU-wide user connection concept (B.1.3.)

In collaboration with the University of Hannover a concept for a quick and easy EU-wide access to EC-DB-COST was elaborated and all technical solutions with specification of necessary equipment were examined.

In the reporting period some major questions about an EU-wide use of the data base were discussed, such as:

- security aspects of the data
- maintenance of the network and of the data
- effort for the administration
- access to the data
- consistency of the data.

Moreover the acquisition cost for the necessary equipment to realise or facilitate the connection to the EC data base are determined. For this purpose a meeting was held in Brussels on 4 June 1996 with all partners.

After careful considerations, the partners selected two of before elaborated solutions which satisfy best the requirements of the contract taking into account the existing network connections and the acquisition cost for necessary equipment and first of all consistency and security of the data:

- central data base with data exchange via Internet
- central data base with data exchange via network.

In cooperation with the University of Hannover a translation form has been designed for each partner in charge of the translation of the vocabulary into his language. Clear and understandable screen masks will make the data base more user-friendly.

3. <u>Connection of EC-DB-COST and EC-DB-TOOL (B.1.5.)</u>

In cooperation with the University of Hannover, an interface between the two data bases was defined and a common menu was designed. Table TOOL2COST was set up, that assigns a task-Identifier (ID) to a combination of the working step and working package.

A form to make the assignment has been designed. At this stage this form can be called from the"Dismantling Task" by pressing a button. The data base user will be provided with a starting form from which he can either switch to EC-DB-COST or to EC-DB-TOOL.

4. <u>Continuation of data collection (B.2.1.)</u>

The data collection is the main tasks for all partners involved in the EC-DB-COST project. The data collection concerns:

- informations about the strategy on decommissioning of representative nuclear installations
- national regulations, acts and rules
- list of nuclear installations in operation as well as decommissioning projects, their main characteristics, start-up date and their present status
- decommissioning activities in individual plants and data arising from decommissioning projects, including relevant decommissioning masses and specific costs related to a working step or working package and relevant decommissioning masses.

To coordinate and improve the data collection a meeting was held in Hanau on 21 October 1996 with the partners.

5. <u>Improvement of the data collection (B.2.2.)</u>

To improve and accelerate the data acquisition, the data collection sheets were revised and simplified.

Moreover an instruction was prepared for the completion of the decommissioning data collection sheets. In this instruction is described the background of a working step and a working package.

The detailed information on decommissioning activities will be described in individual working steps which are the smallest calculable units of the decommissioning task.

This information is very extensive, because the underlying idea is to make a decommissioning process understandable and transparent from the first planning activities to the disposal of the dismantled components.

The data describing a working step consists of:

- description of the component dealt with in the working step
- description of the working step
- description of the technique used
- description of the workload, number of persons involved in this working step and duration of the work
- description of radiation exposure, local dose, internal and external exposure
- description of equipment and expendables used and their costs
- description of the disposal method for this component

- volume and disposal of secondary waste
- indication of the source of information.

All working steps will be assigned to working packages, because of the classification of the work. These working packages are:

- Planning and project management
- Licensing procedure
- Operation
- New installation
- Decontamination
- Dismantling
- Waste management
- Radiation protection
- Site recovery
- Research and development

The report „Collection of decommissioning cost data-Instructions for the completion of decommissioning data collection sheets" shows the content of each working package.

The data base was extended to decontamination and recycling techniques. For this purpose new data questionnaires were created.

In the following contractual period the main task for all partners involved in the EC-DB-COST project remains to provide decommissioning project data. Because most of the specific costs in EC-DB-COST are related to a mass, it is necessary to add relevant data on decommissioning waste masses. A concept to add the decommissioning masses should therefore be created in the next reporting period.

List of Contracts bv number

List of Contractors and Partners

AEA Techn.	AEA (UKAEA), Dorchester, Harwell, Risley, Winfrith	GB
AIB-VIN	AIB-Vinçotte Nuclear asbl-DAS, Brussels	BE
AITEMIN	Asociación de Investigación Tecnológica de Equipos Mineros, Madrid	ES
ANDRA	Agence Nationale pour le Gestion des Déchets Radioactifs, Chatenay-Malabry	FR
ANPA	Agenzia Nazionale per la Protezione dell'Ambiente, Roma	IT
ANSALDO	Ansaldo SpA, Finmeccanica - Div. nucleare, Genova	IT
ARMINES	Ass. pour la Recherche et le Développement de Méthodes et Processus Industriels, Paris, Fontainebleau	FR
BAM	Bundesanstalt für Materialforschung und Materialprüfung, Berlin	DE
BATTELLE	Battelle Ingenieurtechnik GmbH, Eschborn	DE
BELGOP.	Belgoprocess NV, Dessel	BE
BfS	Bundesamt für Strahlenschutz, Oberschleibheim	DE
BGR	Bundesanstalt für Geowissenschaften u. Rohstoffe, Hannover	DE
BGS	British Geological Survey, Keyworth	GB
BN	Belgonucléaire S.A., Brussels	BE
BNFL	British Nuclear Fuel Plc, Risley, Sellafield	GB
BRGM	Bureau de Recherches Géologiques et Minières, Orléans	FR
CAMFIL	Camfil SA, Pont Ste. Maxence	FR
CEA	Commissariat à l' Energie Atomique, Paris	FR
CEA-Cadarache	CEA, Centre de Cadarache	FR
CEA-FAR	CEA, Centre de Fontenay-aux-Roses	FR
CEA-Grenoble	CEA, Centre de Grenoble	FR
CEA-Saclay	CEA, Centre de Saclay	FR
CEA-Valduc	CEA, Centre de Valduc, Is-Sur-Tille	FR
CEA-Valrhô	CEA, Centre de la Vallée du Rhône	FR
CEA-IPSN	CEA, Inst. de Protection et de Sûreté Nucléaire, Clamart, Cadarache, Saclay	FR
CEA-UDIN	CEA, Unité Démantellement des Installations Nucléaires, Valrhô	FR
CEN/SCK	Centre d' Etude de l' Energie Nucléaire / Studiecentrum voor Kernenergie, Mol	BE
CERN	Conseil Européen pour le Recherche Nucléaire, Genève	CH
CIEMAT	Centro de Investigaciones Energeticas, Medio Ambientales y Tecnologicas, Madrid	ES
CIMNE	Centro Internacional de Métodos Numéricos en Ingeniería - UPC, Barcelona	ES
CIRTEN	Consorzio Interuniversitario per la Ricerca Tecnologica Nucleare, Pisa	IT
CISE	Centro Informazioni Studied Esperienze, Milano-Segrate	IT
CLAY TECH	Clay Technology Lund AB, Lund	SE
CNRS	Centre National de la Recherche Scientifique, Orsay, Grenoble, Strasbourg & Paris	FR
CONTERRA AB	Conterra AB, Uppsala	SE
CROSFIELD	Crosfield Ltd., Warrington	GB
EA	Empresario Agrupados Internacional SA, Madrid	ES
ECN	Stichting Energieonderzoek Centrum Nederland, Petten	NL
ECPM	Université Louis Pasteur - Ecole Européenne de Chimie, Polymères et Matériaux, Strasbourg	FR
EDF	Electricité de France, Paris	FR
EDF-Septen	Electricité de France - Service Etudes et Projets Thermiques et Nucléaires, Paris	FR

ENEA	Ente per le Nuove tecnologie, l'Energia e l'Ambiente, Bologna, Casaccia, Roma	IT
ENEL	Ente Nazionale per l'Energia Elettrica SpA, Roma, Milano	IT
ENRESA	Empresa Nacional de Residuos Radioactivos SA, Madrid	ES
ETREMAT	Etude-Recherches-Matériaux, Civaux	FR
EWN	Energiewerke Nord GmbH, Lubmin	DE
FRAMATOME	Framatome SA, Paris La Défense	FR
FZK	Forschungszentrum Karlsruhe GmbH, Karlsruhe	DE
G.3S	Groupement pour l'Etude des Structures Souterraines de Stockage, Palaiseau	FR
GKSS	Forschungszentrum Geesthocht GmbH, Geesthacht	DE
GNS	Gesellschaft für Nuklear-Service mbH, Essen	DE
GRS	Gesellschaft für Anlagen und Reaktorsicherheit mbH, Köln, Braunschweig	DE
GSF	Forschungszentrum für Umwelt und Gesundheit GmbH, Neuherberg	DE
GSFIN	Geological Survey of Finland, Espoo	FI
HSK	Hauptabteilung für Sicherheit der Kernanlagen, Villigen	CH
INMF	Istituto Nazionale per la Fisica delle Materia, Legnaro	IT
ISMES	Istituto Sperimentale Modelli e Strutture Spa, Bergamo	IT
ISTec	Institut für Sicherheitstechnologie GmbH, Garching	DE
IVO	Imatran Voima Oy International Ltd, Vantaa	FI
JRC	European Commission, Joint Research Centre	EC
JRC-EI	Environment Institute, Ispra	
JRC-IAM	Institute for Advanced Materials, Petten	
JRC-ISIS	Institute for Systems Informatics and Safety, Ispra	
JRC-ITU	Institute for Transuranium Elements, Karlsruhe	
KEI	Kraftanlagen.Energie-und-Industrieanlagen GmbH, Heidelberg	DE
KEMA	Keuring van Elektrotechnische Materialen BV, Arnhem	NL
KFA	Forschungszentrum Jülich GmbH, Jülich	DE
KRB-A	Kernkraftwerk RWE-Bayernwerk GmbH, Gundremmingen	DE
MAGNOX	Magnox Electric Plc, Berkeley	GB
NAGRA	Nat. Cooperative for Disposal of Radioactive Waste, Wettingen	CH
NCSR	National Centre for Scientific Research "Demokritos", Aghia Paraskevi	GR
NERC-BGS	Natural Environment Research Council - British Geological Survey, Swindon	GB
NIS	Nuklear Ingenieur Service - Ingenieurgesellschaft mbH, Hanau	DE
NNC	National Nuclear Corporation Ltd., Knutsford	GB
NUCELEC	Nuclear Electric Plc, Barnwood	GB
NUCON	Stork Nucon BV, Amsterdam	NL
ONDRAF/NIRAS	Organisme National des Déchets Radioactifs et des Matières Fissiles, Bruxelles	BE
PSI	Paul Scherrer Institut, Villigen	CH
PTB	Physikalische-Technische Bundesanstalt, Berlin	DE
QUANTISCI	QuantiSci (former INTERA), Henley-on-Thames, Cerdanyola	GB
RADIUS	Radius Engineering NV, Gent	BE
RMCE	RMC Environmental Ltd., Abingdon	GB
Rolls-Royce	Rolls-Royce and Associates Ltd, Derby	GB
SCK/CEN	Studiecentrum voor Kernenegie / Centre d' Etude de l' Energie Nucléaire, Mol	BE
Siemens	Siemens AG - Erlangen	DE
Siemens-KWU	Siemens AG - KWU, München	DE
Siempelkamp	Siempelkamp Giesserei GmbH, Krefeld	DE

SIET	Soc. Informazioni Esperienze Termoidrauliche SpA, Piacenza	IT
SINCROTRONE	Soc. Consorzio per Azioni Sincotrone, Trieste	IT
SKB	Svensk Kärnbränslehantering AB, Stockholm	SE
SMHI	The Swedish Meteorological & Hydrological Institute, Norrköping	SE
SSI	Swedish Radiation Protection Institute, Stockholm	SE
STUDMAT	Studsvik Material AB, Nyköping	SE
STUK	Finnish Centre for Radiation and Nuclear Safety, Helsinki	FI
TAYWOOD	Taywood Engineering Ltd., Southall	GB
Tecnatom	Tecnatom SA, Madrid	ES
TVO	Teollisuuden Voima Oy, Olkiluoto	FI
UNESA	Unidad Electrica SA, Madrid	ES
Univ. Aachen	Rheinisch-Westfälische Techn. Hochschüle, Aachen	DE
Univ. Antwerp	Universiteit Instelling Antwerpen, Wilrijk	BE
Univ. Århus	Århus University	DK
Univ. Athens	University Athens	GR
Univ. Autonoma	Universidad Autonoma, Madrid	ES
Univ. Bath	University of Bath	GB
Univ. Birmingham	University of Birmingham	GB
Univ. Chalmers	Chalmers University of Technology, Goeteborg	SE
Univ. Dresden	University of Technology, Inst. of Power Eng., Dresden	DE
Univ. Exeter	University of Exeter	GB
Univ. FUB	Frei Universität Berlin	DE
Univ. Hannover	Universität Hannover	DE
Univ. Helsinki	University of Helsinki	FI
Univ. Innsbruck	Leopold-Franzens-Universität, Innsbruck	AT
Univ. KUL	Katholieke Universiteit Leuven, Leuven	BE
Univ. KTH	Kungliga Tekniska Högskolan, Stockholm	SE
Univ. La Coruña	Universidad de Coruña, La Coruña	ES
Univ. Lappeenranta	University of Technology, Lappeenranta	FI
Univ. "La Sapienza"	Università degli Studi "La Sapienza", Roma	IT
Univ. Liège	Université de Liège	BE
Univ. Lough.	Loughborough University	GB
Univ. Mainz	J. Gutenberg Universität, Mainz	DE
Univ. Nantes	Université de Nantes	FR
Univ. Oviedo	Universidad de Oviedo	ES
Univ. Parma	Università degli Studi, Parma	IT
Univ. Pisa	Università degli Studi, Pisa	IT
Univ. Politèc.	Universidad Politècnica, Madrid	ES
Univ. Politèc.	Universidad Politècnica, Valencia	ES
Univ. "Politecnico"	Politecnico, Milano	IT
Univ. "Provence"	Université de Provence, Aix-Marseille	FR
Univ. Reading	University of Reading	GB
Univ. Ruhr	Universität Ruhr, Bochum	DE
Univ. Salford	University of Salford	GB
Univ. Stockholm	Stockholms Universitet	SE
Univ. Strasbourg	Université L. Pasteur, Strasbourg	FR
Univ. IKE Stuttgart	Universität Stuttgart - Inst. Kernenergie (IKE)	DE
Univ. Thessaloniki	University Aristotle, Thessaloniki	GR
Univ. TUM	Technische Universität, München	DE
Univ. Twente	Universiteit Twente, Enschede	NL
Univ. UPC	Universidad Politècnica de Cataluña, Barcelona	ES
Univ. Uppsala	Uppsala University	SE
Univ. UWCC	University of Wales College of Cardiff	GB

Univ. Valencia	Universidad Politècnica de Valencia	ES
Univ. "Victoria"	Victoria University - School of Engineering, Manchester	GB
VAMCE	Voest Alpine Machinery Construction and Engineering AG, Linz	AT
VE	Vattenfall Energisystem AB, Stockholm	SE
VTT	Technical Research Centre of Finland, Espoo	FI

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CORDIS represents a central source of information crucial for any organisation - be it industry, small and medium-sized enterprises, research organisations or universities - wishing to participate in the exploitation of research results, participate in EU funded science and technology programmes and/or seek partnerships.

CORDIS makes information available to the public through a collection of databases. The databases cover research programmes and projects from their preparatory stages through to their execution and final publication of results. A daily news service provides up-to-date information on EU research activities including calls for proposals, events, publications and tenders as well as progress and results of research and development programmes. A partner search facility allows users to register their own details on the database as well as search for potential partners. Other databases cover Commission documents, contact information and relevant publications as well as acronyms and abbreviations.

By becoming a user of CORDIS you have the possibility to:

- Identify opportunities to manufacture and market new products
- Identify partnerships for research and development
- Identify major players in research projects
- Review research completed and in progress in areas of your interest

The databases - nine in total - are accessible on-line free of charge. As a user-friendly aid for on-line searching, Watch-CORDIS, a Windows-based interface, is available on request. The databases are also available on a CD-ROM. The current databases are:

News (English, German and French version) - Results -
Partners - Projects - Programmes - Publications -
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In December 1994, the Council of Ministers of the European Union adopted the specific programme on nuclear fission safety for the period 1994-98 covering the following areas: A: Exploring innovative approaches; B: Reactor safety; C: Radioactive waste management and disposal and decommissioning; D: Radiological impact on man and the environment; E: Mastering events of the past.

This report covers the progress of the 62 multi-partner research projects running in 1996, for the areas A, B and C of the programme, under the responsibility of European Commission Unit XII/F/5 'Nuclear fuel cycle, radioactive waste'.

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