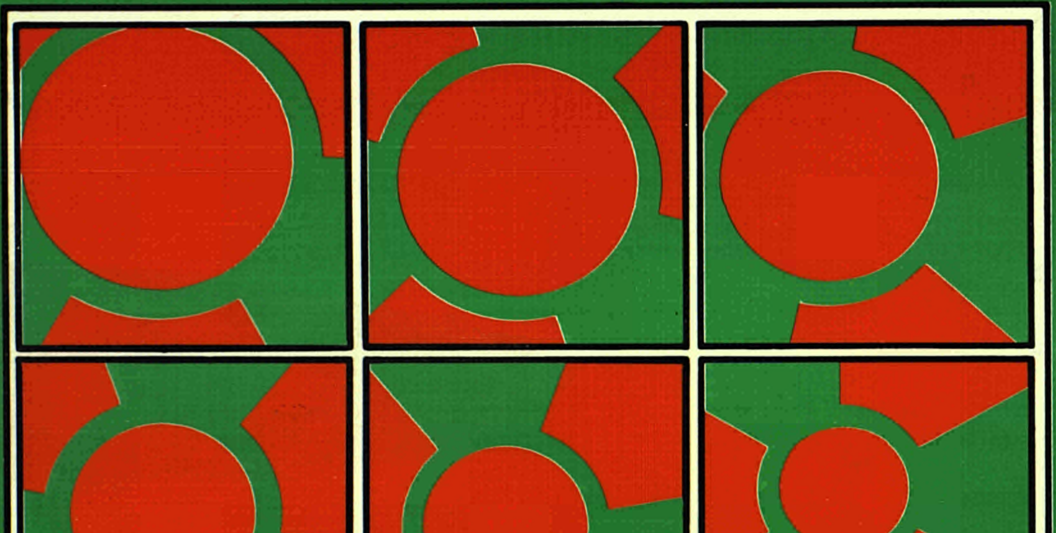
Research and Development on Radioactive Waste Management and Storage

Second Annual Progress Report (1981) of the European Community Programme 1980-1984



RADIOACTIVE WASTE MANAGEMENT
A Series of Monographs and Tracts
Volume 8

harwood academic publishers
for the Commission of the European Communities

RESEARCH AND DEVELOPMENT ON RADIOACTIVE WASTE MANAGEMENT AND STORAGE

Second Annual Progress Report (1981) of the European Community Programme 1980-1984

RADIOACTIVE WASTE MANAGEMENT

A series of monographs and Tracts

Editors : D. R. Anderson, Sandia Laboratories, A. M. Platt, Battelle Pacific Northwest Laboratories, F. Girardi, Joint Research Center, S. Orlowski, Commission of the European Communities

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Preface to the Series

Radioactive waste management is not a new question -- it arose at the very beginning of nuclear energy. Supporting research and management concepts were exposed and discussed within the worldwide scientific community as early as 1955, during the first Conference on the Peaceful Uses of Atomic Energy in Geneva. Since then several management techniques have been developed and put into practice. Specific topics may be found in the proceedings of the numerous national and international conferences held during the last decade.

However, today radioactive waste management has become a matter of great public concern. An acute awareness of the pollution and other potential hazards generally associated with industrial development has to a great extent crystallized during the last years around nuclear energy and waste management. Its final step, the disposal of the waste, introduces a new dimension, i.e., safety over long periods of time. Radioactive waste management is therefore to be viewed as multidisciplinary, involving scientists, engineers, industrialists, lawyers and even specialists in the social sciences and ethics.

It is our hope that this series will provide a permanent record of the development of the various aspects of radioactive waste management which will provide an international overview of the following areas :

- Materiel research
- Waste treatment conditioning and disposal technologies and practices
- Safety and risk assessment
- Economics
- Administrative, legal and financial controls
- Societal aspects

CONTENTS

FOREWORD

This is the second progress report of the European Community's second programme (1980-1984) of research on radioactive waste management and storage (shared expense action). It covers the year 1981 and shows the status of the programme on 31 December 1981.

The Council of the European Communities adopted the second programme in March 1980*, as a follow-up of the first programme (1975-1979), considering:

> "The (first) programme has yielded positive results and opened up couraging prospects of attaining the desired objectives**. The particular nature of the waste is such as to require monitoring of its potential effects and reinforcement of the project and research activities undertaken to ensure the protection of the environment".

The aim of the programme is the joint development and improvement of a management system of radioactive waste produced by the nuclear industry which, at its various stages, ensures the safety and protection of both man and his environment.

The programme covers :

a) Work to solve certain technological problems involved in the processing, storage and disposal of radioactive waste.

   Processing :

   - immobilization of low andintermediate level waste; development of processes and operation of pilot installations;

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* OJ N° L 78, 25.3.1980, p. 22.

** See Annual Progress Reports (EUR 5479, EUR 6128, EUR 6650) and Proceedings of the First European Community Conference, Luxembourg, May 20-23, 1980 (EUR 6871) for the first programme and EUR 7652 for the second one.

- conditioning of high level waste: fuel claddings and residues from dissolvers;
- processing of medium level liquid waste;
- processing of waste contaminated by alpha emitters;
- examination and evaluation of high level solidified waste;
- immobilization and storage of gaseous waste.

Storage and disposal :

- burial of low level solid waste at shallow depth;
- storage and disposal in geological formations.

b) Work to define the general framework for the projects relating to the storage and disposal of radioactive waste.

The programme is carried out by contracts on an expense sharing basis (shared expense action) with qualified public or private bodies in the Community; the Commission's financial participation amounts to 43 millions European Currency Units (ECU)*.

The Commission of responsible for managing the programme and is assisted in this task by an Advisory Committee on Programme Management, which consists of experts appointed by the Member States' governments and of Commission officials **.

The programme is closely coordinated with the activities related to radioactive waste management conducted by the Joint Research Center of the Commission within its pluriannual research programme.

During this second year, the research proposals carefully selected in 1980 for implementation have been worked out into contractu al commitments and the relevant work has begun

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* Conversion rate on 1.7.1981:

1 ECU = 41.3 BFR/LFR = 0.691 IRL
= 7.92 DKR = 1256 LIT
= 2.52 DM = 2.80 HFL
= 61.6 DRA = 0.548 UKL
= 6.02 FF = 1.05 USD

** See Appendix.

accordingly. Most of these contracts being pluriannual, only partial results have been obtained in 1981. They are reported in the following pages as well as the progress of the work. Delays were observed in some countries, as far as in situ experiments for geologic disposal are concerned, due to non-technical difficulties at national level.

A special effort has been devoted to the setting up of concerted activities on special topics like waste form characterisation, container corrosion or coordinated projects like the assessment of the performances of geologic isolation systems. These activities are involving several national laboratories (including the JRC) or research bodies; they are implemented using the appropriate ways and means like round robin tests, common basic materials, common methodologies or a sharing of the work under the control of sub-project leaders.

It is hoped that such an approach will further increase the usefulness and efficiency of the R&D cooperation at Community level.

The professional staff in charge of the management of the programme during 1981 were:

N. CADELLI (part time)
L. CECILLE
B. COME (part time)
A. CRICCHIO (part time)
W. HEBEL
F. REICHARDT
R. SIMON
P. VENET

with the assistance of G. COTTONE, C. EID, J.M. GANDOLFO, B. HAIJTINK, W. KRISCHER and W. FALKE.

S. ORLOWSKI

Head, Division
"Nuclear fuel cycle"

1. WASTE TREATMENT AND CONDITIONING

1.1. CHARACTERIZATION OF CONDITIONED LOW AND MEDIUM ACTIVITY WASTE FORMS

In the original programme description for the current five year programme, activities on testing and evaluation of conditioned waste products other than high level waste, were assigned to separate R&D actions.

During the first phase of the programme all these characterization and evaluation studies were regrouped. Seen the increasing importance of these studies, characterization and evaluation of low and medium activity waste forms are now grouped and described under this chapter. (Process development studies previously included under this heading are reported in chapter 1.3.).

The current investigations concentrate on the testing of the more common industrial waste forms obtained by fixation of reactor and reprocessing concentrates, sludges and ion exchange resins in bitumen, cement, polymer resins and slagging incinerator granules.

In order to allow intercomparison ten reference waste compositions were chosen (Table 1). A coordinated test programme is carried out on these reference waste forms and on six other variants (Table 2).

TABLE 1

REFERENCE WASTE FORMS

WASTE TYPE/REF.	MATRIX	WASTE FORM N°	LEAD LABORATORY (FOR WASTE FORM SPECIFICATION)
BWR - Evaporator concentrate (Garigliano)	Cement	1	CEA - Cadarache
PWR Evaporator concent. (Chooz)	Cement Polyester	2 3	CEA - Cadarache
PWR Ion exchange resin (Chooz (4), EDF (5))	Polyester Polystirene	4 5	
Magnox fuel pond sludge(CEGB)	Cement	6	UKAEA - Harwell
Reprocessing concentr. (Eurochemic (7) WAK (8))	Bitumen Cement	7 8	SCK-CEN, Mol KFK - Karlsruhe
Reprocessing sludges (Marcoule)	Bitumen	9	CEA - Cadarache
Mixed solid waste (SCK-CEN)	Incinerator Slag	10	SCK - CEN , Mol

TABLE 2

EXPERIMENTAL PROGRAMME

1. Effects of waste composition : Homogeneity, waste/matrix compatibility
2. Radiation effects : Radiation damage, radiolysis
3. Leaching :
   - Study of basic mechanism
   - Simulation of disposal conditions
   - Full scale active tests for varying temperature and pressure, leachant composition
   - Characterization of leached nuclides
4. Environmental/climatic influences
   - Microbiological attack
   - Freeze/thaw cycling
   - Ageing effects (excl. radiation)
5. Mechanical properties and shrinking/swelling behaviour
6. Fire consequences and thermal effects
7. Evaluation of test methods : Representativity, reproducibility, scale factors.

1.1.1. ACTIVITY RELEASE FROM CEMENT AND BITUMEN MATRICES

Contractor : KfK, Karlsruhe (240-81-13 WAS.D)

In collaboration with the Institute for Construction Materials Technology comparative testing of the corrosion resistance of various cement products containing concentrated salt solutions runs since 1979. A longterm test programme is to show the effects of cement type, waste/matrix ratio, additives, pre-storage time and temperature on the corrosion resistance in quinary salt brine.

Fig.1 shows the Cs-release versus time of uncompressed cement samples at 40°C and at 1 and 100 bar. Similar results have been achieved for compressed samples. The measurements show that the resistance of the cement under the given conditions is not negatively affected by pressure. At higher temperatures (100°C) only corrosion tests at 1 bar with samples manufactured at various conditions have been conducted. Because during earlier tests it was already noted that the corrosion resistant of cement at elevated temperatures would not be negatively affected by pressure.

The results of investigations carried out so far suggest that the corrosion resistance of cement is independent of the manufacturing conditions.

A series of experiments with mortar products in quinary salt brine and at 90°C was concluded. All tested products were heavily attacked. Before good explanations for this behaviour could be given, further examinations have to follow. A series of experiments with cement samples conducted at 55°C and at 1 and 100 bar, confirmed that the corrosion resistance and Cs-release are not negatively affected by pressure. This implies that the numerous results available from tests carried out below 1 bar are reliable and can serve for safety analysis.

The full scale leaching and corrosion tests of cement products started at KFK in April 1980. The test samples consist of inactive and simulated LLW/MLW-evaporator concentrates immobilised in Portland or blast-furnace slag cement and contained in 200 l drums. Two of the drums were in addition enclosed in lost concrete containers. Leaching tests are being carried out in quinary salt brine at 40°C and 1 bar with six samples in total. The samples are tracered with inactive Cs-salt. The pH-value and the Cs-concentration are measured in the leachant. In addition the Fe concentration is being determined as a measure for drum corrosion.

Neither a change in pH nor any Cs-release have been observed during the first 16 months. However, a precipitate of $Fe(OH)_3$ increasing with time (9-24g/drum) has been identified.

The experiments carried out so far on the activity release during fire test are limited to the determination of Pu release from $NaNO_3$/bitumen product. They comprised Lab-and full scale (200l) tests. Lab scale experiments have been carried out with samples containing approximately 4 wt.% PUO_2. In parallel inactive samples containing Eu_2O_3 as a simulate for PUO_2 were tested and the Eu and Na-releases in the off-gas were measured. Similar tests were performed with 200l drums.

During the fire tests the aerosols are filtered in the off-gas system. The Na and Eu-contents in the filter are being determined. In parallel the Na-content is measured in the ash residues. By comparison of these results with those obtained for Eu, Na and Pu-release in Lab-scale tests, the Pu-release for 200 l drums can be calculated.

The drum samples were exposed in oil fire for 30 minutes. The Eu and Na-content was measured before and after the test by activation analysis.

The lab-scale tests with active samples are meanwhile completed. Final conclusions can only be drawn when the Na-release has been established. However, according to lab-scale tests with Pu-containing samples a maximum Pu-release of 15% can be assumed for unshielded drums.

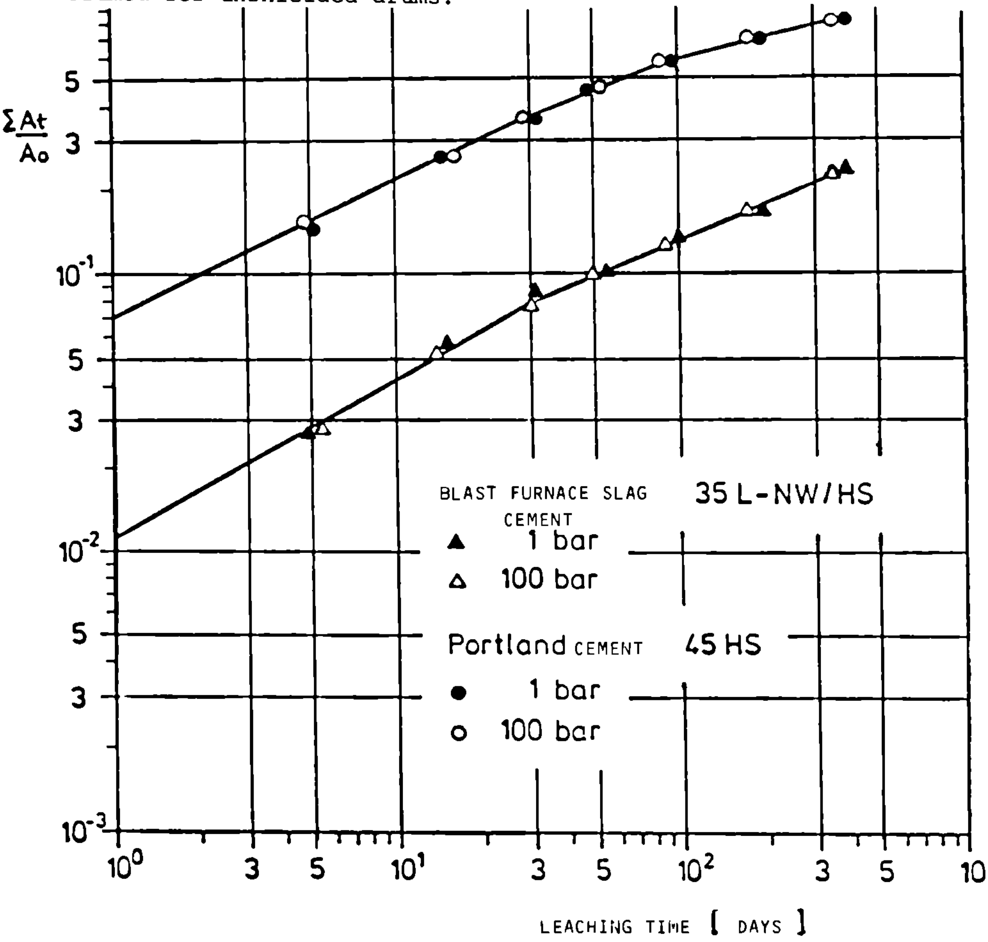

Fig. 1. : Cs-release versus time of uncompressed cement products (W/M=0.32 ; 8 wt% salt) stored in quinary salt brine at 40°C .

1.1.2. LEACHING MECHANISMS OF CEMENT MATRIX

Contractor : University of Aberdeen (241-81-13 WAS.UK)

The activities have initially concentrated on building up apparatus, commissioning equipment and commencing the scientific programme. As many of the experiments are of long duration, no phases of the experimental work are as yet finished.

Construction of a "squeezer", designed to express pore fluids from set cements, is well advanced. It is expected that the equipment will be ready for commissioning trials in February, 1982.

Apparatus based on nitrogen adsorption, for the determination of surface areas should be commissioned in mid-January, 1982 ; its ancillary gas mixing system has been constructed.
The mercury intrusion porosimiter should be commissioned by late January, 1982.

Literature Review and Experimental Studies

A literature review aimed at describing the physicochemical environment prevailing in cements, is being concluded. The search has disclosed a lack of systematic data concerning the behaviour of many important species in the cement environment, and orienting experiments are therefore being conducted to fill in these gaps. In these experiments, soluble salts of some of the principal radwaste species including Sr, Ba, rare earths, Mo, etc..., are being aged in contact with excess $Ca(OH)_2$ in order to determine what fraction of the radwaste species are precipitated, and if extensive precipitation occurs, what the mechanism of precipitation is.

Adsorption Studies of Cs on Cement Constituents

Cement clinker has a complex mineral composition. Some of the minerals present, especially Ca_2SiO_4, have been reported to show adsorptive properties for heavy metal ions even when these ions are in dilute solutions. Cs is one element known not to be chemically precipitated in the alkaline environment of cement liquids. Nevertheless, Cs is absorbed on the cement phases. Therefore, adsorption studies of Cs from solution onto the individual mineral components are being conducted in order to ascertain relationships between mineralogy and adsorption. The relevant mineral phases, including calcium silicates, aluminates and ferites, have been synthesized in phase-pure form and experiments are in progress using non-radio-active Cs. In the near future, these will be complimented by measurements using radioactive Cs. Results obtained thus far show that the adsorptions can be divided into two types of behaviour. Fig. 2 shows an example of Type I adsorption, where the quantity of Cs adsorbed varies smoothly with time. A quasi equilibrium value is attained in ca 30d. A type II adsorption curve is shown in Fig. 3. The adsorption passes through a maximum at relatively short ages, following which desorption occurs. The values obtained for ordinary Portland cement more closely resemble Type I adsorption than Type 2, although they may have some Type 2 character.

Inorganic Ion Exchangers in Cement

The reaction between ion exchange materials and cement is being studied with a view to assess the suitability of cement matrices for their immobilization. These include zeolites of the mordenite, clinoptilolite, ferrierite and chabazite types. These zeolites also affect the rheology of cement-zeolite composites by increasing the water demand ; experiments are

in hand to control the water demand using superplasticizers. A series of cubes made without plasticizer, have been cured for various periods ranging between 30-60 days.
Physical examination and leaching characteristics can now be measured, at least at short ages, and the examinations will be carried on at least over the next six months. The zeolites are being used in their as-received (hydrogen) form, as well as after loading with Cs or Sr ; moreover, the effects of curing conditions, of temperature and of time are being assessed.

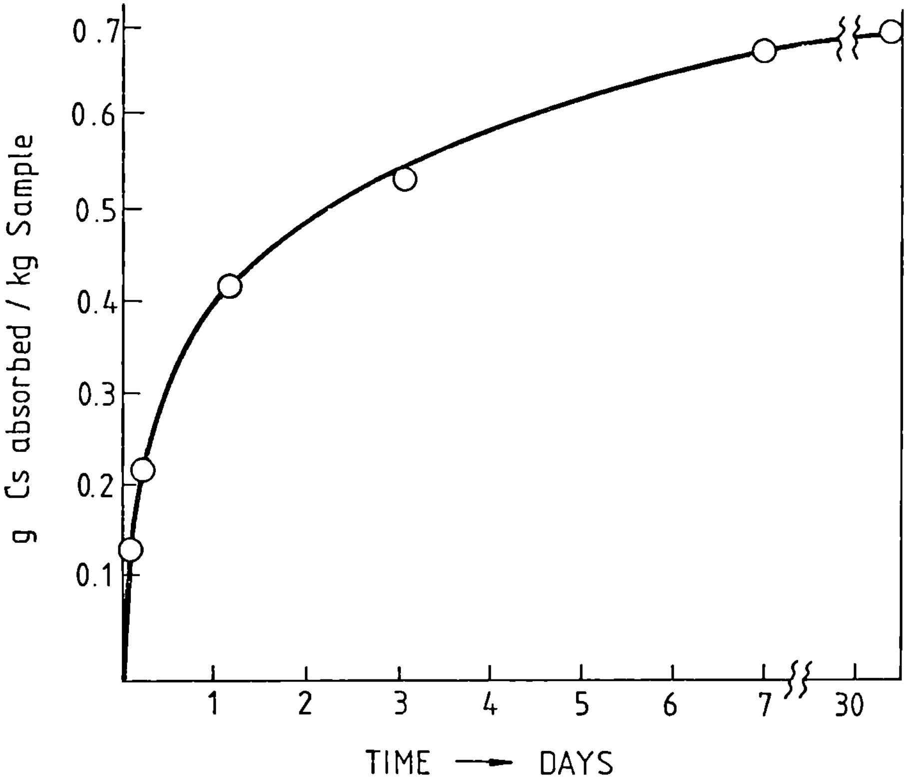

Fig.2 : Absorption of Cs^+ from agitated aqueous solution onto tetracalcium aluminoferrite (4 $CaO.Al_2O_3.Fe_2O_3$). The initial $\{Cs^+\}$ concentration of the solution was 7.8×10^{-4}M and did not change significantly during the course of the experiment. The temperature was 18°C, and the initial surface area of the solid phase 1040 cm^2/g.

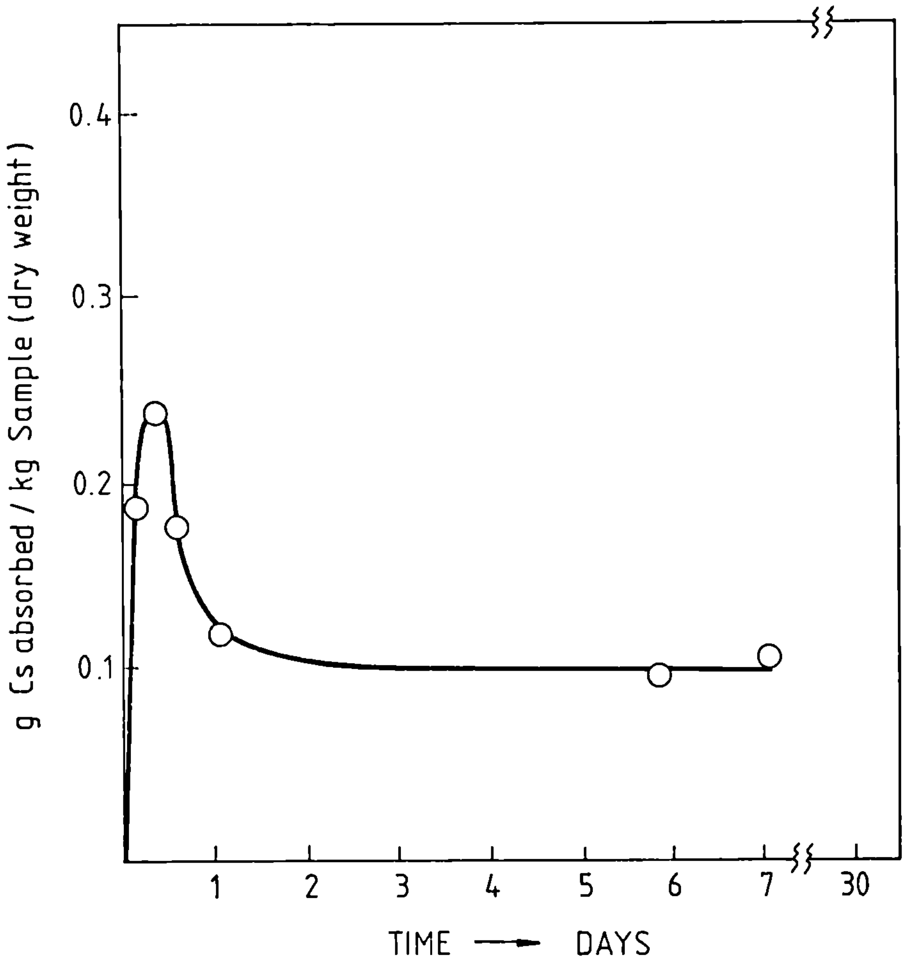

Fig. 3 : Absorption of Cs^+ from agitated aqueous solution onto γCa_2SiO_4. The initial Cs^+ concentration was 7.8 x 10^{-5} M and did not change significantly during the course of the experiment. The temperature was 18°C, and the initial surface area of tne solid phase 1850 cm^2/g.

1.1.3. COMPARATIVE EVALUATION OF ALPHA AND BETA/GAMMA IRRADIATED MEDIUM LEVEL WASTE FORMS

Contractor : AERE Harwell (243-81-13 WAS.UK)

The waste forms to be studied in this joint investigation have not yet been specified in detail. However, the matrices employed are known, viz cement, bitumen, polymers and slag ceramic and preliminary work on the first three has started.

BACKGROUND TO RADIATION DAMAGE IN WASTE FORMS

Classes and ceramics

A large proportion of the work reported in the literature on radiation damage of inorganic waste forms is concerned with high level radioactive wastes incorporated in glass or ceramic materials. Radiation damage in glasses and crystalline solids arises from the displacement of atoms from their normal lattice sites. In crystalline solids this is manifested as an increase in volume and ultimately metamictisation.
In glasses devitrification can occur, and the phase change can also lead to a reduction in volume. The extent of damage by atomic displacement varies between the different types of radiation. The volume changes arising from atomic displacement damage are not apparent at doses less than 5 X 10^9R and generally saturate at 2 X 10^{11}R and 1% vol. change for glasses and 2-20x 10^{11}R and 10% volume increase for crystalline materials. However all three types of ionising radiation can give rise to chemical effects in a waste form or its surroundings by radiolysis, producing free radicals or aggressive chemical species by the recombination of these radicals. Studies have shown that for several specific glass or ceramic waste form simulates displacement damage does not

affect the leach rate of various radionuclides at doses lower than would cause volume changes (1).

Hydraulic Cements

The chemistry of hydraulic cement is complex, and its interactions with encapsulated waste materials introduce a further degree of complexity. It is likely that the products of radiolytic decomposition could interact with the chemical environment in the waste form. Several examples of radiolytic effects have been reported in the literature (2,3,4) :

- Cement contains large quantities of water as water of hydration, absorbed on its internal surfaces, and possibly as bulk liquid retained in the pore network. Radiolytic decomposition of this can yield H_2 and O_2 as gaseous products. Measurements of rates of evolution of these gases have been made for a few fairly specific wastes (4). It has been demonstrated that variations in waste form composition, water content and temperature affect the quality and composition of the gases envolved.

- The effect of radiation dose on the leaching rate of various radionuclides (particularly Cs, Sr and actinides) has shown that a significant increase in the leach rates arises from the production of aggressive species from radiolysis of the environment surrounding the solids of the waste form. For example, if leaching and irradiation are carried out in the presence in the air, nitric acid can be produced. This can increase leach rates by almost an order of magnitude. The presence of air within the pore structure of a cement grout could mean that for a given volume of waste form a much greater surface area of immobilised species is exposed to a chemical etching process as a result of internal radiolysis.

There may be other forms of damage which could occur to certain waste forms and which may be enhanced by irradiation, for example structural damage resulting from the build up corrosion products on encapsulated metals. Each of the materials specified in this programme must therefore be examined independently for possible modes of damage.

Organic Matrices

Recent work at Harwell has shown that some organic materials can swell condiderably under irradiation, and that the extent of swelling depends on both the dose rate and the size of the irradiated sample. A theoretical approach to the prediction of the swelling of waste forms in large storage drums from small scale tests has been developed in another Harwell project.

EXPERIMENTAL

Preliminary experiments have been initiated to investigate effects of gamma radiation on the strength, microstructure and chemistry of Ordinary Portland Cement, bitumen and relevant polymers e.g. epoxide, polyester. In the case of cement, samples have been exposed to ^{60}Co γ-radiation to a total dose of 4 X 10^7R at a dose rate of 0.1 MR/hr. They are at present under examination. It is not possible to draw any conclusions from the work at this stage. Samples of cured polyester both with and without cantained water have been γ irradiated to doses $\sim$ 5 x 10^8R. The dry samples show no measurable swelling at this dose but the wet samples, containing $50^v/o$ water began observable swelling at $\sim 10^8$R and have reached 4% volume increase at $\sim$ 5 x 10^8R.

During this period facilities have been made available for the manufacture (if necessary) and characterisation of real or tracer active simulates spiked with a suitable short-lived alpha emitter for the study of effects due to in situ alpha radiation damage.

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1.1.4. EVALUATION AND CHARACTERIZATION OF IMMOBILIZED AND SOLIDIFIED WASTE PACKAGES OF LOW AND MEDIUM ACTIVITY

Contractor : CEA France (244-81-15 WAS.F)

A. The leach resistance properties of full-scale waste packages with a cement, bitumen or cement/bitumen matrix (1)

In 1981, five waste blocks were subjected to long-term leaching tests : two cement matrices, two bitumen matrices and one cement/bitumen matrix.

They present the following characteristics, which are representative of waste types "WF2, "WF7,"WF8,"WF9" and "M1", as defined elsewhere.

Matrix	Basic constituents	Waste	Main radionuclides
WF2	Cement	Concentrate - borates	^{137}Cs, ^{90}Sr
WF7	Bitumen	Concentrate - sludges,nitrates	^{239}Pu, ^{241}Am
WF8	Cement	Concentrate - nitrates	^{239}Pu, ^{241}Am ^{137}Cs, ^{90}Sr
WF9	Bitumen	Sludges	^{239}Pu, ^{137}Cs
M1	Cement + emulsified bitumen (EB)	Precompacted solid wastes	^{239}Pu, ^{241}Am, ^{137}Cs ^{60}Co

The experimental procedure, which is derived from IAEA recommendations, uses drinking water maintained at 23° ± 3°C and renewed periodically (sequences of 15, 2 X 30, 3 X 90 then n X 180 days). The leachant is continuously recycled during each sequence.

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(1) Work done by CEA Saclay

The waste blocks are removed from their container before the test.

Each leachant is subjected to α, β and γ counts and physico-chemical analyses of the ions released from the waste blocks (dry extract, anions and cations).

At the end of 1981, the five waste blocks tested yielded the results given in the following table.

Matrix	Leaching time (days)	Cumulative fractions released ($\Sigma \frac{ai}{Ao}$)			
		^{239}Pu	^{241}Am	^{137}Cs	^{90}Sr
WF1	615	Not present	Not present	4.8×10^{-2}	1.5×10^{-3}
WF7	885	1.4×10^{-4}	1.3×10^{-5}	Not present	Not present
WF8	885	8×10^{-6}	2×10^{-6}	5×10^{-2}	9×10^{-3}
WF9	525	Not detect	Not detect.	7.5×10^{-4}	1.5×10^{-3}
MI	165	Not detect.	3.3×10^{-7}	4.6×10^{-4}	Not measurable

It was observed that the cumulative fractions released are substantial especially for cesium-137 and in the case of certain cement matrices ; with respect to α-emitters, the releases are low, regardless of the type of matrix considered.

Physico-chemical analyses have made it possible ot observe developments that differ according to the type of matrix. It was noticed that cement based packages underwent dissolution and erosion phenomena, which were imperfectly compensated by incipient carbonation. Creep was considerable in the case of bitumen. As regards cement/bitumen matrices, the observation period is still too short for any changes to be detected.

The following table illustrates the comments made above.

Matrix	Constituents	Leaching time (days)	Weight loss (%)	Amount of sodium released (%)	Amount of nitrates released (%)
WF2	Cement/ borates	615	< 0.2	5	Not present
WF7	Cement/ nitrates	885	1.3	10	1.2
WF8	Bitumen/ nitrates	885	< 0.2	< 0.1	2.2
WF9	Bitumen/ sludges	525	insignificant	insignificant	insignificant
M1	Cement/EB, solid wastes	165	Not detectable to date	5*	Not present

* Theoretically from the cement.

The cement/bitumen matrix that is frequently used for encapsulating precompacted solid wastes of the α-type is utilized on a large scale by the STE at the CEN-Cadarache establishment. It corresponds to the solidification matrix referred to as M1.

Initially envisaged for protecting the inside of the metal container while at the same time producing a matrix of low porosity and high leach resistance, the bituminous structure has been subjected to characterization and optimization tests.

Work carried out to date has related essentially to the following points :

- Verification of the stability and injectability of the slurry;
- Measurement of the physical and mechanical characteristics of the material (fluidity, setting time, heat of hydration; shrinkage, weight loss and mechanical strength).

The results obtained show that :

- The dimensional stability and homogeneity of the mixtures considered depend on several parameters connected with the butuminous emulsion content, the size of the aggregate and the water/cement ratio.
- The capacity to be injected into a precompacted solid medium varies primarly with the aggregate/bitumen ratio.
- The mechanical properties vary around an optimum which depends on the bitumen content of the structure.
- The bituminous emulsion under consideration is compatible with most hydraulic binders of the siliceous or aluminous type.

B. Study of the diffusion of radionuclides in polymers (1)

In order to gain a better understanding of diffusion phenomena in composite materials based on thermosetting resins, a study (Ref. 1) has been conducted in particular on the laws governing the diffusion of water and ^{137}Cs in pure and composite resins. The research that has been undertaken in the context of the Community programme consists in developing a method for the rapid determination of the diffusion coefficients of various radioelements in different thermosetting resin waste matrices.

Two types have been studied :

- A polyester resin which is reticulated by copolymerization of the styrene with unsaturated propylene glycol maleophthalate chains : the resulting product is a hard material with reticulation meshes of approximately 10 Å in size.
- An epoxy resin which is reticulated by the addition of a diamine or diether. The structure of the polymer consists of modules of high reticulation density embedded in a matrix of low reticulation density.

Diffusion in thermosetting resins is a very slow process; it is therefore necessary to ma nifacture extremely thin membranes to obtain significant results within reasonable periods of time. The diffusion coefficient of Cs radioactive tracer in polyester or epoxy resins was found to be approximately 7×10^{-13} cm^2/s. In the same thermosetting resins, the diffusion coefficient of water is approximately 3×10^{-9}cm^2/s. These results are obtained after very long periods of time. The duration of the experiments in the case of pure membranes approximately 100 µm thick are of the order of several hundred days.

A new, more rapid technique uses a diffusion cell with a thermosetting resin membrane and after a period of a few days or a few weeks the diffusion process is stopped and a sample of the membrane is taken.

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(1) Work done by CEA Grenoble.

The profile of the concentration of various ions in relation to their depth of penetration in the membrane is measured by laser mass spectrometry.

The initial results obtained demonstrate that :

- It is possible to establish profiles over depths from 100 to 200 μm ;
- The removal and analysis can be carried out micron by micron for volumes of 10 μm^3 to 500 μm^3;
- It is possible to analyse several ions simultaneously;
- It is possible to analyse organic and mineral materials simultaneously ;
- The technique has an extremely low sensitivity limit (10^{-16}g)

Fig. 4 gives a schematic graph of the apparatus.

It has been possible to obtain concentration profiles for Sr and Cs in a membrane with a thickness of approximately 150 μm. The diffusion time was 30 days and the analysis lasted about one day. The initial electrolytic solution consisted of $SrCl_2$ and CsCl in the proportions of 1 g/l.

References

1. Diffusion dans des matériaux composites à base de résines thermodurcissables.
   B. MORIN - Thèse 17.03.1981
   Université Scientifique et Médicale de Grenoble.

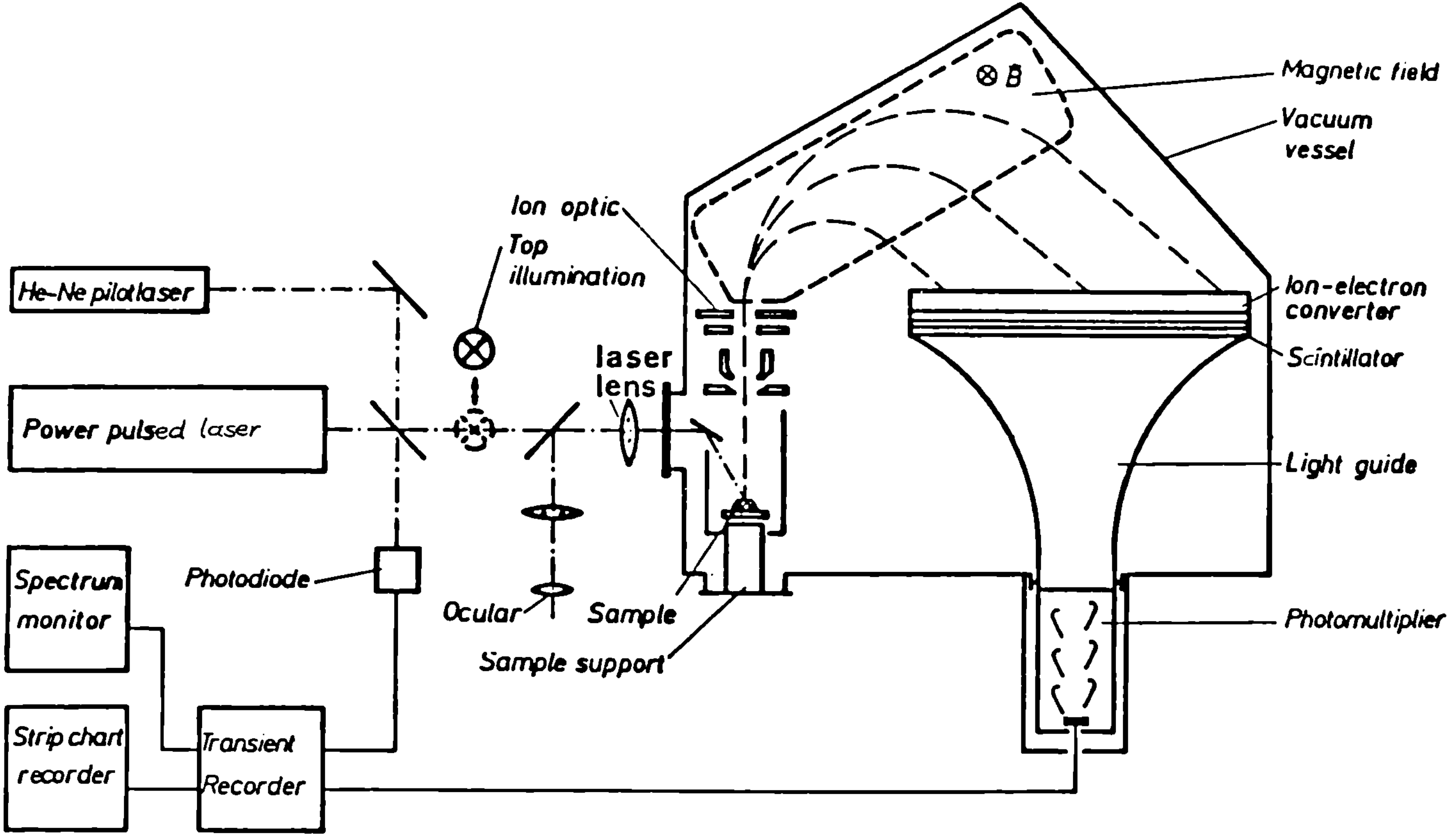

Fig. 4: LASER PROBE MASS SPECTROGRAPH (LPMS) - C.E.A.-C.E.N. - Grenoble - GSF- München-Neuherberg

C. STUDY OF THE CHEMICAL FORMS OF THE RADIOELEMENTS RELEASED BY LEACHING (1)

In order to identify the chemical form of Cs and of the trans-uranium nuclides the assumption was that the cationic forms are retained on an H^+ ion exchange resin, but not retained on an anion exchange resin.

The technique therefore consists in comparing the activities of the leachate solutions before and after the exchanges have taken place.

Three real leachates from Saclay (cf.A) have been subjected to $\alpha$and λ spectrometry and liquid scintillation investigations before and after being passed through cation or anion exchange resins. These leachates came from a cement evaporation concentrate reprocessing type (FAR L17.4), a cement /PWR evaporation concentrate (C4007-LII-6.5) and a bitumen waste product from Marcoule (COGEMA LIV-5.2).

The results of the measurements are given in Table I.

The behaviour of Cesium follows a clear pattern : Cs^+ is largely retained by an H^+ cation exchange resin and not at all by an anion exchange resin . In the latter case, the form of the resin (except for OH^-) has little importance. The Cs^+ content of the SENA and COGEMA leachates is too low to obtain a significant activity yield after passing through the H^+ resin column. In addition, the liquid scintillation measurements are not specific to Cs^+ because α-emitters and ^{90}Sr are present (It should be stressed that ^{90}Sr is absent in the FAR leachate).

An additional special experiment allowed to check that the Cs can only be present in the ionic form.

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(1) Work done by CEA Fontenay-aux-Roses

Plutonium appears to pass through the H^+ -ion exchange column without being retained.

Part of the americium appears on the contrary to be retained on the H^+ - resin (Factor of 5 between the activities measured before and after passing through the resin column).

The Chooz leachate is not supposed to contain any transuranics. Therefore we have no initial data. The values given in Table I are, however, significant.

For the COGEMA leachate the initial α-emitter content (mainly ^{239}Pu) is not well known but should be of the same order of magnitude as that of the FAR leachate. The values for the activity measured after the leachate has been passed through an H^+ - resin column are consequently comparable.

The main difficulty encountered in the initial stages of the study comes from the extremely low actinide contents, which require sensitive but not necessarily analytical methods (scintillation and α-spectrometry) to be used.

TABLE 1

	FAR	CHOOZ	COGEMA
Identification of the leachate	LI 7.4	LII 6.5	LIV 5.2

Technique : Liquid scintillation

Units : i p m (impulses p/min.)

(a) initial activity	12.035	56	16
(b) after passing through H^+ resin	95	24	4
(c) after passing through Cl^- resin	12.132	16	3

Technique ; γ- spectrometry on the ^{137}Cs peak

Arbitrary unit : number of strokes in 1800", 10 ml solution

(a) initial activity	15.194	102	142
(b) after passing through H^+ resin	297	-	-
(c) after passing through Cl^- resin	14.893	113	109

Technique : α-spectrometry (surface-barrier semiconductor)

Unit : pCi/ml

(a) initial activity•	^{233}U	-		
	^{239}Pu (1)	0.055		
	^{241}Am (2)	0.090		
(b) after passing through H^+ resin	^{233}U	0.037	0.014	0.053
	^{239}Pu (1)	0.047	0.020	0.037
	^{241}Am (2)	0.017	0.005	0.010

• comprises ;

(1) $^{239}Pu + {}^{240}Pu$

(2) $^{241}Am + {}^{238}Pu$

D. RESISTANCE OF IMMOBILIZED WASTE FORMS TO THE ATTACK OF MICROORGANISMS OF THE SOIL (1)

A number of microorganisms, bacteria and yeasts that are capable of breaking down heavy hydrocarbon are known to exist in nature. It is therefore logical to envisage the possibility of microorganisms attacking the bitumen matrix during the storage of radioactive wastes immobilized in bitumen in the soil. To this end, the waste products are placed in samples of biologically active soil, which are contained in glass tanks, protected from the light and maintained at constant temperature and humidity.

The waste products tested consist of STE 3 La Hague type reconstituted - therefore inactive - wastes, which are encapsulated in Mexphalte R 90/40 bitumen.

Three types of waste products are used :

- MA (medium-activity) sludges and TBP ;
- Sludges + granular resins;
- Sludges and crushed resins.

The samples were previously subjected to a γ-irradiation. This was carried out by ORIS at Saclay in the Poseidon plant using an external ^{60}Co source of 205 000 Ci.

The dose received by the samples corresponds to the integrated dose estimated on completion of storage, namely 10^9Rad. On the other hand the imposed dose rate of 2.8×10^6Rad/h by far exceeds that derived by the radioelements contained in the waste product.

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(1) Work done by CEA Cadarache

The prior irradiation therefore resulted in releases of radiolysis gases which caused deformations of the samples.

The soil came from the actual storage site at La Hague. It is poor in humic matter and therefore contains little biological activity. The soil was therefore enriched by adding gardening peat in the proportion of 50/50 by weight.

The glass tanks containing the waste packages have a useful volume of 20 litres, the soil is humidity controlled.

In october 1981, the bitumen waste products were placed in soil under the following different experimental conditions :

- In aerobic conditions ;
- In anaerobic conditions ;
- In a sterile soil under aerobic conditions.

Three samples of each of the types of waste products (MA sludges + TPB, granular resins and crushed resins) were placed in aerobic conditions. Air was introduced regularly at the bottom of the tank and at the surface.

Three samples of each of the types of waste products were placed in anaerobic conditions. A regular sweeping with argon gas made it possible to maintain an inert atmosphere.

A sample of each type of waste product was placed in sterile soil. The soil had been sterilized in a sterilizer and the humidity was maintained by sprinkling with sterile water.

A set of non-irradiated bitumen waste products was placed under aerobic conditions and used as a control.

The spectrum of the microbe population in the soil was plotted by large functional families in order to be able to follow over the course of time the quantitative evolution and the qualitative changes in distribution.

In addition, in order to assess the overall biological activity of the soil, the amount of ATP (adenosine triphosphate) in the soil was measured.

The results of the microbiological analysis of the La Hague soil and the mixture of the La Hague soil and peat are summerized in Table III.

The same analysis will be carried out after the microorganisms in the soil have developed for six months in contact with the samples and at the surface of the samples of bitumen waste products.

TABLE III : MICROBIOLOGICAL ANALYSIS OF THE SOIL

	La Hague soil	La Hague soil + peat
ATP µmoles/g of dry matter	1.8×10^{-5}	2.4×10^{-5}
Total germs	3.1×10^{7}	5.7×10^{6}
Number of germs/g of dry matter		
Azotobacters	4.7×10^{3}	3.2×10^{2}
Clostridia	0.9×10^{2}	3.2×10^{3}
Nitrosating bacteria	93	191
Nitrifying bacteria	41	3.2×10^{3}
Ammonizing bacteria	4.6×10^{5}	5.7×10^{5}
Nitrate-reducing bact.	4.7×10^{2}	3.2×10^{4}
Aerobic cellulolytic bact.	-	-
Anaerobic cellulolytic bact.	1.2×10^{3}	2.6×10^{5}
Fungi	1.0×10^{4}	1.3×10^{4}
Actinomycetes	7.2×10^{5}	6.3×10^{6}
Algae	-	-
Iron bacteria	-	-

1.1.5. CHARACTERIZATION OF SOLIDIFIED MEDIUM ACTIVE AND α - BEARING WASTE FORMS

Contractor : S.C.K./C.E.N. Mol (245-81-15 WAS.B)

Two waste forms are investigated :
- The basaltic slags from the FLK incinerator
- The bitumen from both the Eurochemic and the SCK plants.

The work programme covers :

Incinerator slags

- Characterization of some representative slags produced during 1981-1982
- Investigation of the corrosion stability of the slags and of some reference glasses in conditions relevant for disposal in clay media.

Bitumen-conditioned waste products

- Leaching studies in clay media and ageing studies on Eurobitumen
- Leaching studies in sea-water conditions on SCK-bitumen

The waste form samples being investigated are as produced samples. The slag samples are monolithic hot pressed FLK granules (Ref. 1). The four reference glasses (two without Pu and two containing 2 wt% UO_2 + PuO_2) have the compositions given in Table 1. One glass (WG 124) has a composition similar to the average of several representative FLK runs.

The results available at this moment concern the following points :
- Stationary corrosion tests in distilled water, clay-equilibrated water and wet clay at 90 and 40°C (for a surface area

to volume ratio of $1 cm^{-1}$) are underway. Results of specimen weight and leachant pH are available after leach times of 3, 7, 14, 28, 42 days (see Table 2). Autoclave tests, also yielding values of weight loss and leachant pH were carried out at 200°C in distilled water and clay-equilibrated water (1, 7, 28 days).

- Dynamic Soxhlet tests
- Thermal stability : thermal analysis (DTA, DSC, TMA) data on four FLK reference glasses are given in table 3.

Previous results (Ref.1) indicate that the iron in the FLK granules is primarily in the ferrous state. For the reference glasses used in this programme, the fraction of iron in the ferrous state is about 50% for WG 122, 123 and 124 and about 25% for WG 119. Using a different melting atmosphere, a WG 122 glass will be cast in which this fraction is about 80%, thus more nearly representing the FLK slags. One Pu-containing glass (PWG 119) has been prepared. Results can not yet be reported.

References :

1. R. De Batist et al. : "Structure and Stability of incinerated α-waste products", Proc. of Int. Symp. Scientific basis for Nuclear Waste Management (Boston), Vol 2 (1980) p. 351-359

Table 1 : Composition in mol % of the four inactive FLK reference glasses

	WG 119	WG 122	WG 123	WG 124
SiO_2	66	66	66	70
Al_2O_3	-	-	5	2
Fe_2O_3	10	-	-	-
Fe_2O_2	-	10	5	6
Na_2O	6	6	6	4
K_2O	6	6	6	1
MgO	6	6	6	5
CaO	6	6	6	5
BaO	-	-	-	2
MoO_3	-	-	-	1
Cr_2O_3	-	-	-	1
CuO	-	-	-	1
NiO	-	-	-	1
TiO_2	-	-	-	1

Table 2 : Weight loss per surface area (x 10^{-5}g cm^{-2}) after 28 days corrosion at 90°C for a surface area to solution volume ratio of $1cm^{-1}$ for the reference glasses (WG 119 - 124) and FLK compacts.

Specimen	Distilled water	Clay-equilibrated water	Clay
WG 119	6.53	92.96	279.44
WG 122	8.73	108.64	298.48
WG 123	-1.38	85.40	129.64
WG 124	-2.53	56.37	258.72
FLK 77/33-40	-5.96	37.61	167.72
AFLK 78/11-14	-3.42	44.61	63.00
FLK 79/2	-3.34	23.99	31.64

Table 3 : Results from thermal analysis on the FLK ref. glasses

Specimen	DSC		TMA		DTA		
	T_g (°C)	M_g (°C)	T_g (°C)	M_g (°C)	T_g (°C)	M_g (°C)	T_c (°C)
WG 119	545	577	537	610	545	577	846
WG 122	520	542	520	580	537	554	791
WG 123	-	-	635	682	633	664	840
WG 124	-	-	620	715	671	705	765

DSC Differential Scanning Calorimetry
TMA Thermo-Mechanical Analysis
DTA Differential Thermal analysis
T_g Glass Transformation Temperature
M_g Dilatometric Softening Point
T_c Crystallization Temperature

1.1.6. TEST METHODS FOR BITUMINIZED AND OTHER LOW-AND MEDIUM-LEVEL SOLIDIFIED WASTE MATERIALS

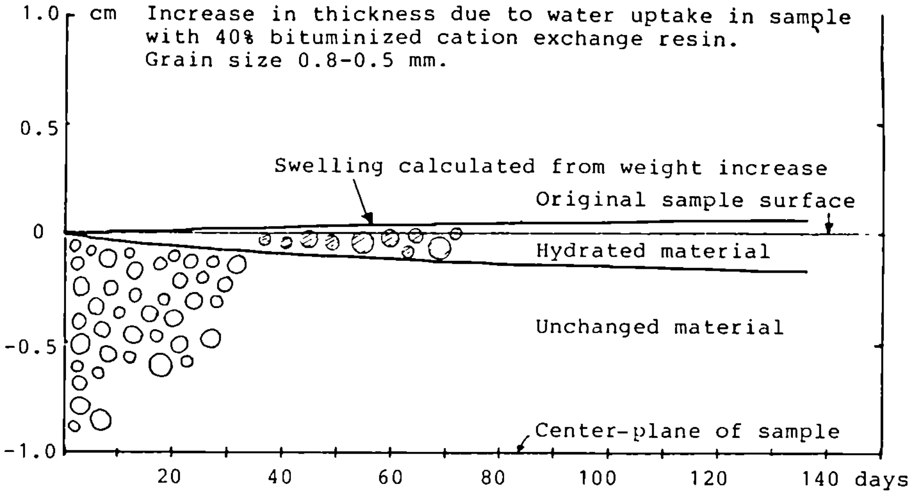
Contractor : RNL Risϕ (235-81-13 WAS.DK)

It is well known that waste materials such as sodium sulphate or ion exchange resin which have been dehydrated during bituminization may swell considerably due to formation of hydrates if they get in contact with water.
A weighing method has been developed which permit the measurement of water-uptake and swelling as function of time for samples with two plane-parallel surfaces exposed to water.
The method has primarily been used to study bituminized cation exchange resin but preliminary investigations of bituminized sodium sulphate and sodium nitrate have also been made.
A strong increase in rate of water uptake with decreasing particle size of the ion exchange resin has been demonstrated, but after sufficient time the final result may possibly be the same, i.e. swelling of more than double of the volume, also for ordinary grain size resin. It must be concluded from the amount of water taken up by the samples that the stresses developed in the material due to swelling result in the formation of an open, waterfilled structure probably with interconnected pores. This is important for the understanding of leaching from such materials. Some preliminary experiments to investigate whether it is possible to prevent the phenomenon have been made. Heat-treatment of the ion exchange resin in non-oxydizing atmosphere to temperatures above 400°C seems to be a possibility but not without special problems. Development of the necessary technology for such a treatment is outside the scope of this work. Bacteriological attack in a humid oxydizing environment is a possible mechanism for increased activity release from bituminized waste materials. An experimental technique, which attempts to compare leaching

of activity from identical samples with and without the presence of bacteria, is under development but has so far not given reproducible results. Measurement of oxygen consumption by bacteria growing on bitumen-coated sand particles indicates that the rate of attack under these circumstances are comparable to typical leach rates from bituminized materials i.e. about 10^{-6}cm/day.

When the old IAEA standard method for leach rate measurements is used on samples of cemented waste the exposed surface area is somewhat undefined since contraction of the material may lead to formation of cracks between the sample and the container. An improved method has recently been proposed which circumvents this difficulty but, in contrast to the new ISO standard, maintains the geometrical advantage of a semi-infinitely thick sample with a single plane surface exposed to the leaching medium. The method will be used to investigate whether the presence of atmospheric carbon dioxide, the orientation of the sample surface or the partial covering of the surface are of importance for the leach rate measurements. Some preliminary results obtained with leach rate measurements from cemented sodium nitrate using a modified IAEA procedure, motivate such an investigation.

The use of silica fume i.e. about 0.2 μ large spherical particles of SiO_2 as an additive to cemented waste seems to improve the leach rate, must be investigated further due to the phenomena mentioned and other possible complicating factors.



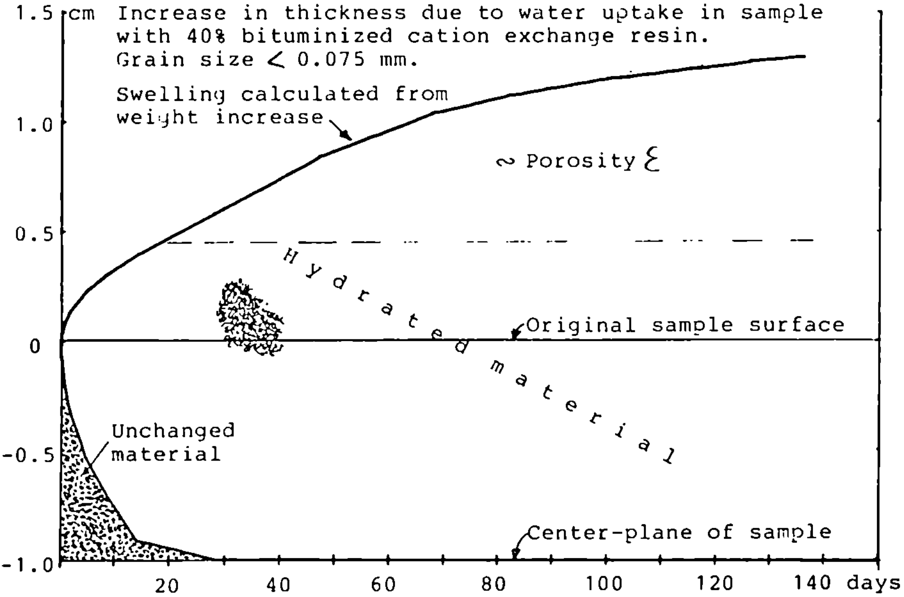


Fig. 5: Swelling of bituminized ion exchange resin.

1.2. CONDITIONING OF HIGH ACTIVITY SOLID WASTE : FUEL CLADDINGS AND DISSOLUTION RESIDUES

All of the 8 experimental activities comprised have started or continued with work during the reporting period.

Three main topics of research can be distinguished. They concern first the development of methods for embedding the waste scrap into a solid and resistant matrix material in order to confine the radioactivity and to prevent it from dispersion. The matrix materials being investigated, are lead alloys, ceramics and compacted graphite or aluminium powder. Next to the known concrete matrix, they constitute potential embedding matrices with improved properties. However, none of them has yet been tested under representative radioactive conditions, the methods being in their exploratory stages of development.

A second topic concerns the conversion of the fuel hulls' waste by melting or chemical treatment. These methods aim at concentrating the zircaloy scrap into a massive inert form while making use of a possible decontamination from the adherent fuel and fission product residues.

Under a third heading, those projects are grouped which deal with the determination of the properties and characteristics of the waste materials themselves or the conditioned waste products. The two experimental works being undertaken at present, concern the characterisation of cemented hulls and the pyrophoricity of Zircaloy fines.

In January 1982, the Commission is planning to organize an E.C. specialists meeting on the methods for conditioning storage of spent fuel element hulls. The intention is to make a review of the state of the art based on a joint study drawn up by KfK and CEA in the scope of the first Community programme.

1.2.1. EMBEDDING INTO MATRIX MATERIAL

1.2.1.1. PRESS COMPACTION AND EMBEDDING OF HULLS INTO LEAD ALLOY MATRIX

Contractor : SCK-CEN Mol (167-81-2 WAS.B)

Inactive laboratory and technical scale studies, made under a previous contract have shown that considerable volume reduction can be achieved by compaction and that the densified material can be incorporated into a matrix waste form with low porosity. The corrosion resistance of lead alloys, especially to groundwaters, should thus assure a long-term isolation of the waste.

The present programme aims at the assessment of the product quality with respect to the immobilization of the contaminating nuclides, in tests with hulls from irradiated fuel rods. Laboratory and field experiments are undertaken in addition, to further explore the corrosion resistance and bonding properties of candidate matrix materials.

Small scale compaction and embedment units, adapted to hot-cell operation, have been constructed in preparation of the work with active hulls. The 0.6 MN-press is fit with a compaction form of 50 mm diameter which accomodates a hulls' batch of about 100 cm^3. The embedment of the compacts is made in such a way that an appropriate exposure surface for leach tests is created.

This equipment has been tested with inactive zircaloy tube cuts in order to determine suitable work conditions for the active experiments and to have reference data for the evaluation of irradiation effects.

Compaction tests have shown that compared to previous data for larger press forms, a 10% lower density is obtained at equal pressure. Densification to 58-59% TD is achieved at 156MPa, to 67-69% at 300MPa.

Different procedures have been tried for the embedding of the compacts into the reference Pb 1,5 Sb-alloy. The samples were found to be completely filled with the matrix material when the embedment was made at 723 K under a vacuum of about 100 Pa and the specimen was allowed to cool-down under atmospheric pressure. Microscopic examination of sample sections show however that small contraction gaps are present at some places. The active experiments will be made with zircaloy hulls from PWR fuel rods (BR 3) irradiated to about 30.000 Mwd/tHM and with stainless steel hulls from FBR-elements (DFR) irradiated to about 52.000 Mwd/tHM.

In view of the leach tests to be made with the active products, the Zry hulls have been analysed for specific contaminants. Data obtained for two different hull sections were respectively: total Pu-content 314 and 326 $\mu g\ g^{-1}$, 134 Cs-activity 0.195 and 0.453 $mCig^{-1}$, 137 Cs-activity 1.31 and 2.13 $mCig^{-1}$. The average tritium content, determined by analyzing 15 sections, amounted to 523 μCi per gram of hull material.

In completion of earlier work on the corrosion resistance of candidate matrix materials, selected lead alloys are exposed in the laboratory to interstitial clay water and to saturated salt brine, as well as to direct contact with the clay in field experiments.

Results from long-time exposure tests with interstitial clay water correspond in general to the data found for Antwerpian ground water during the previous study. At 283 K the samples exhibit small positive weight changes, slowly increasing with time. A weight loss was obtained at 373 K but this stabilizes after 4 months of exposure and would correspond to the dissolution of only 4 mm of material in 1 000 Years. In electrochemical corrosion tests it was found that the Pb 2 Sn 0.6Ca alloy present the highest resistance.

The bonding between the zircaloy hulls and the matrix is evaluated in wetting experiments. Tests with Pb 0.8 Sb and Pb 0.1 Zr 0.2 Mg indicate that the wetting is improved by a heat treatment at 823 K.

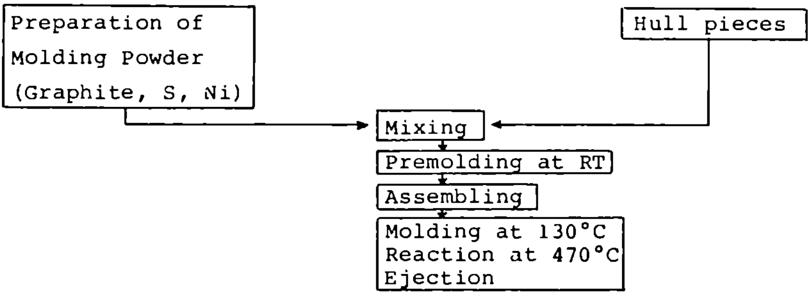
1.2.1.2. EMBEDDING OF HULLS INTO GRAPHITE AND ALUMINIUM

Contractor : NUKEM Hanau (169-81-WAS.D)

Two embedding materials for the fixation of spent fuel hull pieces and the corresponding processes were developed and tested. The materials are :

- Graphite with an anorganic binder and
- Aluminium

The process for embedding the hull pieces in graphite-matrix is schematically the following :



The physical properties of fabricated graphite compounds as well as the leach and corrosion resistance in saturated brine solutions (and/or demineralized water) were investigated. The results for three different matrix compositions are given in table 1 and 2.

Table 1 : Physical properties

Material Properties	80% Graphite 20% Sulphur	43.7% Grap. 41.3% Nickel 15.0% Sulp.	43.3%Grap. 36.7%Nick. 20.0%Sulp.
Density (g/cm^3)	2.18	3.28	3.09
Th. Density (%)	>98	94.8	91
Heat conductivity (W/cm·K)	⊥0.69 ∥0.29	⊥0.92 ∥0.29	⊥1.16 ∥0.42
CTE (μm/m·K)	-	-	⊥4.27 ∥10.10
Compression strength (MN/m^2)	⊥54.2 ∥59.1	⊥70.7 ∥99.6	⊥71.9 ∥107.4
Tensile strength (MN/m^2)	-	-	-
Bending strength (MN/m^2)	-	-	⊥78.5 ∥25.5
Young modulus (KN/m^2)	-	⊥76.8 ∥17.7	⊥86.3 ∥21.4

Table 2 : Corrosion and Leaching Resistance

Material Properties	80% Graphite 20% Sulphur	43.7%Graphite 41.3%Nickel 15.0%Sulphur	43.3%Grap. 36.7%Nick. 20.0%Sulp.
Leaching Rates (cm/d) for Cs in demineralized water at RT	$<3.10^{-7}$ $<3.10^{-9}$	-	3.10^{-4} 5.10^{-6}
Corrosion rates ($G/cm^2.d$) in saturated Quinary Carnallite-Solut. at 100°C	-	$4.2 .10^{-6}$	$1.5 .10^{-5}$
Corrosion rates ($g/cm^2.d$) in saturated Quinary Carnallite-Solut. at 150°C	-	$2.8 . 10^{-5}$	$<5.6 . 10^{-5}$

For embedding the hull pieces in aluminium powder two processes have been studied:

- A high pressure-process (working at room temperature with a pressure of about 500 MN/m^2)
- A high-temperature-process (working with a pressure of 50 MN/m^2 in the range of 400-500°C)

For both processes similar steps, as shown above (except the heating in the case of the high-pressure-process and the reacting) are necessary.

By using the high-pressure/low-temperature-process all fabricated samples show numerous cracks on the surface (content of hull pieces 10 wt.-%), whereas the low-pressure/high-temperature-products do not show any defect up to a charge of 26wt.-% of hull pieces.

The physical and chemical properties of the aluminium-samples have not been investigated till now. During the following months these data will be measured.

1.2.1.3. EMBEDDING OF HULLS AND DISSOLUTION RESIDUES INTO ALUMINO-CERAMICS

Contractor : CEA Saclay (172-81-2 WAS.F)

Simple or complex oxides, based mainly on Al_2O_3 or SiO_2, show a good corrosion behaviour and particulary good oxydation behaviour in most media.

Alumina ceramics have been chosen as coating matrix, because of the acquired experience (from laboratory to industrial stage) in the porous ceramics field, as well as in non-porous, dense ceramics.

A bibliographic research, showed that high purity and high density Al_2O_3, has good mechanical, physical and chemical properties for permanent storage of radioactive wastes. The chemical stability of Al_2O_3 is high and the leaching behaviour is good, compared to other oxides.

The embedding process comprises the following steps :

- Pieces of hulls, possibly crushed, are embedded in an alumina powder. The lot is isostatically compacted at ambient temperature and at pressures of 1000 to 2000 bars. A cylindric block called core or matrix is obtained.

- The core, is then again embedded in alumina powder, which will constitute, after sintering, the final envelop of the compound. The whole is once more compacted, isostatically, at pressures of 1000-2000 bars and at ambient temperature. A cylindric block is obtained.

- This cylindric block, (core + envelop) is sintered by the usual sintering method : that is to say at atmospheric pressure, and at temperatures in the range of 1200°C.

Two main difficulties inherent in the procedure are the

- Cracking during the two compacting steps and the
- Breaking of the pieces in the final sintering phase. As a matter of fact, sintering leads to a densification of the material, that is to say a dimensionnal reduction of the outer envelop which has to be made " adaptable" to the central part of the block i.e. the core.

The adjusting of the procedure, started with "blank experiments" and with a core without hull. These experiments have led to choose the suitable alumina powders, in order to succeed in the two compacting operations, and also in the final sintering.

The final dimensions obtained after sintering, were compounds with 100-200 mm diameter and 200-300 mm height, in agreement witn the values previously envisaged.

Preliminary tests, with non irradiated hulls are under way at present. Non crushed hulls, of cylindrical shape have been used. The filling ratio is 30 to 40% of the vòlume of the core part. Optimisation of the filling ratio is in progress.

As to the dissolution residues, preliminary tests consisted in trying to reconstitute a liquid, non active solution, being as close as possible of solutions, obtained in the reprocessing of nuclear fuels.

The cyclindrical blocks, obtained by the process, are very hard, and diamond tools are necessary to cut the blocks and to sample and machine test pieces for characterization.

Characterization measurements are in the initial phase.

1.2.1.4. SOLIDIFICATION OF ALPHA BEARING WASTES IN A CERAMIC MATRIX

Contractor : KfK Karlsruhe (173-81-2 WAS.D)

The investigations into the immobilisation of alpha-bearing wastes in a sintered alumosilicatic matrix were continued by experiments :

- To improve the quality (leach resistance) of the ceramic matrix,
- To determine the release of volatile waste compounds like Ru and Mo;
- To solidify original dissolver residues from the reprocessing of spent fuel elements,
- To optimize the homogenisation process of the ceramic raw materials using radioactive tracer elements,
- To convey simulated dissolver residues by air-lift technique

The leach resistance of the ceramic matrix is controlled by the solubility of its mineral phases depending on the ratio of the matrix phases, as they are :

- corundum (Al_2O_3)
- mullite ($Al_6Si_2O_{13}$)
- quartz (SiO_2)
- glassy phase

The leach resistance decreases from corundum to the glassy phase. A significant reduction of the glassy phase from 30 to less than 5% had been achieved by increasing the $\frac{Al_2O_3}{Al_2O_3 + SiO_2}$ ratio from 0.28 to 0.78 using the equivalent amounts of Kaolin and reactive corundum as starting matrix materials. Mixed with 20 wt% of simulated alpha-bearing dissolver residues the following mineral phases are identified by X-ray diffraction after the heat treatment : corundum, mullite, RuO_2, $CaMoO_4$, ZrO_2 and Pd.

The leach rate for Pu in distilled water was determined, it is 10^{-8}g/cm^2.d after 28 days leaching time ; the equivalent value using a matrix with an $\frac{Al_2O_3}{Al_2O_3 + SiO_3}$ - ratio of 0.30 yields 10^{-7}g/cm^2.d.

The lowest Ru-volatility was measured with 1.6% relative to the initial Ru contained in the starting material under air-atmosphere at 1300°C/1.5 h. It seems that the chemical form of the Ru-starting material (Ru, RuO_2,$Ru(NO_3)_3$, $RuNO(NO_3)_3$) has no influence on its volatility.

The volatility of Molybdenum can be reduced to 1.4% by adding the stoichiometric amount of $Ca(OH)_2$ to form $CaMoO_4$ which is stable up to 1450°C.

Also the solidification of original dissolver residues into a ceramic matrix has been experimented using 50wt% Kaolin, 25 wt% Feldspat, 25 wt% Quartz as ceramic raw materials. A waste loading of 20 wt% has been achieved. The properties of the products are similar to the equivalent products with simulated dissolver residues.

The equipment for the control of homogeinity of the starting materials by means of radioactive tracers has been installed.

Experiments refering to the air lifting of simulated dissolver residues suspensions with 40 wt% of solid compounds were succesful.

1.2.2. MELTING AND CONVERSION OF ZIRCALOY

1.2.2.1. CLADDING WASTE CONDITIONING BY EUTECTOIDIC MELTING AND BY EMBEDDING INTO GLASS

Contractor : CEA Marcoule (170-81-2 WAS.F)

The investigations concern the conditioning of zircaloy hulls by two techniques : melting and embedding in glass.

Because the work started recently, report can be given only on the first mentioned technique.

The main experimental equipment consists of a furnace and a trap for tritium.

The furnace of the VULCAIN cell devoted usually to the annealing of radioactive glasses was used after some modifications were made. The furnace lies in a cylindrical muffle made of inconel, heated by electrical resistance. The muffle which is held vertically contains an alumina crucible. The muffle is closed at its bottom with a lid which enables to move the crucible up and down and to provide for Argon supply. The tightness is ensured by a Viton gasket.

The device for trapping the tritium consists of a CuO bed kept at 450°C associated with 2 water bubblers. In addition 2 cellulose acetate filters are placed on the gas stream line. One is located just at the outlet of the muffle, the other one at the inlet of the tritium trap.
Three tests were made successively :

- The first one was completely non-radioactive and performed in a "cold" laboratory,
- The second one envolved non-radioactive hulls together with a tritium capsule. It was carried out in VULCAIN,
- The third test concerned the melting of real hulls coming from tne BORSSELE reactor (Zircaloy4). It was also carried out in VULCAIN.

For all these tests, about 350 to 400 g of zircaloy were used and copper was added in ratio enabling a Cu weight content of 20% in tne final product. In the second test, copper was provided by the tritium capsule which was made of Cu.

As outcome of the tests one may summarize :

The tightness of the muffle was found defective.
This drawback hindered to evaluate the efficiency of the tritium trapping. The tightness was however good enough to avoid large oxidation and to enable the fabrication of an ingot at 1150°C. (See Figure 6.)

In the near future the radioactive ingot will be leached with water.
A radioanalysis of this material will be performed afterwards in order to check the informations about the characteristics of the hulls which were provided by the FONTENAY AUX ROSES CENTRE as well as to calculate various leach rates and

possibly to evaluate roughly the volatilisation rate of Cs and Ru.

Another ingot will be made after improvement of the tightness of the muffle furnace.

Fig. 6 : Bottom view of the radioactive ingot.

1.2.2.2. CONVERSION OF ZIRCALOY CLADDING INTO ZIRCONIA

Contractor : AERA Harwell (171-80-2 WAS.NK)

In the conversion process being studied, Zircaloy is dissolved in 3M ammonium fluoride solution and precipitated as hydrous zirconium oxide using ammonia. Most of the ammonia fluoride can be recycled. The hydrous oxide can then be immobilised in massive form. The present work includes studies on incorporation into cement, and hot pressing.

Experiments continued on a small scale have confirmed that a five stage countercurrent wash process can give 85% removal of the total process fluoride (Zr:F ratio 1.08:1) with 7 litres wash/mole Zr. Larger scale (1 litre per stage) equipment with 3 stages has now been built and runs with batch sizes of 40 g Zr, have shown that approximately 13 litres wash/mole Zr are required to achieve 80% fluoride removal. With 3 litres wash/mole Zr (equal volumes of wash and precipitated slurry) approximately 65% removal of fluoride was obtained.

A filtering centrifuge operating at 8000 rpm (4500 g) has been found to give 75% removal with approximately 5 litres wash/mole Zr.

The pilot plant equipment was commissioned during this year and has been mainly used to produce material for subsequent washing and solidification experiments. However, experiments have shown that on this scale of operation, the basket containing the Zircaloy must have holes of about 5 mm diameter in order not to extend the total dissolution time beyond that found in small scale experiments.

Analysis of the recycle solution has shown that approximately 5% of the zirconium would be recycled to the dissolver following batch washing of the slurry to remove 80% of the fluoride. After 9 dissolutions using recycled fluoride solution, the tin and iron concentrations in the recycle liquor have reached approximately steady values of 150 and 170 μg/ml respectively (Zircaloy - 4 contains 1.5 wt% tin and 0.2 wt% iron).

Experiments have been performed using ammonium hexacyancobalt ferrate (ACFC) to remove inactive caesium from the dissolver liquor. The absortive capacity of the material has been found to be approximately 60mg caesium/g ACFC under neutral conditions. However, at higher pH values the capacity reduced to 10 mgCs/g ACFA, and the ACFC appeared to break up.

The washed and dried precipitate produced from the ammonium fluorozirconate solution is mainly amorphous, but with some crystalline material which was probably $NH_4ZrOF_3.2H_2O$. On heating to 300°C this crystalline material decomposes and volatile material (~ 30% by weight) is lost from the precipitate (probably as H_2O and NH_4F). The result is a fine (10μm grain size) powder having the same structure as monoclinic ZrO_2, but containing an amount of fluoride which is controlled mainly by the washing of the original precipitate.

The hot-pressing characteristics of the oxide material were investigated using 12 mm diameter graphite dies and pressure of 22.5 MPa applied both during heating and cooling. Materials with F/Zr atom ratios in the range 0.005 to 1.4 were studied and it was found that the F/Zr ratio has to be greater than about 0.04 for densification to be obtained at temperatures below 1000°C. Material calcined at 300°C was hot-pressed to 90% of theoretical density at 700°C rising to 98% theoretical density at 980°C.

The material could be pressed without calcination to yield products of slightly lower density.

Larger blocks weighing up to 400 g have been produced.

The ceramics contain two major phases : one of composition ZrO_2 with the monoclinic ZrO_2 structure, and an oxyfluoride of composition close to $Zr_{10}O_{13}F_{14}$ with the orthorhombic αU_3O_8 structure. Minor phases closely related to the two major ones are also formed. The grain size is typically a few microns, with some oxyfluoride grains up to 100 μ in size.

The ceramics have good mechanical properties with a rupture strength of ~ 150 MPa.

The solid solubility of radwaste ions Cs^+, Sr^{2+} and U^{4+} was investigated by doping batches of hydrated zirconium oxide (radwaste-ion/Zr ratio ~ 0.03) individually with this ions. Sr^{2+} and U^{4+} were found to be accomodated in both the ZrO_2 and oxyfluoride phases whereas Cs^+ could only be accomodated in the oxyfluoride and with $Cs/Zr < 4 \times 10^{-3}$. This latter restriction is not a problem in practice however since Cs will arise at only 20 ppm by weight in Zircaloy. Within the above limits, the ceramics are therefore suitable hosts for these radwaste ions.

The interaction of ceramics with water has been investigated in Soxhlet tests at 100°C and in standard static tests (the MCCL test developed at Battelle N.W.) at 90°C. The experiments have shown that initially the oxyfluoride phase is preferentially attacked by water to release ZrF_4 into aqueous solution. However, the reaction soon becomes very slow probably because of the formation of a protective layer of ZrO_2 at the surface of the oxyfluoride grains. Samples containing ∿ 0.1 m Ci of ^{85}Sr have been hot-pressed and leach tested. Leach rates for ^{85}Sr at 90°C are $\sim 5 \times 10^{-3}$ cm d^{-1} averaged over the first 3 days falling to $\sim 5 \times 10^{-6}$ cm d^{-1} after 10 days as the protective layer develops.

Blocks have been produced using dried precipitated oxide with ordinary Portland cement on their own, and also with replacement of 20% of the cement by pulverised fuel ash. In each case, setting was retarded (compared to normal concrete), but after 3 days rapid curing there was no significant difference in the strengths of the concretes produced. Additional tests using high alumina cement gave weak products, however, indicating this material to be intolerant of the hydrous oxide.

1.2.3. WASTE PROPERTIES AND CHARACTERIZATION

1.2.3.1. CHARACTERIZATION OF THE FINAL HULLS-CONCRETE-PRODUCT

Contractor : KfK Karlsruhe (168-80-2 WAS.D)

The work during 1981 was related to the determination of the radioactivity content of spent KWO fuel hulls, the investigation   leach rates of hulls-concrete-products in various leaching media and the completion and calibration of a calorimetric device for the determination of heat release from 200 l-barrels filled with concrete embedded radioactive hulls.

The specific total radioactivity (Ao) was determined by α-and γ-spectrometric investigations of dissolved Zry hulls from the same batch as those used for the leaching tests. The results are in good correspondence with those gained before by I.L. Jenkins (AERE Harwell) with hulls of similar origin and history.

The leaching tests were carried out on laboratory scale with samples consisting of several hulls embedded in concrete and similar unembedded reference-samples.
As leaching media demineralized water (H_2O), salt brine(NaCl)

and carnallite brine (Q.L.) were used.
Some of the results are shown in the Fig. below.

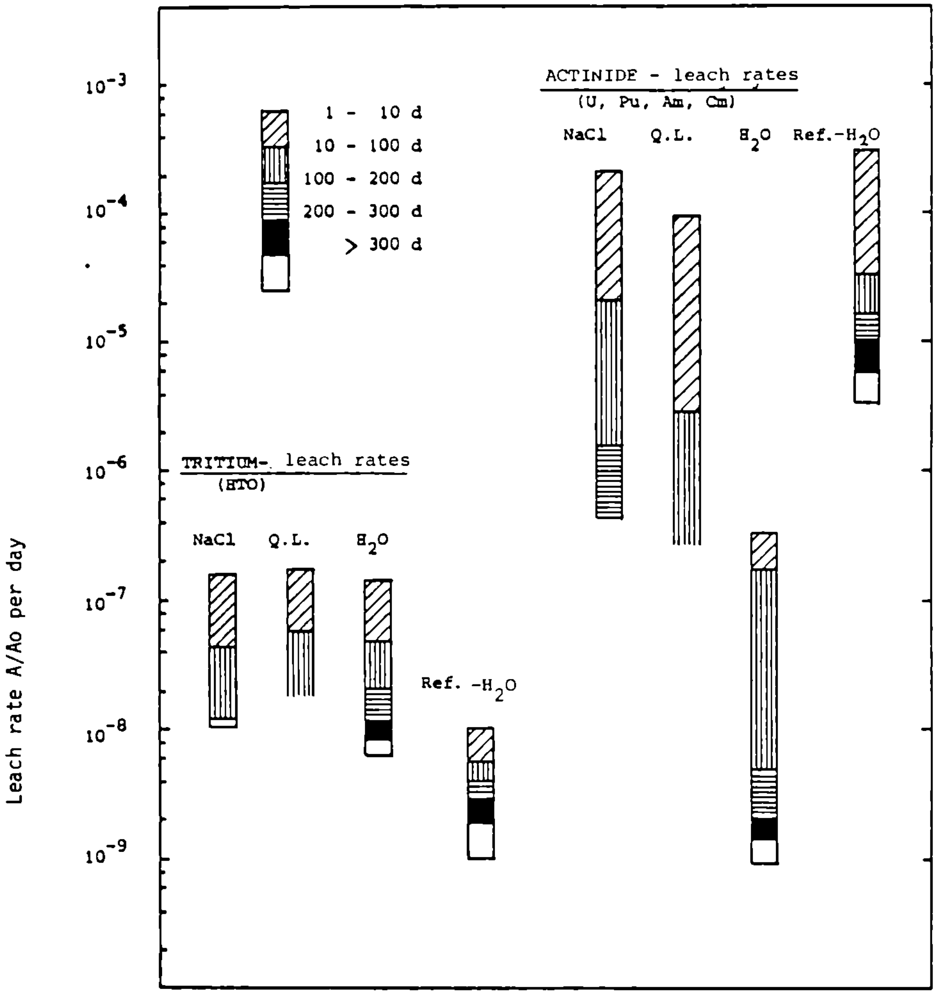


Fig. 7 : Leach rates for Tritium and Actinides of concrete embedded Zry hulls from irradiated fuel elements submerged in salt brine, carnallite brine and demineralized water.

For the determination of the radioactive decay heat from 200l-barrels filled with nuclear waste such as Zry hulls from spent fuel elements, a special calorimetric device had been developed. This device was completed as to allow remote transport, operation and maintenance in a hot waste interim storage cell.

The system was optimized for a total heat release of 10 W and can be applied as well in an isothermal as in an adiabatic type of operation.

A code was established which describes the temperature development within the device using a multi-zone model. It comprises all important physical properties of the system (e.g. specific heat, heat transfer coefficients, thermal conductivities, starting temperatures).

The calibration was performed using an electrically heated barrel. The responsiveness was determined to be 0.51 K/W average temperature difference between the inside and outside insulation surface in the case of isothermal operation and 0.0069 K/h/W average surface temperature rise for a typical metal-concrete filling of the barrel.

1.2.3.2. PYROPHORIC BEHAVIOUR OF ZIRCALOY CHIPS AND FINES

Contractor : KfK Karlsruhe (246-81-2 WAS.D)

A literature survey made by KfK in 1977 has shown that there are no data available on the pyrophoric behaviour of real irradiated Zircaloy. The examination of fines from irradiated fuel hulls as expected to arise in a reprocessing plant, is the main objective of the experimental programme.
However Zry-fines as they will be generated in situ during shearing of the fuel assemblies, chopping of fuel pins or

compaction of leached hulls will not be available. For the forseen investigations, fines will be produced from hulls of irradiated fuel pins by a tool especially developped for the purpose. Leached hulls as well as hulls from which the fuel has been removed mechanically will be used in order to get material representative for fines before and after the dissolution step. The "Bundesanstalt für Materialprüfung" (BAM) will as Kfk-subcontractor define the procedure and equipment to characterize the explosion and fire behaviour of irradiated Zry-fines. For their experiments on non-radioactive Zircaloy fines fractions < 200 and < 63 µm were required. Filings and curlings were produced by several tools and compared by means of scanning electron microscopy. The best results were obtained by a filedisk. Based on this principle a tool was developped which is capable to generate filings of the required particle size and in an acceptable rate. The device is adapted for operation in hot cells and equiped for operation under inert atmosphere.

The shape of the files was irregular and high discrepancies of length, width and heigth was noticed.

In order to determine the most suitable sieves to obtain the demanded fractions , the particle size of fractions from sieves with various mesh have been compared. Provided the particle size, defined by $\frac{\text{length + width + height}}{3}$ is smaller than 200 or 63 µm, the highest yield has been achieved by a 300 respectively 150 µm mesh size. The demanded 1.5 kg fines of each fraction was produced and the BAM has recently started the experiments.

1.3. TREATMENT OF LOW AND MEDIUM ACTIVITY LIQUID WASTE

This programme action includes the treatment processes for low and medium activity liquid wastes and the corresponding conditioning processes. The number of research projects belonging to this task amounts to 13. However, hardly half of them have been subject to noticeable development in 1981. Due to administrative problems, delays have to be expected in the implementation of the four CNEN-contracts as well as for the KFA-contracts. Moreover the contract with SENA only started in late 1981. The other contracts have progressed normally and some promising results have been achieved.

Basically, for the treatment processes, emphasis has been placed on the reprocessing wastes in considering either each stream, or a concentrate which results from the mixing of part or all of the different MAW arisings. For the wastes which can not be treated by evaporation (because of its prohibitive treatment cost or the high salt content) different processes are under development. These processes include conventional (chemical precipitation) or improved methods (combination of membrane with floc processes) as well as new ones (new organic and inorganic ion-exchangers - electrical processes - catalytic reduction of NO_x).

As to an overall MAW management strategy some orientations should result in the future from the implementation of these research activities. Particularly, two flow-sheets concerning the treatment of the same MAW-concentrate solution are under development and deal with the actinide separation or with the radioactivity concentration using chemical precipitation techniques.

For the conditioning processes, only results related to incorporation of various MAW streams in cement are, so far, available.

The scale to which each of these research activities is, at present time, being conducted is varying : from the lab-scale tests on simulates for the new processes (electrical processes, organic and inorganic ion exchangers) up to the pilot scale with fully or traced active solutions for the other ones (chemical precipitation combined or not with ultrafiltration). Moreover, it is anticipated to construct, in 1982, an industrial flocculator prototype for the treatment of real utility liquid wastes arising from PWR operation.

Since the contracts with CNEN have not yet been signed by the Italian authorities, reporting of the work performed in 1981 was not possible. This concerns four contracts (two conditioning processes and two treatment processes).

1.3.1. PRECIPITATION AND MEMBRANE PROCESSES

1.3.1.1. Separation of Actinides and Fission Products from MAW-Solutions

Contractor : KfK Karlruhe (177-81-31 WAS.D)

The liquid MAW from reprocessing plants contains actinides, fission products, inorganic and organic salts.

The present treatment process proposes the separate precipitation of actinides and fission products. It would thus be possible to reduce the long-term risk of waste and, by separating the β/γ-emitters, the shielding of the solidified waste.

The decontamination factor (DF) for actinides should be greater than 100. The DF for tne fission products was calculated on the basis of the shielding of solidified MAW-concentrates from a 350 t/a reprocessing plant (Table 1)

If the concentrates are solidified with cement and packed in 400 l drums [1], each drum contains an activity of 86 Ci. This needs a concrete shielding with a minimum thickness of 34 cm. If no shielding is used, a separation of the main radionuclides is required. The decontamination factor (DF) should then be for Cs-134/137 = 280, Sb-125 =12, Ru-106 = 6.

In the case of bitumenization and packaging in 200 l drums, each drum has an activity of 162 Ci which requires a concrete shielding of 41 cm. No shielding is required, if it is possible to reach DF's of about Cs-134/137 = 900, Sb-125 = 40, Ru-106 = 15, Co-60 = 5, Eu-154/155 = 5. If a typical 200 l drum concrete shield is used only Cs-134/137 must be separated with a DF of 20.

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(1) Actual DWK-concept.

If the bituminized concentrate is packed directly in a 200 l drum concrete shielding a separation of Cs-134/137 with a DF of 60 and Sb-125 with a DF of 14 is necessary.

The actinides are precipitated in acid solutions by oxalate-anions, which are added as oxalic acid or dialkyloxalates. Precipitation experiments were carried out with different simulates.

The precipitation with oxalic acid was done during the denitration of the waste solutions with formic acid. The complexing agents (citrates, tartrates, EDTA,...) had no significant influence on the denitration reaction. Because of the high salt-content in MAW-solutions, the non-washed precipitates contained parts of inactive salts and fission products.

Dialkyloxalates form oxalate-anions in acid solution, therefore the crystal growing of metaloxalates will be slow. Precipitation experiments with dialkyloxalate were carried out with simulates with different H^+-ion concentration. If Cerium is present, only small amounts of inactive salts were co-precipitated. This could be reduced by washing the precipitates.

Based on literature data, a process for the fission product precipitation was elaborated. It will be optimized by making experiments with simulated solutions.

Table I: Concrete shielding for MAW-concentrates solidified with cement or bitumen.
350 t/year reprocessing plant [1].

	Solidified with		
	bitumen 400 l-drum	bitumen 2001-drum	bitumen 310 l [2]
Total β/γ activity per container	86 Ci	162 Ci	250 Ci
Salt-content per container	74.5 kg	140 kg	217 kg
Thickness of concrete shielding	34 cm	41 cm	42 cm
container per year	7000	3700	2400
Volume of shielding + waste prod. per year	18 900 m^3	9 700 m^3	7 420 m^3
Volume of shielding per year	15 800 m^3	8 850 m^3	6 670 m^3
Weight of shielding + waste prod. per year [3]	43 300 t	22 400 t	17 100 t
Weight of shielding per year	38 000 t	21 300 t	16 000 t

1) MAW concentrate per year 1 500 m^3
Total activity per year $6 \cdot 10^5$ Ci
Total salt-content per year 520 t

2) in a 200 l-drum concrete-shielding

3) Density concrete (shielding) 2.4 g/cm^3
Density cement product 2.1 g/cm^3
Density bitumen product 1.4 g/cm^3

1.3.1.2. Decontamination of Low Activity Liquid Wastes from Fuel Fabrication Plants by Ultrafiltration

Contractor : NUKEM Hanau (230-81-35 WAS.D)

Purpose of the work is to investigate wether ultrafiltration is a suitable method for the decontamination of aqueous waste streams from the fuel element fabrication plant.

These waste streams belong to two categories :

- Process waste solutions with fairly constant compositions and activity levels (Table 1)
- Floor cleaning and laundry waste waters with less predictable characteristics.

In all cases a total activity level in the final effluent of 10 pCi/ml is desired. This means that decontamination factors in the range of 50 to 500 have to be achieved.

All experiments were made with a bench-scale unit having the familiar Romicon hollow fiber cartridge with a filter area of 0.1 m^2 and a nominal molecular weight cut-off of 50.000, corresponding to a pore size of about 10 nm.

First series of parametric studies were made to test whether ultrafiltration could replace some established treatment methods. Authentic waste solutions (Table 1) were subjected to ultrafiltration under varying conditions of temperature and pressure, and without pretreatment other than a prefiltration through a 0.8 μm pore size filter, in the case of the highly acidic raffinate, a partial neutralization to protect the membrane.

Although it had for long been suggested that the uranium in these solutions might be present in a colloidal form, the test results show that the waste waters listed in Table 1 are true solutions. Hence the decontamination factors achieved differed little from unity and did not vary significantly with temperature and pressure. A chemical pretreatment was therefore required.

With laundry waste water, however, a decontamination factor of 25 was obtained without pretreatment.

Long series of test runs were related to the decontamination of several types of floor cleaning waste with activity levels between 300 and 1 500 pCi/ml. Contrary to the experience made with laundry waste, the ultrafiltration without chemical pretreatment was unsuccessful. Even conventional flocculation with calcium phosphate, calcium sulfate and ferric hydroxide with amounts of calcium and iron as high as 2 g/l, followed by ultrafiltration, was surprisingly ineffective. The highest decontamination factor was obtained with the phosphate flocculation (DF = 6).

Uranium, the principal contaminant in all waste streams so far mentioned, is almost invariably present in the form of the tricarbonato uranylate complex. The stability of this complex ($K = 2 \times 10^{18}$) prevents uranium flocculation.

In a next stage it is intended to convert the uranium into a colloidal suspension of an extremely insoluble compound so that the suspension might be decontaminated by ultrafiltration.

Waste stream	Components (g/l)					pH	Activity (pCi/ml)		Enrichment (%)
	U	NH_4^+	F^-	NO_3^-	CO_3^-		α	β	
Filtrate from AUC precipitation ex UF_6	0.3	140	76		114	10.1	400	320	2.0
Fitrate from AUC precipitation ex UNH	0.4	99		70	112	10.0	950	3570	2.3
Raffinate from U-extraction from scrap solutions	0.07	0.004	0.7	210		⩽ 0.5	75	4040	2.3
Laundry waste	0.55					8.8	375	390	

Table I: Composition of U-bearing waste waters.

1.3.1.3. Active liquid Treatment by a Combination of Precipitation and Membrane Processes

Contractor : AERE Harwell (179-81-31 WAS.UK)

The aim of this programme is to develop improved processes for the treatment of medium and low active liquid waste, by the use of ultrafiltration (UF) to remove the activity in the form of colloidal or larger particles, which either already exist or are deliberately formed within the stream. The desired result is processes which have improved decontamination factors (DF's), and/or reduced volumes of final active solids, compared existing methods.

The fission products ^{137}Cs and ^{90}Sr, because of their relatively long half-lives, constitute the major activity in some medium level wastes, such as water from the ponds used to store magnesium alloy clad fuel. Conventionally the activity has been removed from the pond water by ion exchange.

Under the conditions of the pond water, it was found that addition of $TiCI_4$ solution gave a colloidal $Ti(OH)_4$ precipitate which would remove efficiently Sr activity from the effluent. Experiments were carried out on a laboratory scale using litre samples of simulated effluent spiked with ^{85}Sr. After forming the $Ti(OH)_4$ precipitate, the effluent was filtered through small 9 cm^2 flat discs of UF membrane. The $Ti(OH)_4$ precipitate behaved as an ion exchange material, giving almost complete ^{85}Sr removal from the effluent, provided its capacity was not exceeded. Under normal conditions, an addition of 4 mg/l of Ti (i.e. approximately 10 mg/l Ti $(OH)_4$) was sufficient to ensure a ^{85}Sr DF in excess of 100.

Alternative methods of filtration, including centrifugation and sedimentation, were unable to separate the colloidal precipitate from the stream and gave much lower ^{85}Sr DF's. The combined precipitation/UF process was scaled up to operation with 20 litres batches of effluent using laboratory (Amicon) ultrafiltration modules. Such modules contain 600cm^2 of membrane area and are composed of a bundle of plastic hollow fibres with 0.5 mm internal diameter. The modules were operated in batch, with volume reduction factors up to 40. The concentrate produced in such operations was always less than 20 times the concentration of the starting feed because the bulk of the $Ti(OH)_4$ precipitate deposited on the membrane surface during the volume reduction.

Tne deposits could, however, be completely removed by backwashing the module with nitric acid, and then soaking the module in such acid for an hour. The volume reduction factor (VRF) during UF was 30. The concentrate and neutralised acid wash liquor were combined, and settled rapidly to a sludge, which was drawn off, (giving an overall VRF of 500), and centrifuged (overall VRF = 1700), the supernates from the processes being recycled to the UF plant. If the compacted sludge were ultimately to be incorporated in bitumen at 50wt% solids, the overall VRF would rise to 20 000.

This work has since been scaled up further to enable processing at a rate of 200 litres per day, using commercial (Romicon) modules. These Romicon modules contained larger diameter hollow fibres and were operated with turbulent crossflow. Under such conditions the rate of deposition on the membrane was several times lower, and as a result it became possible to operate the plants continuously rather than in batch. A pilot unit has been operated continuously at a volume reduction factor of 10-20 over 5 working days (of ~ 8 hours) with only a moderate decline in flux, stable ^{85}Sr DF's (~100) and little sign of fibre bore blockage.

At the same time, comparative experiments are now being carried out with a different type of UF module. In this module the membrane is inorganic and is formed inside a tube of very much wider bore (6mm).

Radioactive ^{137}Cs was removed from the permeate from the above process after reducing the pH to 8 and adding a small quantity of a colloidal ferrocyanide precipitate.

The tendancy of the chosen copper ferrocyanide precipitate to deposit on the membrane was much less strong than for $Ti(OH)_4$, and concentration factors over 200 were achieved during volume reduction of batches of effluent.

Because of the much lower deposition rates, the used Amicon modules could even be operated continuously, over several working days, at a volume reduction factor of 10-20, with little apparent blockage of the fibre bores. The process has now been scaled up to operation with commercial Romicon modules.

Important Pu containing effluents arise during the production of plutonium oxalate (i.e. the filtrate after Pu oxalate precipitation and filtration), and during the concentration of Pu nitrate by evaporation (i.e. the evaporator overheads). Laboratory scale simulant experiments, carried out prior to the current CEC programme, had suggested that by rising the pH of the stream the Pu could be precipitated as a colloidal hydroxide and removed by UF.

Experiments have now been carried out with Pu and Am containing streams, and have confirmed that excellent αDF's can be achieved by this process. For a feed representing Pu evaporator distillate, which is essentially Pu in dilute nitric acid, DF's for Pu and Am of 10^4-10^5 were achieved by raising the pH to 11.5.

In a number of fuel reprocessing plant effluents, e.g. solvent extraction raffinates, α activity is associated with considerable concentrations of iron. By raising the pH of such effluents, a much greater amount of ferric hydroxide is brought down with the α activity. It has been shown that UF can be applied to such processes also, to increase the overall α DF by application as a polishing step after the bulk of the precipitate has been removed by sedimentation.

1.3.1.4. Treatment of Low and Medium Activity Liquid Wastes by Flocculation

Contractor : SENA Chooz (181-81-31 WAS.F)

The objective of an industrial flocculation prototype is the treatment of low and medium activity liquid wastes arising during PWR reactor operation. These wastes are the laundry drains, the decontamination flushing wastes, the cavern and floor drains.

The power station of CHOOZ uses for the treatment of liquid wastes, only one decontamination system: : evaporation. This treatment is used as well for low and medium activity liquid wastes as for wastes arising from the primary coolant. The recuperation of Boron is thus impossible.

The evaporation system price is roughly twice the flocculation/precipitation system price and one of the advantages of the second solution is also the reduced size for construction.

The flocculation/precipitation process occurs in two phases : The first one is the adsorption of radioactive isotopes by a solid product, the second one is the precipitation and separation of the liquid and solid phases.

Principal radioisotopes to be eliminated are Mn54, Cs 137, Co 58, Co 60 and Cs 134.

In earlier tests best adsorption was obtained when using a ferrocyanide of copper and with a liquid solution pH between 8.6 and 9.4.

The results have shown the following decontamination factors.

DF of 2 for the Mn 54
DF of 5 for the Co 58 and Co 60
DF greater than 10 for Cs 134 and Cs 137.

These results were obtained with a static decanter : the homogeneity being assured with an agitator.

Subsequent tests were undertaken in CHOOZ with a lamellar decanter : the maximum flow was 300 l/h. The decontamination factors obtained during a 24 hours test are :

DF of 2 for Mn 54
DF of 10 for Ce 144
DF of 10 for Co 60 and Co 58
DF of 20 for Cs 134
DF of 20 for Cs 137.

The treatment of 20 m^3 liquid wastes, gives only 120 l of muds with 97% of water or a concentration factor of 165 . With addition of "Drew Floc 306", a flocculator agent, the concentration factor reaches 825.

Based on the satisfactory results, SENA have decided to build an industrial flocculator prototype with a flow of 5 m^3/h.

Engineering of the prototype was done in 1981 and the construction will begin in the Power Station of CHOOZ in early January 1982 .

1.3.2. EXCHANGE PROCESSES

1.3.2.1. INORGANIC ION-EXCHANGERS PREPARED VIA SOL-GEL PROCESS

Contractor : Agip Nucleare (231-81-31 WAS.I)

Different forms of hydrous titanium oxydes are currently considered as ion-exchangers for separating and/or immobilizing radionuclides contained in liquid, medium and high activity radwaste streams.

At present, the exchangers are only available in the form of fine powder agglomerates not optimal for fixed and/or continuous bed operation.

Efforts are being made therefore to prepare the exchangers in more compact forms, for example by impregnation of macroreticular resin beads with sodium titanate.

The work was undertaken assuming that sol-gel techniques earlier developed in Italy by Agip Nucleare and CNEN for preparation of nuclear fuel particles might offer advantages.

Main achievements during the present reporting period were :

- Definition of a modified gel supported precipitation flow sheet for preparation of xerogel microspheres of hydrous titanium oxyde (HTiO) and/or multicomponent IX microspheres of elemental composition corresponding to the formulation of SYNROC-B immobilization matrices (SYNROC precursors)

- Characterization of the ion exchangers by means of determination of the physicochemical properties of the prepared IX materials and batch extraction tests of selected radionuclides (IX = Ion Exchange).
- Carrying out of scouting tests aimed at demonstrating the feasibility of converting loaded IX microspheres to dense ceramic bodies by cold pressing and sintering.

HTiO xerogel microspheres (250-400 microns diameter size), fired at temperatures between 100 and 500°C, were characterized by means of batch extraction tests of strontium, americium and cesium from aqueous solutions of different pH value, and specific surface area determination.

Specific surface area characteristics of the glassy highly mechanically resistant xerogel microspheres, even when fired at relatively high temperatures, were found to be well above those reported in literature for friable powder agglomerates of similar chemical composition.

For example, specific surface areas of $600 m^2/g$ and $200 m^2/g$ were measured respectively for HTiO xerogel particles fired at 200 and 600°C.

Batch extraction tests of Cs^+, Sr^{++} and Am^{+++} from aqueous solutions of pH value higher than 2.5 using HTiO xerogel microspheres fired between 120 and 400 C, showed similarly IX capacities comparable or superior to those reported in literature for sodium and titane powder agglomerates.

For example, maximum IX capacities of 5 meq. Sr^{++}/g and 1.3 meq. Cs^{+}/g were determined in extraction tests from 1.5 M NH_4OH solutions, using HTiO xerogel microspheres dried at 180°C, 60% of the total absorption taking place in the first five minutes of contact.

HTiO microspheres fired at 400°C, conserve a considerable fraction of the initial IX capacity. For example 1.5meq.Sr^{++}/g in extraction tests from 1.5 M NH_4OH solutions.

Distribution Coefficients (Kd) of Cs^{+} and Am^{+++} in extraction tests from very dilute aqueous solutions increase sharply with increasing pH equilibrium value. Maximum Am^{+++} and Cs^{+} Kds of circa $3/10^{4}$ and 10^{3} were measured respectively at pH 3.5 and 11. In these cases equilibrium was reached within the first three minutes of contact.

The extraction behaviour of SYNROC-B precursors is still under investigation.

Scouting tests made during the present reporting period in the effort to convert loaded xerogel HTiO and/or SYNROC-B precursor microspheres into dense pellets by cold pressing and sintering were succesful.

Xerogel particles, pretreated in air and Ar-5%H_2 at temperatures between 400 and 800°C, were pressed without addition of lubricant into green 1.2 X 1.2 cm cylindrical pellets of excellent mechanical resistance and densities varying between 1.4 and 2.2 g/cm^3 depending on calcination temperature and formation pressure applied (2-7 t/cm^2).

The green pellets were then fired at 1 350°C in Ar-5%H_2 yielding sintered compacts of quasi theoretical density (4.2g/cc).

In conclusion, the experimental data collected during this reporting period, show that Gel Supported Precipitation is applicable to the preparation of mechanically resistant HTiO and/or multicomponent xerogel microspheres of good ion-exchange (IX) characteristics.

1.3.3. PARTICULAR TECHNIQUES

1.3.3.1. Liquid Wastes Treatment by Electrical Processes

Contractor : AERE Harwell (176-81-31 WAS.UK)

The potential attractiveness of electrical methods stems from the fact that they can be controlled remotely and automatically by the extra reaction variable of applied voltage, which is available in addition to the normal process variables of temperature, pressure, concentration, mass transport, etc. Also, the use of electrons in place of inactive chemical reagent additions to perform redox and pH changing reactions is of considerable importance in helping to reduce the amount of material needing controlled disposal.

The processes can be broadly grouped into three classes :

1. Removal of particulates and colloids by electrokinetic effects, including electro-osmosis, electrofiltration, and electrocoagulation ;

2. Removal of dissolved ionic material by faradaic deposition as metallic elements (either as solids or in amalgams), or as insoluble compounds ;

3. Removal of dissolved ionic material by adsorption onto

high surface area electrodes and electrochemically controlled ion exchangers.

As a prelude to the experimental programme, literature surveys that formed the basis of a review completed in 1979 have been updated. Experimental investigations were then started on topics 1 and 3.

The well defined model system of colloidal ceria has been used to define the mechanism of operation of such processes as electro-filtration and electrocoagulation because of its physical similarity to plutonia colloids and also because it is available in a convenient and reproducible form. In a simple laboratory cell, Millipore filters of various pore sizes were used as the separation membranes. Under the influence of a potential gradient, particle-free liquid is pumped from the colloid in the cathode compartment into the anode compartment. EDAX analysis of the membrane showed that it has become coated with a thin layer of the colloid which effectively gave the surface a positive charge. It is the diffuse double layer (nitrate counter ions plus associated solvent) adjacent to the pore walls that is transported through the membrane under the field gradient. In order to balance the overall movement of charges, acidity is generated in the anode compartment and alkalinity at the cathode by local electrolyte decomposition. After the establishment of the membrane pre-coat; the system reaches a steady state with no evidence of pore blockage, due to electrophoresis of the particulates away from the membrane surface. The rate of passage of liquid increases with voltage by several orders of magnitude over simple filtration ($\sim 10^3$ at 40V), and the concentration of particulates in the effluent is reduced by factors of $\sim 10^4$. The pumping rate also depends on solution conductivity and the particulate ζ potential as well as the membrane pore structure. Typical flow rates of $10^3 lh^{-1}m^{-2}$ were achieved at a power consumption of 0.1 kWh l^{-1} in the non-optimized cell. At the cathode, due to a local pH rise, some degree of aggregation of the colloid occurred, producing an easily removable precipitate. A denser deposit with a very low water content was formed on the electrode surface.

The general performance parameters obtained thus far have demonstrated that wastes containing particles down to 0.01μm diameter can be efficiently dewatered with a DF of 10^4.

In comparison, evaporation has a power demand of 2 kWh $\cdot$ l^{-1}. Also the sludges have been concentrated up to ~ 19% solids- even in the simple laboratory cell. There is promise that with further developments higher solid contents could be obtained with even less power consumption.

Electrode structures have been fabricated that display potential dependent ion-exchange properties. Two types of electrode have been examined- one based on carbon powder that has been activated-either by electrochemical oxidation to generate surface "oxide" groups, and the second involving a conventional weak acid ion exchange resin.

Current densities of 1-5 mA cm^{-2} (geometric area) were used in this work. Of the various combinations of absorber, binders, current collectors and techniques of maufacture are investigated.

The most successful ion-exchange electrode investigated thus far was a PTFE-bonded large surface area carbon electrode originally intended for use in a battery application for the reduction of oxygen.

The principle of electrochemically activated ion-exchange electrodes has been successfully demonstrated in neutral solutions, and cationic concentrations can be easily reduced to below the 1 ppm level. With activated carbon adsorbers capacities of ~ 1.3 meq./g were achieved without any evidence of saturation.

The preliminary results of electrochemical adsorption have shown that electrodes containing weak acid ion-exchange groups can be made to adsorb strontium ions from a dilute solution of $Sr\ (NO_3)_2$ at a cathodic potential and elute them at anodic potentials.

1.3.3.2. Denitration and chemical precipitation of MAW-concentrate and conditioning of the highly radioactive residues by utilizing a drum-dryer

Contractor : KFA Jülich (178-81-33 WAS.D)
(180-81-33 WAS.D)

The aim of this programme is to adapt a process for the conditioning of medium active liquid wastes which has originally been developed for the vitrification of fission product solutions.

Fission product solutions removed in the first step of the solvent extraction during fuel reprocessing contain about 98% of the total activity concentrated in a comparatively small volume. The amount of medium active liquid wastes generated in the fuel cycle and by decontamination processes is significantly higher. By adapting the vitrification technique which includes :

- denitration
- separation of radionuclides by chemical precipitation
- drying of the sludge with suitable additives on a drum-dryer
- vitrification,

the quality of the product could be improved and the expenses for transportation as well as for storage could also be reduced by separating a highly radioactive fraction from a larger amount of low activity waste.

The activities, started late in 1981, have mainly been focused on technical preparations, like the testing of analytical methods. Formaldehyde determination has been cold tested obtaining good results. Only preliminary work has been done on nitrate detection by means of a nitrate ion-specific electrode. The results suggest this method to be susceptible to disturbances in waste solutions. Gamma-ray spectroscopy, alpha-spectrometry and X-ray fluorescence had been adapted earlier to the process control of reprocessing. Their applicability to the present process of waste conditioning, especially the sample preparation, has still to be proven.

The hot cell apparatus together with technical documentation has been completed and cold testing has begun.

The life time of the equipment is essential for hot cell operation. In particular suitable materials have to be found for the drum-dryer.

The drum-dryer of the laboratory cold test equipment has been coated with a ceramic layer and the blade holder has been supplied with two dynamometers. This will allow better control of abrasion during experiments for product development.

1.3.4. IMMOBILISATION METHODS

1.3.4.1. Incorporation of Low and Medium Activity Wastes in Cement

Contractor : AEE Winfrith (166-81-15 WAS.UK)

The laboratories and rig area for small and large mixing and testing of simulated radwaste/cement composites were completed in March 1981. The range of equipment now operational for laboratory scale work includes conduction calorimeter, viscometer, apparatus for compressive strength, permeability and dimensional stability, simultaneous thermal analyser and other standard BS testing equipment. Equipment for mixing waste with cement using in-drum batch and continuous processes up to 220 l scale is operational, together with facilities for the evaluation of 220 l size waste/cement products.

A range of mixes up to 40% (m/m) sodium sulphate decahydrate (17.6% anhydrous) have been prepared with Ordinary Portland cement (OPC) and are undergoing extended evaluation.
Sodium sulphate has an accelerating effect on the hydration of cement. This is observable both in the exotherm, obtained by conduction calorimetry, and the faster rate of strength development. The specimens are being assessed for long term stability by observing the dimensional changes that occur with (a) storage under normal curing conditions-air at high relative humidity, (b) storage under water, and (c) freeze - thaw cycling. Under normal curing conditions there is a concentration of sodium sulphate (around 17% m/m) that gives a dimensional change. However, under water all concentrations above 5% (m/m) show marked deterioration with time. Samples prepared using a 90% replacement of the OPC with ground Blast

Furnace Slag (BFS) have shown much superior stability under all test conditions. The leach rates for caesium are also significantly reduced.

A range of mixes up to 26% (m/m) loading of sodium nitrate in OPC and BFS/OPC have been prepared and assessed in the same way as the sodium sulphate samples.
In contrast to sodium sulphate, sodium nitrate retards the setting of OPC. The results obtained on dimensional changes indicate that, under all conditions of storage, sodium nitrate OPC blocks are more stable than sodium sulphate/OPC - with the BFS/OPC samples showing even smaller dimensional changes.

A simple pretreatment of organic cation exchange resins, to alter the equilibrium concentrations of sodium and calcium ions present, has enabled the preparation of stable waste forms with OPC and particularly, BFS/OPC mixes. Samples have been under test for 6 months with no observable deterioration.

The use of clays available in the UK have been investigated with the objective of reducing leach rates, mainly of caesium from cement/waste products. Of the clays tested bentonite and vermiculite gave lowest leach rates ; typical concentrations of clays required is between 5 and 10% of the total solids (2% for vermiculite). The best results again being achieved with 90% BFS/OPC mixes.
Three mixing systems based on different principles are available at Winfrith. These are :

(i) Batch mixing, based on high speed shear mixer.
(ii) Continuous mixing using a barrel containing two mixing/ conveying agitators.
(iii) In-drum mixing, using a 10 HP hydraulic piston motor capable of speeds between 1 and 200 rpm.

Mixes with the batch mixing equipment have shown reasonable homogeneity with respect to strength. The energy input during mixing can raise the mix temperature to 40° but this does not affect the subsequent hydration exotherm.

The continuous mixer used in these experiments is a laboratory scale (2"diameter) continuous mixer designed to mix viscous products using a combined mixing and conveying action.
Cement powder is continuously metered, using a variable speed vibrating screw feeder, into the top of the mixer barrel. A helical screw transfers the cement towards the agitators. Liquid or slurry wastes are continuously metered, using a peristaltic pump, into one of two injection ports in the mixing chamber. The product is discharged from the bottom of the barrel. 220 l mixes prepared with simulated sodium nitrate wastes in OPC show drum centre temperatures during setting in excess of 100°C. Subsequent core samples show that the high temperature zones have higher caesium leach rates than the samples that hydrated at lower temperatures.

1.4. PROCESSING OF ALPHA-CONTAMINATED WASTE

Most alpha wastes originate from "mixed oxide" fuel production and from fuel reprocessing.
The quantity of such wastes will increase in function of the growth of electricity generation in nuclear power stations. It is, therefore, important to improve those processes and methods which will enable alpha waste management and storage to be optimized in the future. To this end, a best possible compromise must be searched for between economic constraints, technical and technological know-how, safety of the population and conservation of the environment.

The more advanced these processes and methods become, the easier it will be to achieve this compromise. These different, sometimes contradictory aspects have been taken into account in drawing up the programme for research on the treatment of this type of waste.

In the previous Community programme the current state in the field had to be taken into consideration and the selection of works to be sponsored was based on initial results obtained in laboratories.

The present programme, which reflects the desire to be consistent and effective, takes account of the directions taken and the selections made in the first programme, of the first results obtained and of the latest technological knowledge in the field. For the various processes under consideration significant progress may be stated.

As to volume reduction of wastes by high temperature incineration giving a final product with good leaching properties suitable for disposal, a pilot plant has been put in operation. For the recovery of Plutonium in order to be reintroduced into the fuel cycle, a pilot plant has given satisfactory results with inactive material and the construction and putting into operation of a pilot plant with active material has just been completed. Also the optimization of pyrolysis processes, the selection of materials for the molten salt technique, the electrolysis or the relatively new, but interesting, method of "washing-leaching" have continued to advance during the reporting period.

Another important aspect of the management of this type of waste, i.e. the measurement of the plutonium contained in drums of waste, has been the subject of a joint research programme involving five laboratories in the European Community The problem of comparison of techniques and equipment for the interpretation of the results obtained could find a solution by this cooperation. It also proved to be the quickest way of developing a method with sufficient accuracy and reproducibility for industrial utilization. The production of sources and drums for this programme has practically been completed and the first measurements have begun in one of the laboratories concerned.
It should also be pointed out that the products obtained from the development work on the processes are subjected to "characterization" tests which are carried out in parallel .

1.4.1. INCINERATION AND PYROLYTIC METHODS

1.4.1.1. Recovery of Plutonium from Combustible Wastes

Contractor : AERE Harwell (185-81-43 WAS. UK)

The aim of this research is to provide information on the recovery of plutonium by the leaching of residues from incineration or pyrolysis of specific waste materials (plutonium compound), and to compare the recovery efficiencies achieved with those from alternative processes such as washing and acid digestion.

Most of the work carried out during the 1981 calendar year has concerned the preparation and leaching of incinerated / pyrolysed residues.

Preliminary washing studies have allowed an initial comparison to be made.

The alpha-active model incineration/pyrolysis furnace that was commissioned towards the end of the previous Programme has been used to prepare a large number of ashes/ residues for leach testing. Individual as well as mixtures of combustible materials typical of those found in plutonium contaminated wastes have been heat treated in the furnace. The various wastes have been either incinerated in air, or pyrolysed in pure nitrogen or pyrolysed and the residual char subsequently oxidised in air, at various temperatures from 550° C to 900° C.

The majority of experiments in this period have used waste mixtures contaminated with plutonium oxide prepared by calcination of the oxalate at 500° C. Other contaminants have included 800° C calcined plutonium oxide, solid solution and physically blended uranium - 30 % plutonium oxides, and plutonium nitrate and uranyl plutonium nitrate solutions. The quantities of contaminants added were such as to give ca. 6gPu per kg of waste.

The three waste mixtures studied have all contained polyvinylchloride, polythene and cellulose tissue, together with either Neoprene, low or high ash Hypalon as the glove-box qauntlet constituent.

The percentages of incinerated ash or oxidised char obtained with the three mixtures show significant differences (up to a factor of 4), reflecting the differences in inorganic filler content of the constituent contributing most to the mixture residue. The percentage residues for a particular material or mixture are comparable whether that material has been incinerated or charoxidised, suggesting little difference in residual carbon content. Similarly the change in percentage residue varies only marginally with temperature over the range 550-900° C, suggesting that even at the extremely low temperature of 550° C it is possible to produce an ash of relatively low carbon content. These suggestions are confirmed by carbon analyses which generally are below 2 % by weight.

Similar low values are also found for the chlorine content of the residues, with surprisingly higher values (1-2 %) resulting from the treatment of Neoprene or Hypalon compared with polyvinylchloride ($\leqslant$ 0.1 %), despite comparable levels (32-36%) in the start materials.

Leaching studies have been carried out on most of the ashes and chars prepared as described above. As far as possible a standard leach test has been used to enable a comparison to be made between the various treatments given to the contaminated materials. Only a limited range of pyrolysis chars have been prepared and leach tested as the leaching mixture tends to froth and the carbon present may in some cases protect the plutonium from attack by the acid. Most experiments have been carried out on the incinerator ashes or pyrolysis chars after oxidation.

Leaching of pyrolysis chars followed by char oxidation has shown that the oxidation temperature rather than the pyrolysis temperature plays the key role in determining the leachability of the plutonia. The plutonium becomes less soluble above a temperature in the region of 700-750° C for both incinerator ashes and oxidised pyrolysis chars, but for a given temperature the plutonium is less easily leached from the incinerator ash than from the corresponding oxidised char. This is presumably because the char oxidation is a relatively slow process and, unlike the burning which can take place during the initial decomposition of materials in air, is not likely to lead to a rise in temperature. The differences are least at 550° C and despite the relatively low temperature an essentially carbon free residue is obtained.

As reported before the quantity of inorganic filler present in the combustible materials plays a key role in the leachability of the plutonium from the ash. Low ash content materials lead to a much easier dissolution of the plutonium. The type of contaminant also plays a role in the leachability of the plutonia; if the contaminant is $(U,Pu)O_2$ or $UO_2(NO_3)_2$/$Pu(NO_3)_4$ solutions (30 % $Pu/_{(U+Pu)}$) at least at low temperatures up to 90 % of the plutonium can be dissolved in 10 M nitric acid with no added calcium fluoride in a single leach.

Comparative studies on washing using 1M NaOH have shown that recoveries of ca 90 % can be achieved for many plutonium contaminated materials, e.g. PuO_2 or Pu $(NO_3)_4$ contaminated rubber but recoveries of 90 % are difficult to achieve with some materials, e.g. PVC. Mixtures containing cellulose give much poorer recoveries. Thus it appears that, if very high recoveries are required, incineration or pyrolysis followed by char oxidation may be a better process.

1.4.1.2. Incineration of Plutonium Contaminated Waste

Contractor : UKAEA Springfields (186-81-44 WAS.UK)

The kinetics of pyrolysis at 600-800° C of various solid simulated plutonium contaminated materials (PCM) have been determined experimentally. The behaviour of individual materials and mixtures was assessed using sample sizes of up to 1000 g. The products of pyrolysis such as chars, tars and noncondensible gases were determined.

Individual solid PCM waste commence significant pyrolysis at different temperatures. Pyrolysis of polythene starts at about 350° C whereas chloroprene (neoprene) only needs to be heated to 200° C. Other materials such as hypalon and PVC have intermediate initial decomposition temperatures. The rate of decomposition and hence the volatile generation rate of the solid materials considered vary by a factor of two. The time for 90 % of the volatiles to be liberated has been used to compare the various pyrolysis rates. In the case of 1000 g packages of polythene and polyurethane, pyrolysed at 700° C for example, these times were 35 and 17 minutes respectively.

The distribution of pyrolysis products depends on the original material's chemical composition. Broadly, the PCM solid wastes can be devided into two categories :

(a) Those that produce significant quantities of char, and

(b) Those which form little or no char.

Hypalon, PVC, neoprene and paper tissues yield 32.4, 13.9, 24.7 and 19.4 % wt char respectively, whereas polythene and polypropylene yield no char and polyurethane only 5.3 % wt. The volatiles are primarily non-condensible hydrocarbons and HCl (in the case of the chlorinated plastics) and tars.

Pyrolysis data on individual materials can be used to predict, to a first approximation, the char yields and also the amount and composition of volatiles from mixtures of solid PCM waste. However, the rate of pyrolysis of mixtures of PCM wastes cannot readily be predicted. The time to achieve complete char formation with 132 mm dia. packages (1000 g) at 700° C for one mixture was 26-28 min whereas another with 30 % polythene took 48-50 min. This increased pyrolysis time is probably related to the slow pyrolysis kinetics of polythene.

The oxidation rates of chars derived from individual materials and mixed packages are being determined. Packages containing about 1 kg of PCM waste have been pyrolysed at 700° C and oxidised again at 700° C with air flowing over the upper surface of the undisturbed material. In general, the oxidation rate decreases with time as ash accumulates on the upper surface. Therefore, in order that meaningful comparisons can be made, the initial oxidation rates have been used. These oxidation rates varied from 6.0×10^{-3} g cm^{-2} min^{-1} for hypalon char to 9.1 x 10^{-3} g cm^{-1} for chloroprene char. The chars derived from two mixtures of PCM waste oxidised at 6.5 x 10^{-3} and 8.5 x 10^{-3} g min^{-1} cm^{-2}, the higher rate is probably related to the high chloroprene content of the original mixture. In order to achieve 90 % oxidation of the available carbon in the chars from these mixtures oxidation times of 120 min were required. Increased oxidation rates will only be achieved if the char is mixed with the oxidising gas, for example in a rotary kiln.

1.4.1.3. High Temperature Incineration of Radioactive Waste

Contractor : SCK-CEN Mol (190-81-42 WAS.B)

The work concerns the exploitation and further development of the FLK-60, high temperature incineration installation.

During 1981, most of the equipment has been completed and two incineration campaigns have been carried out, one in March and another in the last quarter of the year.

The second compaign lasted 10 weeks and allowed to check the working of all components of the installation. Most emphasis was put on the gas purification line and especially on the bag filters which were used for the first time.

About 13 ton of solid waste has been shredded and mixed during the campaign. It consisted of 80 % paper, plastic and wood; 9,5 % recycled granules, 5 % sand, 4,5 % glass and 1 % metal. No sludge was added.

The burnable part of the waste was slightly active, the radiation intensity being limited at 5 mR/h at contact for a 15 l-bag. All other constituents were inactive.

During the working time, the temperature in the main combustion chamber was kept between 1450 and 1500° C; that in the auxiliary combustion chamber between 1150 and 1250° C in the first four weeks and between 1300 and 1350° C thereafter; that in the postcombustion chamber between 950 and 1100° C.

The mean combustion rate during the campaign was about 30 kg/h; a value of about 50 kg/h was reached during the 5th working week. Those values are far below the maximum capacity.

The off-gas temperature at the inlet of the bag filters has to be carefully controlled at 205° C to avoid thermal destruction of the Teflon fabrics or water condensation and corrosion of the filter walls and internals.

The bag filters have been put in operation in the 2nd working week.

No measurement of the dust concentration has yet been carried out.

The bags can be cleaned by automatic blowing of compressed air at 5 bar. This can be carried out during working of the filter and the time interval between two cleaning operations is adjustable.

Without bag filters, the pressure drop through the venturi is to be kept above 1000 mm WG to avoid too frequent interventions at the active HEPA filters.
With the bag filters working, the venturi plays a minor part and the pressure drop can be kept low.

Dust from the "main" bag filter is slightly acid and for 87 % soluble in water.

Detailed analyses results of those samples are expected in the first months of 1982.

1.4.1.4. Incineration of Alpha-Waste in Molten Salts

Contractor : Agip Nucleare (191-81-44 WAS.I)

During the first six months of the year 1981, several tests of treatment of non-active synthetic mixtures of materials simulating the wastes (paper, rubber, wood, rags, PVC, etc.) have been carried out in the pilot plant for incineration of wastes in molten salts (20 kg incinerated material).
The plant proved to be functional on the whole, with a capacity between 0.5 and 1.5 kg/h waste.
However it seemed convenient to introduce in the plant some changes concerning the feeding and conveyance system for granulated waste, in order to improve its efficiency.
Some troubles occurred in the course of the next tests :

- a heating section of the gas post-combustion electric furnace broke and the maximum operating temperature provided for (950 to 1000° C) could not be attained;
- the gas post-combustion (Inconel 600) chamber, due to some corrosive phenomena which could not be fully cleared up, got splitted at several spots and the damage could not be repaired immediately.

Therefore the operations of the plant had to be temporarily interrupted.
During the second half of 1981 the following works have been carried out :

- planning, manufacturing and installation of e new stainless steel tank for the waste feeding, with a reduced capacity and being provided with a different type of stirrer and a different bottom opening for the passage of waste to the drop tube;
- replacement of the exhaust-fan for the pneumatic conveyance of waste by one with improved capacity, and placing of new conveyance lines;
- re-manufacturing of the gas post-combustion (Inconel 600) chamber, by making use of some parts of the old chamber. Also the air/gas mixing has been modified which now takes place inside the chamber by means of two concentric inlets;
- installation of a gas absorption unit above the post-combu-

stion in replacement of the scrubbing bottles. The unit consists of a glass filled Rashig ring column and a circulation pump for the neutralizing liquid (sodium hydroxide 5 % solution);

- purchase of a new electric furnace for the post-combustion of gases (25 kW, kanthal resistors and refractory spark plugs) for a maximum temperature of 1100° C.

A schematic view of the plant, without the changes under way, is shown on Fig. 8.
Tests on the behaviour of Inconel 600 under high temperatures and in the presence of post-combustion gases have been done as well as experiments on the treatment of ion exchange resins, mineral oils and tributylphosphate. Salt samples have been prepared with a known content of waste combustion ashes and uranium, in order to be sent to CEA Fontenay-aux-Roses for uranium electrolytic recovery tests according to the existing cooperation agreement (cf. 1.4.1.5. below).

Tests on the behaviour of materials being consistent with the molten salt in acid environment have been carried on in laboratory, by submitting ceramic crucibles vitrified on the surface, and graphite crucibles to a tightness test at 600° C in presence of molten sulphates containing sulphuric acid.
Also the vitrification of filtered residues, consisting of combustion ashes included in salt, has been tested. The chosen compositions were silico-titanates glasses which have been submitted to leaching and have shown good results.

1.4.1.5. Recovery of Alpha-Wastes by Molten Salt electrolysis

Contractor : CEA Fontenay a.R. (192-81-44 WAS.F)

Several processes are presently being investigated to recover alpha wastes, specially plutonium, from various materials such as rubber, cotton, paper or plastic products.

AGIP NUCLEARE (Bologna, Italy) is conducting tests to

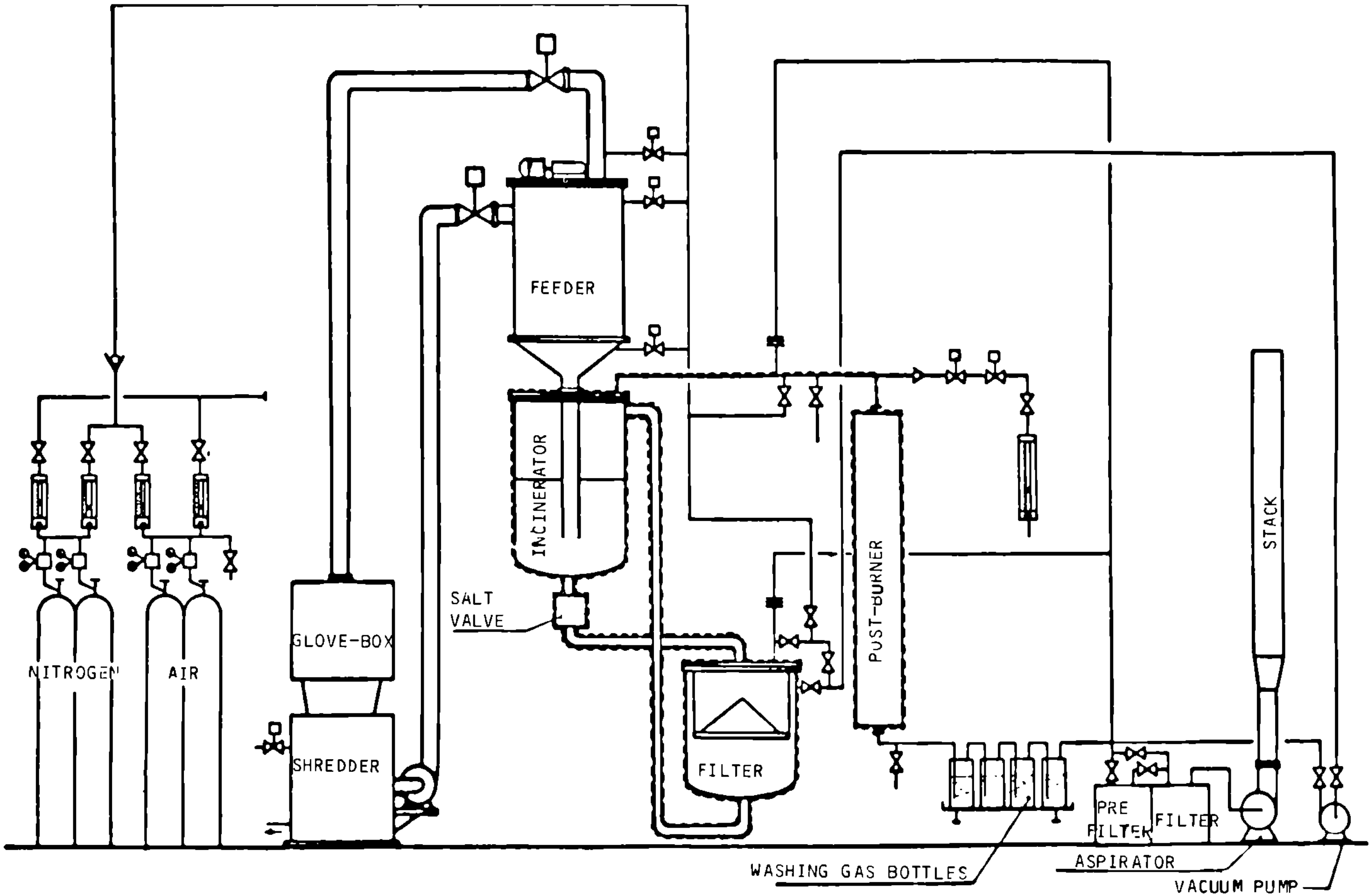

Fig. 8: PILOT PLANT FOR RADIOACTIVE WASTE INCINERATION IN MOLTEN SALT

"solubilize" the wastes by pyrolysis in molten sulfates, i.e. to reconcentrate these wastes by volume reduction.

The four steps of the AGIP process are :

a) pyrolysis ("incineration") of wastes in a molten sulfate bath, at 600° C, under an air-nitrogen mixture;

b) treatment of pyrolysis gases by combustion at 1,000° C under air atmosphere;

c) dissolution of the U-Pu ashes present in the molten salt by a sulfuric acid treatment at 550° C, and then electrolysis with recovery of UO_2-PuO_2;

d) filtration of the salt bath.

The pyrolysis tests carried out by AGIP (mostly on uranium) brought satisfying results mainly on laboratory scale.

On the contrary, the recovery tests for UO_2 and PuO_2 by electrolysis presented several difficulties so that AGIP wished to solve them with the help of our own experience in the field of molten salt electrolysis.

The principal results were obtained from tests at 580° C in special electrolysis cells of the CEA, under pure argon, in three types of baths :

- pure solvent;
- an approximately 0.5 % U bath (essentially from AGIP) referred to as "diluted";
- an approximately 10 % U bath prepared by CEA through sulfuric dissolution and referred to as "concentrated".

Before beginning the tests with uranium-containing baths, a few preliminary determinations in the form of polarization curves were conducted in pure ternary solvent K_2SO_4 - Li_2SO_4 - Na_2SO_4 (MP = 512° C) and with different electrodes. A rather large difference was evidenced between the decomposition voltage of platinum electrodes (U = 2.6 volts) and the decomposition voltage of graphite electrodes (U = 3.5 volts).

Therefore the graphite electrodes can advantageously be used instead of the platinum electrodes initially selected by AGIP.

As to the tests *with diluted bath (about 0.5 % U from AGIP)* the electrodes were either platinum or graphite and current densities (anode or cathode) about 2 $A.dm^{-2}$ (voltage up to 3.5 volts). The duration was about 10 h.

Under such conditions, only thin and slightly adherent deposits were obtained (e<1 mm), while a black deposit was often also recovered on the bottom of the crucible.

The electrolyses with these diluted baths are complex and several complementary tests are necessary to ensure a valuable process.

In "concentrated" baths (about 10 % U, prepared by CEA) the electrodes were also either platinum or graphite; the current density was 10 to 80 $A.dm^{-2}$ (voltage : about 1.5 volt); intensity was 600-800 mA at the start of operation and the test duration about 10 h.

Under such conditions, the deposits are much better. They show "honeycomb" morphology (Fig. 9) and are much thicker (a few mm) than those of diluted baths. Moreover, the adherence and the mechanical strength are excellent. The reproducibility of results is sitisfactory.

A further investigation of the various characteristics such as faradic yield and current intensity was carried out particularly by analyzing their variations versus the depleting rate of the bath in "UO_2^{2+} uranium ions".

In that case we obtained relevant results showing clearly a

Fig. 9: Uranium oxide deposit obtained by molten sulfate eleltrolysis, at 580° C ("concentrated" bath, approximately 10 % uranium)

high yield of about 90 % when electrolysis is starting, down to about 10 % at the end when U concentration is lowered to about 1 %. Intensity decreases, like wise from 650 mA down to 50 mA. We find here again approximatively the values of 30 mA given by AGIP for diluted bath.

This study of yield and intensity characteristics confirm the operations with concentrated uranium baths. Of course we have also to study wether these results give a satisfactory "extraction" yield and a good purity. (Tests presently carried on).

Almost all the investigations were conducted in "synthetic" baths prepared by CEA. They have to be checked with pyrolysis AGIP baths in particular to examine a possible influence of pyrolysat elements on intensity potential curves, on faradic and extraction yield as well as on purity level.

1.4.2. WASHING AND LEACHING, CHEMICAL TREATMENT

1.4.2.1. Conditioning of Plutonium-bearing Waste by Acid Digestion

Contractor : KfK Karlsruhe (187-81-45 WAS.D)

On the basis of data and experience gained with the inactive operation of a test facility at KfK (1), a prototype plant for active process demonstration called ALONA has been constructed at Eurochemic in 1981. The plant is part of a set of installations for conditioning the plutonium-bearing waste stored at Eurochemic. The other parts are the unit for waste pretreatment (waste sorting and preshredding) and the unit for plutonium separation and purification. The nominal throughput of the plant is 10 kg of waste per day.

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(1) H. Wieczorek et al.; EUR 7052 d (1981)

The plant has been constructed according to the process flowsheet shown on Fig. 10. The material of construction is technical glass. The flanges are sealed with gaskets made of PTFE.

As far as criticality problems are concerned, all the installations along the plutonium path are designed to be criticality safe by geometry. The process components have been placed according to their function in seven glove boxes of a total volume of 60 m^3. They include : final waste shredding (cutting mill), waste digestion (ring reactor), off-gas treatment (3 boxes), acid recovery and residue separation (containing plutonium). A view of the plant is shown on Fig. 11. Included in the plant are also the process control instruments, a cooling and underpressure system and radiation shielding (against americium).

Since autumn 1981 the phase of inactive tests is continuing. This phase also includes the training of the personnel in plant operation and maintenance. The active operation is planned to start in May 1982. It is foreseen to operate the plant in three shifts. During the first shift the waste processing, residue separation, feeding and discharging of the plant will take place. In the two other shifts the plant will be operated (stand-by operation at 250° C) to complete the conversion of plutonium oxide to plutonium sulphate and also for cooling down the reactor acid (2 h).

1.4.2.2. Washing Processes for Plutonium Recovery from Solid Wastes

Contractor : ALKEM Hanau (188-81-44 WAS.D)

Aim of this R+D-programme is to evaluate the potential of mechanical washing processes for the recovering of Plutonium and Uranium from primary wastes generated during fabrication of Plutonium containing fuel.

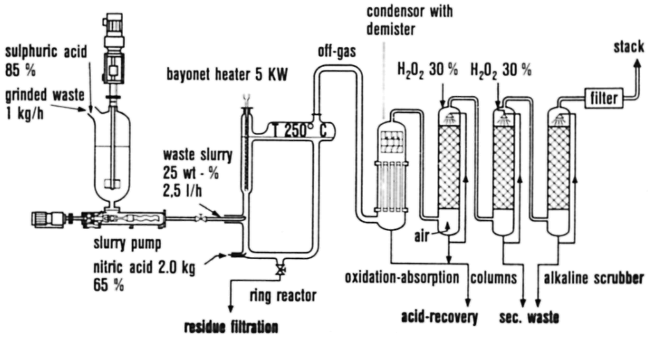

Fig. 10 : Active lab-scale plant ALONA process flowsheet

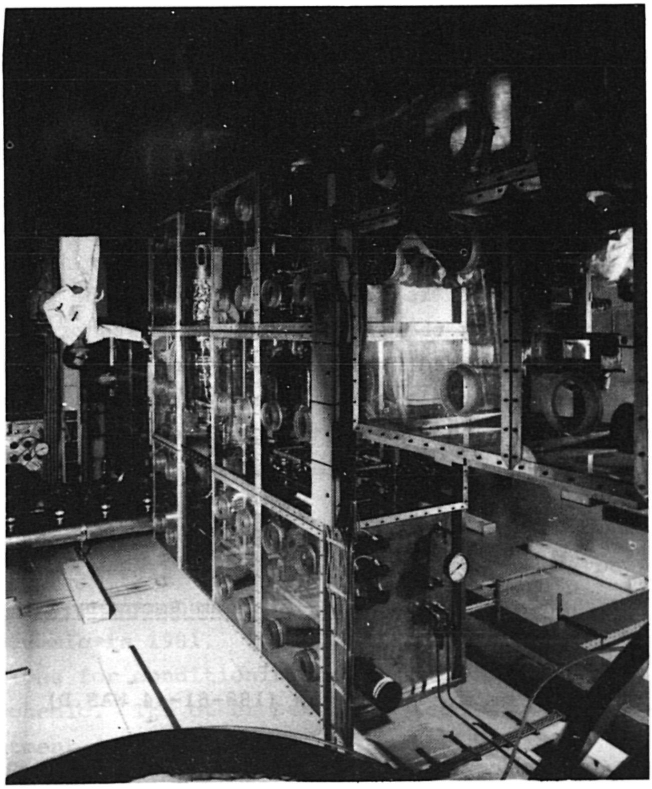

Fig. 11 : Active lab-scale plant ALONA
left : final waste shredding box,
center : waste digestion box

Objectives of the R+D-programme are :

- to establish a washing process with sufficient decontamination effect applicable for the decontamination of solid alpha bearing waste,
- to recover fissionable elements from washing solutions in order to avoid secondary wastes of Plutonium.

After preliminary investigations on laboratory scale for the determination and optimisation of processes and process parameters the selected washing process will be tested on a technical scale and its efficiency will be determined with original wastes.

The characteristics of the existing waste components have been identified and a significant number of waste packages were measured by neutron interrogation. From the material evaluated 55 w/o of the box wastes are organic materials with different contamination levels.

Approx. 30 % of all packages contain more than 0,5 g Pu (> 250 mg Pu/kg).

In order to assess the influence of the structure on the washing results, scanning-electron-microscope photos of non-treated waste materials were made. For using aqueous wash solution, some work was devoted to select a suitable detergent. Various detergents have been examined with respect to foam production between 20° and 100° C, to the dry/glow residues, to the pH-value in solution and to the Pu-decontamination factor in conjuction with various waste materials. (For example, cellulose waste material do not allow the use of aqueous washing solution).

Before starting washing experiments with original wastes generated during MOX-fuel fabrication, tests were carried out with U-contaminated wastes in a flask shaker and a small lab-washing machine, built for that purpose at our workshop. It was found out, that dilute nitric acid (< 1 m) as well as aqueous solution with an additive detergent are suitable for the cleaning of UNH-contaminated polyester-wipes. The cleaning rate was in the range of 95 - 99 %. UO_2-contaminated materials can be cleaned in a neutral aqueous solution with nearly the same efficiency (94 %) in the lab-washing machine.

Cleaning with trifluortrichloräthan, a carbon-hydrofluoride-compound, in a stirred bath in combination with the use of ultrasonic energy seems to be a good alternative, especially for UO_2-contaminated hard materials like metal, glasses, ceramic and some synthetics. UO_2-purification of up to 95 % has been reached.

This medium is also the only possibility to clean contaminated cellulose-wipes, e.g. filters. The best washing result was 73 % up to now.

Since the end of 1981 the lab-washing machine tested with U-bearing simulated wastes is working in the active area to check the good U-washing results with original Pu-bearing waste. For the first test series neutral aqueous wash solution has been chosen for cleaning PuO_2 bearing polyester wipes and neoprene box gloves. The results from UO_2-waste washing tests are confirmed in these preliminary investigations on Pu-purification (up to 93 % for polyester-wipes). The results of these first tests also indicate a possible correlation between washing efficiency and the life time of the organic material within the active area.

1.4.3. ALPHA-MONITORING OF WASTES

1.4.3.1. Measurements on Standard Waste Drums

Contractor : CEA Cadarache (182-81-42F)

The programme should enable different European Laboratories participating in the development of techniques for the monitoring of plutonium-contaminated waste, to compare their results by conducting measurements on the same set of waste drums, to exchange information on the techniques employed and to examine jointly and in detail the reasons for discrepancies.

The activities of the programme comprise :

- Preparation of the material to be measured (active samples, waste drums),
- Calibration of the active samples,
- Definition, measurement and interpretation of the different configurations to be composed,
- Exchange of information between the participants,
- Destructive analysis of the real waste,
- General conclusions.

The first four items are covered by the present contract. A supplementary contract covering the years 1983-1984, is to be drawn up with regard to the last two points.

In 1981, the work covered the definition of the configurations for measurement and preparation of the material to be measured.

The chosen configurations concern three major categories of light-matrix waste drums :

- Modular synthetic-waste drums for parametric studies,

- Sealed drums containing known samples of synthetic waste to test the measuring equipment,
- Real waste drums for which non-destructive and destructive measurements will be compared.

A modular concrete drum is planned to be used for parameter studies concerning heavy matrices.

For the parameter studies on synthetic-waste and modular drums, three types of contaminant, four sample sizes, three light-matrix densities and three drum sizes were selected, giving in total 83 different configurations.

As to the testing material light polyethylene-foam matrices have been prepared in the form of homogeneous cylinders having the same dimensions as the container drums. Four longitudinal holes will be made in each cylinder to allow the contaminants to be inserted.

A concrete cylinder has been ordered also with 4 longitudinal holes.

The contaminants are in the form of small cylinders made of stainless steel which contain powdered plutonium oxides.

They have been produced and the measurements of neutron and gamma radiation is currently in progress.

Three or four samples of each of the three different types of contaminant were selected to serve as standards. They were sent to a reference laboratory to be calibrated by means of calorimetric techniques and for the measurement of neutron and photon emission.

1.4.3.2. Comparison of different Monitoring Techniques for alpha bearing Waste

Proponent : AERE Harwell (ref. 183)

The work is a contribution to the abovementioned programme, aimed at comparing different techniques and instruments for the measurement of plutonium in solid wastes.

The experiments will have to show whether the existing methods and equipment are satisfactory or need improvements.

The contribution to the common programme will involve the measurement of a series of 200 litre drums containing different waste materials and known plutonium samples.

The work is planned to start in 1982.

1.4.3.3. Comparative Measurements on Waste Drums

Contractor : KfK Karlsruhe (184-81-43 WAS.D

The work will begin in January 1982. In the scope of the abovementioned programme measurements will be done on a complete set of drums provided by CEA - Cadarache and KfK(Mol).

The measurement work will be followed by a joint evaluation.

1.5. TESTING AND EVALUATION OF SOLIDIFIED HIGH ACTIVITY WASTE

Based upon the results of the previous five-year programme (1975-79), research in the present programme (1980-84) is almost entirely devoted to the study of reference borosilicate glasses. In addition to 6 materials (5 glasses and 1 glass-ceramic) already tested in the frame of the previous programme, the AVB and the (new) PAMELA glass compositions, (to immobilize EUROCHEMIC waste) were included. In the following the results achieved during 1981 of six participating laboratories (Hahn-Meitner-Institut, AERE Harwell, CEA Marcoule, SCK/CEN Mol, Fraunhofer Institut and University of Leiden) are reported. As CEA Saclay has only recently joined the programme, its contribution cannot yet be reported.

The emphasis of the research programme is directed towards a better understanding of the basic leaching mechanisms and towards testing under simulated repository conditions. The main items of the joint test programme are :

- Investigation of basic leaching mechanisms by analyses of the leachant, the gel layer and the chemical form of dissolved radionuclides.
- Isotope-specific leaching tests and leachant analysis.
- Small-scale simulation of repository conditions, i.e. leaching tests in the presence of canning and backfill materials and in ground water according to the various candidate formations.
- Influence of temperature, pressure, pH, leachant flow rate and composition and leaching mode : complementary to previous programme (elevated temperatures and pressures).
- Long term evolution of the leach rates.
- Alpha radiation stability : continuation of tests on Pu-doped samples started previously and leaching of heat treated doped samples. Testing and crystallographic

examination of 4 active block samples at the Hahn-Meitner-Institut.

- Thermal stability : effects of devitrification.
- Mechanical stability and development of cracks.

The University of Leiden is examining in a limited programme the radionuclide retention capacity of the mineral compounds of the "Synroc" material.

Provisions to carry out a Round Robin Leaching test during 1982/83, with the aim to develop a standard method for quality control purposes, were elaborated during 1981.

1.5.1. LEACHING MECHANISMS OF THE GLASS MATRIX

1.5.1.1. Testing of the Radiochemical Resistivity of Glass Products

Contractor : CEA Marcoule (123-80-55 WAS.F)

The effects of various factors (time, temperature, pressure, pH, leachant composition, thermal pre-treatment) upon the leach rates of several glasses were investigated. Both radioactive and non radioactive glasses were involved. The glass compositions tested are given in table 1.

Preparation of glasses

Radioactive blocks

Radioactive glasses were made in the VULCAIN cell by a pot vitrification technique which provides glass blocks weighing approximately 1 to 2 kg with an activity ranging from 100 to 1500 Ci beta/gamma or 150 Ci alpha. Studies of pH-value were carried out on active samples (50 g) taken during casting of the blocks.

Non radioactive blocks

Non radioactive glasses were manifactured in a platinum crucible at 1250° C and cast in a graphite mold.

As to the effect of time with leaching at room temperature, under static conditions and in tap water, the beta/gamma active block SON 58 showed no marked effects after 500 days. For the alpha active blocks SON 61 the leach rate decreased drastically from 10^{-6} to 10^{-8} g/cm^{2}d after 160 days of leaching and did not fluctuate with time. The equivalent radiation dose amounted to 6.5×10^{17} disintegrations/g. The total beta yield decreased during the first 300 days and increased between 300 and 400 days. The Cs and Sr releases proved very sensible to the test temperature, even between 20 and 30°C. The Ce and Ru releases decreased during the first 200 days and increased slightly thereafter; in general showing almost no temperature dependence.
Tests with inactive samples are in progress.

For evaluating the temperature effect, leach tests were carried out on the inactive block SON 64 and on the active block SON 60/20 in tap water (renewed every week) and at static conditions in the temperature range of 25° to 180° C. For Cs and Sr the leaching rates increased largely between 25° and 50 ° C, only slightly between 50° and 70° C and not at all between 70° and 150° C. Some increase, however, was observed at 180° C. After returning to ambient temperature the leach rate raised initially remarkedly and fell then back to the value it had at elevated temperature. The leaching rates for Ce and Ru increase by a factor of 5 at 70° C and by a factor of 24 at 180° C. The leaching rate for Si increases scarcely between 70° C and 100° C. The rates for B and Na are larger, those for Al smaller than the total weight loss. The rates for Cs at 100° C were observed to be twice as high for inactive compared to the active species.

Concerning the pressure effect, only tests on the inactive block SON 61 have been carried out so far. The active tests are in preparation. Leaching with tap water (weekly renewal) at static conditions and at room temperature yielded the following results :

- Up to 100 bars no significant change in leach rate was observed, except for Na which decreased.
- At 200 bars a slight increase in weight loss was measured, but no change of leach rate for single elements was found.

The effects of the pH on the leachability was tested on the SON 64 glass block at room temperature and with daily renewal of the leachate.

The active tests were carried out under dynamic conditions, one sample with acid (tap water + HNO_2) one with basic leachants (tap water + NaOH). The inactive tests were performed under static conditions. Even though the test procedures are not directly comparable, the results of active and inactive tests are in good agreement. In general the active constituents are much more susceptible for acid compared to alkilnie pH's. The glass formers show a very divergent behaviour. Si and B encounter virtually no change in leachability between a pH of 3 to 10. In this context it should be kept in mind that dissolved borosilicate glass leads to a significant decrease of the pH down to 11 in stagnant water.

As to the different leachants the tests were carried out with active samples type SON 62 at room temperature and under dynamic conditions (water renewed every day), with the following leachants :

- tap water
- siliceous water
- granitic water
- sea water
- synthetic clay water

The reference leachant was industrial tap water, of which tests have been conducted before and after each test with the other leachants. Analyses of Cs, Sr, Ru, Ce and Sb as well as total beta measurements have been carried out in each case. The leach rates in total beta expressed as multiples of the reference leach rate are for :

- siliceous water (type Mont Dore) = 0.5
- granitic water (type Charrier) = 1.6
- sea water = 2.0

It should, however, be stressed that in the case of sea water, subsequent leaching with tap water did not lead to the starting value, not even after 200 days. This phenomenon will be investigated more thoroughly.

Crystallization effects testing is now completed. The tests were conducted with active blocks of the following compositions :

- SON 45 : low Si, high Na content,
- SON 60/30 : high Si, low Na content,
- SON 65 : low Na, high B and Al content.

The test conditions were : dynamic, tap water (renewed every day), room temperature. The maximum effect on the leach rate at 800° C does not exceed one order of magnitude. The leach rate for Ce decreases with crystallization in all three cases. The rates for Cs and Sr increase for SON 45 and 65 and decrease for SON 60/30. The crystallization effects were little pronounced for Ru.

Table 1 : Compositions of the glasses tested.

GLASS Oxides	SON 453014U2 (A 65)	SON 583020U2 (A 8L)	SON 602018F3 (A 100)	SON 603014U2 (A 79)	SON 611920F3 (A 97)	SON 613014 (A 53)	SON 622024F3 (A 98)	SON 641920F3 (A 96)	SON 641920 (A 87)	SON 652324A20 (A 75)
SiO2	36,91	43,3	45,11	48,27	44,96	50,82	43,41	47,16	43,8	45,24
A/2O3	1,78	1,0	1,69	1,62	1,70	-	1,70	1,69	1,2	4,83
B2O3	12,86	18,8	16,50	13,09	18,43	13,39	22,11	18,42	17,15	21,98
No2O	19,49	9,3	15,04	8,05	14,74	8,25	12,60	12,53	11,4	8,68
CaO	0,03	-	0,01	0,03	0,01	-	0,01	0,03	0,01	0,03
NiO	-	-	0,26	-	0,24	-	0,24	0,24	-	-
Fe2O3	0,66	0,6	5,44	0,73	5,07	-	5,07	5,07	5,86	0,60
Cr2O3	0,13	0,2	0,47	0,05	0,43	-	0,43	0,43	0,49	0,10
P2O5	-	0,6	0,38	-	0,36	-	0,36	0,36	0,4	-
	-	-	-	-	-	-	-	-	-	-
F -	0,06	-	0,04	-	0,04	-	0,04	0,04	-	0,06
SO4	-	-	0,02	-	0,02	-	0,02	0,02	-	-
AmO2	-	-	-	-	-	2,83	-	-	-	-
MgO	0,24	0,1	-	0,15	-	-	-	-	0,06	0,18
Oxides PP+Act.	26,39	26,10	15,03	27,97	14,01	24,68	12,01	14,01	19,58	18,7

1.5.1.2. Corrosion Mechanisms of HAW-Containing GLASS

Contractor : Fraunhofer-Institut Würzburg
(232-81-53 WAS.D)

The investigations were carried out with a model waste glass containing ~15 wt. % of inactive "waste" and SiO_2, B_2O_3, Al_2O_3 and BaO as main components. Temperatures in the range of 80-200° C and a pressure of 130 bar were applied as well as a salt brine composition of the quinary system NaCl-KCl-$MgCl_2$-$MgSO_4$-H_2O.

Several leaching experiments were performed during periods up to 30 days. The glass samples were each exposed to the brine in a cylindric teflon container closed by a teflon foil. Five of these containers were placed in one autoclave, the autoclaves being heated to the specified temperatures.

The main measuring methods are apparent weight loss measurements and ESCA (electron spectroscopy for chemical analysis) measurements supported by a few microprobe measurements.

The first striking result was the occurrence of a rather loose corrosion layer on the glass which can easily be removed. The layer consists of elements from the glass as from the solution. It was not regarded as belonging to the glass sample but as a layer of precipitation products of unsoluble parts of the brine. Therefore the layer was completely removed before further investigation of the glass.

The total weight loss of the samples was then determined. Figure 12 shows the total weight loss per surface area as a function of corrosion time and temperature.

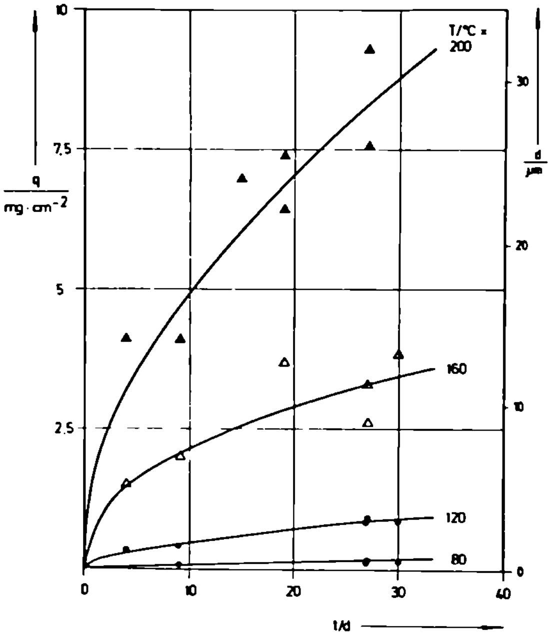

Fig. 12 : Total weight loss (per surface area), q, and dissolution depth, d, of the corroded model glass as a function of corrosion time, t, and temperature, T.

Figure 13 shows the concentration profiles of the elements Si, Ca and Al in a semiquantitative presentation as weight percents.

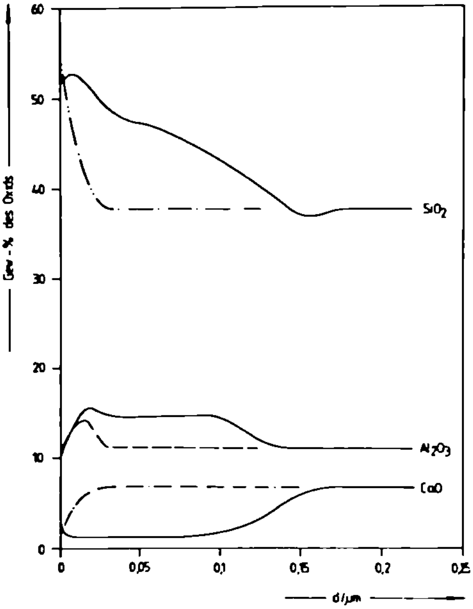

Fig. 13 : Concentration profiles of the elements Si, Al and Ca, in a corroded (————) (30 d, 200° C, 130 bar) and an uncorroded (---,-.-,-..-) sample; both samples were pretreated for 1 h with H_2O at 60° C; semiquantitative presentation in wt. % of oxides; d = o corresponds to the actual sample surface.

The glass has a surface layer enriched in Si and Al, but with poor Ca content. Other measurements show that Ba, Na and Nd have decreasing concentration profiles as well, while Ti behaves similar to Al. The profiles never exceeded 0,2 µm For all temperatures the profile depths were neglegibly small compared to the calculated dissolution depths.

Microprobe measurements confirm that concentration profiles are constant at least beyond 5 µm. The range between 0,2 and 5 µm could not yet be investigated in a satisfactory way.

The results obtained so for lead to the conclusion that the major corrosion process is a glass dissolution. Provided that no unexpected concentration changes occur within the range between 0,2 and 5 um one should expect an overall corrosion process approaching a linear kinetic.

Crystallization properties of tempered glass were looked at by DTA (differential thermal analysis). The maximum of nucleation is found to occur between 600 and 620° C. By X-ray diffractometry celsian, hyalophan, Ba-molybdate and -titanate could be detected.

During the investigation period the waste glass corresponding to the model glass described here was rejected by other institutions for technical reasons. That is why the model glass will not be further investigated though there are still many open questions. A new model glass with a borosilicate frit was accepted as an appropriate successor. In the meantime a series of leaching experiments with assigned leaching times up to 1 year could be started.

1.5.1.3. Migration Study on Actinides and Fission Products in HAW-glass Matrices

Contractor : CEA Saclay (268-81-55 WAS.F)

During 1981, measurements concerning profiles of the interdiffusion of hydrogen and sodium in the surface regions of leached glasses were carried out using nuclear analyses methods specially developed for the purpose.

The work involved investigations of the dynamic behaviour of the erosion of two AVH (Atelier de Vitrification La Hague) glasses in water as a function of certain parameters (pH, temperature, time, etc.).

The hydration of the glass, essentially an interdiffusion process of Na coming from the glass and H or H_3O coming from the water, is being studied by activating the Na in the glass through an incident ion-beam of some 1000 KeV intensity. The dealkanised layer thickness was measured as a function of the leaching temperature. An accumulation of Uranium and lanthanides was observed in the dealkanised surface layer.

The concentration profiles of various elements (so for U, La, Th and Ce) are determined, before and after leaching for various time intervals and at different temperatures, in the micro-region of the sample surface. This is performed by measuring the back-scattering ions produced by a Van de Graaf accellerator. For the back-scattering spectra a computing programme has been developed. Migration studies for actinides(in particular Pu and Np) with alpha-spiked samples and for longlived fission products (Ce, Cs, Ru, Sr) as a function of aqueous leaching conditions will be carried out next.

In order to accomplish the programme, highly accurate nuclear analysis methods will be developed, to enable the examination of superficial layers in a non-destructive manner.

1.5.2. TESTING OF VITRIFIED WASTE FORMS

1.5.2.1. Evaluation of Properties of Materials for Immobilisation

Contractor : AERE Harwell (121-80-53 WAS.UK)

The programme comprises work on various European waste solidification compositions, as follows :

- Theoretical studies of the leaching mechanism backed up by experiments
- Specific isotope leach tests
- Leach tests carried out in contact with granite at low water flow rates to simulate the conditions that will occur in a granite repository
- Studies of the α-radiation stability of the materials
- Studies of the mechanical stability of the materials.

Leaching experiments on glass UK189 at 20° C and 30° C in leachants of various pH have been continued. The leachates were analysed for Si, B, Na, Li and Mg. It is well known that in simple binary glasses the network modifiers (e.g. Na, Li) are leached by ionic exchange with protons (or H_3O+ ions) in the leachant and this is governed by a diffusion mechanism. Network formers are leached at a constant rate, as the network breaks down.

Some results for UK189 are shown in Fig. 14, where the leached quantities $Q(g.m^{-2})$ of the major constituents of the glass, normalised by dividing their concentrations in the glass $C(g.m^{-3})$, have been plotted against time. For silica the relationship is a linear one : ${}^{Q}Si/{}^{C}Si = 2.3 \times 10^{-7}t$ where ${}^{Q}Si/{}^{C}Si$ is in μm and t is in seconds. For the other four elements, Li, Na, B and Mg, the equation is :

$$Qm/Cm = 2.3. \times 10^{-7}t + 2.6 \times 10^{-4}t^{1/2}$$

Note that the linear part of this latter equation is the same as for Si, presumably dependent on the rate at which the glass matrix is destroyed. The fact that the other four elements leach at the same rate suggests that the same factor is controlling their release, perhaps the diffusion of H_3O+ into the glass. The four ions have different sizes and valencies and would not all be expected to diffuse at the same rate. Obviously, the coefficients are different at different temperatures and different pH.

The leach rates of samples have been measured spiked with

Sr-90, Tc-99, Cs-137, Np-237, Pu and Am-241, using two techniques (a) with a very slowly flowing leachant and (b) in a repository simulation where the leachant is static apart from that used for sampling.

The results for samples of glass UK189 leached at 60° C by the first of these techniques are shown in the Table below. The leach rates obtained span over 4 orders of magnitude with Tc leaching fastest and Am slowest.

Isotope	Leach rates ($g.cm^{-2}day^{-1}$) at 60°C			Rate limited by
	10mℓ/day	1 mℓ/day	1mℓ/week	
Sr-90	5×10^{-5}	1.8×10^{-5}	2×10^{-6}	Solubility
Tc-99	7×10^{-5}	7×10^{-5}	6×10^{-5}	Leaching
Cs-137	6×10^{-5}	4×10^{-5}	1.6×10^{-6}	Solubility
Np-237	2×10^{-5}	9×10^{-6}	6×10^{-6}	Leaching
Pu	(5×10^{-7})	1.4×10^{-7}	2.5×10^{-8}	Solubility
Am-241	4×10^{-9}	(5×10^{-10})	(1.4×10^{-9})	Leaching

The leach rate of Tc-99 is unaffected by flow-rate which suggests that the rate of attack on the glass is the limiting factor. This is probably also true for Np-237 and Am-241. The leach rates for Sr-90, Cs-137 and Pu however decreased by factors of 25, 38 and 20 when the flow rate was reduced by a factor of 50 which suggests that solubility of the resulting species in the water may be a factor.

Doped samples of UK209 have been tested at a flow rate of 1mℓ/week. These results show a much smaller spread than for glass UK189. Indeed, for Pu and Am-241 the leach rates from UK209 are higher than from UK189; for Cs-137 they are about the same and for Tc, Sr and Np they are higher for UK189.

In the repository simulation experiment, a sample with a a surface area of $3cm^2$ is held in a water-filled specimen chamber above a column of crushed granite. A 1mℓ sample of leachate is withdrawn from the specimen chamber at intervals and is replaced by water that has flowed up the column. This intermittent sampling provides the only flow in the system which is thus very slow, about 1 mℓ per month at present. Initial results do not show a consistent pattern compared to those from the low flow experiment described above; the leach rate determined by counting Tc-99 is lower in the repository simulation experiment whilst those obtained by counting Pu and Am are higher.

The six glass compositions doped with Pu-238 to simulate the irradiation that will occur in real solidified waste due to incorporation of actinides are still being monitored. The doses have now reached about 2×10^{18} α-disintegrations per gram, equivalent to about 200,000 years for real waste for the UK glasses, 25,000 years for the German compositions and 800 years for the French glasses, the differences being due to the different waste concentrations in the glasses and the different reactor systems considered (Magnox for the UK glasses and PWR for the others). The samples are still quite unchanged in appearance but the densities are following the exponential equation :

$$\Delta_{\rho} = \Delta_{\rho sat} \{1-\exp(-\alpha D)\}$$

where ρsat is the density change after infinite time, and D is the dose in disintegrations per gram. The predicted values of the constants are :

Glass	$\Delta_\rho$sat (%)	α(x 10^{17}g.disint.$^{-1}$)
UK 189	- 0.44 ± .01	0.122 ± .008
UK 209	- 0.88 ± .05	0.043 ± .003
F SON 58.30.20.U2	+ 0.62 ± .06	0.061 ± .009
G VG 98/3	- 0.81 ± .02	0.102 ± .006
G Ceramic B1/3	- 0.49 ± .01	0.115 ± .007
Phosphate beads	+ 0.41 ± .03	0.095 ± .015

The largest predicted density change is equivalent to a linear expansion of about 0.3 % for UK209. The French glass and the Pamela phosphate glass contract on irradiation whilst the others expand, but the reason for this difference is not known.

1.5.2.2. Characterization and Comparison of HAW-Products

Contractor : HMI Berlin (122-80-53 WAS.D)

A second joint programme of the European Communities on characterization of various potential waste forms was started in 1981. The aim of the research work at the Hahn-Meitner-Institut is to produce data for calculating the activity release from waste products under the attack of different solutions and conditions expected for final repository. The programme comprises investigation on simulated inactive, as well as, highly radioactive samples :

- Studies of hydrothermal leaching and surface attack of waste glasses and glass ceramics
- Studies of thermal effects and their impacts upon leachability of simulated inactive waste products
- Leach tests of highly radioactive samples
- Leach tests of actinide spiked samples
- Studies of the mechanical properties of waste products.

The hydrothermal leaching of simulated high-level waste boro-

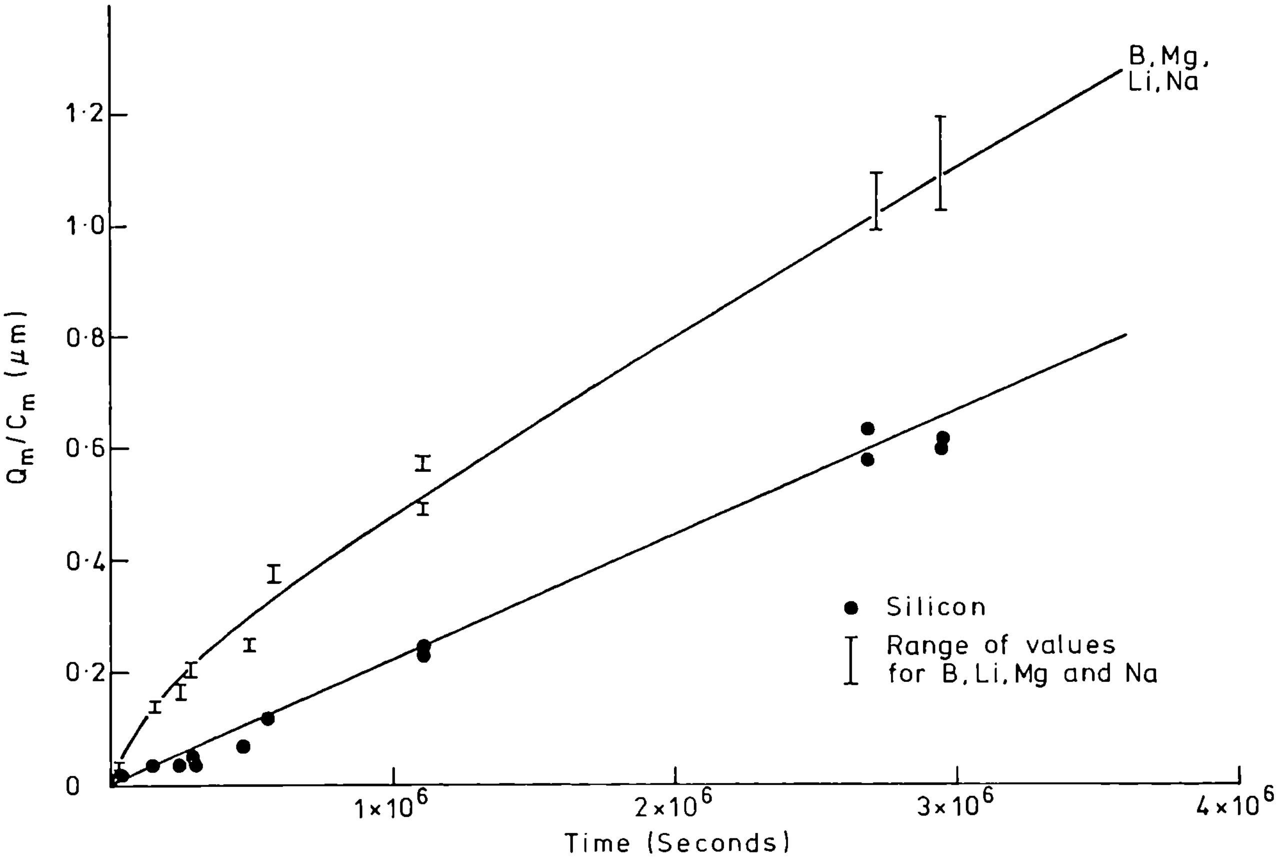

Fig. 14 : LEACHING OF UK 189 AT 20°C IN DISTILLED WATER AT pH7

silicate glass was investigated in water, rock salt and $KCl-MgCl_2-CaCl_2-MgSO_4$ solutions at 200° C.

The samples were glass beads having a diameter of some 0,5 cm and a surface area of about 1 cm^2. The leaching time was 3, 10 and 30 days. The specific weight loss (g/cm^2) was measured as a function of time, the leachates were analysed by inductive ccupled plasma emission analysis and the leached glass surface by microprobe analysis. Leachability droped below detection limits after a few days. Surface layers formed on the glass beads during leaching, they yielded after 30 d about 20 - 30 µm. The layers appeared inhomogeneous under the scanning electron microscopy.
At least two successive layers could be seen and microprobe analysis showed considerable differences in their chemical composition. The drop in the leach rate suggests, that a surface layer protects the underlying glass against further attack.

The studies of thermal effects have started with the borosilicate glass, which was primarily developed to transform it into a celsian-type glass ceramic. The X-ray diffraction patterns of the glass ceramic changed after heating, indicating a thermal instability of the material.

Actinide leaching experiments planned in addition to the inactive investigations, use spiked glasses and glass ceramics. The emphasis is placed on the evaluation of the leaching characteristics of actinides and on its comparison with other glass constituents. Am-241 and Pu-239 doped samples have been prepared using a mixture of powdered glass and nitric acid solution. The sludge was denitrated by heating in a graphite crucible. After melting, the glass was stress-relieved and slowly cooled to room temperature.

Whereas autoradiographs taken from polished surfaces showed no inhomogeneities of the doped material, microprobe analysis yielded some enrichment in the neighbourhood of precipitations.

Preliminary leaching experiments were performed using different times and leachate volumes. Development of the typical surface layer was observed and detected by the energy spectrum of the α-particles emitted from the near surface region.

1.5.2.3. Physico-Chemical Characterization of Conditioned Radioactive Waste Products

Contractor : SCK-CEN Mol (124-80-55 WAS.B)

The work relates to two main topics, i.e. the physico-chemical characterization of two glass compositions designed for the conditioning of Eurochemic waste concentrates and the study of the corrosion behaviour of six representative waste forms (four from the previous CEC programme and the two "Eurochemic" glasses) under conditions relevant to clay disposal.

Till now only a restricted number of preliminary experiments was carried out using simulated waste forms prepared at Mol. After the necessary glass samples have become available, the first results from the corrosion experiments using these materials are being obtained.

The waste forms to be investigated in this programme are the SON 58.30.20 U2, SON 64.19.20 F3 and UK 209 glasses and the C 31.3EC glass-ceramic from the previous CEC indirect action programme and two "Belgian" glasses, relating to the two vitrification units for the Eurochemic site. The compositions of the four materials from the previous CEC programme have been described earlier (ref. 1); the PAMELA composition which is to be used for the LEWC (low enriched waste concentrate) of Eurochemic has been developed in Germany and is given in ref. 2, whereas the AVB composition which is to be used for the HEWC (high enriched waste concentrate) is developed by CEA and is given in ref. 3.

The characterization of the AVB and PAMELA glasses consists in the investigation of the thermal stability (devitrification) and of the chemical stability (by means of Soxhlet tests and long term static leach tests in distilled water).

The Corrosion studies on the 6 materials under conditions relevant for disposal in clay media, use static leach tests at various temperature, pressure and sampling time and various corrosion media (deminaralized water, water at controlled pH, clay equilibrated water and synthetic interstitial clay water).

Part of the results so far obtained have already been presented at the KFA ,Jülich ,International Seminar in 1981 (ref. 4).

The corrosion experiments were carried out on polished, thin glass plates (surface area ~5 cm^2, thickness ~1 mm). These glass plates were fixed in teflon cells, with a surface area to leachant volume ratio of ~2 cm^{-1}.

Corrosion rates (based on the weight losses, after removal of the surface layer) of the different glasses after one day of corrosion as a function of temperature, both in distilled water and in clay water were determined. The experiments at 250° C were done by simply heating the teflon corrosion cells, the pressure of the fluid thereby reaching 50 bars. At 150° C, two pressures were applied; 7 bars were obtained by simply heating up to 150° C, 50 bars were reached by additional argon pressure. The few results obtained far do not allow conclusions about a possible correlation between corrosion rate and pressure, or between corrosion rate and temperature.

In Fig. 15a, the cumulative weight loss per unit surface area of glass BWG 1 is plotted as a function of time. The corrosion test was conducted in distilled water at 95° C, and each point in the figure corresponds to a distinct sample. One may conclude that after one day of testing the corrosion rate has nearly become zero. Furthermore, the pH value has increased up to 9.1 after one day and remained between 8.8 and 9.5 between the first day and the sixth month. In Fig. 15b the infrared reflection spectra of some of the samples are shown. One can observe a net decrease of the 900 - 700 cm^{-1} (related to non bridging oxygen-alkali or alkaline earth ion bonds) and of the ~ 1000 cm^{-1} (related to the silicon-oxygen bond) reflections. No definitive conclusions can be drawn yet from these observations. Nevertheless, it seems to be clear that saturation effects are occurring, caused by the fairly high surface area to solution volume ratio. Using a lower surface area to solution volume ratio (e.g. 0.1 cm^{-1} instead of 2 cm^{-1}) will presumably lead to much slower change from diffusion over dissolution to the saturation state.

Corrosion tests for the glasses BWG 2 and 4 were conducted at 95° C in distilled water, clay-water (pH = 8.05) and interstitial claywater (pH = 7.80).

In contrast to BWG 1 and 4, the corrosion rate of BWG 2 in distilled water has not reached zero, although the corrosion rate decreases as a function of time.

The glasses BWG 2 and 4 behave differently in distilled water as compared to the clay media, and also between each other. It is remarkable that in contact with interstitial clay water the weight of BWG 2 is still decreasing after six months, while the weight of BWG 4 is still increasing after this period.

References

(1) J.A.C. Marples et al. : "Testing and evaluation of the properties of various potential materials for immobilising high activity waste" - EUR 7138 EN (1981).

(2) R. De Batist et al. : "Physico-chemical characterization of conditioned radioactive waste products" - research contract n° 124-80-55 WAS.B - 1° semestrial report January-

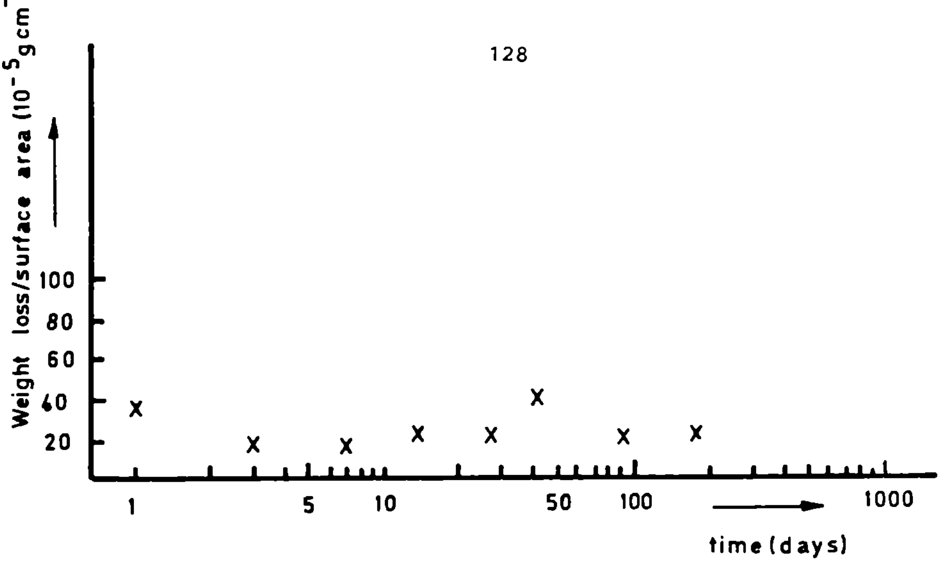

Fig. 15a, Weight loss of glass BWG 1

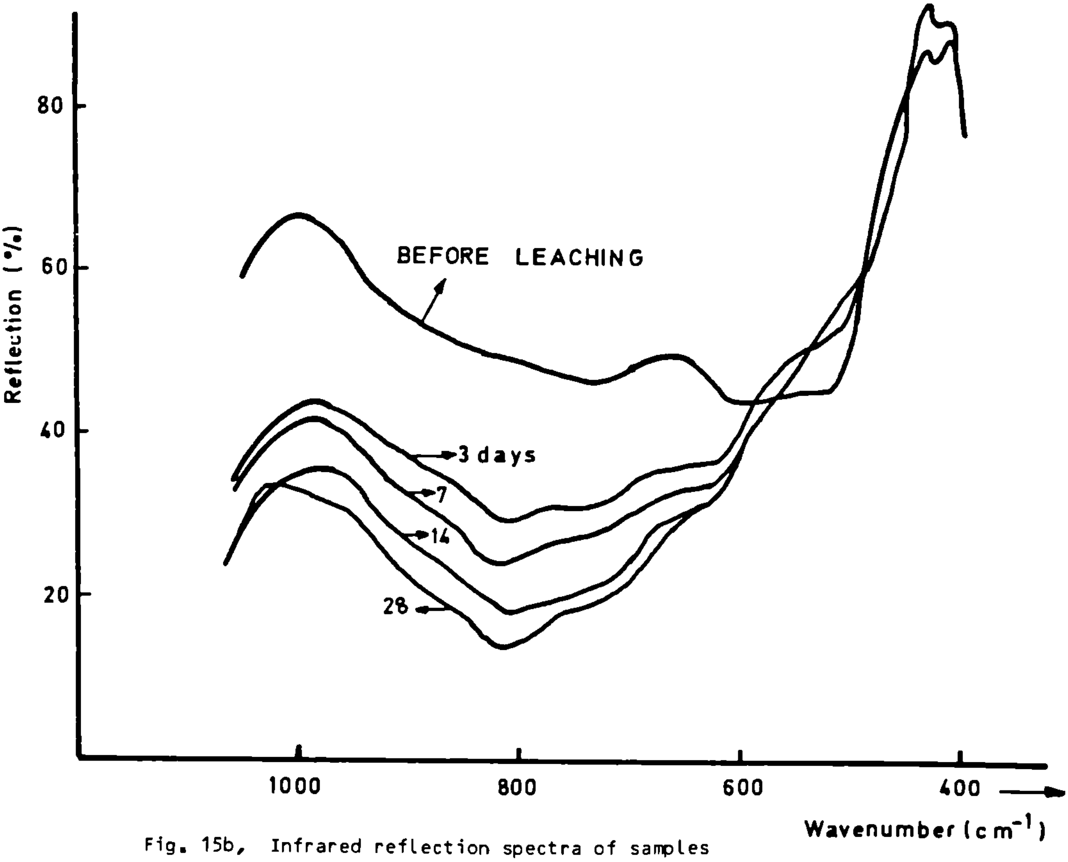

Fig. 15b, Infrared reflection spectra of samples

June 1981 - S.C.K./C.E.N. Mol, Belgium.

(3) R. De Batist et al. : "Physico-chemical characterization of conditioned radioactive waste products" - research contract n° 124-80-55 WAS.B - 2nd semestrial report July - December 1981 - S.C.K./C.E.N. Mol, Belgium.

(4) P. Van Iseghem et al. : "Interaction of vitrified high level waste with clay environment", International Seminar on Chemistry and Process Engineering for high-level liquid waste solidification, KFA Jülich, June 1981, in press.

1.5.2.4. Testing and Evaluation of Ceramic Materials for HAW-Immobilisation

Contractor : University of Leiden (125-80-55 WAS.N)

The concept of immobilization of radioactive nuclear waste into a compound of mineral phases was suggested by Hatsch(1). According to recent literature there are ceramics that are stable in water or NaCl solution at elevated temperature (300° C under pressure) (2). The aim of the research reported here is to prepare model compounds and study their thermal stability, phase relations, crystallographic properties and behaviour in water and 10 % NaCl solution in the temperature range 300-400° C and under pressure. The model compounds are the hollandite and perovskite phases containing Cs, Sr and Ru.

Starting materials for the preparation of the hollandites are $CsNO_3$ 99.9 %, Al_2O_3 99.99 %, TiO_2 (anatase) 99.8 % and earth-alkaline carbonates 99.8 %. For compounds containing Cs an appropriate mixture was heated for one hour at 750° C in an open crucible in order to decompose the nitrate and carbonate. Weight loss was checked and X-ray powder patterns were recorded to determine whether all nitrate and carbonate

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(1) L.P. Hatsch, Am.Scientist 41, 410 (1953).

(2) A.E. Ringwood, Safe Disposal of High-Level Nuclear Reactor Wastes : A New Strategy, A.N.U. Press, Canberra, Australia (1978).

was decomposed without any loss of Cs. Hereafter the mixture was sealed in Pt capsules to prevent Cs loss and placed in a furnace. To determine solubility and leaching about 0.3 gr of a compound is sealed in a Pt tube together with approximately 0.4 ml of water. The tube is placed in a hydrothermal apparatus and heated at 300° C while the pressure is maintained at 2 kbar. After a given time the tube is opened, the liquid is tested for Cs or Sr and an X-ray powder diffraction pattern of the dried solid phase is recorded.

Cs-hollandite, $Cs_xAl_xTi_{4-x}0$ (x=0.5-0.8)

At temperatures above 1150° C the compound decomposes :

$Cs_xAl_xTi_{4-x}0_8 \rightarrow xCsTiAl0_4+(4-2x)Ti0_2$

$CsTiAl0_4$

The crystal structure of this new compound was determined with neutron powder analysis a=9.9572(10), b=5.7300(6), c=8.9655(9) Å, space group probably $Pna2_1$. In water of 300° C (2kb) the compound disintegrated completely to $Ti0_2$ (rutile), Al0OH (boehmite) and (some) additional phase(s).

Ba(Cs) hollandite phase, $Ba_{x-y}Cs_{2y}Ti_{4-2x}Al_{2x}0_8$ (x+y=0.66)

A number of compositions were prepared with y=0.0083; 0.0167; 0.025; 0.033; 0.111; 0.167; 0.222; 0.333, all adopting hollandite-like structures. However, no pure phase could be prepared. From the lattice constants it is concluded that Cs is incorporated in the tunnels of the hollandite structure. Hydrothermal treatment loses the additional phase(s) present ($CsTiAl0_4$) but the hollandite phase retains.

Strontium hollandites

Sr hollandite, $Sr_xTi_{4-2x}Al_{2x}0_8$ could not be prepared at 1300° C, therefore a series of compounds $Ba_{1-x}Sr_xTi_{2.8}Al_{1.2}0_8$ with x=0; 0.1; 0.2......1.0 was investigated. A hollandite phase was formed for $x \leqslant 0.25$. For $x > 0.25$ the X-ray pattern show (Ba,Sr)-hollandite, $SrTi0_3$, $Ti0_2$ and another unknown phase. In order to find the composition of this unknown phase both the systems $BaO-TiO_2-Al_2O_3$ and $SrO-TiO_2-Al_2O_3$ were investigated.

System $BaO-TiO_2-Al_2O_3$ (1200° C)

This system has recently been reported by Guha (3). Three ternary phases were found $\simeq Ba_{0.6}Al_{1.2}Ti_{2.8}O_8$, $BaTiAl_6O_{12}$ and $Ba_3TiAl_{10}O_{20}$. Of these compounds crystal systems and the lattice constants were determined. For all three compounds both the crystal systems and lattice constants differ from the results reported by Guha. The structure of $Ba_3TiAl_{10}O_{20}$ was determined (4). Ba hollandite is stable in H_2O at 300° C (2kb) but the other two compounds decompose completely under formation of AlOOH (boehmite).

System $SrO-TiO_2-Al_3O$ (1300° C)

The system is under research now. A ternary compound was found of about the composition $SrTi_3Al_7O_{17.5}$. The same phase is found in the mixture of compounds resulting from an effort to prepare $Ba_{1-x}Sr_xTi_{2.8}Al_{1.2}O_8$ $(x>0.25)$.
In the determining of the phase diagrams the experience of recent determination of the system $BaO-Fe_2O_3-SnO_2$ (1200°C) (5) was used.

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(3) J.P. Guha, J.Sol.State Chem. 34, 17-22 (1980).

(4) M.C. Cadée, D.J.W. IJdo, G.Blasse, J.Sol.State Chem. 41, 39-43 (1982).

(5) M.C. Cadée, D.J.W. IJdo, J.Sol.State Chem. 36, 314-323 (1981)
M.C. Cadée, D.J.W. IJdo, J.Sol.State Chem. 40, 290-300 (1982).

1.6. IMMOBILISATION AND STORAGE OF GASEOUS WASTE

The radioactive gaseous waste considered under this chapter mainly arise from the reprocessing of nuclear fuel and in particular from the dissolution of the fuel at the head end of the process.

The research activities concern the known volatile radionuclides Krypton-85, Iodine-129, Tritium and Carbon-14 as well as the management of aerosols.

All of the nine different research projects comprised in this chapter have continued or started with work in 1981. A substantial part of the supported research concern the immobilisation of Kr-85. Four projects are dedicated to this problem. They deal with the krypton inclosure in metals or metallic matrix and in the porous structure of zeolites. They are at different stages of process development but none of them has reached yet industrial maturity nor has been tested under active operation.

The retention of tritium from aqueous effluents is studied by a laboratory experiment on technical scale. The tritium is concentrated in the waste liquid by a combined isotope separation process using electrolysis and catalytic gas liquid exchange. Process development has advanced now to the state of hot (active) testing.

As to Iodine-129 and Carbon-14, two desk studies are running in order to improve or to draw up respectively, the assessment of possible management methods. In September 1981, an E.C. specialists' seminar was organised by the Commission on the question of Iodine-129. It gave a review on the state of the art, based on a collaborative study made by UKAEA, NRPB and CEA in the scope of the previous Community programme (cf. proceedings EUR 7953 (1982)).

Finally two experimental works are under way to study the performance of regenerative prefilters for aerosol retention. The objective is to spare the high efficiency particulate (HEPA) filters in off-gas systems in order to reduce the waste volumes to be stored or disposed of for renewing the filter elements.

1.6.1. IMMOBILISATION OF KRYPTON-85

1.6.1.1. Incorporation into a Metal Matrix by Ion Sputtering

Contractor: AERE HARWELL (157-80-8 WAS.nk)

In the previous European Community's R&D programme a half scale pilot plant was designed, built, and operated with inactive gas to demonstrate the process on a scale representative of an industrial plant. A copper deposit 14mm thick, containing over 200 litres of krypton was built up in a half scale cylindric vessel.

The main aim of the current programme is to assess potential candidate metals for the long-term storage of krypton arising from the re-processing of nuclear fuel.

Deposition of copper was continued in the current programme until the deposit reached a thichness of 22 mm. The deposit weighed over 23 kg and contained over 300 litres of krypton.

The assessment of the thick copper layer is being carried out, in order to determine the suitability of the krypton filled matrix for the long term storage of gas. Long term tests of the thermal stability of the gas are being made by sealing samples in silica ampoules and holding them at temperatures between 150° C and 500°C for periods up to 1 year. Gas release measurements of samples annealed for up to 4 months have just started. Only 5 ppm of the gas was released over this period at 300°C. Samples have also been annealed for periods up to 400 hours at 400°C and the release monitored continuously. Extrapolation of these measurements indicates that, at 400°C, samples may eventually lose up to 2% of their gas content. The subsequent release rate at

150°C is still very low.

A systematic study has been made of the effects of annealing for 1 hour at temperatures up to 800°C on the microstructure, hardness, and lattice parameter. Transmission electron microscopy of the deposit before annealing showed gas bubbles of diameter 10-20 Å at a density estimated to be 5.10^{18} to 1.10^{19} per cm^3. The bubbles in the grain boundaries were larger and there was evidence of large strains. In material deposited at 150°C, the bubbles and grain size were larger. Samples were annealed either in the electron microscope or as bulk samples. Gross swelling of the samples was observed in those annealed at or above 625°C and the surface showed pin-holes or sponge-like defects, caused by the intersection of grain boundary porosity with the surface. The intragranular bubble size increased with annealing temperatures above 400°C, reaching 200 Å at 700°C. However, the effect on the bubbles in the grain boundaries was more marked. Between 550°C and 600°C in-situ annealing experiments revealed the coalescence of bubbles, and in thick regions large pipes of interlinked bubbles of gas developed and intersected the surface. Samples were also bombarded with 600 Kev electrons in the High Voltage Electron Microscope. No dislocation movement or bubble growth was observed.

The lattice parameter has been measured by X-ray diffraction. The deposited material has a lattice parameter 0.4% greater than that of normal copper, and the diffraction peaks are extensively broadened. The lattice parameter decreased with annealing temperature until 500°C, when it acquired the value of pure copper.

Some physical measurements of the deposit have been made. The thermal conductivity is 80 $Wm^{-1}K^{-1}$, similar to many copper-based alloys. The metal is hard (340 VHN) and brittle as deposited. The specific heat and melting point are similar to those of copper.

The stability of the gas in the presence of beta and gamma irradiation has been investigated both by irradiation with 600 KeV electrons in the high voltage electron microscope and by sealing samples in ampoules and immersing them in the spent fuel pond for times to give gamma doses from 10 to 1000 M rads. In the high voltage electron microscope, no changes in structure were seen for an irradiation dose equivalent to that of storage for 100 years. The amount of gas released into the sealed ampoules was less than 0.2 ppm of the total content for a gamma dose of 100 M rads, which corresponds to storage for 100 years.

Leach tests at 100°C in distilled water, (Soxhlet leach test) have been carried out for 1 year. The measured leach rate of 4 micrograms/sq mm/year represents a negligible release of gas should it occur during storage. The corrosion rate at 90°C in strong brine has been measured for normal metallic copper and deposited material in argon and oxygen. The sputtered material was a factor of 5 more resistant to corrosion in the presence of oxygen than normal copper.

Small angle neutron scattering and positron annihilation studies have also been carried out and indicate that the majority of the gas is in small bubbles or other defect clusters at a spacing of about 50 Å which agrees with the electron microscope observations. However, calculations of the gas content of the bubbles seen in the electron microscope indicate that if the bubbles are at equilibrium, they can contain only a small fraction of the total amount of gas. The remainder of the gas may be in bubbles too small to be resolved in the microscope, or in other defects, or the bubbles may be severely over-pressured.

Apart from copper, Incaloy 825 and stainless steel AISI 316L were chosen for testing.

An improved design of process vessel has been used for the preparation of these samples. The vessel is slightly smaller than that used for the copper deposits and uses many more standard vacuum components and standard stock size source material, so that the cost is reduced significantly. The gas incorporation efficiency for Incaloy was found to be about $0.8.10^{-3}$ l/kWh, compared with at least 1.10^{-2} for copper. An efficiency of 1.10^{-} has been achieved with stainless steel. However, the adhesion of these deposits to the stainless steel vessel is not very satisfactory when the thickness of the deposit is greater than 1 to 5 mm and the deposit tends to peel off. Improved preparative cleaning methods are being tried, in order to improve the integrity of the interface between the substrate and deposit. A copper interface layer is also being tested.

Preliminary studies for the design of a small, transportable, active test facility to demonstrate the process with active gas have shown that active operation can be simulated by using only 100 Curies of radioactive krypton to produce the saturation beta levels inside the vessel, together with an externally applied gamma irradiation source of 2000 R/hour to simulate the gamma levels which build up continuously as the thickness of the deposit containing active gas increases. This combination will reproduce the maximum levels of both beta and gamma fluxes, which will be attained within the process vessel without the need for large quantitites of radioactive krypton.

1.6.1.2. The long term Storage of Kr-85 by Fixation in Zeolite 5A

Contractor: KfK Karlsruhe (158-80-8-WAS.D)

The fixation process is based on the experimental observation that Kr can be trapped efficiently and securely in zeolite 5A in the temperature range of 500 - 650°C and at pressures between 100 and 2000 bar. The fixation involves a transformation of the original crystal framework into an amorphous glassy state. Several experimental observations demonstrate the change in phase:
i) the characteristic x-ray spectra of unloaded zeolite 5A disappears after noble gas fixation,
ii) the specific surface area decreases from about 450 m^2/g to less than 3 m^2/g upon gas trappping
iii) the transmission electron microscope pictures of loaded samples (enlargement 440 000) show no α-cages,
iv) the typical neutron scattering spectra of zeolite 5A disappears after loading with a noble gas
v) the electron scattering spectra indicate that a vitreous phase is formed at the fixation temperature, and a derivative of $CaAl_2Si_2O_8$ at about 100°C above that temperature,
vi) the scanning electron micrographs demonstrate that upon sin-

tering in the presence of high pressure noble gas the 5A crystals melt together.

All experimental evidence obtained so far show that zeolite 5A in the vitreous state is an adequate matrix for the long-term storage of radioactive krypton. The gas-zeolite system is resistant to high temperatures (no significant leakage was observed after 18 month at 450°C), to liquid Rb at 150°C, to several salt solutions as well as to γ-radiation exposure (0.3 - 3.10^8rad).

At high specific Kr-85 loading of zeolite increased temperatures are expected, caused by the decay heat generation. Because the Kr release rate out of vitreous zeolite is a temperature controlled process. it was important for a reliability assessment, to know the temperature profile in the final storage vessel as a function of the design of the containment and the krypton loading of the zeolite. Therefore the thermal conductivity of zeolite 5A in several aggregation forms and in various fluid media (krypton, argon, helium, air, etc.) were systematically measured as a function of temperature. It was observed that activated zeolites have a lower conductivity than samples loaded with a noble gas or not previously activated. Compaction of binderless 2 mm spheres or drying of a zeolite powder/water slurry, brought about a substantial increase in volumetric loading of the autoclav as well as a significant improvement of the thermal conductivity. Conditions in a final storage vessel were simulated with stainless steel cylinders provided with several heating coils and numerous radially as well as longitudinally arranged thermo-couples. The results, obtained with unloaded 2 mm 5A zeolite spheres, indicate that natural convection will remove the heat generated by radioactive decay of Kr-85, and will maintain the maximum center-line temperature in a 134 mm ⌀ final storage vessel at temperatures below 80°C, when the specific loading of the zeolite spheres is kept below 40 cm^3STP Kr/g. A good correlation between the calculated and the experimentally obtained temperature profiles was found.

Much work was devoted to the design of a demonstration plant. Process and equipment sizes were selected on a viable size basis

for the required throughput. The facility includes a lag-storage of feed gas and two separate areas for gas fixation by remote operation. The major supporting equipment involves compressors, vacuum pumps, (eventually cooling systems) and a welding unit. The immobilization of radio-krypton is carried out at 300 bar and 520°C in a one-way autoclav with a capacity of 10 liters. The substrate container consists of a thick walled cylinder provided with a quickconnector, filled with compacted zeolite 5A. Gas compression is thought to take place by one of three alternatives:
i) freezing and expanding of Kr,
ii) sorption/desorption of Kr in zeolite 5A at above atmospheric pressures or
iii) sorption/desorption of Kr in zeolite 5A at low temperatures and atmospheric pressure. For the chemifixation the one-way autoclav needs to be heated for at least one hour.

The one-way autoclav concept considerably simplifies the fixation procedure and provides a final storage product which complies with high safety standards.

1.6.1.3. Screening of Materials for Embrittlement by Rubidium

Contractor: AERE Harwell (227-81-8 WAS.UK)

A one year programme has been conducted to assess the likelihood of Kr 85 container flask materials based on iron and nickel being degraded by Rb (decay product of Kr-85) induced liquid metal embrittlement (LME). The work was motivated by the probable future requirement to store gaseous Kr 85, liberated during fuel reprocessing, for long periods before its activity has fallen to levels consistent with release to the biosphere. One method being considered is containment in high pressure gas flasks. No systematic study of possible degradation by Rb induced LME has been undertaken prior to this work although a few American experiments have been reported.

LME differs from the other degradation processes evaluated previously in that it does not depend on any chemical interaction. It is caused by intimate contact, wetting, between a molten embrittler and a stressed solid so decreasing the interatomic bonding of the solid, that micro cracks propagate by cleavage rather than being blunted by slip. Thus normally ductile materials can fail in a brittle manner and at a somewhat decreased stress when deformed in the presence of low melting point metals. Pinchback, in the USA, reported that Monel 400 suffered LME when deformed in Rb at 127°C.

The susceptibility of eleven potential constructional materials to Rb induced LME have been assessed. Six of the materials were suggested by Harwell and five by other laboratories. The materials fell into three categories.

Ferritic, BCC, steels

BS970/070M20, contains 0.32 Mn, 0.03 C
BS970/817M40-T, contains 1.25 Cr, 1.0 Ni, 0.4 C
DIN 1.4922, contains 12.0 Cr, 1.0 Mo, 0.20 C

Austenitic, FCC, steels

BS304, contains 18.1 Cr, 9.0 Ni, 0.03 C
BS316, contains 17 Cr, 13.5 Ni, 2.32 Mo
BS321, contains 17.1 Cr, 9.0 Ni, 0.26 Ti
BS347, contains 17.3 Cr, 9.5 Ni, 0.76 Nb
DIN 1.4439, contains 17.5 Cr, 13.5 Ni, 4.5 Mo, 0.15 Ni

Nickel, FCC, alloys

Monel 400, contains 30.0 Cu, 2.5 Fe
Nimonic 80A, contains 20.0 Cr, 2.0 Ti, 1.5 Al
Nimonic 105, contains 15.0 Cr, 20.0 Co, 5.0 Mo, 5.0Al, 1.2 Ti

Rods of these alloys were machined into tensile test pieces and tested to destruction in Ar and Rb environments using specially designed capsules. The Nimonic alloys were heat treated before being tested but the others were used in the as-received condition, generally "bright finished", as they would be in flask construction. The Ar and Rb impurity levels were reasonably low, less than 15 ppm and 0.02% respectively. The tests were conducted at a high extension rate, 50 $mm.min^{-1}$, to maximise the likelihood of producing LME and at temperatures of 50°C, close to the melting point of Rb, and 150°C.

It was found that Rb readily wetted all the materials at 50°C and above, although there was some indication that the wetting of the ferritic steels was marginally inferior to that of the austenitic steels and nickel alloys. If the Rb became contaminated by prolonged exposure to a glove box Ar atmosphere, wetting characteristics were degraded. However care was taken to achieve good wetting and hence alloy-Rb contact when immersing the test samples by filling the capsules with fresh molten metal.

The use of the Rb rather than Ar environments had no significant measurable effects on the mechanical properties of the ferritic steels tested at either 50 or 150°C. However, the ductilities of the FCC austenitic steels and nickel alloys, as defined by the elongations at which the tensile test samples began to neck were increased during tests at both temperatures, but particularly those at 50°C. The elongations at which austenitic steels fractured were also increased by Rb environments, very substantially in some cases. Thus elongations at which necking and fracture occurred with BS304 samples were increased from 50 to 64% and from 58 to 74% by using Rb rather than Ar environments at 50°C. The change had no discernible effects on strength parameters except for the ultimate tensile strengths of austenitic steels tested at 50°C which increased by 2 to 10% to yield a ranking BS304 > BS321 > DIN 1.4439 > BS316.

Optical and scanning microscopy of the test samples showed that failure produced a typical ductile cup-and-cone topography. Optical microscopy did not reveal any evidence or corrosive attack produced by the 0.5 to 1.0 hour exposures to Rb, nor were any other environment related microstructural effects identified by examination of the ferritic steel and nickel alloy samples. However, there was partial transformation in the gauge lengths of the austenitic steel test pieces to a distortred lathe like morphology characteristic of BCC matensite. The extent of this transformation appeared to be most extensive in samples that had been tested at 50°C, and particularly in those tested in Rb. X-ray diffraction analyses of cross-sections confirmed that martensite had been formed in at least some samples. The peak intensities produced by low index FCC and BCC planes suggested that the extent of transformation of BS304 and BS321 samples tested at 50°C were about 10-15 and 30-70% when Ar and Rb environments were used. The CuKα X-rays used penetrated only about 6 μm and analyses of the unsectioned BS304 samples that had been tested at 50°C suggested that the extents of transformations in the surface regions produced by tests in Ar and Rb at 50°C had been about 60 and 80%. However, examination of cross-sectioned BS321 samples that had been tested at 150°C suggested that less than 5% had transformed and a similar observation was made for samples of BS316 tested at 50°C.

A few experiments were conducted to assess the effects of prolonged stress free exposure to Rb on mechanical properties. Samples of BS970/817, BS321 and Monel 400 were treated in air or Rb for 2060 hours at 150°C before being stressed to destruction in Ar or Rb at 50°C. No environmental effects were observed for BS970 or Monel 400, but BS321 tested in Rb was more ductile than that tested in Ar regardless of the exposure environment. There was also a slight strengthening when Rb environments were used in tests with BS321 samples.

The prime conclusion than can be drawn from the work is that a wide range of potential flask materials do not seem to suffer LME when deformed in Rb. While the testing conditions were far from comprehensive, some confidence is felt about the validity of the conclusion because the test conditions used should have maximised the chance of observing LME. Thus no restrictions on the choice of flask materials have been identified.

Far from embrittling, the Rb environments enhanced the ductilities of the FCC austenitic steels and nickel alloys. The results of the tensile tests on the pre-exposed samples show that these enhancements are due to effects occurring when the samples are being exposed. It is suspected that these beneficial effects are due to the action of Rb in wetting the alloys and stripping the oxide films from their surfaces.

Not only the ductilities but also the low temperature ultimate tensile strengths of the austenitic steels were enhanced by using Rb test environments. These materials also partially transformed to martensite and substitution in published correlations between the compositions and temperatures at which plastic deformation can induce the lattice shearing transformation from an FCC to a distorted BCC structure yield a very similar ranking for transformation susceptibility to that cited above for Rb environmental strengthening. These temperatures are about +40°C for BS304, BS321 and BS347 and about -40°C for BS316 and DIN 1.4439. Hence it is not surprising that little transformation was observed for BS316 that had been tested at 50°C or for BS321 samples tested at 150°C.

1.6.1.4. Occlusion and Storage of Krypton in Solids

Contractor: University of Antwerp (234-81-8 WAS.B)

A new immobilisation procedure for gases and vapors in solids has been investigated for encapsulating Krypton in zeolites. The procedure is based on an adsorption process in a solid, followed by a structural modification of the substrate, which is responsible for the encapsulation.

The modification consists of a chemisorption of e.g. silanes or boranes, on zeolitic hydroxylgroups in a primary reaction.

This reaction can be followed by secondary reactions.
The chemisorption is followed by an oxidation of the chemisorbed groups with O_2, H_2O, CH_3OH, etc.

These reactions create obstructions in the zeolitic channels, reducing the effective pore size in a controlled way so that a desorption of the adsorbed gas molecules (Kr, Xe, etc.) is prevented.

The encapsulation procedure, including the physical sorption of Kr, the chemisorption and the oxidation, operates at low pressures (≃ 0.5 atm) and moderate temperatures (0°-50°C) and has the advantage of low energy, running and installation costs. However, higher pressures (1-10 atm) and temperatures (100°C) can be applied for increasing the gas storage capacity and modification degree.

In the studied pressure- and temperature range (0.5 atm. 0-120°C) the best results were obtained on mordenite and on clinoptilolite. Diffusion- and percolation experiments showed that higher modification degrees are required for clinoptilolite than for mordenite. The smaller windows of clinoptilolite are blocked more efficiently by implantation of obstructions but the two-dimensional channel system requires a larger number of windows to be blocked than the one-dimensional system of mordenite.

Silane and diborane have been found to be very suitable for modification of H-zeolite. A higher reactivity of diborane towards zeolitic hydroxyl groups compared with silane, gives the advantage that higher chemisorption degrees can be obtained within reasonable time at low temperatures (0°C).

Encapsulations of Krypton were carried out by silanation, by boranation and by treatments with chlorosilanes ($Si(CH_3)_2Cl_2$ and $SiCH_3Cl_3$). When the formed Kr-zeolite systems were heated gradually, the main release was observed around 200°C. The zeolite loading is strongly dependent upon the temperature and pressure at which the gas fixation and modification process are performed.

A typical Kr-encapsulation experiment is shown in Fig. 16. The Krypton was adsorbed in silanated clinoptilolite at 0°C and at a pressure of 0.5 atm. Under these conditions Krypton loadings of 9.45 liters STP ker kg zeolite were achieved. Oxidation was carried out with water. Finally the sample was treated with dichlorodimethylsilane (coating procedure) in order to create an additional barrier for preventing desorption phenomena. Furthermore, the encapsulation of gases was found to be stable towards H_2O. Storage experiments under water for a long time reveal no detectable gas desorption.

Improvement of the thermal stability is expected by increasing the modification degree of the substrates.

In addition, the change in sieving properties of zeolites by chemical modification, has resulted in interesting separation possibilities. Narrowing the pore size gradually in a controlled manner by successive boranation, oxidation and thermal treatments resulted in exclusion of a gradually increasing number of gases as Xe, Kr, N_2, O_2 until finally all of them were excluded (Fig. 17). Application of this pore size engineering technique can lead to a separation of Krypton from a gas mixture by selective adsorption.

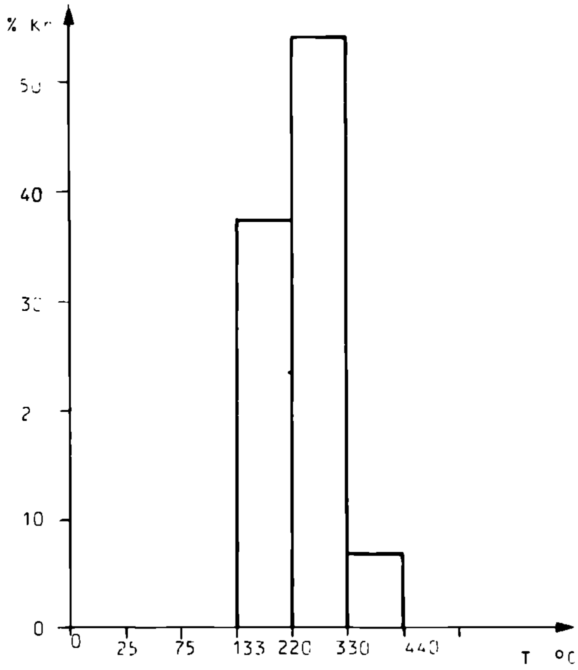

Fig.16: Stability of Krypton encapsulation in clinoptilolite performed by silanation and DDS treatment.

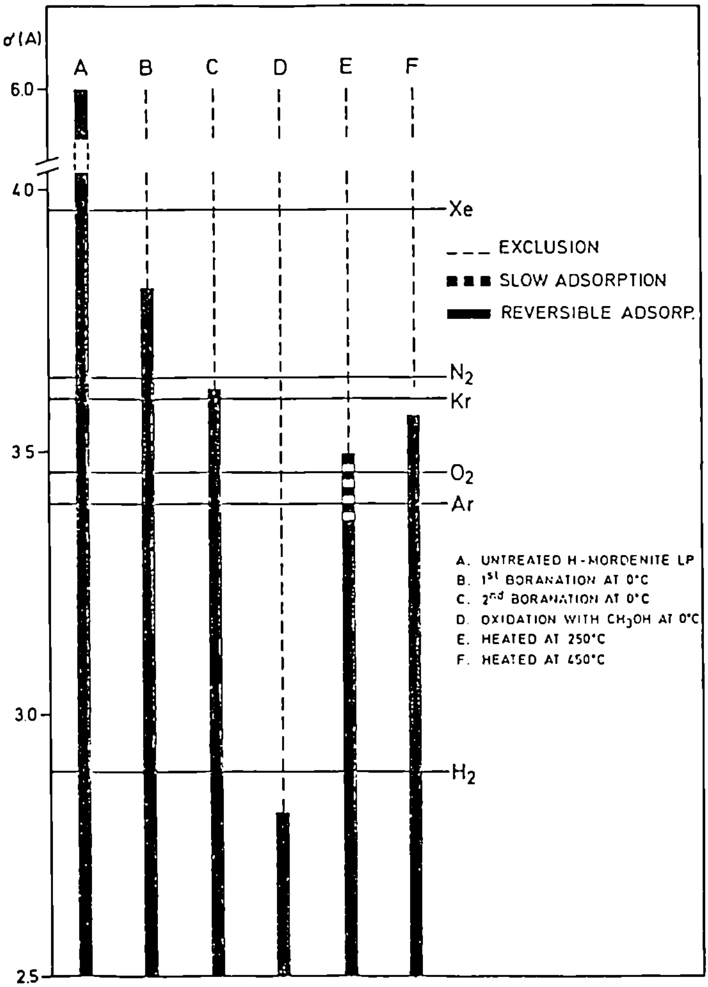

Fig. 17: Correlation between effective pore size of mordenite LP at different modification stages, with the kinetic diameter (σ) of Xe, N_2, Kr, O_2 Ar, H_2.

1.6.2. RETENTICN OF TRITIUM

1.6.2.1. Separation of Tritium from Aqueous Effluents

Contractor: SCK-CEN Mol (159-80-8 WAS.B)

Research was continued on the development of the ELEX process for the removal of at least 90% of tritium from the aqueous effluents of a commercial reprocessing plant (tritium content: about 100 Ci m^{-3}). The ELEX process uses a combination of water electrolysis and tritium exchange between hydrogen and water. The exchange is promoted by a hydrophobic catalyst. Recent work concentrated on increasing the overall tritium exchange rate, construction and testing of an integrated detritiation unit for process demonstration and optimisation at bench scale, and design of a pilot detritiation installation (80 kVA).

The fabrication of hydrophobic Pt-C-PTFE catalysts for tritium exchange between hydrogen and liquid water was optimised. The porosity of the catalyst was increased in such a way that a higher specific platinum surface was obtained despite a lower total platinum content.

The overall tritium exchange rate between hydrogen and liquid water was studied in 2 and 3 cm diameter counter-current packed bed reactors.

The combination of a more active catalyst, a more appropriate packing material and a suitable wetting procedure before any start-up of the installation resulted in a drastic increase of the over-all tritium exchange rate constants. At atmospheric pressure, at a gas flow rate of about 10 mol.$m^{-2}s^{-1}$ and at liquid flow rates of 2.5 to 10 mol.$m^{-2}s^{-1}$ heights of a transfer unit from about 0.2 m to less than 0.1 m were obtained with a 1 wt % platinum catalyst at temperatures from 20 to 80°C. This corresponds to over-all exchange rate constants from about 50 to more than 100 mol.$s^{-1}m^{-3}$.

An integrated bench scale detritiation unit has been constructed and installed in walk-in processing hoods. This installation consists of a 1.4 kVA electrolyser, an oxidation adsorption installation which is used as a recombiner and in-between two appropriate counter-current packed bed reactors.

After leak testing and cold testing of the installation two active, integrated and long duration experiments were carried out at feed concentrations of respectively about 0.4 and 2 mCi tritium per dm^3 water. In the first experiment, which lasted 553 hours, the water to be detritiated was fed directly to the electrolyser. 81 dm^3 water was detritiated. The decontamination factor was mostly around 100 and the tritium concentration in the electrolyser increased linearly with time until the planned value of about five times the tritium concentration in the feed was obtained. In the second experiment which lasted 1661 hours, one exchange column was used for stripping and the other for enrichment. 235 dm^3 water were detritiated with a decontamination factor between 50 and 100. The concentration factor was about 10.5.

A larger pilot detritiation unit has been designed. It will mainly consist of an 80 kVA electrolyser and a suitable counter-current packed bed reactor, i.e. an externally thermostated (max. 80°C) stainless steel exchange column with an enriching and a stripping part. An 8 kVA presaturation column is foreseen under the enrichment part and a cooler condenser will be placed on the top of the stripping part.

No recombiner will be used as tritium free water will be added at the top of the exchange column.

The 80 kVA electrolyser was delivered and preliminary installed.

The electrolyser, which is provided with built-in deoxo units and molecular sieve driers, produces max. 12 m^3h^{-1} dry H_2 (20°C, 1 bar) and 6 m^3h^{-1} dry O_2 (20°C, 1 bar). Its main operation is automatically controlled.

The first tests of the electrolyser were aimed at verifying the stability of operation of the unit at a rate of about 10 m^3h^{-1} H_2 (20°C, 1 bar). During these testing runs a few technical problems arised with some sub-systems which are now being adjusted repaired or replaced. With regard to possible tritium losses, the adaptation of this electrolyser for use in the ELEX process is being prepared.

Also a cooling unit with a thermal capacity of 30 kW and other peripheric equipment have been deliverd.

All tritium (and hydrogen) containing parts of the loop will be mounted in a vented room under depression with respect to the rest of the building.

1.6.3. ASSESSMENT OF I-129 AND C-14 RELEASE

1.6.3.1. Radiological Assessments of Waste Management - Modes for Iodine-129

Contractor: NRPB Chilton (161-81-8 WAS.nk)

The objectives of this study are to continue and conclude the 1979-80 study on 'Management Modes for Iodine-129' (1) particularly in the areas of radiological modelling and assessment of the relative merits of the various iodine-129 management options.

If iodine-129 is released into the environment, either as a liquid or airborne effluent, or subsequent to its disposal in solid form, the manner and radiological impact of its initial dispersion depends very much on the chosen management option. In contrast, doses due to the subsequent global circulation of iodine-129 prior to its eventual decay (with a half-life of 17 million years) depend much less on the initial mode of release. It is therefore convenient to have a range of initial-dispersion or 'regional' models, one for each management option, each of which is carefully interfaced to a common global-circulation model.

The consequent requirements for a satisfactory global-circulation model for iodine-129 have been identified, and a suitable model adopted. A model proposed (2) by the Oak Ridge National Laboratory (USA) was thoroughly reviewed, using a data-base on the environmental behaviour of iodine-129 prepared in the framework of the previous study, and modified where necessary for the purposes of this work. The structure of the model (Fig. 18) facilitates interfacing to a range of different regional models, which will assist in comparisons between the radiological impacts of the respective iodine-129 management options.

The model has provided new insights into the behaviour of iodine-129 in soil, which will also be incorporated into more detailed regional models, and into the fate of iodine-129 on the million-year timescale. At such times after release the model predicts that about 99% of the iodine-129 will have been incorporated into ocean sediments (Fig. 19 , line 7), but that it will be almost entirely re-released before it decays. Therefore ocean sediments appear to constitute a major long-term reservoir of iodine-129, but there appears to be no true 'ultimate sink' for this extremely long-lived radionuclide.

Revision and development of regional models continue, with the objective in 1982 of making an improved and unified radiological assessment of the relative merits of the management options for iodine-129. Work being carried out by NRPB under the Commission's Biology and Health Protection Programme, on the use of cost-benefit analysis as an aid to waste management decisions will also contribute to this study.

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(1) McKay, H.A.C. (Ed) Management modes for iodine-129, EUR 7953 (1982)

(2) Kocher, D.C. A dynamic model for the global iodine cycle for the estimation of doses to the world populations from releases of iodine-129 to the environment; ORNL/NUREG-59 (1979)

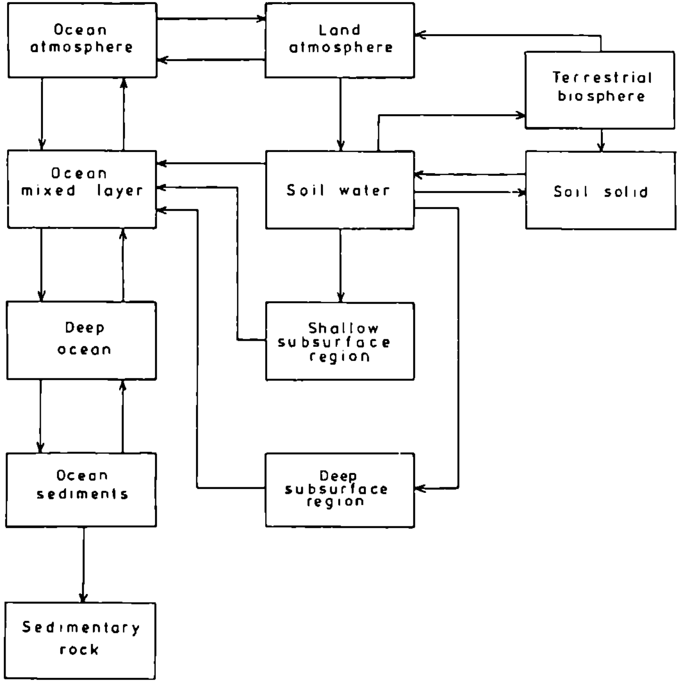

Fig. 18: Proposed model for the global circulation of iodine, on to which releases of ^{129}I may be superimposed.

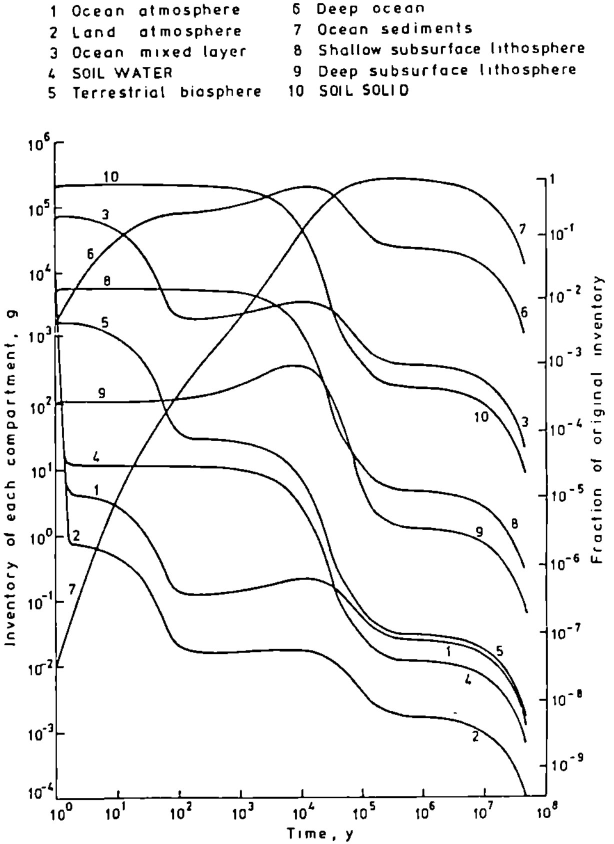

Fig. 19: The inventory in each compartment following a release of 2 TBq (305 kg) of ^{129}I in one year to the land atmosphere, as predicted by the proposed model.

1.6.3.2. Assessment of Carbon-14 Waste Management

Contractor: AERE Harwell (163-81-8 WAS.UK)

Carbon-14 arises in nuclear reactors mainly as a result of neutron activation reactions of the nitrogen-14, carbon-13 and oxygen-17 nuclei present in core materials, either as impurities or as constituent elements. Depending on the type of reactor system, carbon-14 can therefore be produced in the fuel, the cladding, the coolant, the moderator and the structural materials. Both the amounts produced, and the chemical species as which the isotope is present, depend strongly on the details of the system, as do the subsequent behaviour of carbon-14 and the pathways by which it can be released to the environment.

Carbon-14 present in the coolant is usually released to the environment in the reactor off-gases, either as hydrocarbons or as carbon dioxide. That present in the graphite moderator of Magnox and advanced gas-cooled reactors (AGRs) and any present in core structural materials of other reactors will remain in the reactor until decommissioning takes place. Carbon-14 within the fuel elements, either in the fuel itself or in the cladding, will normally pass to the reprocessing plant. There the inventory in the fuel will mainly be released as carbon dioxide into the dissolver off-gases; and the cladding will normally constitute as separate solid arising.

For most types of reactor system, representative calculations of carbon-14 production rates have been published in the open literature. In some cases, experimental measurements of releases to the environment have also been reported. A critical analysis of such evidence has led to a reliable picture of carbon-14 flows within the various reactor types and fuel cycles. A summary of typical quantities arising, based on all the evidence, is given in Table 1.

TABLE 1. Summary of Carbon-14 Waste Arisings (Ci/GW(e)year)

Reactor type	Total Production	Releases to off-gases (a)		Solid wastes	
		Reactor	Reprocessing plant	Reactor	Reprocessing plant
BWR	95	10(b)	20(c)	45(d)	20(e)
PWR	70	5	20(c)	25(d)	20(e)
Magnox	380	30	100	240	10(f)
AGR	230	15	20	145(g)	50
LMFBR	25	small	5(h)	(i)	20
HTGR	150	small	150	small	small
HWR	275(j)	200	40	(i)	35

Notes

(a) It is assumed that no steps are taken to reduce carbon-14 emissions.

(b) Calculations give values of 5-6 Ci, similar to those in PWRs. However, measured releases seem to be consistenly higher for BWRs.

(c) Based on a nitrogen level of around 25 ppm in the fuel.

(d) The arisings in stainless steel structural components calculated by Davis are assumed mostly to remain in the reactor.

(e) It is assumed that production in other parts of the fuel element bundle is small compared to that in the zircaloy cladding.

(f) Represents arisings in the Magnox cladding. If chemical decladding is used rather than mechanical decladding, or if the Magnox swarf is dissolved in acid as part of a waste treatment procedure, then this quantity will probably arise as a gaseous rather than a solid waste.

(g) Includes 10 Ci arising in the graphite sleeves of the fuel elements. Although they are actually transferred to the reprocessing plant, these can be regarded as a decommissioning waste for the purposes of waste management studies.

(h) Based on a nitrogen level of 10 ppm in the fuel.

(i) No calculations have been reported. Probably small in LMFBRs.

(j) Based on Bonka's calculations for the Atucha reactor. It is assumed that nitrogen gas is not used to fill the annulus.

All carbon-14 produced in the fuel and its cladding in any reactor passes to the reprocessing plant. For metal clad oxide fuels (and probably for Magnox fuel) the available evidence is that essentially all the carbon-14 in the fuel is evolved as carbon dioxide into the dissolver off-gases, when dissolution in nitric acid takes place. The cladding will generally constitute a solid waste arising at the reprocessing plant.

The quantity of carbon dioxide in dissolver off-gases arising from the carbon-14 content of the fuel will, in general, be very much less than the quantity in the air cover gas. In a plant handling 1500 t of LWR fuel per year at a burn-up of 33,000 MWd/t , the annual carbon-14 arising in the dissolver off-gas would be about 900 Ci, corresponding to 660 g of $^{14}CO_2$. For an illustrative air flow rate of 200 m^3/h and 300 days operation per year, the total flow of carbon dioxide would be about 800 kg/year, implying an isotopic dilution factor of 1200. These figures will vary significantly from plant to plant.

In selecting a method for trapping carbon dioxide, it is desirable to choose a method that converts the gas to a form suitable for subsequent immobilisation and disposal. Most workers in the field consider that an alkaline earth carbonate (probably calcium or barium carbonate) will be the best compound for immobilisation, although the suitability of such compounds needs to be confirmed. (In particular, leach rate studies of composites (eg with cement) of metal carbonates are required).

Three alternative trapping processes that convert carbon dioxide to such insoluble carbonates have been suggested. Any of them could be used as a preliminary to krypton removal, as part of the product purification system of a fluorocarbon separation for krypton, or to treat gas regenerated from a molecular sieve bed or a cold trap. They are:

(a) The double alkali process. Carbon dioxide is absorbed by sodium hydroxide solution. The resulting liquor is transferred to a vessel in which it is treated with calcium hydroxide to form calcium carbonate and regenerate sodium hydroxide for recycle. Calcium carbonate slurry is drawn-off and concreted. (A modification of this process could be used to treat the liquor from a caustic scrubber).

(b) The direct process. Carbon dioxide reacts with an aqueous slurry of calcium hydroxide to give calcium carbonate directly.

(c) The barium hydroxide octahydrate process. Carbon dioxide reacts with a fixed bed of solid barium hydroxide octahydrate to give barium carbonate and water, which volatilises.

Any of these processes is capable of giving a high decontamination factor for carbon-14. Each has its advantages and disadvantages.

The barium hydroxide octahydrate process is dry, so that the potentially troublesome filtration or sedimentation step is avoided. However, the process requires a feed gas of very carefully controlled humidity and of low carbon dioxide content. The necessary humidity control may be difficult to achieve in an operating plant.

The double alkali method can be regarded as established technology. It has the important advantage over the slurry reactor method, (b), that no solids are present in the absorption column. No blockages can occur, and the process is relatively insensitive to fluctuations in pressure and mass flow rate. As to the radiological impact of carbon-14 release to the environment, work is under progress and will be reported subsequently.

1.6.4. IMPROVED AEROSOL FILTRATION TECHNIQUES

1.6.4.1. Removal of Solid and Droplet Aerosols on a Packed Fiber Mist Eliminator

Contractor: KfK Karlsruhe (162-81-8 WAS.D)

By means of a remotely handled packed fiber mist eliminator (Fig. 20) it is intended to remove droplets < 10 μm with a removal efficiency ≥ 99%. Moreover, it is to be used as a prefilter for particulates in reprocessing off-gases so as to increase the service life of the down stream HEPA filter.

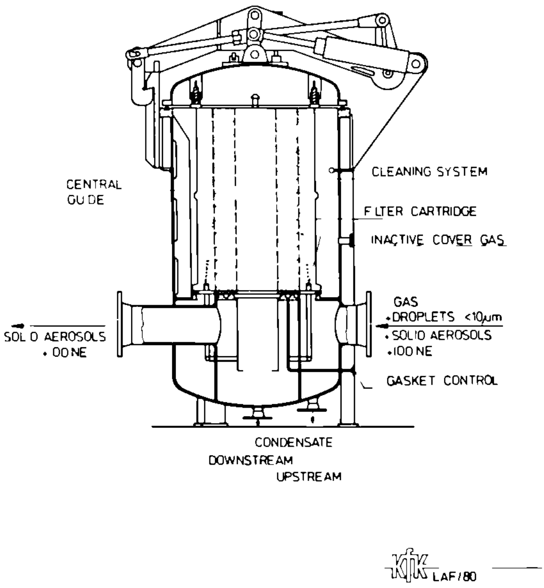

Fig. 20: PASSAT packed fiber mist eliminator for droplet removal.

The demister consists of packed glass fibers of about 20 µm diameter with a statistically vertical orientation. Impinging droplets are retained by the fibers, combine into large droplets and due to gravity and pressure difference they migrate from top to bottom in the fiber layer to reach ultimately the sump of the filter vessel. As soon as an unacceptable high pressure difference over the fiber layer due to the salt loading is obtained a spraying device is actuated. It consists of a spray ring with six nozzles so as to wash out the crystallized salts and the solid aerosols deposited with diluted nitric acid.

The dual feed nozzle for droplet generation was operated in all the tests at 1 l H_2O/h and 1 l of solution/h, respectively. In the measurements on the untreated and clean air sides of the packed fiber mist eliminator, droplet spectra were obtained in the ranges shown in Fig. 21. The removal efficiencies were determined at 30°C and 50°C and 100% r.h. at different flows of 75 - 150 m^3/h. No dependence on temperature was found. All bulk removal efficiencies exceeded 99.99%. However, large droplets are removed predominantly and are responsible for the high removal efficiencies. For this reason, also the frequencies of identical droplet sizes on the untreated and clean air sides have been compared.

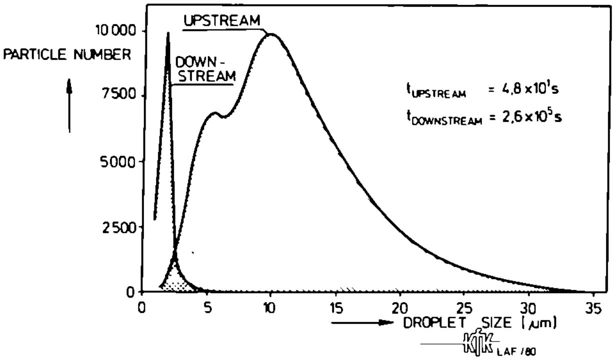

Fig. 21: Droplet size distribution for the calculation of the removal efficiency.

For small droplets (1 - 10 µm) the removal efficiency as a function of diameter was mostly between 99.8 and 99.99995 %, which corresponds to decontamination factors of 500 to 2.10^6. No dependency on the volume flow of the retention factors for droplets was found. With increasing droplet diameter the removal efficiency rose steeply (Fig.22).

Since evaporation will have to be taken into account especially for the small droplets mainly on the clean air side, a 5% so sodium nitrate solution was sprayed. The mass of dried $NaNO_3$ particles put onto nucle pore filters was determined by electrodes selective to sodium ions.

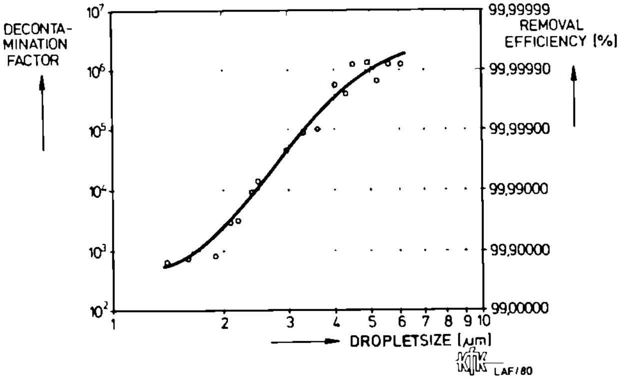

Fig. 22: Decontamination factor as a function of droplet size.

This resulted in a bulk removal efficiency of > 99.9% for the same aerosol spectrum in a packed fiber mist eliminator (packing density 300 kg/m^3, fiber diameter 10-20 µm) with 50 mm thickness of the layers and a gas flow of 75 $std.m^3.h^{-1}$. Comparison of this result with the result obtained from scattering light measurements indicates that some evaporation of small water droplets on the clean air side cannot be excluded. The packed fiber mist eliminator was exposed to sodium fluorescein particles of an average particle diameter of 0.12 µm. Tab. 1 lists the decontamination factors found as a function of the volume flow and the average velocity in the fiber packing, respectively. The decontamination factor is in excess of 1000. Moreover, it is seen from Fig.23 that removal decreases with increasing velocity $\bar{v}$ in the filter packing.

Tab. I: Dependence on volume flow and mean velocity $\bar{v}$, of the (DF) decontamination factors of packed fiber mist eliminators (thickness of layer: 50 mm, test aerosol: airborne sodium fluorescein particles.

$V \left[m_N^3 \cdot h^{-1}\right]$	75	100	125	150
$\bar{v} \left[cm \cdot s^{-1}\right]$	2.9	3.9	4.8	5.8
DF	$4.5 \cdot 10^3$	$1.9 \cdot 10^3$	$1.4 \cdot 10^3$	$1.2 \cdot 10^3$

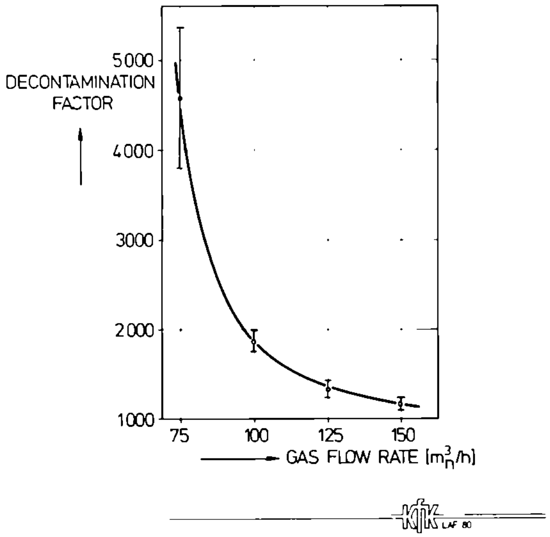

Fig. 23: Decontamination factor as a function of flow-rate.

1.6.4.2. Aerosol Filtration at Elevated Temperatures by Regenerative Metal Fibers

Contractor: SCK-CEN Mol (228-81-8 WAS.B)

This experimental investigation of aerosol filtration is aimed at the development of fibre metallic prefilters to be located up-stream of HEPA-filters in view of reducing the consumption of the latter. Moreover, in situ regeneration of these prefilters will be studied in order to minimise the arising of secondary waste. The metallic nature of these prefilters allows their use in exhaust gases at elevated temperature such as in the case of waste incinerators or waste vitrifiers.

As a first task, screening tests at ambient temperature have been performed on different metallic fibre materials. Therefore an experimental test unit, called AFLT (Aerosol Filtration at Low Temperature) has been put in operation. Its gas throughput is 15 m^3h^{-1} allowing to test flat metallic filters at superficial velocities in the range of 3.5 to 19 cm s^{-1} by variable by-pass of the filter.

Monodisperse latex aerosols in the size range from 0.091 to 2.2 μm are sprayed by means of a Collison atomiser. The concentration of aerosols in the carrier gas is measured alternatively before and after the filter material using two laser spectrometers having each 60 channels. The useful filtration surface is 133 cm^2.

In this AFLT-unit, the filtration performance of porous webs and sintered mats were measured. The fiber diameter of the porous webs was changed in the range from 4 to 12 μm and the number of filter layers from 1 to 5. The three different sintered mats tested had an absolute filter rating for liquid filtration quoted at 10, 25 and 40 μm respectively. As an example of the results obtained, the decontamination factors in a porous web with metallic fiber diameter of 4 μm at a superficial velocity of 3.5 cm s^{-1}, are given in Fig. 24.

The study has led to the optimum choice of the working parameters of the best combination of filtermaterials while keeping the working pressure drop lower than 80 Pa.

On this basis candle type filters of three different compositions have been designed. They will be tested in the future using a three-candles-holder, increasing the filtration surface in this way by a factor of ten. Their filtration performance and regeneration possibilities will be determined for monodisperse and for polydisperse soluble or insoluble aerosols.

Finally, the performance of these metallic filter-candles will be measured in a second unit, called AFHF (Aerosol Filtration at High Temperature). This unit has been designed and is under construction. Its aerosol generator is a high temperature spray calciner which can generate simultaneously water vapour and nitrous oxides.

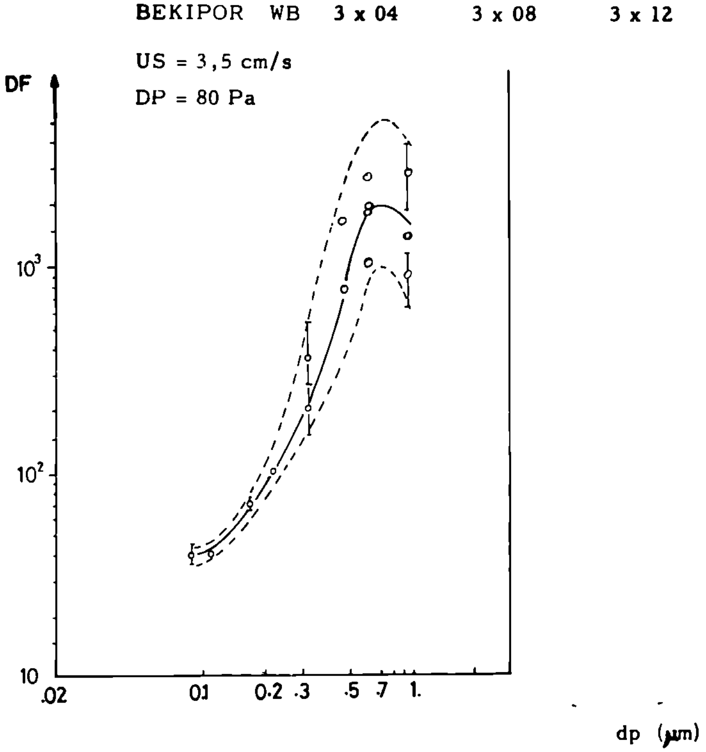

Fig. 24: Decontamination factor of a porous web as a function of particle diameter.

WASTE STORAGE AND DISPOSAL

2.1. SHALLOW LAND BURIAL OF SOLID LOW-ACTIVITY WASTE

This sub-programme covers the following three topics :

- Assessment of current experience.
- Improvement of burial techniques.
- Study of radionuclide migration and safety aspects.

As of end of 1981, a total of 5 research or study contracts covering these subjects have been concluded on the basis of the proposals described in the previous annual report (Ref. EUR 7652).

2.1.1. STATUS OF EXISTING EXPERIENCE

2.1.1.1. Present Situation and Prospects for the Burial of Low-Activity Solid Waste at Shallow Depth within the Community

Contractor : CEA, Cadarache (267-81-6 WASF)

The aim of the study, to be carried out in 1982, is to review the state of the art and the prospects in the Community's Member States in the field of shallow land burial of low-activity waste.

The study being mainly based on an earlier questionnaire, will point out the following subjects :

- regulations and licensing conditions ;
- selection criteria for the burial sites ;
- characteristics of the waste to be buried ;
- conditioning and handling of the waste to be buried ;
- operation of the burial sites ;
- operational and post-operational surveillance of the sites ;
- economic aspects.

2.1.2. IMPROVEMENT OF BURIAL TECHNIQUES

2.1.2.1. Improvement of the Radioactivity Confinement by Soil Barriers

Contractor : CEA, Fontenay-aux-Roses (196-81-6 WASF)

The task of selecting and implementing processes for improving the confinement properties of soil barriers comprise two main stages. The first one is related to the compilation and classification of techniques and industrial processes for rendering soils impermeable. The second stage deals with experimental studies, on laboratory and at full-scale, concerning the retention properties of soils and filler materials.

A technical documentation on the construction of impermeable barriers and on injection processes has been compiled. This documentation comprises the following headings :

- theoretical studies ;
- optimum geological application conditions;
- implementing modes ;
- characteristics of the used materials ;
- examples of practical applications ;
- costs.

As far as the experimental stage is concerned, a testing device has been set up (see Fig. 25) and tested for tightness under pressure. The tests were carried out without samples in a preliminary step. No leackage was observed during 24 hours at 12 bars. A successive test with a sample showed some leackages. After having slightly modified the device, a new tightness test has been carried out with a sandstone sample of 10 cm height. This test showed a normal percolation rate of 50 cm^3 of water in two hours at a surface of about 35 cm^2 and at a pressure of 6 bars. The corresponding permeability is 5.10^{-8} $m.s^{-1}$. Future tests will be carried out with a Cs-137 labelled caesium chloride solution.

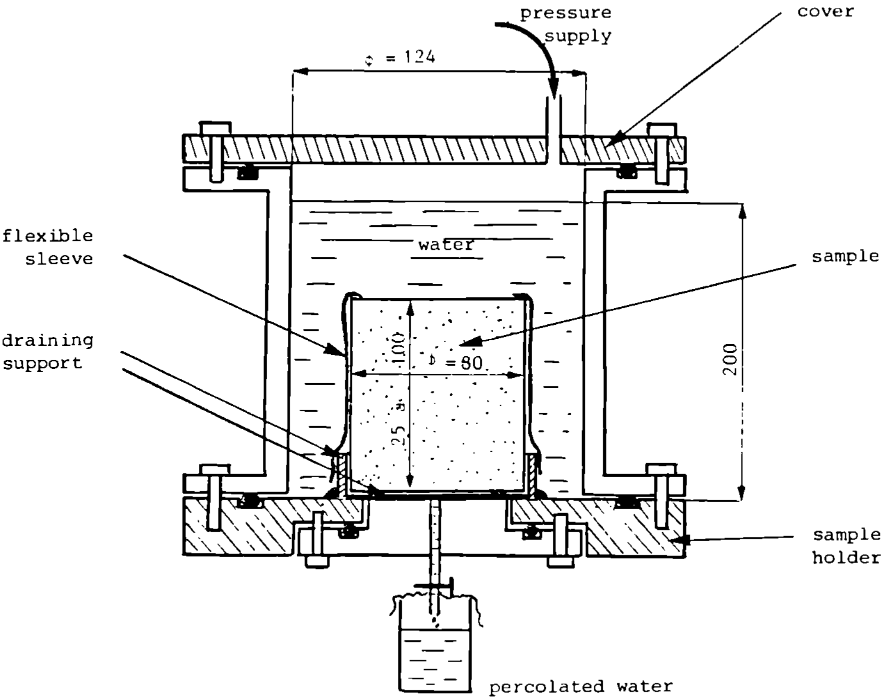

Fig. 25: Testing device for water percolation rate

2.1.2.2. "Development of Waste Unit for se in Shallow Land Burial."

Contractor : Risø Nat. Lab. (195-81-6 WAS.DK)

The investigation concerns the improved safety of disposal of low- and medium-level radioactive waste in suitable man-made constructions placed below ground but near the surface.

The repository under development is based on the use of standardized waste units of hexagonal shape which are placed closely together in the repository .

The units are made of concrete and the narrow intervals between them are filled with bitumen. The soft impermeable bitumen layers prevent contact between the concrete units containing the waste and the ground water even if cracks should develop in the constructions of the repository. On the other hand the high pH values associated with concrete reduce the risk of bacterical attack on the bitumen.

The hexagonal waste cells can be used directly as containers for solidified waste or as an over-pack system for ordinary 210 l standard steel drums.

Moulds for casting of the containers have been constructed at Risø and a few units have been made at the Cement and Concrete Laboratory of Aalborg Portland Cement Factories which acts as a consultant for the project. No difficulties with the casting procedure were encountered and a large number of the units will be made at Risø in the near future. These containers will be used to study the durability and quality of the units under various more or less extreme circumstances. Information about long-term behaviour which can be attained within the time-frame of the contract is of course limited.

Two different types of concrete for the construction of the units will be compared. One is an ordinary sulphate resistant high quality concrete and the other is a special type of concrete called DENSIT developed at the Cement and Concrete Laboratory. This material is also based on sulphate resistant Portland cement but with an addition of up to 20 weight % of the cement of silica fume powder, i.e. a waste product from ferro-silicium production consisting of very small - about 0.2 μm spherical particles of nearly pure silica. A small amount of organic superplastiziser is used. This to make easily castable concrete with low water/cement ratio.

The DENSIT material has extremely high compression strength. Measured values of the material used in the first units were 125-140 MN/m^2 after 28 days curing. This can be compared with typical values of about 50 MN/m^2 for ordinary good quality concrete.

The DENSIT has also other properties which make it an interesting candidate material for the construction of barriers around disposed radioactive waste. Due to the structure of the material the permeability is extremely low. Diffusion coefficients for dissolved ions in DENSIT tend to be two or three decades lower than in ordinary concrete. Reactions possibly due to carbon dioxide from the atmosphere also may reduce the apparent diffusion coefficients for ordinary concrete considerably.

The electrical resistivity of DENSIT is in the same way two or three decades higher than the resistivity of ordinary concrete. This is important for the use of the material as corrosion protection for embedded steel in form of reinforcement but also as protection for the steel plates when the system is used as over-pack for standard drums. A draw back as far as corrosion protection is concerned is the somewhat lower pH in DENSIT compared with ordinary concrete. Reaction between the added silica and calcium hydroxide from the hydration of the cement minerals is an important mechanism in connection with the long-term behaviour of the material.

The properties of the new material are investigated experimentally in connection with this project as a cooperative effort between Risø, the Cement and Concrete Laboratory and the Danish ATV Institute: Korrosionscentralen.

2.1.3. RADIO-NUCLIDE MIGRATION AND SAFETY ASPECTS

2.1.2.1. The Influence of Ground Heterogeneity on the Dispersion of Radio-Nuclides

Contractor: Soil Mechanics Laboratory Delft (193-81-6 WAS.NL)

The work concerns the calculation of the dispersion of radionuclides from a shallow burial site into a heterogeneous soil system. As in any pollution problem the three aspects source, path and target have to be identified.

For the purposes of this study the source can be identified as a release of chemicals into the soil. The results of a literature survey indicate that in an actual or proposed site almost any chemical element can be present and simplification is therefore necessary.

It is proposed to classify the migrating species according to the following scheme:

Type	Characteristics	
I	simple anion	adsorbed neither on mineral nor organic soils
II	simple cation	typically from group I or II of the periodic table. Can be involved in cation exchange reactions on mineral and organic soils
III	metal cation	specifically adsorbed on soil components
IV	protonium ion H^+ (tritium)	exchange reactions in acid-base systems high mobility
V	organic metal complex, or oxide complex	behaves as an anion but possible unstable to Red-Ox changes or pH-changes
VI	clay complexes	migration of clay colloid with associated strongly adsorbed species. Filter effects possible

This classification is not exclusive of course. Whereas potassium for example would be assigned to type II in normal soils its behaviour in illite clays is analogous to type III metals. It is to be expected that the important migratory species will only be present in trace concentrations. Two extreme types of background leachate composition have been identified. These are ordinary soil solution and leachate that strongly resembles that from domestic landfill sites, characterised by high specific conductivity, COD and BOD in an anerobe system.

The migration path can be considered to consist of two sections an unsaturated system followed by a saturated part. At this time it would appear that the convective water flow along both paths can be considered to be at steady state, although this conclusion requires checking in detail for the unsaturated part of the path. Liquid phase convection, dispersion molecular diffusion, adsorption and desorption are the important transport mechanisms in the system.

Two targets can be identified. First the ground water as a whole at the end of the unsaturated section, and second, a remote location, say 1000 m, from the site at the end of the saturated section.
Characteristic for the target is that the concentration on the spot is only a small fraction of the concentration at the source. The physical model of geosphere is based upon the principle of dividing the relevant flow field into volumes whose properties vary in space but not in time. The net effect of the leachate at the target is then the sum of all the effects in the volumes on the path. The mathematical formulation of the problem is simple. The difficulty is the identification of the transport frunction.

Different transport functions are appropriate to different porous media, transport mechanisms and scales of the problem. For the homogeneous case there is only one flow field volume, and one transport function.

For the heterogeneous case the equations can be generalised in one of the following ways which can also be combined by dividing the field into volumes:

- By assuming that the parameters are spatially constant but adjusting their value to reflect the heterogenity.

- By allowing the parameters to vary determinately over the field
- By allowing the parameters to vary randomly over the field.

These three cases may give very different forecasts of the concentrations along a path.
More detailed models will be evaluated in the cours of the program.

2.1.3.2. Assessment of Radiological Protection Aspects and Review of Siting Principles

Contractor: NRPB Chilton (233-81-6 WAS. UK)

This assessment aims to provide information which will allow some conclusions to be drawn about the types and quantities of wastes potentially suitable for disposal, site selection, burial facility design and the time period for which the site should be monitored and maintained and access restricted.

A review of current and future shallow land disposal operations indicated that several different waste types should be considered. A set of reference inventories has therefore been compiled, detailing the radionuclide content, the physical and chemical forms of the waste and the quantities which have been or may be disposed of the burial sites. The first inventory consists of low level wastes, including general trash, of the type which are currently disposed of by shallow land burial in the UK. The second inventory consists of the major types of intermediate level wastes which arise at Magnox reactors. The third inventory consists of LWR operating wastes.

One of the aims of the assessment is to provide some information which can be used in designing appropriate facilities for given sites and wastes. In order to cover a wide range of disposal practices two reference design facilities have been defined. The first design incorporates minimum engineering being an 8 m deep trench, the bottom of which is lined with a thin layer of gravel, capped with 1 m of soil. No waste conditioning is assumed. The second design is a fully engineered trench which has a total depth of 18 m and is lined with concrete. The wastes are assumed to be incorporated in cement and packaged in 200 1 drums. After emplacement the gaps between the drums are infilled with cement and the trench sealed with a 10 m cap consisting of puddled clay and concrete.

Three different periods can be identified for a shallow land disposal site:

Stage 1. Operational (i.e. while wastes are being buried)
Stage 2. Post-operational; site still maintained and monitored and access restricted.
Stage 3. Post-operational; site returned to 'green fields'.

Possible release mechanisms during each of these stages were reviewed. This review indicated that for the purposes of the assessment three initial release mechanisms should be considered:

- Trench fire (Stage 1 only)
- Contact by percolating rainwater and/or rising groundwater (all stages)
- Human intrusion (Stage 3 only)

Work is underway to define the methods, assumptions and parameters to be used in calculating the risk arising from inhalation of contaminated material dispersed by a fire in the minimum engineered trench.

For a minimum engineered trench, coefficients defining the release of radionuclides from the wastes to the percolating water have been estimated from data obtained for the release of metals from ordinary landfill wastes. A simple layer model has been developed to calculate radionuclide migration through the unsaturated zone. An existing one-dimensional migration model will be used to calculate lateral movement towards the stream. This model is currently being modified to include non-linear sorption

Due to the greater depth of emplacement it is likely that the engineered trench would be positioned below the water table. Hence, it is important to locate the trench in strata which have good sorption properties and a generic clay site has therefore been chosen. Because of the configuration in which clays are found with harder, more permeable rocks, most clay outcrops are under artesian conditions with groundwater tending to rise vertically on a regional scale. However, nearer the surface minor relief features will have considerable influence over groundwater movement. The relief assumed for the generic site is shown in Fig. 26. A two-dimensional finite element model is being used to calculate the detailed groundwater flow pattern at the site for trench permeabilities in the range 10^{10} - 10^{-5} ms^{-1}. These results will be used to divide water passing through the trench into a series of one-dimensional flow paths. Radionuclide migration in each of the paths will then be calculated using the existing migration model.

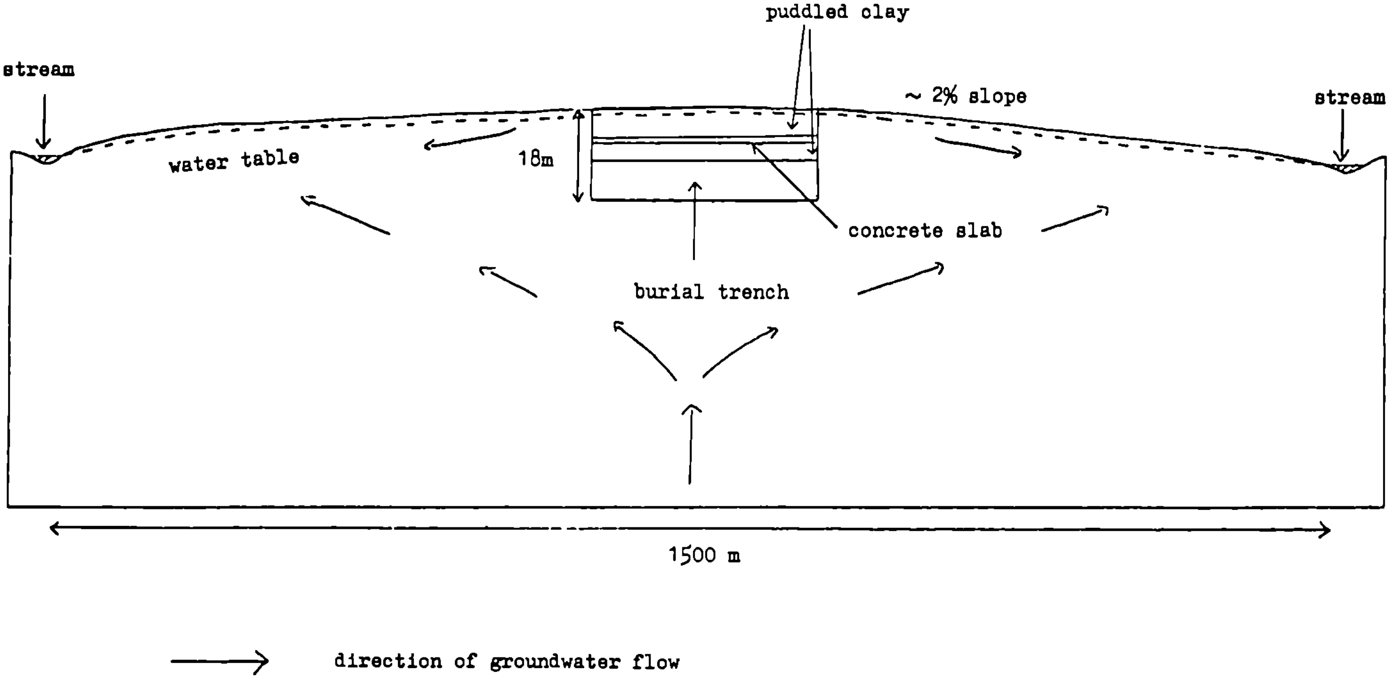

Fig. 26 : Expected groundwater flow pattern at clay generic site (not to scale)

2.2. STORAGE AND DISPOSAL IN GEOLOGICAL FORMATIONS

The main objective of this sub-programme is to determine the optimum conditions for the disposal of high level radioactive waste in geological formations, so that no harm results neither for man nor for the environment. Four geological formations are being studied resp. clay, granite, salt and ocean sediments. Considering the complexity of the subject a large number of aspects have to be studied. They have been grouped into a limited number of tasks according to their operational logic as shown by the headings of this chapter.

Again during the year 1981 scheduled field research suffered from delays due to non technical reasons. (failing of authorisations from local or national authorities). The borehole studies resulting from the drilling campaign in Altnabreac (UK) were completed , however additional scheduled drillings at other sites were postponed.

The sinking of the access shaft for the underground experimental room at Mol (B), which started at the end of 1979, was completed just before the end of the year 1981. The final depth of the shaft is now 227 m and the excavation of the horizontal experimental room will start in early 1982. It is expected that the room will be operational by mid 1983 .

The research programmes in the Asse II salt mine (D) have continued. The thermophysical tests at the temperature test field IV were finished by the end of November and the results are being analysed. The temperature test field V is almost completely equipped and experiments will start early in 1982.

With regard to the research programme performed in the abandoned iron mine Konrad (D), the acquired scientifical and technical results are presented in a final report. This report will be used as a basic document for a so called "plan" which is needed for the submission of a licensing procedure for the construction of a final repository for low-level and decommissioning waste in the mine.

The laboratory and in-situ experiments on the characterisation of the internal equilibria, before and after disposal of radioactive waste, of the three continental geological formations considered, have continued. In particular much emphasis has been put on the study of the impact of heat on the geotechnical as well as hydrogeological properties of the host rocks. A review study was performed which aimed at collecting the existing knowledge and assessing the admissible thermal load. It was concluded that within the present state of knowledge heating effects are predictible with an acceptable degree of confidence and can be coped with by applying an appropriate waste management (cooling time) and/or repository design (disposal density).

During the year 1981 a coordinated action was launched on the evaluation of the isolation performances of the four geological formations considered (PAGIS). The action is divided into three phases. The first phase to be started at the beginning of 1982 aims at collecting, discussing and selecting data, range of parameters and variants in view of the subsequent evaluation (phases 2 and 3).

2.2.1. CONTINENTAL FORMATIONS

2.2.1.1. DEEP DRILLING PROGRAMMES

Drilling campaigns and field research area studies have suffered in some Member States from delays due to licensing problems. Only the drilling campaign around the Mol site (B) for hydrogeological research could be performed on schedule.

The borehole studies resulting from the drilling campaign in Altnabreac (UK) were completed, however additional drillings at other sites could not be performed.
The drilling campaign scheduled to start in France was delayed because of a review of policy by the government.

2.2.1.1.1. Investigation into the Hydrogeology of the Mol Site and the Mol Area (B)

Contractor: SCK / CEN, Mol (144-80-7 WAS.B)

The aim of this project is to install a network of observation wells in an area around the Mol site.
The well observations have to supply the data and information necessary for the modelling of groundwater movement in the aquifers above and beneath the Boom clay layer.
Observation wells were installed at 20 locations. A total length of 5,325 m was drilled in 1981, 7,535 m of piezometric tubes and 52 screens were installed in the different aquifers. The screens are placed between the -5m and -572 m level depending upon the location and the aquifers encountered. The basin of the "Kleine Nete" between Lommel-Postel (east of Mol) and Herentals (west of Mol) can now be monitored for the various aquifers above the Boom clay layer and one aquifer just beneath this formation.

Borehole loggings were performed in the deepest borehole of each drilling location and soil samples taken. This allowed to identify the geological sequences and to correlate the results with those previously obtained during boring operations and local seismic prospection campaign at the Mol site. It was also abserved that 15 km east of Mol the top of the Boom clay layer is about 80 m deeper than expected.

The water levels (piezometric heads) have been measured every week. A first tentative to determine the water flow direction into the under-Rupelian aquifer indicated a westwards water flow.

A mobile data logger unit was made available for continuous data recording on all screens of each location. The records can be computerized and directly plotted.

2.2.1.1.2. Geological Assessment of the Crystalline Rock Formation at Altnabreac (UK)

Contractor: NERC/IGS , Harwell (128-80-7 WAS. UK)

Hydrogeological testing of the three deep boreholes at Altnabreac was completed. The work was carried out using a packer system developed by IGS which is capable of measuring in-situ hydraulic conductivity and hydraulic potential within isolated borehole sections between packers. Hydraulic conductivities from 1×10^{-12} to 2×10^{-6} m/sec were measured with an approximately log normal distribution skewed towards higher values, as a result of fracturing. The average bulk rock value is 7×10^{-10} m/sec, whilst the fracture zones average 1×10^{-6}m/sec. Storativity values measured using slug test techniques, average 1×10^{-6}. Hydraulic head differences have been measured in the boreholes, usually in the order of 0.12 m, but variations of up to several metres have been encountered. These head differences induce groundwater to flow vertically in the boreholes from one fracture zone to another. However, the volumes of water involved do not exceed 0.25 m^3/day. It is clear that only about 10% of the boreholes contribute to the total transmissivity and that a small number of fractures or fracture zones provide the bulk of the water flow through the rock.

In addition to the hydrogeological testing, water samples were collected from selected depth zones in the three deep boreholes using the packer system and also from the shallow boreholes using a specially designed lift pump. These samples were collected after sufficient volume had been removed to ensure that the water was being drawn from the body of the rock rather than from borehole storage. Water samples were also collected from 60 of the 140 springs successfully identified using thermal infrared linescan techniques earlier in the year. These water samples are being analysed for major and trace elements and also for D, ^{18}O, ^{13}C, ^{3}H, ^{14}C and U/Th. In some selected samples dissolved inert gas contents and the ^{222}Rn and ^{226}Ra contents are also being measured. The results to date indicate that the groundwater is low in total dissolved solids, has low chloride, is bicarbonate dominated and iron rich.

Helium contents from some of the deep groundwaters indicate water ages in the range 10 000 - 30,000 years. None of the spring samples contain significant amounts of older water. Since the primary route for the movement of groundwater in crystalline rocks will be via the fracture system, the chemistry is likely to be controlled by reactions which occur between migrating fluids and minerals infilling or coating the fracture surfaces.

The geophysical borehole logging has been completed including an assessment of the experimental "Darcylog" for the determination of in-situ permeabilities. The work has covered the assessment of a wide range of logging techniques and their interpretation in terms of geology, hydrogeology and geomechanical properties of the rock mass.

The computer programme NAMMU developed by the UKAEA, is being used to model the groundwater flow systems at Altnabreac. The results of these model predictions are being assessed relative to the information gained on the actual flow systems gathered during the detailed hydrogeological packer testing in the bore holes and also the spring surveys using the thermal infra-red linescan technique. The results will also be used to help interpret the hydrochemistry.

Two additional research sites were sought, one in the Loch Doon Granite in Strathclyde Region, Southern Scotland, and the other within the Cheviot Granite in Northumberland, Northern England.

Applications to drill in these areas were turned down by the local Planning Authorities and led to Public Inquiries. The Inquiry covering the Loch Doon application took place in February/March 1980, while the Inquiry covering the Cheviot application took place in October/November 1980. No decisions were forthcoming until December 1981 when the Government announced that following a review of the research programme into the long-term options for disposing of high level radio-active waste, the appeals for planning permission at these two sites were being dismissed.

2.2.1.2. UNDERGROUND EXPERIMENTAL ROOMS AND SHAFTS

Within this chapter are presented the projects concerning the

- underground experimental room excavated in the Boom clay layer beneath the Mol site in Belgium.
- the selection and adaptation of an existing mine in Italy for use as an underground test facility.
- the research going on in the Asse II salt mine in Germany.

2.2.1.2.1. Construction of an Underground Laboratory in the Boom-clay Layer beneath the Mol Site

Contractor: SCK/CEN, Mol (129-80-7 WAS. B)

The digging works for the shaft construction were continued and completed just before the end of the year. The final depth of the shaft is now 227 m. The outer concrete lining is foreseen to resist to the ground pressure (lithostatic pressure), its thickness is about 40 cm. At the -200 m level a circular stainless steel crown was placed in the concrete. It is the connecting point for the 3 mm thick polyethylene sheet which will be installed up to ground level in order to ensure the water tightness of the shaft. This sheet and the inner concrete lining, also about 40 cm thick, will be built up in 1982.

During the shaft digging works, using freezing technique, frozen soil samples were taken every two meters in the aquifers between ground surface and the top of the Boom clay layer. These samples were analysed for their granulometric composition and glauconite content. Interpretation and results are given graphically in Fig. 27.

Between -190 m and -227 m, at 5 different levels, various geotechnical testing devices and thermoprobes were placed into the clay in view of ground pressure and temperature measurements. A total of about 100 measurement points were regularly checked starting July 1981. Fig. 28 shows the oil ducts of a pressure cell going through a steel sleeve before pouring concrete. Just before starting the monolithic construction of the lower shaft and connecting room some cracks appeared in the concrete lining due to clay creep. As a consequence, it was decided to reduce the diameter of the connecting room from the originally planned 6 m to 4 m, enlargment being foreseen later on.

Experiments on heat transfer and corrosion in the underground gallery were further developped, allowing the final design of the cast iron lining. Fig. 29 shows the first complete ring of segments ready for testing in the factory. The study of the machine to dig holes of 70 cm in diameter into the plastic clay starting from the gallery at -220 m was undertaken and some preliminary tests performed in the clay quarry of Terhagen (in collaboration with N.V.SMET).

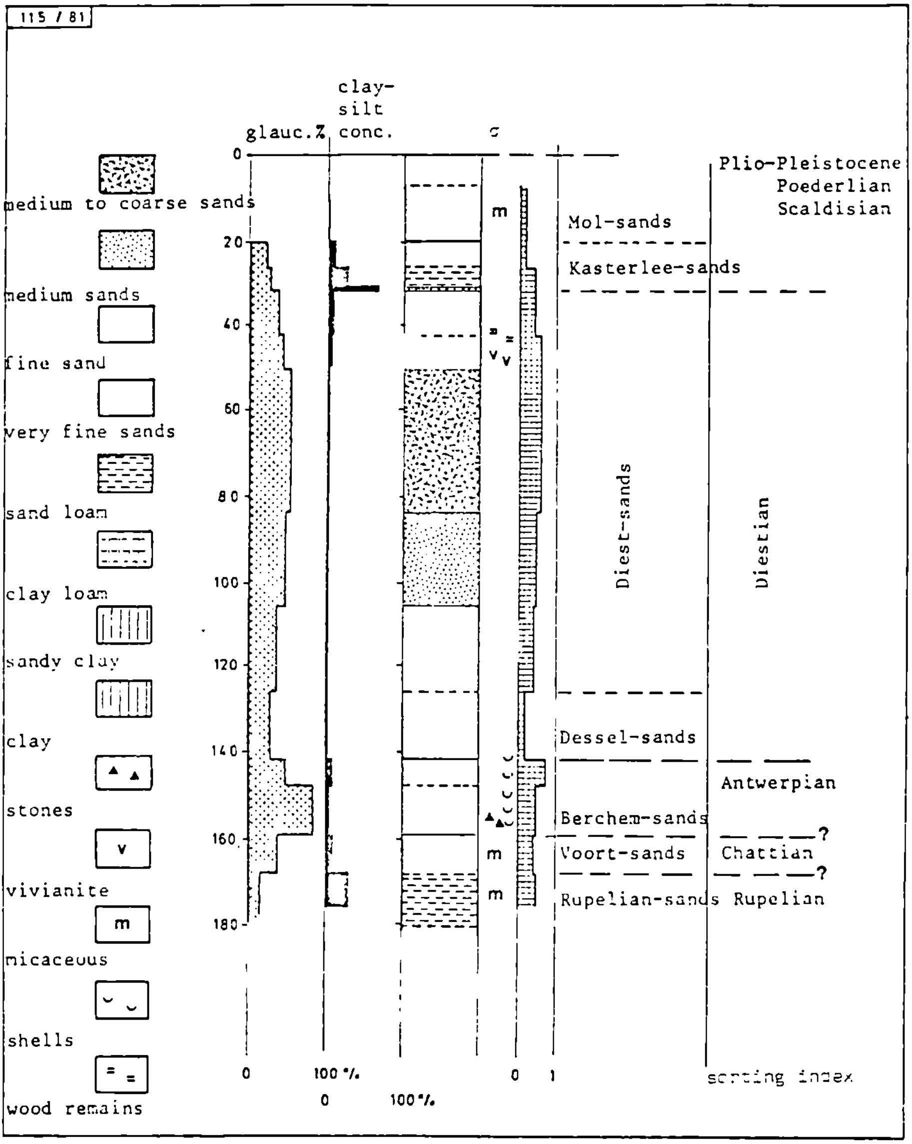

Fig. 27 : Lithostratigrafic sequence of the underground of the Mol site

Fig. 28 : Oil ducts of a pressure cell and sleave before concreting

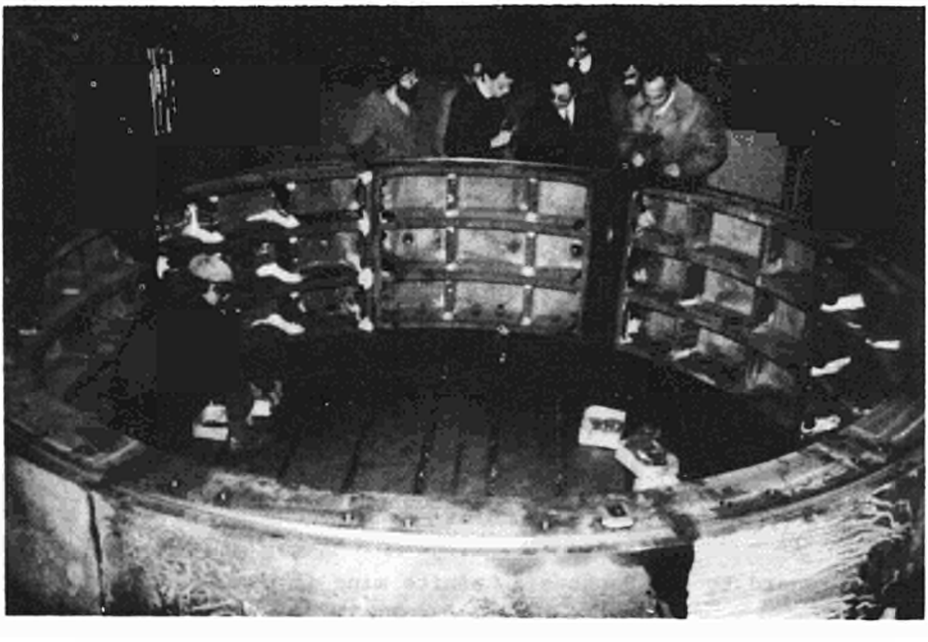

Fig. 29 : Cast iron ring of 8 segments for gallery lining

2.2.1.2.2. Use of an Underground Cavity as a Test Facility for Radioactive Waste Disposal in Clays

Contractor: CNEN, Casaccia (140-80-7 WAS. I)

The aim of this project is to select and adapt an already existing cavity into an underground test facility in order to study the modifications that clay formations can undergo due to the building up of a H.L.W. repository in these formations.

In order to select the most suitable site (mine, quarry, natural slope and civil engineering gallery) and to define the general geological and geotechnical features of the involved clay formation, the first step carried out has been a bibliographic and field research.

Until now, the following have been considered:

- Gypsum quarries (Cocconato Piemonte; - Vezzano - Emilia).

From a geological and mining point of view, both localities are similar: gypsum bodies included in a pliocenic formation mined by large galleries, through which big trucks can proceed. Little clay zones outcrop in the deeper levels (approx. 90 m.) of the quarries. Beginning from these zones, experimental cavities can be mined at a not too expensive cost.
The clays, close to the gypsum bodies, are probably affected by a local tectonic. The geotechnical analyses carried out on clay samples have shown a high content of carbonates.

- Openwork lignite mine (S.Barbara-Tuscany).

A clay stratum, about 100-120 m. thick, overlays the lignite levels.
The mining in progress is a great impediment to the excavation of the cavity.

- Kainite and rock-salt mines (Pasquasia, Milena, Realmonte-Sicily)

With regard to the Pasquasia kainite mine it can be expected that the pliocenic clay levels of more than 180 m will be reached in 1982. There are only clays with gypsum affected with water circulation in the kainite and rock salt Milena mine. Clays, for the most part with gypsum and not in evidence due to the covering of the walls of the galleries are present in the kainite and rock-salt Belmonte mine.

- Limestone quarries (Sacile-Friuli)

In this area the geology is characterized by a very pure cretaceous limestone in a tectonic contact with miocenic clays. The bibliography refers to clays which have been met when the stopin was carried out by galleries. It has not been possible to verify this, since the present openwork mining has destroyed the old galleries.
Clay, rich in sand, was found in the one and only small outcrop seen.

- Civil engineering galleries

Some galleries where it is easy to reach pliocenic clay formations have been visited, even though the thickness of the overlaying rocks is only suitable sometimes for this purpose. Bureaucratic and administrative difficulties are an impediment to the use of these galleries.

- Natural slopes (Lubriano, Bagnoregio, Calcata - Lazio; Orvieto, Botto - Umbria)

In all these localites the pliocenic clays are covered with tuff strata. It is possible to get a cavity, with about 150 m of overlaying rock, mining a gallery, about 150 m long, in Lubriano, which is the most suitable site.

- About 15 different geotechnical analyses have been carried out on clay samples.

2.2.1.2.3. Thermophysical In Situ Investigations on HLW Disposal in Rock Salt

Contractor: GSF, Clausthal Zellerfeld (130-80-7 WAS. D)

Special test galleries on the 750 m - and 775 m - level of the Asse II salt mine are equipped for in-situ tests on salt.

Temperature Testfield 4 (750 m -level)

The temperature testfield 4 (TTF4) is an 80 m long gallery, 6m wide and 6m high. The field is located in the centre of the Asse anticline structure in the so-called Older (Strassfurt-Halite Na2β(U).

The so-called 'Simulation Experiment in the Older Halite' started in January 1980, is a brine migration test using electrical heaters as a heat source for the simulation of HLW. Five cylindrical heaters, 1 m long each with a diameter of 0,2 m have been placed in a 15 m deep vertical borehole in the floor cf the test gallery in a depth of between 9,6 and 15 m. The total power output of those five heaters was 9000 Watt.

The main goal of this experiment is the investigation of the heat induced water liberation into the heated borehole. The upper third of the borehole has been sealed using a cemented standpipe with a flange mounted cap. The lead-in of cables for measurements and heater supply was made gastight.

The temperature distribution in the surroundings of the borehole is measured three-dimensionally in two fan-shaped arrays of temperature gauges.

Measuring Results

The measurement of the water liberation with the specially developed cool trap device was already started in the underheated borehole immediately after it was closed on November 6, 1979. A certain water inflow ranging about 0,8 g/day was observed during the first two months. This water inflow is considered to be due to small traces of brine included in some small anhydrite layers which are intersecting the borehole at a depth of 5,6 m and 7,9 m, marked by salt efflorescences.

When the heaters were turned on in January 1980, a sharp acceleration of water inflow was observed (Fig. 30).During the first twenty days an inflow rate of 40 to 50 g/day was measured which then decreased continuously until standard day 50 and then remained nearly constant with 8 g/day until standard day 148. Between standard day 160 and 180 an intermediate higher rate of 13 to 14 g/day occured. This is believed to be due to a higher permeability of the salt formation as a result of a certain microfracturing caused by the failure of heaters 2/3 on standard day 148 (40% of total power input) and a subsequent cooling and contraction of the salt. After standard day 180 the inflow rate stabilized at 2 to 3 g/day. This is certainly an effect of the now lower power input and reduced temperatures within the salt formation.

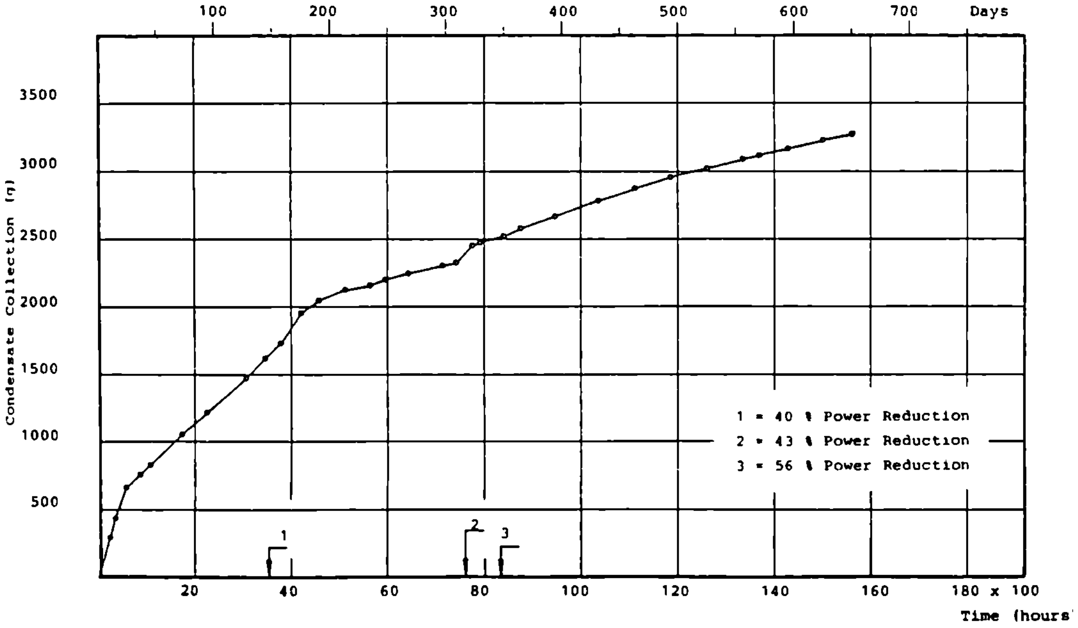

Fig. 30 : Water Liberation Curve in Borehole 2 of TTF 4

On standard day 315 and 346 in 1981 two further power reductions occured, probably due to corrosion of the heaters. After standard day 315 the inflow rate increased to 5-6 g/day and stabilized after standard day 346 with 3-4 g/day. As a long time effect the inflow rate decreased due to the decreasing temperature. On standard day 650 a rate of 1.8 g/day was measured.

The maximum temperature of 280°C was registered on the borehole wall in the midplane of the heated zone. The corresponding temperature gradient ranged between 8.5°C/cm at the borehole wall and 6.5° C/cm at a distance of 0.2 m from the borehole wall. The heating test was shut down on Nov. 24th 1981 as no further information was to be expected.

Temperature Testfield 5 (775 m-level)

The temperature test which will be performed in this testfield is quite similar to the described test in the temperature testfield 4. The main difference exists in the heating procedure. Instead of using a heater with constant power, the heater power in this case will be increased stepwise in order to evaluate the temperature dependent beginning of the release of hydration water of minerals like polyhalite and kieserite.

In 1981 the main objective was to install all technical components and measuring devices. It is planned to start the experiment early in 1982.

2.2.1.2.4. Liberation and Diffusion of Water and Gas Components within the Rock Salt

Contractor: GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

Release and Migration of Water in Rock Salt

The determination of the water content of the rock salt has shown that most of this water belongs to the hydrated minerals polyhalite and kieserite. This water will be liberated into the intergranular spaces by the elevated temperature of the disposed high-level waste and it may migrate at crystal boundaries and through microfissures as a result of total pressure partial pressure, concentration and temperature gradient.

A mathematical model of this transport phenomena has been developed in order to determine the amount of water which will be released into the disposal borehole. This model is based on the consideration of a porous medium, in which the pores and intergranular spaces are filled with the two components air and water in a gaseous phase.

The different transport phenomena have been investigated in the laboratory. The parameters which are necessary to calculate the amount of water being liberated into the borehole have been determined by means of rock salt samples from different depths and different stratigraphic layers. The results are

- permeability within the range of less than 0,05 to 120 µD
- porosity within the range of 0,01 to 0,74 vol.%
- diffusivity within the range of $0,1.10^{-4}$ to $2,1.10^{-4} cm^2/sec$
- diffusivity of temperature and partial pressure gradient ($k_p - k_T$) within the range of -5 to -225.

The diffusivity depends on the temperature and water concentration within the pores. At the same partial pressures of the water within the pores it decreases with increasing temperature.

The permeability which has been determined according to the Darcy law for compressible media, shows that it depends on the triaxial pressure surrounding the sample. It decreases by the factor of 30 to 100 if the pressure increases from 20 to 140 bar.

Gases in Rock Salt

The thermogravimetric investigation has shown that besides the water the following gascomponents are in the rock salt.

- H_2S within the range of 0 to 5 ppm
- CO_2 within the range of 0 to 200 ppm

- Thermally generated HCl by decomposition of rare and trace minerals within the range of 0 to 150 ppm.
- Gaseous hydrocarbons within the range of 0 to 60 ppm.

The relationship between these different gas components and the mineral composition as well as the generation- and liberation temperature has not yet been investigated.

2.2.1.2.5. Investigation of the Processes During Flooding with Water and Brine of a Repository for High-level Wastes in a Salt Dome

Contractor: GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

The effects of natural groundwaters which may intrude into a high active waste repository were further investigated. In 1981 the investigations concentrated on field measurements within flooded shafts and laboratory studies of the transition of ions from highly saturated solutions into less concentrated phases as well as solubility measurements of Am-241 within different solutions.

Field Measurements in Flooded Shafts

These field measurements are conducted in order to understand the processes occurring in flooded mines. The chemical reactions depend upon the temperatures, the minerals and the present solutions. This system is not closed and there is no thermal nor mechanical equilibrium within the liquid and solid phases. A chemical equilibrium is achieved only locally.

Temperature, conductivity- and flow logs and water samples were measured from eight different shafts. The liquid-liquid boundaries move in vertical directions with a velocity of 0,5 to 3 cm per day. It could also be demonstrated that the composition of the solutions clearly depends upon the adjacent minerals, e.g. carnallite, sylvite, polyhalite or kieserite.

Laboratory Experiments

Several experiments with glass laboratory scale models were set up and performed. These efforts concentrated on experiments which tried to simulate certain aspects of transport phenomena within brines. A cylindrical tube of 15 cm diameter and 225 cm length was filled with $MgCl_2$-saturated solution at the bottom and NaCl saturated solution at the top.

The lower part was kept at 30°C by thermostats, the upper part at 24°C, thus simulating by simple means the conditions within a flooded shaft. The concentration of Mg^{++}-ions in the upper layer was observed. The lower layer was additionally spiked with Zn^{2+} to a concentration of 10 mg/l. Zinc was chosen because of its excellent solubility and detectability with a hanging mercury drop electrode (HMDE).

The increase in Mg^{++} and Zn^{++} was observed for more than 8000 hours. After a steep increase at the beginning, the concentration versus time flattens. It looks like the speed controlling step in this system is the diffusion via the sharp boundary layer between the $MgCl_2$- and NaCl-solutions.

In-Situ Tests in the Asse Mine

The field test plan for the 1st flooded tunnel experiment as well as a detailed geological mapping of the test field and mineralogical investigations of the different stratigraphical horizons occurring within the field is finished. Insertion of a heat source of 2 m length and 38 cm diameter 3m below the tunnel floor is foreseen. The assembly will be backfilled by a salt-concrete-slurry. At a number of points in the rock salt, at the surface and in the brine temperature, concentrations, flow and dissolution will be monitored. Direct access to the heated area will be possible.

The main aim of these field tests will be the investigation of material release out of the heated borehole, the development of suitable borehole seals and the comparison of predicted and measured flow and leach rates.

2.2.1.2.6. Measurement of Deformations in Underground Galleries in the Asse Salt Mine

Contractor: GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

Parallel to the heating experiments carried out in the Asse salt mine (cf. 2.2.1.2.3.) extensometer and convergence measurements are performed in order to understand the petromechanical behaviour of the surrounding rock mass. The measuring data are used for a comparison with calculated data in order to estimate the validity of the calculation models.

Measurement of Deformations in the Gallery of Temperature Testfield 4 (TTF 4)

The measurement of the horizontal room convergence (cf. Fig. 31 AB) was started immediately (3 days) after the excavation of this part of the gallery and it was found that linear creep behaviour of about $3,2.10^{-2}$mm/day was achieved within 200 days. After the heater (9000 Watt total power input) in borehole 2 was turned on in January 1980, the horizontal convergence rate changed to 6.10^{-2} mm/day. After measuring day 800 the horizontal convergence rate then decreased, which obviously is considered to be due to the failure of heater 2/3 which was 40% of the total power.

After measuring day 900 the convergence rate is again $3,2.10^{-2}$ mm/day and is still decreasing. After measuring day 1100 it is approximately $2,9.10^{-2}$ mm/day which is a lower value compared to the linear creep period before the start up of heating. It can be assumed that the decreasing convergence behaviour is the result of stress relaxation induced by creep.

Measurement of Borehole Convergence

Also, the convergence in the nonheated borehole 3 of TTF 4 which had an initial diameter of 350 mm and has been drilled parallel to borehole 2 at a distance of 20 m was measured. One of the measuring levels was nearly at the same borehole depth (12.8m) as in borehole 2. A slight increase of the convergence rate from

0.15 . 10^{-2}mm/day to 0.17 . 10^{-2}mm/day was observed after the heater in borehole 2 was turned on. 160 days after start up of heating, the rate is again 0.15 . 10^{-2}mm/day. The rate is still decreasing and is now 0,13 . 10^{-2}mm/day 633 days after the start up of heating in borehole 2.

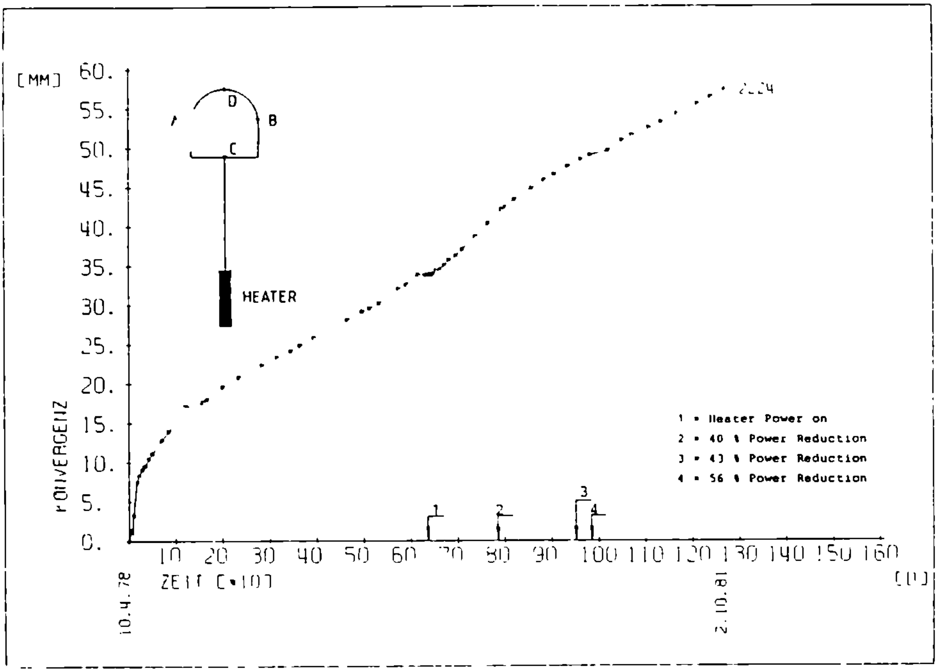

Fig. 31 : Horizontal Room Convergence $\overline{AB}$ - TTF 4

2.2.1.2.7. Determination of Absolute Stress in Rock salt under Ambient and Higher Temperature Conditions

Contractor: GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

In 1981 in the Asse salt mine two methods of stress measurements were used to render information on their applicability in deep salt structures, first the hydraulic fracturing technique and second the method of hard inclusions (monitoring station). The first one basically gives more information on the state of stress in geologic structures, whereas the latter is used for measuring the change of stresses versus time.

The results of the hydraulic fracturing tests which were carried out in the secondary stress field of the mine on the 800m-level show that the state of stress is not lithostatic, and that there might be a rotation of the axis of the main principal stress from the vertical to the horizontal.

The same tendency show global computer calculations for the Asse salt mine. The first results of the frac-tests are summarized in Fig. 32.

The monitoring station consists of flat hydraulic pressure cells cemented in a borehole with a suitable material. The material in total length of the borehole is reloaded in a range which is equivalent to the state of stress in the solid rock.

Laboratory investigations for the borehole replenishing material and the installation of one measuring system in a borehole were also conducted this year.

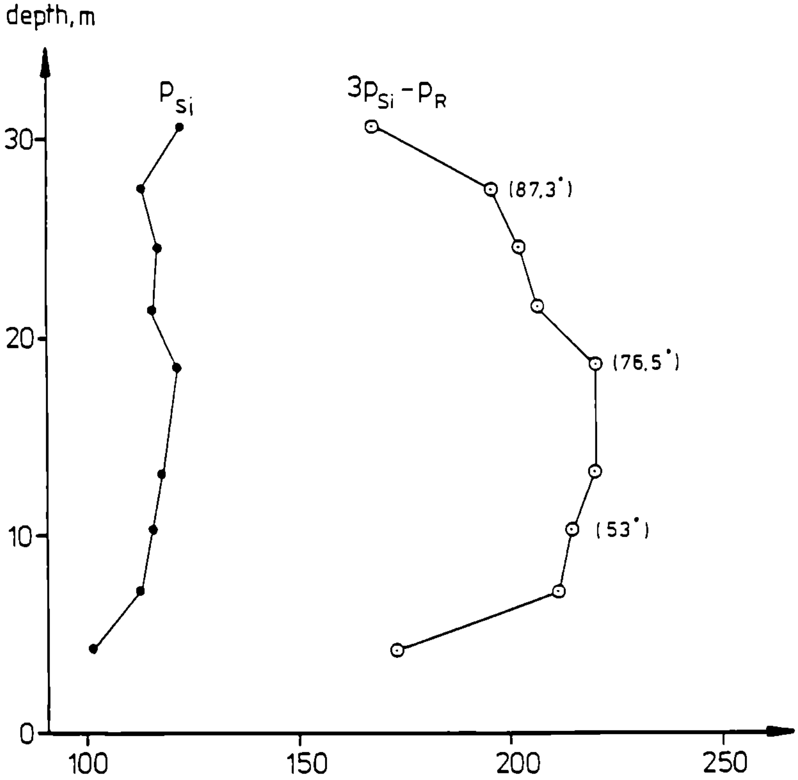

Fig. 32 : P_{si} (mean principal stress) and 3 P_{si}-P_R (maximum principal stress) as a function of the borehole depth in FB1 - 800 m-level. The angle gives the orientation of the pressure-induced fracture from the vertical direction

2.2.1.2.8. Laboratory Investigations of Rheological Salt Rock Properties under Various Conditions including Elevated Temperatures and Radiation

Contractor: GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

The laboratory experiments for the determination of mechanical and creep properties of Asse salt rocks are carried out together with the BGR, Hannover.
The test programme in both test facilities contains a series of uniaxial and triaxial experiments in order to furnish mechanical data on the stress-strain behaviour, failure and creep response of different salt rocks under various conditions, which include especially elevated temperatures.

Uniaxial Experiments

After a test period of 485 days a series of 4 uniaxial creep tests with cylindrical samples of Asse-halite from the temperature test field 4 at 300K and 15,3 MPa uniaxial stress have been finished. The primary creep rate $\dot{\varepsilon}_p$ in these experiments deviates by approximately -450% from those values determined with the BGR creep law; the secondary creep rate $\dot{\varepsilon}_s$ deviates in the order of +900%.

In 25 stress controlled uniaxial compression tests with Younger Halite Na3 and a stress rate σ = 1 MPa . min^{-1} a linear decrease of the uniaxial strength by 30% and a progressive increase of failure strain by approximately 600% in the temperature range of 293 to 473 K has been observed.

In addition to the Brazilian tests from 1980 further indirect tensile tests were carried out with Na2 and Na3 (25 bending tests and 10 torsional tests). The bending tensile strength exceeds the uniaxial tensile strength by the factor 3; the tensile strength derived from the torsional test coincides rather well with the uniaxial one (deviation < 5%).

Uniaxial creep tests of BGR at room temperature with Asse rock salt over a period of 3,7 years at present show, that the long term value for the corresponding values of creep rates remains uniform. Nevertheless these creep rates are slightly lower compared with those creep rate data, derived from experiments with a test duration between 30 and 90 days.

The temperature control units of the BGR uniaxial creep test facilities for experiments at elevated temperatures could be improved progressively. Corresponding creep tests at incremental increased and decreased temperature levels are underway. As a preliminary result this test mode yields slightly larger values for the effective activation energy compared with former creep tests at constant temperature levels.

With regard to creep failure a generalized creep failure criterium could be derived in a first approximation.

Triaxial Experiments

As well strain controlled as stress controlled multistep triaxial experiments of BGR with stepwise changes of strain rate or stress rate conditions yielded first data on the different response of rock salt depending on the direction of loading or unloading.

Additional stress controlled triaxial BGR experiments with cubes of Na3 at elevated temperatures up to 423 K showed that the influence of the stress path and of the hydrostatic stress level on the cube strength vanishes at elevated temperatures.

Strain controlled triaxial experiments carried out at GSF with Asse rock salt investigated the triaxial stress-strain behaviour of right circular cylinders at confining pressures up to 20 MPa (with a priority at 10 and 15 MPa) and constant axial strain rates ranging from 10^{-4} to $10^{-2}\mathrm{min}^{-1}$. First of all tests were restricted to room temperature conditions. The results of these experiments carried out up to now show that the octahedral shear strength increases with increasing confining pressure but decreases with decreasing axial strain rate.

During the year these experiments have been extended in a first step to elevated temperatures up to 423 K. Preliminary results from experiments at a confining pressure $\dot{\sigma}_3 = 10$ MPa and a constant axial strain rate $\varepsilon_{10} = 4 - 5,5 \; . \; 10^4 \text{min}^{-1}$ show that the true differential stress $\sigma_1 - \sigma_3$ at constant natural axial strain levels seems to decrease exponentially with increasing temperature. The strength reduction at 423 K is in the order of 60% compared with room temperature conditions. This is connected with large natural axial strains up to more than 50% and a corresponding intensive radial barreling of the test sample.

2.2.1.2.9. Geophysical Investigation and Monitoring of Rock Salt Behaviour in the Vicinity of Underground Heating Experiments in the Asse Salt Mine

Contractor: GSF, Clausthal-Zellerfeld (130 80 7 WAS.D)

Due to the heating of rock, changes in its mechanical properties and in the stress distribution are to be expected. Eventually sudden stress releases and the formation of cracks may occur. It shall be investigated, to what extent such changes and processes can be monitored by non-destructive seismic methods. As changes in the mechanical properties will influence the propagation velocity of compression (P) and shear (S) waves, these velocities will be determined periodically between certain fixed points in the vicinity of planned heating experiments.

In order to detect and locate sudden stress releases a passive microseismic network has been installed.

Microseismic Activity

The microseismic network, consisting of 7 geophones distributed within the Asse mine, is operating since May 1980. The accuracy of source location could be improved by the registration of a number of small blasts at known locations. By this method an unknown source may be located by computing the radius-vector from a nearby known blast, thereby taking into account large-scale inhomogeneities and anisotropy.

An important accomplishment is the possibility to discriminate roof-fall in most cases. These frequently occuring events (about 5 per month) can be identified by its forerunners, which arrive 1.7 s (the falling time in most rooms) before the main event.

Seismic Transmission Measurements

In the vicinity of the hexagon-experiment as well as of the test site V, attachments for the transducers and provisions for the source (hammer-blow on a rod in a borehole) have been prepared at fixed positions. So transmission experiments can be repeated on identical paths.

The first ones of these experiments have been performed. Propagation times may be read directly form the scope, but the signals can also be recorded on tape for further processing in the laboratory. Fig. 33 shows an example.

Measured P-velocities range from 4540 to 4670 m/s parallel to the strike and from 4270 to 4380 m/s perpendicular to the strike (rock salt). The S-velocities are 2500 to 2620 m/s and 2450 to 2620 m/s, respectively. These values correspond to dynamic Poisson ratios of .24 to .27.

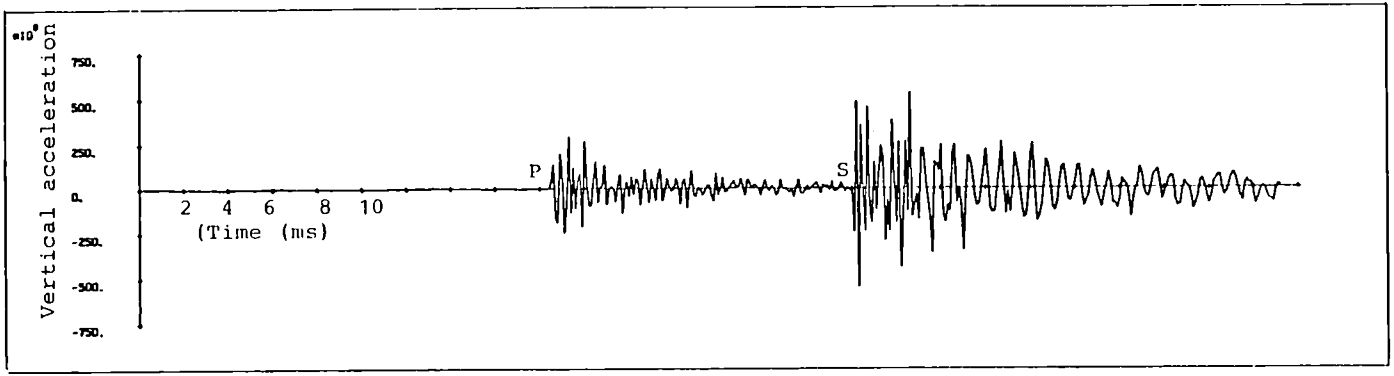

Fig.33 : Example of a seismic transmission registration (Vertical component). Eightfold stack. Source-receiver distance : 80 m. Compressional and shear wave onsets have been marked by P and S respectively.

2.2.1.2.10. Mineralogical Investigation of Thermometamorphic Processes in Salt

Contractor: GSF, Clausthal- Zellerfeld (130 80 7 WAS.D)

A mineralogical-petrographical survey of temperature testfield V has been performed for a documentation of the present state of the salts before heating up.

As it always takes a long time to obtain thermally altered salt samples from in-situ temperature tests, the conceptual design and construction of a special laboratory furnace for the simulation of thermometamorphic processes of smaller cores were made.

Mineralogical-Petrographical Investigation of Temperature Test-Field V (TTF V)

The geological mapping of the 775 m-level by DIEM (1981) revealed the stratigraphical position of TTF V within the socalled 'Polyhalitbänkchensalz' (polyhalite bedded salt) which is in normal stratigraphical contact with the 'Kieseritisches Über gangssalz' (kieseritic transition salt). but outside the zone to be heated up.

The microscopic study of samples showed a fine to mediumgrained halite including irregular layers of a halite polyhalite-anhydrite-mixture. The polyhalite-content of the halite layers ranges between 0,2 and 2,6 (max. 10,0) wt-%. That of the polyhalite-halite layers between 3 and 65 wt-% besides an anhydrite content up to 50%. The more polyhalite is present, the more fine-grained and stronger the rock salt becomes. Within the slightly polyhalitic rock salt layers anhydrite could not be detected until now.

Polyhalite streaks occur up to 1 cm thickness only in special folding areas. Irregular spots and nodiform inclusions in the rock salt could be recognized under the microscope as a very fine-grained felt of polyhalite also. However, the normal occurence of that mineral in the 'bedding salt' is represented by xeno to idiomorphic crystal grains after showing the characteristic polyhalite twinning. Grain size ranges from a few to 5 mm. Practically the same applies to anhydrite which can sometimes be corroded by polyhalite and halite.

Halite as main mineral averages between 94 wt-%.
The texture in the rock salt was also investigated and described. The average constitution of the area revealed by slit samples was: 93,7% halite, 4,3% polyhalite 2,0% anhydrite. Herewith the main characteristics of that test area are known and will have to undergo a thermometamorphic alteration.

Concept for a Laboratory Test Facility

As the performance of in-situ tests and the thermal alteration of salt connected herewith in general takes a lot of time and the marginal conditions can only be changed slightly in the frame of large-scale tests, the concept for a laboratory furnace was developed and layed out for sample diameters of 245 mm, height of 400 mm, and a working-pressure of maximally 10 bar. The central heater borehole has d = 30 mm, the electrical capacity will be 750 W. The stainless-steel tube of that furnace will be surrounded by a second heating element to ensure the setting of different temperature gradients.

This furnace is now under construction and will be available for installation at the beginning of 1982.

2.2.1.2.11. In-Situ Investigation in the Asse Salt Mine of the Stability of Borehole Casings and Development and Testing of a Standard Convergence Probe

Contractor: KfK, Karlsruhe (266-80-7 WAS.D)

In-Situ Investigation of the Stability of Borehole casings

To determine the pressure load on a borehole casing caused by heat induced borehole convergence, a pipe section was constructed and instrumented with two types of strain gauges.

The test was started in May 1980. The pipe section was heated with a power of 1.5 kW (corresponding to a maximum temperature at the pipe surface of 190°C). This is roughly the maximum salt temperature of about 200°C as defined in the previous concept for the German HLW repository.

Parallel laboratory tests were performed in a high pressure test rig in order to evaluate the drift behaviour of the strain gauges.

In the course of an in-situ experiment in the Asse salt mine it was found in case of a 1.5 kW power supply that an unexpected low convergence of the rock salt occurs which is insufficient for measurements. For this reason, the power was increased to a maximum value of 3.6 kW (maximum temperature at the pipe surface about 310°C). The salt entered into contact with the pipework after about 320 days of measurement and hence allowed measurement of the pipe deformation.

For evaluation of the experiment three points of measurement were used which are located at the hottest points of the pipe section. The average of the three measured values is a pressure of 360 ± 50 bar.

In-Situ Application of the Standard Probe

The probe was used in three in-situ tests performed at the 775 m level in the Asse salt mine. The result of the first test can be seen in Fig. 34. The average borehole diameter before the beginning of the test was about 186 mm, the depth of the borehole 4.90 m. The borehole was heated with a constant power of 1.2 kW over a period of 104 days, except for failures of the power supply system of the mine. The change of the borehole diameter as a function of time is represented for the five points of measurement. The electrodes 1 and 5 exhibit nearly no rise in convergence. By contrast, a marked rise in convergence at the beginning of heating (up to 0.8 mm/day) can be observed at the electrodes 3 and 4, corresponding to the higher temperature.

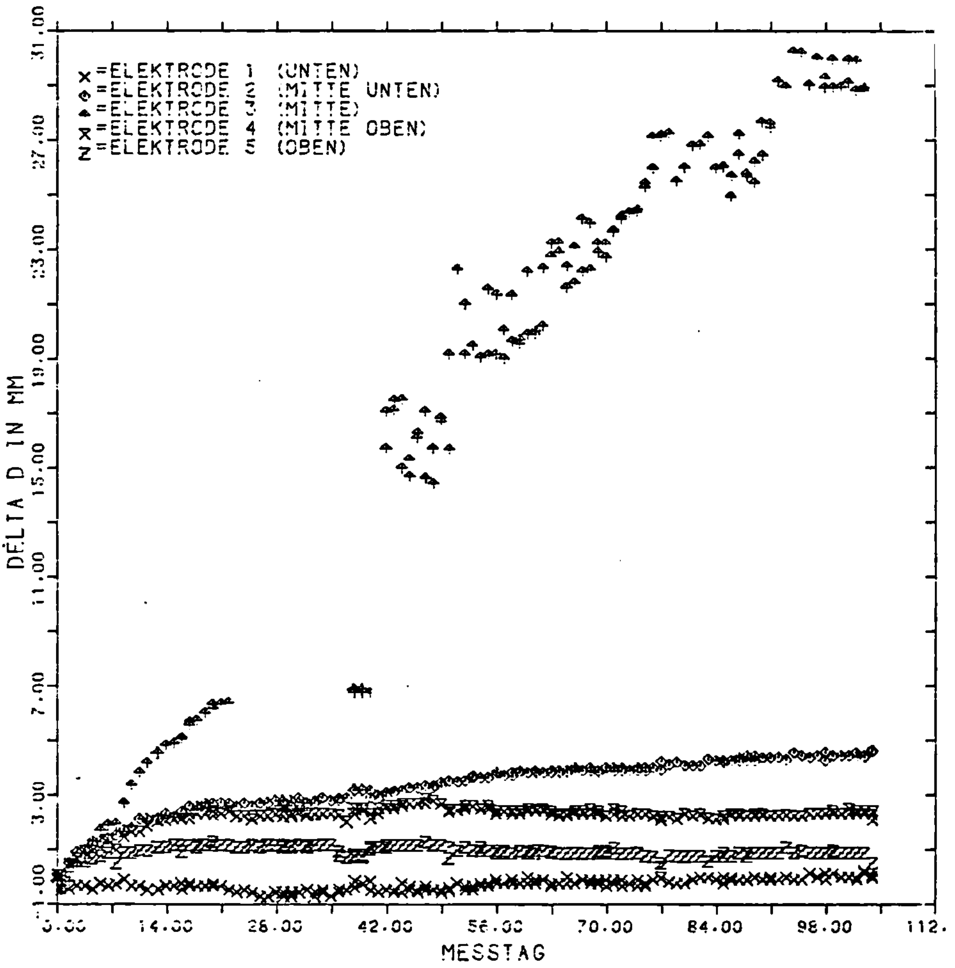

Fig. 34: Change of borehole diameter as a function of time.

This rise falls to a value of 0.01 mm/day after about 20 days of measurement. The values measured at the electrode 2 were compared with the values determined by the ADINA computer code.

With the material parameters and the boundary conditions chosen a good agreement of the results was found.

An explanation for the measured values at the electrode 3 was found after the test had been terminated. By efflorescende in the borehole caused by brine the external electrode was pushed inwards at the overlapping ends which means that no convergence of the salt was measured but the growth of brine efflorescence.

2.2.1.2.12. Convergence and Pressure Measurements in the Dry-drilled 300 m Deep Borehole in the Asse Salt Mine

Contractor: ECN, Petten (142 80-7 WAS. NL)

The convergence measurements in the dry-drilled hole in the ASSE mine, that started end 1979, have been continued in 1981. The measurements were executed at a depth of 292 m of the 300 m deep hole which corresponds with a depth of 1042 m from the surface level. At the end of 1981 measuring data of 560 days have been collected, the results are shown on Fig. 35.

A creep law for this curve on the basis of a measuring period of 250 days has been derived. An extrapolation of the derived creep law up to 560 days shows that the law still fits the curve very well.Mid 1981 one of the 6 measuring clocks has been removed due to mal functioning (corrosion). These measurements will be concluded in the spring of 1982.

In order to investigate the change of diameter in the time over the length of the hole, an auxiliary probe has been constructed. The investigation of the accuracy of the measurements is still going on.

With regard to measurements t be performed in heated areas in

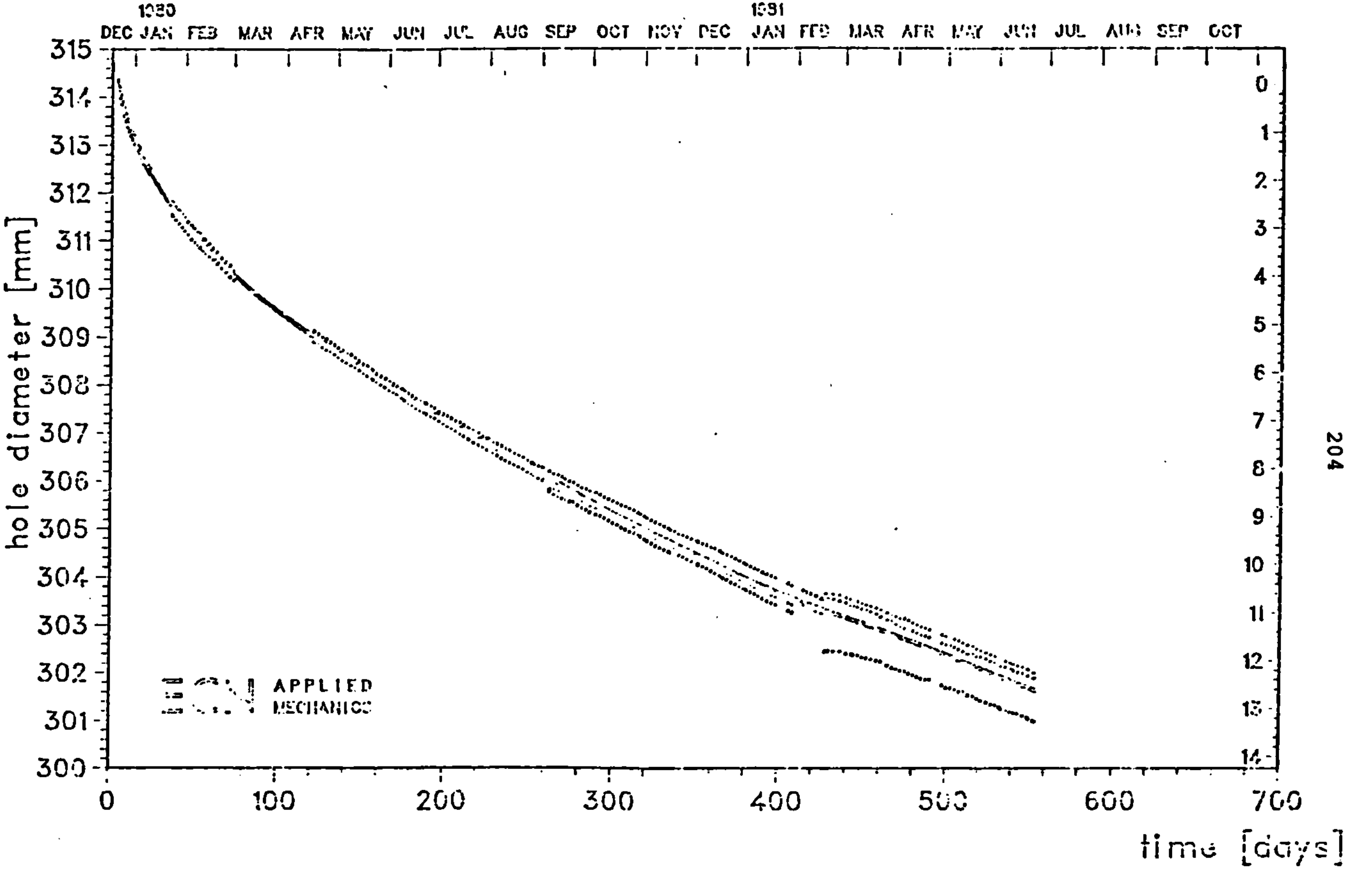

Fig. 35 : Convergence ECN-hole at 292 m below 750 m floor.

the bore-hole the main parts of the equipment have been delivered. The testing phase of parts of the measuring system has started end 1981, also the software for the data collecting and control system is partly finished. This also applies for the manufacturing and assembly of the Heated Pressure Probe.

2.2.1.2.13. Laboratory and in-situ Measurements of Cataclastic Effects in Rock Salt

Contractor: ECN, Petten (142-80-7 WAS.NL)

The work is to determine the influence of the presence of a vertical bore hole of 30 cm diameter on the structure of the surrounding rock salt at about 1000 m depth. The main point will be to find out the size of an eventually cataclastically effected zone, i.e. a zone containing micro and/or macro cracks. The next problem will be to follow the cataclastic events during a period of heating the inside of the borehole.

Structural changes by cataclasis can be discovered by means of the acoustic traveltime.

The underground measuring device

The investigations on cataclastic structural changes have been planned to be done by means of the "acoustic measuring tube". The first experiment with only one tube (the one-tube-method) has been carried out in the laboratory. To that purpose a concrete model of 600x600x2400 mm has been designed and constructed. However, the one tube method failed. In order to reach the aimed purpose the two-tube-method was developed. In the laboratory this method proved to be successful. It will be tested underground next.

The device consists of two rubber acoustic measuring tubes, 2000 mm length, Ø 66 mm outside; each tube provided with seven metal boxes to contain accellerometers i.e. pulse generating and receiving transmitters.

In order to receive and to hold the traveltime graphs and to read them, an advanced technique of sampling and averaging has been chosen. A transient Recorder VK 220-4 of the latest design has been tested in the laboratory. This apparatus will be used in connection with an x-y recorder.

2.2.1.3. ENGINEERED BARRIERS

Under this heading are classified the contracts concerning:

- selection and testing of suitable materials for high level waste containers and for repository structures.
- studies and research on backfilling and sealing of disposal holes, galleries and shafts.

WASTE CANISTERS AND STRUCTURAL MATERIAL

The objective of this project is to identify container materials with sufficient resistance to general and localised corrosion in order to isolate high level nuclear waste from the disposal environment for 500 to 1000 years.

Laboratory and field experiments have been undertaken in various Member States on a rather large number of materials under different test conditions.

During the year 1981 a critical review study was performed (see 2.2.1.3.1.) in order to identify a coordinated programme on a small number of materials. Such a programme, to be started in 1982, was defined and will be focussed on three materials.

- Hastelloy C 4
- Ti-Pd (0,2 % Pd)
- Mild Steel

2.2.1.3.1. Review Study on Research Projects concerning the Selection of Materials for Waste Containers

Contractor: University of Clermont-Ferrand (236-81-7 WAS.F)

During 1981 a critical review study was performed on the work, undertaken by various laboratories in the Member States in the framework of the CEC research programme, aiming at the selection of suitable materials for waste containers and for repository structural components.

A methodology was defined which should permit within a reasonable time to ascertain whether the candidate materials can withstand to the corrosion conditions imposed on them during the specified service time.

Finally a coordinated programme was identified in which all the participants (i.e. SCK/CEN, CEA, CNRS, KfK, UKAEA, JRC-Ispra) could participate according to their specific abilities.

2.2.1.3.2. Corrosion Studies on Candidate Canister and Borehole Casing Materials in a Clay Environment

Contractor: SCK-CEN Mol (144-80-7 WAS. B)

The corrosion resistance of candidate canister and borehole casing materials are being evaluated by means of corrosion tests carried out in direct contact with clay, in humid clay atmospheres, in interstitial clay water and Antwerpian ground water.

Corrosion rates in direct contact with clay have been obtained after "in situ" corrosion experiments carried out in an open clay pit at Terhagen at 13°C and 150°C. The results are in agreement with experimental data obtained from corrosion experiments carried out in laboratory in thick-walled cylinders.
Unprotected ductile iron (grade 60) corrodes in direct contact with clay at a mean rate of 6.10^{-6}m.a^{-1}. Localized attack proceeds along the graphite nodules - ferrite matrix interface and areas with a pearlitic structure.
Corrosion rates of cast iron have also been derived from post-

corrosion analysis of underground structures of cast iron in operation for 60 to 90 years under similar conditions.

The corrosion layer is in the two cases build up by an outer and an inner reaction layer and a zone characteristic for internal oxidation. The different sublayers and transition zones have been characterized by metallographic and X-ray analytical techniques.

The archeological objects of cast iron corrode in a similar way as the specimens during short time exposed in direct contact with clay.
Different anti-corrosive coatings (metallic and non metallic) have been applied on grade 60 ductile iron and tested in direct contact with clay at 13°C, in a humid clay atmosphere at 50°C and in a gas corrosion chamber at 25°C. Most of the anti-corrosive coatings based on paintings and plastics give more promising results than the galvanization treatment. The application of a duplex system based on the superposition of a metallic (Zn) and a non-metallic anti-corrosive layer presents the best anti-corrosive protection against blistering phenomena starting from artificial defects.

Stressed specimens (bent beams, U bends, small size canisters) of selected candidate canister material (AISI 304, 304L, 316, UHB 904 L, 1803 MoT, Inconel 825, Hastelloy C, IMI 115, commercial pure Titanium) have been exposed in the as-received and heat treated condition in the humid clay atmosphere at 50°C, Antwerpian ground and interstitial clay water at 98°C. Crack initiation has been found in an heat treated AISI 304 U-bend sample after 4500 h of exposure tested in humid clay atmosphere and in AISI 316 tested in the as-received condition in Antwerpian ground water.

In the second case crack initiation has been found after 1,050 h of exposure. Most of the experiments lasted up to 20,000 h.

The corrosion rate and corrosion phenomena of candidate canister materials has been determined after corrosion testing in interstitial clay water at 25°C and 98°C and potentiodynamic measurements at 25°C, 40°C and 90°C.

Hastelloy C, Inconel 825 and IMI 115 possess the best corrosion resistance. Precipitates present in Hastelloy C and 1803 MoT are nucleation sites for preferential attack. The nickel rich alloys Ni 200 and Inconel 600 tested at 25°C suffer from a bulging attack. S, Mg and Si play at 98°C a predominant role in the corrosion attack.

Potentiodynamic corrosion testing resulted in the determination of the intensity of the passive current and the mechanism of corrosion attack for the various candidate canister materials.

The "in situ" corrosion tests carried out in an open system (in inert furnace placed in boreholes in a clay quarry at Terhagen, Boom) and the characterization of the environment at three different temperatures are continuing. CO_2, SO_2 and HCl are the main gaseous products escaping from the clay heated up to 13°C, 45°C and 62°C.

Long exposure times ($\approx$10,000 h) result in the complete disappearence of the sulphur and chloride bearing products form the humid atmosphere in equilibrium with clay.

Post corrosion analysis of candidate canister materials corroded "in situ" for 9 months at 13°C and 50°C revealed the predominant role of the relative humidity on the nucleation and growth of deposition and corrosion products. The highest concentration of these products is present on the speciments exposed at 13°C.

2.2.1.3.3. Corrosion Studies on Packaging Materials for High-Level Waste in a Saliferous Environment

Contractor: KfK, Karlsruhe (250-81-7 WAS.D)

Selective corrosion studies and mechanical tests were performed on the materials listed below:

- Cr-Ni-steels, materials Nos. 1.4306 and 1.4417
- nickel-base alloys Inconel 625, Incoloy 825 and Hastelloy C4
- Ti 99.8-Pd (material N°. 3.7025.10)
- chromium steel X1CrMoTi 182 (1.4591 ELA -ferrite)

According to accident considerations saline brines are being considered as a potential corrosion medium. The corrosion tests were made under selective conditions in a brine at T = 170° and p = 1 bar.

The brine had the following composition:

45 wt .% $MgCl_2$; 3.8 wt.% KCl; 3.4 wt.% $MgSO_4$; 1.1 wt.% NaCl; 47.1 wt.% H_2O.

The Cr-Ni-steels which are susceptible to local corrosion, especially stress corrosion, were considered in order to study the time-to-rupture under accident conditions of these materials intended for use in HLW canister design.

To evaluate the influence of welding and annealing treatment on the mechanical and corrosion behaviour of the materials, different material conditions were investigated.

The mechanical tests made on the Cr-Ni-steels and the nickel-base alloys have shown that these materials undergo only slight changes of their mechanical properties as a result of welding and annealing treatment (1100°C at the maximum), respectively.

The most important results on corrosion are summarized in Tables 1 and 2. The following statements can be derived from them:

- The Cr-Ni-steels 1.4306 and 1.4417 prove to be highly sensitive to any type of local corrosion attacks, especially stress corrosion cracking. Due to the short time to rupture these materials, as has been expected, cannot fulfill a barrier function in the repository.

- The materials Inconel 625, Incoloy 825 ELA-ferrite and Ti-Pd resisted stress corrosion cracking, but these materials are susceptible to damage by pitting corrosion and might lose their function as a barrier therefore.

- A high resistance to any type of local corrosion is exhibited by the material Hastelloy C4. It showed a uniform corrosion rate which was less than 30 µm/a after a test period of 432 days.

Final statements about the suitability of the material Hastelloy C4, positively evaluated here, and of other materials investigated so far (e.g., mild steel 1.0566) cannot be made before more detailed investigations will be completed. They include above all the study of the corrosion mechanism and the evaluation of the influence of gamma radiation on the corrosion behaviour of the materials. These studies are under way.

Table 1: Results of the Corrosion Studies after the Latest Sampling (quinary brine, 170°C 1 bar)

Material	Condition	Maximum test Duration (d)	Mass Loss Rates ($g.m^{-2}.d^{-1}$)	Pitting Corrosion	Crevice Corrosion
1.4306	A+) W++)	246	0.08 0.26	x x	x x
1.4417	A W	243	0.97 1.61	x x	- -
2.4856 (Inconel 625)	A W	248	0.23 0.16	x x	x x
2.4858 (Incoloy 825)	A W	235	0.19 0.23	x x	x x
2.4610 (Hastelloy C4)	A W	244	0.87 0.39	- -	- -
1.4591 (ELA ferrite)	A	135	2.7	x	x
3.7025.10 (Ti-Pd)	A	130	0.3	x	-

+) as-delivered; ++) in the annealed condition

x = corrosion

\- = no corrosion

Table 2: Results of Stress Corrosion Investigations of U-size Specimens in a Quinary Brine at T = 170°C and p = 1 bar

Material N°	Material Condition	Time-to-rupture (d)		
		Specimen 1	Specimen 2	Specimen 3
1.4306	S	< 1	< 1	< 1
	SW	< 1	< 1	< 1
	WS	< 1	< 1	< 1
1.4417	S	< 1	< 1	< 1
	SW	broken while being bent		
	WS	< 1	< 1	< 1
2.4856 (Inconel 625)	S	> 248	> 248	> 248
	SW	> 248	> 248	> 248
	WS	> 248	> 248	> 248
2.4858 (Incoloy 825)	S	> 235	> 235	> 235
	SW	> 235	> 235	> 235
	WS	> 235	> 235	> 235
2.4610 (Hastelloy C4)	S	> 432	> 432	> 432
	SW	> 432	> 432	> 432
	WS	> 432	> 432	> 432
1.4539 (ELA ferrite)	A	> 135	> 135	> 135
	S	> 135	> 135	> 135
3.7025 (Ti-Pd)	A	> 130	> 130	> 130
	S	> 130	> 130	> 130

S = welded; SW = welded and annealed

WS = annealed and welded

A = as-delivered

2.2.1.3.4. Corrosion Studies on Candidate Materials for Waste Containment in Crystalline Rock

Contractor: UKAEA, Harwell (248-81-7 WAS. UK)

Laboratory experiments

A survey of existing data identified a number of metals and alloys which appeared to have potential as corrosion resistant containment materials. Consequently the first phase of the laboratory programme consisted of a series of accelerated sorting tests designed to identify the best of these. The Ti-0.2% Pd alloy proved most resistant to both general and localised attack with a general dissolution rate equivalent to a metal loss of about 1.0 mm in 1000 years. Commercial purity titanium was equally resistant up to 90°C, but suffered severe crevice corrosion at 150°C.

Similar sorting experiments have been conducted with less corrosion resistance materials appropriate for corrosion allowance containers (eg copper, lead, cast iron). These have shown that the rate of general dissolution is determined by the rate of transport of oxygen. In geologic disposal media, where oxygen transport is limited by diffusion or slow water movement general metal loss should not exceed about 20 mm in 1000 years.

Field Experiment

This experiment is being mounted in a bore-hole adjacent to the heater being used in the heat transfer experiment in progress at the UKAEA's test site at Troon, Cornwall(Fig.36).By taking advantage of this facility it has been possible to test specimens in a granitic groundwater environment at temperatures between 65 and 90°C. The experiment has now been underway for about 17000 h.

Current results have underlined the low corrosivity of groundwater in comparison to the solutions used in the laboratory test. None of the corrosion resistant metals which included

Fig. 36: Field experiment assembly lowered into the borehole at he UKAEA test site at Troon, Cornwall.

stainless steels, Ni-Cr alloys and titanium alloys, have suffered any localised breakdown and their rates of general corosion are negligible. Cast iron samples have corroded at an average rate equivalent to a metal loss of about 11 mm in 1000 years. This supports the results of the laboratory tests which indicated that the rate of dissolution will be controlled by oxygen transport.

2.2.1.3.5. Study of Corrosion Phenomena

Contractor: CNRS, Vitry-sur-Seine (249-81-7 WAS.F)

The understanding of various corrosion phenomena is necessary for being able to extrapolate results of short term tests.

A literature study has been undertaken concerning the composition and formation of passivation layers on Fe, Cr, Ni alloys and on Titanium in various environments and under different conditions. This study will be completed by a survey on the knowledge concerning the impact of aggressive ions like Cl^-, F^-, $S^=$ on the composition and structure of the passivation layer formed on Nickel alloys.

Laboratory experiments will start in 1982.

2.2.1.3.6. Evaluation of Corrosion Behaviour of Candidate Materials for Waste Containers

Contractor: CEA, Fontenay-aux-Roses (252-81-7 WASF)

The R&D project, started within the previous phase (1978-'79), on the testing in laboratory of the corrosion behaviour of potential materials to be used for the manufacture of waste containers has continued. Test are being performed on eight alloys:

- Titanium and Titanium + 0,2 % Palladium.
- Zircaloy 4.
- Hastelloy C 276.
- 625 and 825 nickel alloys.
- 316 L and 304 L stainless steels.

Test solutions were obtained from a synthetic mineral water by enhancement of some ions concentration (especially Cl^- and H_3O^+). Temperature was 80°C.

Generalized corrosion rate after 3 months of immersion was found to be the same as in pure NaCl solutions of identical concentration, 316 L stainless steel being excepted. This corrosion rate is a little higher for new tested alloys (625 and 825) but the absolute value is very small ($< 0,5$ µm/an).

Pitting corrosion was observed on both stainless steels. Repassivation of 316 L can happen but remains an aleatory process. No pitting appeared on other alloys during a 3 months-immersion but experiments using polarisation curves showed that 825 alloy was not so well protected against this type of corrosion than 625 or C 276 alloys.

Crevice corrosion is a consequence of local chloride enrichment and acidification. Polarization curves on deoxydized specimens were used to assess their susceptibility to crevice corrosion. From the worst to the best, the rating is : 304 L, 316 L, 825, 625 and C 276.

To assess the stress corrosion susceptibility, the constant extension rate technique was chosen. Several facilities were designed and built. After complete setting it will be possible to stress a probe at a very slow strain rate (e.g. $10^{-6}s^{-1}$) in a solution. The electrode potential of the probe will be controlled. The foreseen temperature is 80°C, but new items will be designed for 170°C studies.

BACKFILLING AND SEALING

The aim of this task is to select and test materials for the backfilling and sealing of disposal holes, galleries and shafts. In particular the interaction with the geological environment, the physico-chemical stability and the retention power for radionuclides is to be studied.

A critical review study on the state of the art is underway, which should also come up with recommendations for the design and validation of feasibility experiments.

2.2.1.3.7. Review Study on Backfilling and Sealing of Radioacti Waste Repositories

Contractor: Mott, Hay & Anderson, Croydon (204-81-7 WAS.UK)

A comprehensive review study has been started on the functional requirements, materials, placement and demonstration facilities needed in connection with the backfilling and sealing of waste repositories in different host rocks (granite, clay and salt).

Progress has been made with the following study areas:

- environmental conditions within the repository in relation to backfilling
- backfill/rock and backfill/waste interactions
- the design roles and desirable properties of backfills
- a critical review of international backfilling research studies to date
- identification of particular design problems for each host rock type, and different parts of the repository system.
- identification of an initial list of candidate fill materials and development of a procedure for screening, classification and assessment of priorities for design and materials researc studies.

Preliminary work has been carried out in relation to the more detailed appraisal of relevant material properties and mix formulations, together with outline costs and availabilities and engineering design and placement aspects.

2.2.1.3.8. Studies on Backfilling and Sealing Materials

Contractor: SCK/CEN, Mol (144-80-7 WAS.B)

Twenty different mixtures of Boom clay, bentonite, cement and calcium carbonate in various ratios were tested as potential material for borehole plugging and filling between the cast iro lining in the gallery and the clay massive. Mixtures of thermo-hardening resins and Boom clay will also be investigated.

In the first laboratory tests, specal attention was given to the mechanical properties of the mixtures such as, setting time, shrinkage, density, consolidation factor, permeability, porosity, tensile and compressive strength etc.

The most promising mixtures from this point of view are those containing up to 5% of cement, about 3% of bentonite, 5 to 10% of calcium carbonate, the remaining being dried Boom clay with a granulometry between 100 and 500 µ. Some mixtures will now be tested for their technological applicability "in situ". Attention will also be given in the future to the properties of the interface between the lining materials or the clay and the plugging or filling mixture. The "Institut National des Industries Extractives" (I.N.I.E.X.-Liège) contributes extensively to this research.

2.2.1.3.9. Studies and research relating to borehole plugging

Contractor: CNEN, Cassacia (199-81-7 WAS.I)

The work is to develop the instrumentation as well as the procedure to test "in situ" the effectiveness of sealing boreholes containing the wastes by means of appropriate mixtures of clays and additives.

The main phases of the programme, which has been harmonised with the foregoing one at CEN/SCK in Mol (Belgium) are the following:

- assessment of the critical parameters that can be measured "in situ" to check the effectiveness of the proposed sealing mixture;
- design and construction of the instrumentation and testing of the experimental procedure by means of preliminary laboratory runs on models;
- planning of one "in situ" experiment at Mol for borehole plugging and sealing effectiveness testing.

2.2.1.3.10. Emplacement Techniques and Characteristics of Backfilling and Sealing Materials for Repositories in Granite

Contractor: CEA, Fontenay-aux-Roses (202-81-7 WAS.F)

This programme consists of two phases; first the critical review of existing knowledge and of the properties to be considered, and second,a limited test programme to be carried out on certain materials or components chosen from the 1st step. This experimental phase will focus on the emplacement techniques and the geotechnical, physico-chemical properties of the material in place.

2.2.1.4. CHARACTERIZATION OF INTERNAL EQUILIBRIA OF GEOLOGICAL FORMATIONS

2.2.1.4.1. Characterization of Boom-clay

Contractor : SCK/CEN, Mol (144-80-7 WAS.B)

Analyses of frozen samples

A direct consequence of the excavation work presently under way in the Boom-clay formation was the obtention of fresh clay samples frozen in situ. Stored in the frozen state and quickly analyzed under controlled atmosphere they have revealed a sulphate content similar to the SO_4 concentrations prevailing in the neighbouring aquifers, i.e. 10 to 30 mg $SO_4^=$/l Therefore the Boomclay should constitute a particular hazard from the point of view of corrosion at room temperature by sulphate reduction with iron. Essentially the interstitial medium of "in situ" oxygen free clay appears to be a 0.01 M $NaHCO_3$ solution saturated with $CaCO_3$.
Redox potential measurements and chemical analysis have shown that in its "in situ" state, the Boom-clay constitutes a medium sufficiently reducing to keep long-lived radionuclides like Pu, Np and Tc in the state IV where they are highly insoluble.

Geotechnical properties as function of temperature and pressure

The laboratory tests on geotechnical properties variations of Boom-clay as a function of temperature and pressure cycling were continued. This work was partly performed by the "Rijksinstituut voor Grondmechanica", R.I.G.M.(Gent)and partly by

the "Laboratoires de Génie Civil L.G.C." (University of Louvain-la-Neuve). The clay samples originate either from the Terhagen site or from the shaft in Mol.
The aim of the R.I.G.M. programme was the determination of the influence of temperature variations from -25°C up to 80°C on compressibility, swelling and permeability coefficients measured by oedometric tests and also on the cohesion and the angle of the shearing resistance measured by triaxial tests.

Some results and trends can already be given :

- Increase of shear stress between 0 and -25°C, this increase is a function both of consolidation pressure and temperature (the lower the temperature the higher the shear stress) ;
- While at normal temperature and at -25°C the elongation at failure is around 8%, this value reaches a maximum of 15 to 20% at -5°C ;
- Repeated cycles of temperature between ambient temperature and -25°C has no influence on elongation and failure strenght

The compressibility and swelling (TERZAGHI) values corresponding to samples tested at ambient temperature in an oedometer are respectively :

C = compressibility constant : 20 and 25 ;
A = swelling constant : 45 and 48.

At temperatures around 80°C, measurements indicate an increase of the previous mentioned constants by a factor of 2. The deduced permeability at this temperature (80°C) is nearly the same as at ambient temperature (values of 1 to 5.10^{-9}cm s^{-1}).
The L.G.C. programme was aimed to describe the behaviour of frozen clay under pressure (creep tests at temperatures of -10°C,-20°C and -30°C). The first measurements under uniaxial conditions indicate an increase of shear strength at failure (3,3 MPa at -10°C and 11,5 MPa at -30°C) for elongations

between 5 and 10%. Two deformation rates (at 10^{-2} and 10^{-3}min^{-1}) show no significant difference between those results. The creep tests were performed in uniaxial conditions with a constant vertical stress at 80, 65 and 50% of the strength of failure.

Heat transfer experiment

After 18 months of running, the heat transfer experiment with a mock-up of a high level solidified waste canister burried in a clay pit at Terhagen was definitively stopped. The planned test with an output of 3 Kw could not start due to failure in the electricity supply and the canister was digged out for corrosion examination. The main objectives of the experiment were however reached.

2.2.1.4.2. Thermal Studies and Research relating to Clays

Contractor : CNEN, Casaccia (206-81-7 WAS.I)

The CNEN is carrying out, in situ and laboratory, heating experiments in clay. These experiments are performed :

- To obtain a correlation between the variation of thermal properties and the increase of temperature ;
- To determine the pressure increase of interstitial water in clay in the presence of a thermal gradient ;

The laboratory experimental procedures which seem more suitable for the measuring of thermal properties of clay are the following :

- needle probe method (Von Herzen-Maxwell) ;
- cylindrical probe method (Jager)

In the first method, a probe of small diameter (ϕ= 1mm), which contains a heating resistence and a thermistor to measure the temperature, is used. In the second method a similar probe but with a larger diameter (ϕ= 4mm) is used.

Some preliminary tests have been executed at the laboratory of ENEL with the needle probe method. The measure of heat conductivity (K=0.015 watt/cm.°C) corresponds well with results obtained in clay block tests at the laboratories of CNEN and the University of Rome.

The "in situ" experiment is planned in an open clay quarry, in the area of Monterotondo, near Rome, where large deposits of clays are outcropping and extensively used by brick-works. These clays, whose age is Plio-Pleistocenic and thickness is in the order of 100 m, constitute a part of the Tiber Valley sediments.

The approximate composition of the clay is : chlorite 5-8%, smectite 40-45%, illite 40-50%, kaolinite 3-5%

The cylindrical heater is 60 cm high in diameter, with a maximum power of 500 watt. Ten probes, for temperature measurements, have been installed around the heater at distances ranging from 50 cm to 200 cm. The heater and the probes have been embedded at a depth of 6.4 m in 11 boreholes. The boreholes have been drilled by means of portable equipment "Mobile Drill", equipped with a cutting edge designed to drill clays in dry conditions. After the positioning of the heater and the probes, the boreholes have been filled with bentonite. The experiment will start early 1982.

2.2.1.4.3. Evaluation of Modifications induced in Clays by Intrusive Subvolcanic Bodies

Contractor : CNEN, Casaccia (152-80-7 WAS.I)

The effect of the heat released by a H.L.W. repository into the host rock represents one of the most interesting aspects of near field studies.

Natural phenomena, similar to those in clay formations surrounding a H.L.W. repository, are present in Italy.

Particularly in Tuscany there are magnetic intrusions into pliocenic clays. In one of these zones (Orciatico) a detailed study is planned, into the spatial relations between the intrusive body and the surrounding clay, and the effects caused by the first one on the other. It concerns a laccolitic subvolcanic body, very near the surface, intruded into pliocenic clays.

A detailed geological survey and a refraction seismic exploration has been carried out to determine the limits between the intrusive body (selagite) and the clays modified by the thermal effect (termantite). To obtain samples, a set of drillings of 124 m in total length have been carried out in the termantite and in the clays close to the intrusive body.

The sampling has been made from the non altered clay to the selagite. The thickness of the metamorphic aureole ranges from 12 m to 14 m, on the top of the intrusive body, and from 5 m to 6 m, on its sides. In order to obtain samples to compare with the metamorphic ones, another set of drillings has been carried out in zones where clays are not altered.

On the collected samples, several analyses are being carried out at present.

2.2.1.4.4. Propagation of Seismo-tectonic Displacements through Clay Deposits

Contractor : CNEN, Casaccia (238-81-7 WAS.I)

The knowledge of the mechanical behaviour of clay formations affected by seism-tectonic diplacements is important for engineering studies of a repository.

The particular objective of the study is the mechanics of displacements in an usualyy plastic medium laid on a non-plastic medium. A first step is to define the mathematical model of clay formation, the second one is to verify this model by experimental and field data.

A bibliographic research for data collection with regard to the mechanical behaviour and the constitutive law of clay formation is in progress.

2.2.1.4.5. Experimental Studies on Pore Water Composition in Clay Formations

Contractor : CNEN, Casaccia (151-80-7 WAS.I)

Laboratory tests on clay pressing have been carried out and new analytical techniques for the atomic sorption spectrometer PE 500 have been set up.

Experiments of clay pressing by means of oedometer able to supply a pressure of about 60 Kg/cm^2, have been carried out on consolidated and non consolidated clay samples. For these two types of clays, respectively, 15% and 40% of the total water content has been yielded. These water contents are believed to be insufficient in order to obtain significant analytical results on chemical composition of the pore water. Consequently it has been decided to plan and to build an oedometer capable of reaching a pressure of about 500 Kg/cm^2.

An analytical method has been set up to detect Rb and Cs. By means of this method Rb concentrations less than 0.001 mg/cm^3 and Cs concentrations slightly less than 0.02mg/cm^3 have been detected.

2.2.1.4.6. Interaction of Fluids, Rocks, Grouts and Waste under Repository Conditions

Contractor : NERC/IGS, Harwell (128-80-7 WAS.UK)

Laboratory studies are carried out with a view of the characterization of the physical and chemical properties of solid ateration products and fluids synthesised during the reaction of borosilicate glass waste forms with water and rock. A series of experiments have been carried out using glass-water-granodiorite mixes to understanding the detailed changes in fluid chemistry with time at temperatures in the range 100°-350°C. The results of granodiorite-water experiments have revealed that the chemistry of the resultant fluid phase is favourable for the preservation of stainless steel canister materials (high pH and low chloride and sulphate concentrations) thus obviating the necessity of providing a groundwater conditioning agent in any repository backfill material.

The production of montmorillonite as a secondary alteration phase in these experiments indicates that bentonite would be a geochemically stable backfill material, even at temperatures up to 200°C. Glass-granodiorite-water experiments under closed system conditions have shown that release of waste components from the near-field may be orders of magnitude lower than those implied from weigthloss measurements derived by "dynamic" leach tests (e.g. Soxhlet). Glass dissolution has been shown to be strongly incongruent and leach rates for specific elements decrease with time. Many waste elements are concentrated on the glass surface alteration layer, which is completely crystalline at the higher temperature experiments (350°C). Mineral phases thus produced may concentrate waste elements by factors of up to 10, as compared with the initial glass. New, long term experiments (6-12 months duration) have been initiated to study the solubilities of various waste components in the glass-granodoirite-water system in the temperature range of 100°-350°C. The results from the experimental studies have been used, along with radionuclide scorption data, to develop a simple model of radionuclide release and transport for caesium. The analysis of the experimental data are being aided by use of computer programme packages.

2.2.1.4.7. Thermal Phenomena in Crystalline Rocks

Contractor : U.K.A.E.A., Harwell (154-80-7 WAS.UK)

Heat transfer experiment

The heat transfer experiment was continued during this year at constant 10 kW power to obtain steady, maximum temperature rises up to the greatest distance from the heater at which these rises can be measured with the sensitivity of the resistance thermometers being used. This sensitivity is about $\pm \frac{1}{2}$ °C and steady rises of this amount are being detected up to 30 m from the heater. All recording is done automatically. Occasionally checking of the data show that the heat transfer is still mainly by conduction with a coefficient continuing to be about 3.5 $W.m^{-1} {}^{\circ}C^{-1}$.

Hydraulic Conductivity

In the first half of 1981, an extensive series of detailed pressure and flow measurements were made at the Cornish research site. These were intended to provide effective permeability data for flow through the fractures in the rock, treating the fracture pattern as a permeable medium. The results have, however, shown that this intention cannot be achieved and they have led to the changes in the programme and the progress during the second half of 1981 described below. From measurements of flow from single holes into the rock at 100 to 200 m depth it was found that most of the flow occurs in discrete fractures which are separated on average by about 10 m and which are interconnected and probably randomly oriented to form a three dimensional fracture pattern. Interpretation of these results to determine the effective permeability of the pattern is not possible because the flow from the 10 cm diameter holes into individual fractures with average separation of 10 m between intersection with other fractures is mainly determined by the hydraulic apperture of the fracture locally round the hole and substantially independent of interconnections through the fracture pattern as a whole. Two further points should be noted. Firstly, direct determination of the effective permeability would require flow and pressure drop measurements over distances large compared with the average fracture separation, say 100 m , which would be difficult if not impossible. Secondly, because diffusion of radionuclides from the flow into stagnant water in cul-de-sac cracks is an important retardation mechanism which cannot be evaluated without data for the separation between flow paths permeable medium models will be unable to quantify the radionuclide transport. The following alternative combining theoretical and experimental approach to the fracture hydrology is therefore being developed.

It is assumed that the fractures forming the pattern through which most of the water flows are planer and have the following variables :

- Frequency of occurance or number per unit volume of rock
- Orientation
- Lengths between ends or characteristic linear dimension
- Effective hydraulic apperture.

Theoretical work is being done at Harwell and other establishments to develop percolation models which, given experimental statistics for the means and distributions of the above variables, should be able to predict the flows.

Experimental methods have been developed in Cornwall to obtain these statistics at 200 m depths in granite. Fracture occurance is determined from multi-packer tests in single holes. Radioactive tracers are then pumped into single fractures and detectors are repeatedly lowered and raised in adjacent holes to locate the positions of single or multiple arrivals of tracers - these results provide orientation and topological data about the fractures and their interconnections. Finally, pairs of double packers in adjacent holes are set at positions shown by the tracers to be on the same fractures and interhole flow and pressure drop measurements are made to obtain estimates of their effective hydraulic appertures.

These interhole tracer and pressure drop methods have been proved and are now being used to accumulate experimental statistics for percolation theory.

In addition to the existing 200 m deep vertical holes a 700 m vertical hole and a 200 m long 45° hole are now being drilled. The former is intended to allow the effect of depth on the fracture statistics to be studied. The latter is to investigate a geological thesis that there may be a higher incidence of vertical fractures which are not of course sampled by vertical holes than fracture of other orientation. Experiments in these holes will start about the middle of 1982.

2.2.1.4.8. Mechanical Behaviour of Granite under Pressure and High Temperature

Contractor : CEA, Fontenay-aux-Roses (146-80-7 WAS.F)

Rock cores extracted from boreholes drilled in granite at Auriat (F) were tested in the laboratory. The following measurements were performed during 1981.

a) General characterization of the sampled interval
b) Porosity
c) Volumetric Behaviour under isotropic compression
d) Behaviour under uniaxial compression
e) Behaviour under triaxial compression

General characterization of the sampled interval

The sampled interval lies between 238.8 and 249.2 m depth,i.e. about 10m cores were selected. At normal temperature, this granite can be considered as homogeneous and identified by the following values :

- Bulk specific gravity : 2.62 $g.cm^{-3}$
-Compression wave velocity : 4,900 ms^{-1}

- Uniaxial compressive strength : 150MPa
-"Brazilian" tensile strength : 11 MPa

Porosity

Porosity was measured by 2 different methods :

- Directly, by means of a mercury porosimeter, the maximal injection pressure being 200 MPa
- Indirectly, using the compression wave velocity in the dry and then in the saturated rock.

Both methods were applied to granite in its natural state and after one month heating at 100°C. Similar results were obtained in each case :

- In natural state, porosity lies between 1.5 and 2%
- After heating, it increases to 2-2.5%.

Volumetric Behaviour under Isotropic Compression

Volume changes under compression were measured up to 100MPa isotropic pressure and under three different conditions :

a) Granite in the "natural" state
b) After 10 days heating at 100°C
c) After one month heating at 100°C

For each case, the following measurements were performed :

- Bulk modules, K (GPa)
- Crack porosity, η
- Pressure level at which cracks become closed, Pf (MPa)

Mean values of these parameters, for each of the three test conditions are given in the table below :

Test condition	K (GPa)	$\eta (10^{-3})$	Pf (MPa)
Natural state	51	0.55	50
10 days at 100°C	41.5	0.75	55
1 month at 100°C	43	0.95	60

It can be concluded that heating of the samples enhances the cracking of the rock, thus reducing K and increasing D and Pf.

Behaviour under uniaxial compression

Measurements of axial and lateral strains by extensometers allowed the determination of elastic and strength parameters :

- Young's modulus, E (GPa)
- Poisson's ratio, ν (dimensionless)
- Uniaxial compressive strength, σ (MPa)

These measurements were performed under three types of experimental conditions :

a) Granite in the "natural" state
b) After 8 days heating at 100°C
c) After one month heating at 100°C

In all three conditions, the tested granite shows a marked elastic-brittle behaviour. The enhacement of cracking due to heating can be seen reflected mainly in a noticeable decrease of Poisson's ratio, as summarized below :

Test condition	E (Gpa)	ν	σ (MPa)
Natural state	68	0.27	165
8 days at 100°C	66	0.17	168
1 month at 100°C	55	0.15	155

Measurements of the same mechanical properties (σ, E, ν) but under high temperature (200°C) are underway.

Behaviour under triaxial compression

These tests were performed on granite in the "natural" state only. For increasing confining lateral pressures, P, the axial load causing failure, σ_1 was recorded. Mean results are summarized below :

P (MPa)	10	20	50	75	100
σ_1 (MPa)	320	405	550	685	785

It can be concluded that the compressive strength of this granite increases very quickly with confining pressure. Tests at 200°C are in preparation.

2.2.1.4.9. Study of Deep Fracturing of Granite Formations

Contractor : CEA, Fontenay-aux-Roses (148-80-7 WAS.F)

Collection of literature data

The first part of the programme consisted of :

a) A screening of BRGM's report about loggings of deep galleries, performed in the French Pyrenees, Alpes and Massif Central, between 1942 and 1964. From these reports, only little quantitative informations could be gained about fracturing.

b) A detailed analysis of the "Arc-Isere" gallery log. This gallery, built by the French Electricity Board (EDF), intersects the external crystalline formations of the Belledone Range in the French Alpes. A detailed geological Survey was carried out and reported in a Ph D thesis by D.Gasquet (1979). From this work, an interpretative longitudinal profile was produced, which allowed comparison between fracturing in the gallery and fracturing gained from surface mapping, 1.000 to 2.000 m above gallery level. The main conclusions are as follows :

- A good correlation exists between the orientations of fractures determined from surface mapping and from gallery logging. This is valid for small fractures as well as for large faults.

- The influence of a fault on the small-scale fracturing, having the same orientation, can often be seen reflected in a local increase of the frequency of this small-scale fracturing.

- Minor and intermediate-sized fractures (some meters to some tens of meters) are the most important drains, while mylonitic zones and major discontinuities which limit great tectonic units are remarkably water-tight.

Structural study of the fracturing at the Fanay-Augeres site

Aim of the works performed in 1981 was to establish the tectonic history of the St. Sylvestre leucogranite with special emphasis on the influence upon fracturing. The fracture families were identified, together with their possible evolution from ground surface to greater depth.

To achieve this goal, a geometrical, cinematic and chronologic study of the fracturing was performed.

The St.Sylvestre leucogranite (Fanay-Augeres Site), of Namurian Westphalian age, contains numerous mineralized uranium-bearing veins, which are currently mined by the French COGEMA, in open-pits as well as in underground galleries. Many locations are then available to observe the fracture pattern, aboveground and down to 400 m depth.

The tectonic evolution of this leucogranite, which lead to its present fracturing, was shown to have occured in seven phases. Among these phases, only the two first ones (Stephanian and Autanian age) gave rise to major faults. The third one (Saxonian age), an extension caused new movements of big E-W and NW-SE faults. Subsequent Mesozoic extensions and Caenozoic compressions induced only minor displacements of the existing fractures.

Big faults can be traced from ground surface down to greater depth without alterations of their geometric and cinematic characteristics. Small fractures, well correlated with big faults, can be encountered both aboveground and at depth.

Present conclusions

- It was shown that the orientations of fractures, measured aboveground and in deep galleries, are in good agreement. This is valid for two different crystalline massifs : one in the Alpes district and one, of the late Hercynian age, in the Western part of the Massif Central.
- However, the hydraulic role of these fractures at depth is hardly predictable from surface observations.

2.2.1.4.10. Theoretical Study of Fracture Sealing in Granitic Rocks by Silica Precipitation

Contractor : CEA, Fontenay-aux-Roses (200-81-7 WAS.F)

The introduction of heat-producing waste in a fractured granitic rock may have a self-sealing effect on the fractures, through the combined action of two mechanisms :

(i) Convective movements of the water in the fractures due to density variation of water with temperature,

(ii) Dissolution of silica by water in the hot areas surrounding the waste, and precipitation of this silica when the convective movements have brought it to colder areas above the waste.

The aim of the present study is to quantify the importance of these mechanisms considering the length of time during which significant amounts of heat are produced by the waste. The first part of the study is only theoretical, and will later be backed up by laboratory measurements.

First results were obtained from a literature survey, which allowed to correlate the mechanisms of precipitation and dissolution of silica in moving water.

Depending on the type of silica (quartz or amorphous), the concentration at equilibrium is a different function of the temperature, amorphous silica being five times more soluble than quartz.

Subsequent results were obtained by calculating the actual temperature field in a fracture surrounding a real repository, as a function of time and distance, given the heat input of the waste and an expected density of waste per area. This made it possible to calculate the velocity field in a fracture of given aperture as well.

2.2.1.4.11. Statistical Study of the Determination of the Hydraulic Conductivity in a Fractured Medium

Contractor : CEA, Fontenay-aux-Roses (150-80-7 WAS.F)

The aim of this study is to determine in a statistical framework the hydraulic conductivity and its uncertainty as a function of the available data set.

In order to obtain valid simulations of fractured media, the relevant properties are studied in two different areas, in granitic rocks, where data are available :

- The Fanay-Augères district, near Limoges, where uranium has been mined since the early 1950's,
- The Auriat district, where detailed observations of the fractures are available from the core of a well drilled in 1980-1981

2.2.1.4.12. The Influence of Fluid Rock Interaction on the Rheology of Salt Rock and on Ionic Transport in the Salt

Contractor : University of Utrecht (153-80-7 WAS.NL)

Salt rock deforms by a variety of mechanisms. Different mechanisms prevail when different conditions of mean stress, temperature, deviatoric stress, strain-rate and pore pressure apply.
A sequence of experiments will be performed to study :

- The variation of penetrative dilatancy with deformation at different values of mean stress.
- The effect of pore fluid on the dilatant behaviour of rock salt.
- The transport of ions along grain bounderies.

These experiments will be done with an apparatus specifically designed for this project, permitting pressures up to 500 bars and temperatures up to a maximum value of 400°C. At the end of 1981 this apparatus was being installed and the experiments are scheduled to start in the beginning of 1982.

2.2.1.4.13. Suitability of the Konrad Mine for the Disposal of Radioactive Wastes

Contractor : GSF Braunschweig (145-80-7 WAS.D)

In connection with the geological-hydrogeological feasability studies, tests are carried out in view of assessing the possible disposal of low-level and decommissioning wastes in the

Konrad mine.

Petrographic and Geochemical Investigations

- In tracer-tests with the isotopes Cs-134, Co-60 and Sr-85, the sorption properties, distribution coefficients and KD coefficients, have been determined on 15 samples of selected shale in the rock overlying the iron ore bed.
- Tests to determine diffusion coefficients on Konrad rock samples following the "Sandwich-procedure" with the radionuclides Sr-85, Cs-134, Co-60 and Pu-239, were started.
- Using the γ- spectrum analysis, samples from the overlying rock were tested from the point of view of their natural γ-activity. The oolithic iron core was analysed to determine its content of Ra-226, Pb-214, Tl-208, Ac-228 and uranium.

Hydrogeological and Hydrochemical Investigations

- Up to now, the results of the hydrogeological investigation on the mine waters as well as on the water from surface areas exclude any possibility of infiltration of external water.
- Special geochemical investigations were performed on the secondary salt minerals in the mine : on 47 salt samples Na, K, Rb, Mg, Ca, Sr, Cl, Br, I, SO_4, BO_3, NO_3 were quantatively determined. The results showed no evidence of any participation of brines from subrosion of the neighbouring salt dome in the composition of the minerals. They can be interpreted as crystallisation products of the residual saline moisture of the corallian oolith.
- In a test gallery, further measurements of the migration of residual moisture in the iron ore were performed by means of conductivity measurements. It is clear from all measurements that the rock mass conductivity of the test gallery is by

35% greater than the one of older ventilated galleries.

Rock Mechanics and Seismic Investigation

- The deformation measurements for the determination of the stress distribution in a gallery-pillar system, were continued on 8 extensometers measurement stations. During the excavation phase, two extensometers, each 29 m long, were installed from a parallel gallery for the observation of the geomechanical deformations.
- By means of ultrasonic measurements, the extension of the fractured zone and the stress-redistributions in the rock-mass up to 15 m deep were determined from a borehole for rod extensometer.
- Results from investigations into

a) The shear-strength behaviour of tectonic discontinuities by means of TV-logging in two horizontal boreholes (30 m long),
b) Micro-tectonic cartography and
c) Exploitation of laboratory rock mechanics tests,
eventually resulted in an improved assessment of the stability of disposal room.

- The seismic monitoring of the mine recorded up to now only insignificant events as well from the point of view of the frequency as of their magnitude (apart from artificial events).
- The measurement process for ultra long rock mass movements with a geophysical tiltmeter was further developed.

Mining Investigations

- The development works for the backfilling of disposal rooms was continued by a technical study. In this latter, one took account of the particular safety requirements for the disposed waste. It was shown that adequate processes include pneumatic in-filling and, by elemination, casting processes.

- For the first time, a boring machine was put into operation for the sinking of big diameter ventilation holes by the dry-drilling method as well as a "road-header" type cutting machine for the mechanical excavation process.
- In the framework of the evaluation procedure, the natural activity concentrations were determined in the ventilation air of the mine. The estimated radon activity concentrations show that the values measured in the mine atmosphere are higher than those measured in the environment.

Waste Canister Corrosion Investigations

In the report period, tests on 6 internal and 6 external coating systems were performed in Konrad under simulated, conditions of storage and disposal. Two coating systems based on Epoxy were found adequate. Tests are continued.

Disposal Tests with Inactive Waste Canisters

- The testing of waste bundle maintenance with or without pallets was started. The tests also supplied indications for the lay-out of the technical installations.

2.2.1.4.14. Study on "Admissible Thermal Loading in Geological Formations and the Consequences on Waste Management"

Contractors : SCK/CEN, Mol (111-79-10 WAS.B)
CEA, Fontenay-aux-Roses (110-79-10 WAS.F)
GSF, Clausthal-Zellerfeld (112-79-10 WAS.D)
NERC/IGS, Harwell (118-79-10

The purpose of this study is to review the thermal effects induced by geological disposal of high-level wastes and to assess their consequences for "Admissible thermal load" in particular and for waste management in general.
Three parallel studies dealt separately with the three continental geological media being considered for HLW disposal within the CEC programme: granite, salt, clay. The significance of this selection also resulted from the quite different properties of these geological media. The studies have been performed under the leadership of the Centre d'Etude de l'Energie nucléaire (SCK/CEN) for clay, the Cmmissariat à l'Energie Atomique for granite, the Gesellschaft für Strahlen-und Umweltforschung (GSF) for salt, and with the assistance of the Environmental Protection Unit of the Institute of Geological Sciences (NERC/IGS). A "Synthesis Report" has been elaborated by an Editorial Committee on the basis of the specific detailed reports produced for each of the formations.
The scope of this study was defined as follows :

a) Only vitrified high-level radioactive waste was considered.
b) The"Multibarrier" confinement concept was assumed.
This includes : waste-glass, container (with or without overpack), buffer material, rock formation.
c) Disposal is in a deep mined repository, in an "in-land" formation.

In this framework, only "normal" situations and processes were covered, no "accident" scenario being taken into account. No experimental work as such was performed. The studies however were based on extensive compilation of available literature data, resulting for a large extent from the CEC Programme since 1975. The common content of the three specific studies is shown on the table below.

Main results of the study

For each rock type, best estimate temperature limits were set taking into account :

1. Heat propagation
2. Thermo-mechanical effects inside the rock formations
3. Induced or modified groundwater or brine movement
4. Effects on the buffer material
5. Effects on the waste package (waste glass and canister)
6. Nuclide transport

To comply with these limits, one has to consider :

(a) Waste characteristics, i.e. initial power output and cooling-time prior to disposal

(b) Repository design, such as spacing between heat sources and general repository lay-out.

Particularly, the waste canisters must be surrounded by a large enough "rock-cell" so that the deposited energy brings about a tolerable temperature rise. Indicative values of both wastes and repository parameters resulting in an admissible thermal loading of the formations considered are listed in table 2. The data for granite are those for a "generic" granite formation, whereas those for salt are resulting from a study which was concentrated on a well defined North-German salt formation of the Zechstein age and those for clay are valid for the Boom clay formation beneath the SCK/CEN site at Mol. Although the results show features which are representative for a wide range of host rocks in the three formations considered, the results cannot be translated as such to any other disposal site in a same formation. Moreover these figures must be considered with caution. Many of the data required for assessing the consequences of thermal loading on the ultimate post-closure performance of a waste repository, in terms of long-term safety analysis, are limited or are only now becoming available. Thus the results of this study should be seen as an attempt to deriving "working" criteria to be used in the design of waste repositories.

Subsequent comprehensive safety analyses of specific proposed repository sites may result in adjustements to these limits when the thermally driven processes leading to release and migration of radionuclides are better quantified.

CONCLUSIONS

Heat propagation itself and the majority of associated effects are predictable within existing knowledge with an acceptable degree of confidence.

Further research, and in particular comprehensive safety assessments for the various options, may result in adjustments of admissible temperature levels but disposal of heat-emitting high-level waste will remain feasible in each of the formations studied, as thermal loading is controllable so that any critical limit can be respected.

What has been described as admissible thermal loading with regard to the integrity of man-made and natural barriers is not necessarely economically viable.

An optimisation of the HLW cycle is desirable and should take into account the merits and costs of intermediate storage and in-situ cooling to arrive at a disposal solution which can be considered as technically feasible and economically acceptable.

As a conclusion, it can be stated that, within the present state of knowledge, heat emission does not make the disposal of HLW into deep geological formation an impossible or unrealistic operation.

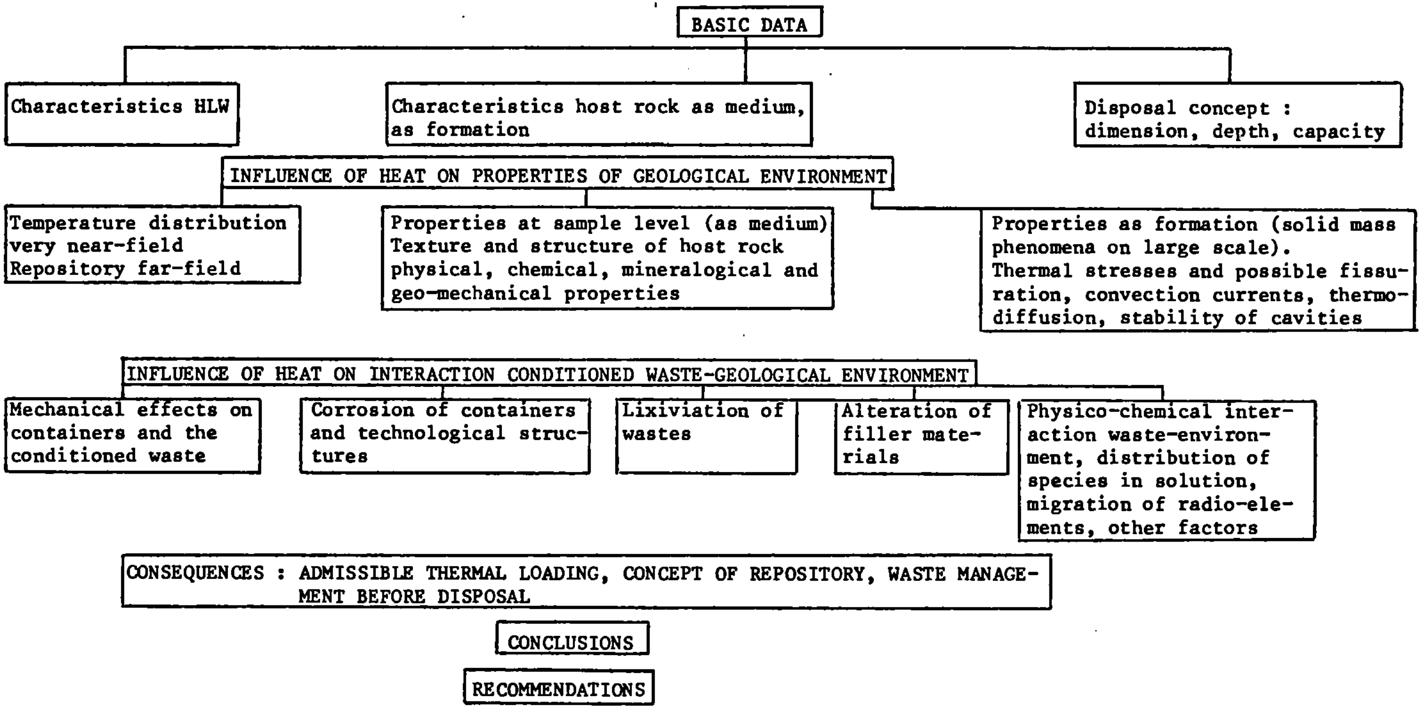

Table of contents for the 3 studies

	Granite	Salt	Clay
Characteristics of waste canisters	ϕ = 35 cm H = 150 cm V = 150 l	ϕ = 20 cm H = 1 m V = 30 l	ϕ = 30 cm H = 150 V = 100 l
Initial power output	935 W/Can 6.000 W/m^3	400 W/Can 12.000 W/m^3	435 W/Can 3.900 W/m^3
Elementary rock cell	15.000m^3/Kw	1.000-2.000 m^3/Kw	Not relevant (flat repos.)
Specific area	150m^2/Kw	10-20 m^2/Kw	400 m^2/Kw
Cooling time	30 years	60 years	60 years

Table 2 : Summary table of most important data to define admissible thermal loading

2.2.1.5. MIGRATION OF RADIO-NUCLIDES

2.2.1.5.1. Migration of Radionuclides in Clay Formations

Contractor : SCK/CEN, Mol (144-80-7 WAS.B)

Migration studies by means of three dimensional reconsolidation cells have shown no decrease in diffusional flux for HTO (water mobility) consecutive to reconsolidation, and a limited decrease for Sr. It seems safe to conclude that consolidation, although favourable, is not central for the capacity of clay to prevent effectively radionuclide migration. Diffusion runs on clay plugs carried out with ^{154}Eu (quasi insoluble at the pH of Boom clay) have revealed how impractical it is to use diffusion runs to assess diffusional properties of insoluble nuclides. Apparently, experiments should be extended over years. Practical estimations of long-lived radionuclides migration are more likely to be obtained from an accurate chemical description of the "source term" (radionuclide effective solubility in media of known composition, buffered Eh and pH). K_D measurements under conditions as near as possible to "in-situ" conditions have shown a limited affinity of Fe^{++} for adsorption. In consequence, corrosion products, limited as they are in their solubility, should not be real competitive for radionuclides in their adsorption on clay.

2.2.1.5.2. Migration or Radio-nuclides in Clay

Contractor : CNEN, Casaccia (208-81-7 WAS.I)

Laboratory tests have been carried out in order to determine :

- The coefficient of apparent diffusion (D) and the coefficient of distribution (Kd) in clays and in synthetic clays.
- The clay minerals that affect the radioisotope diffusion.

Main part of the work consisted in adjusting suitable technologies and instruments. The methods of pressing clay minerals ready for sampling, their sampling, as well as of the most suitable contact for radioactive solutions/clay cores, have been studied in particular.

Synthetic clay has been made by mixing the clay mineral or minerals with water (40% of whole weight), and SiO_2 and $CaCo_3$, with grain size 50-170 mesh (7.5% of each mineral).

Afterwards, these synthetic clays have been pressed by oedometer CBB/15, in a cell with a surface of 38.48 cm^2 and 5.5 cm high.

During these experiments pressures ranging from 0.2 up to 30.0 kg/cm^2 have been used for a period of 7 days.

The decrease in thickness of the clays in the cell (from 2.6 up to 3.8 cm) depends on the kind of mixture.

In order to determine D and Kd, either analytical/chemical and radiochemical methods have been used.

For the determination of D the following three methods have been set up :

- Diffusion of a radioisotopic solution in a core and following effusion in water;
- Diffusion of radioisotopic solution in a core ;
- Diffusion of a radioactive solution, soaking a filter paper, in a core.

Only the last method has given reliable results. According to the type of clay, the following results have been found :

Radioisotope	$Dx10^6$ (cm^2/s)	Kd (cm^3/g)
^{131}I	1-5	1-9
^{85}Sr	1-8	2-180

The wide range of Kd values for ^{85}Sr has to be attributed to the different radionuclide concentrations.

2.2.1.5.3. Nuclide Migration Studies in Granite

Contractor : UKAEA, Harwell (209-81-7 WAS.UK)

Nuclide - Rock Interaction

As in 1980 the studies on nuclide - rock interactions have been mainly concerned with the finding of mobile, non-adsorbed species and determing the chemical and physical properties of those species.

Technetium as pertechnetate (valency 7) and neptunium (val.5) were both shown to be weakly sorbed and in distinction to plutonium and americium (the other nuclides of particular importance) they do not appear to form colloids nor be affected by the presence of colloidal silica.

The technique devised for the measurement of diffusion coefficients in porous media has proved to be well suited to the detection of weakly sorbed species and the measurement of low $K_d$values. In the case of technetium a Kd of 0.06 l/kg has been found for the pertechnetate ion, and it should be noted that the method precludes interference by Tc ^{IV}.
Diffusion tests with neptunium - V have been started, the diffusion is very slow and although it is evident that K_d is greater than 0.5 l/kg no positive results have yet been obtained. (Note that batch tests gave a $K_d$value of 1-2 l/kg).

Studies on colloids have continued, with examination of leachates from simulated waste glass by photon correlation spectroscopy. Samples of glass were leached in mock granite groundwater for periods of 5 to 18 weeks at temperatures of 25°C, 50°C and 75°C. The measured diffusion coefficients were all around $5.10^{-12}m^2/s$ with mean hydrodynamic radii in the range 40 to 70 mm. The distribution in size within individual leachate samples was remarkably narrow. Some of the observed features may be artifacts due to storage of the leachate at room temperature after leaching.

Physico-chemical effects

Previous work established the feasibility of measuring permeability of granite to water by direct observation of flow but showed some anomalous effects of pressure at the relatively high differential pressures used (up to 100kPa).

A new technique has now been developed in which the water flow is measured using an ion-selective electrode with iodide ions as tracer. Measurements have now proved possible on granite with differential pressures as low as 1 kPa. Values of granite permeability of about $10^{-19}m^2$ have been found.

The iodide tracer technique is now well established and a number of measurements have been made on granites from different areas of the U.K. ;

Three Scottish granites have shown diffusion coefficients ranging from 3 to $16.10^{-13}m^2/s$ with corresponding capacity factors (porosity) ranging from 1 to 15.10^{-3}. A Cornish cornmenelli granite had a diffusion coefficient of $3.10^{-14}m^2/s$ - an order of magnitude lower than the lowest Scottish granite. The capacity factor (porosity) was 8.10^{-4}.

Field experiments

Preparation has continued during this year for an underground single fracture water flow and radionuclide transport experiment. The objective of this experiment is to pump water at known rates through a single well defined fracture between pairs of holes. Step and puls changes in concentrations of various radionuclides will be put into the flow and retardation of these by diffusion into stagnant water in cul-de-sac cracks will be measured.

A 3 X 4 X 5 metre cavern for this experiment has been created. The roof of this cavern was found to be dangerous and has been stabilised by rock bolting. As an additional safety precaution it will be shoterected before routine scientific work is started.

A pattern of nine holes, one central and two groups of four on concentric circles of 0.70 m and 1.2 m radius have been drilled. Their direction is the one shown by a fracture survey to be most likely to intersect fractures. Core and impression packing from these holes is now being examined to determine the geometry of the fractures found.

Services are now being installed in the cavern and scientific work is expected to start in about the middle of 1982.

2.2.1.5.4. Integral Migration Tests

Contractor : UKAEA, Harwell (253-81-7 WAS.UK)

In order to provide information on overall radiation factors for radionuclides in advance of the definition of the individual components of the waste isolation system, an integral migration test has been started.

In this test, both the near and far fields are simulated in the laboratory by experiments in which water passes through columns containing consecutive beds of material. These simulate, under geologically realistic conditions, the host rock, backfill, overpack/canister and vitrified waste.
The vitrified waste consists of a typical glass composition to which has been added all the elements expected to be present after 1000 years, including all the principal radioactive nuclides. The host rock and backfill constituents will be an experimental variable in order to represent typical potential disposal sites. The groundwater conditions and flow rates are a further experimental variable, within the realms of geological reality.

A simple pilot experiment is in progress to provide experimental data for these design parameters which cannot be estimated and the rest of the main experiment is being designed. The pilot experiment consists of 5 short columns containing granite, concrete and corroded iron.

A leachate to flow through the column is being prepared by contacting an agitated batch of groundwater with simulated vitrified high level waste doped with Np, Pu, Am and selected fission products. The rate of leaching of the glass is being monitored. So far α-activity has been leached out at constant rates which vary according to the nuclide. Neptunium is being leached at a relative rate about 30 times that for plutonium and americium. This leachate will be fed to the 5 columns during the early part of 1982.

2.2.1.5.5. Study on the Scale Effect in Radio-nuclide Transport in Fractured Media

Contractor : CEA, Fontenay-aux-Roses (147-80-7 WAS.F)

It has been shown recently that, in porous or fractured media the "dispersion coefficient" representing the spreading of a plume of tracer in the medium, is not a constant, but varies with time (or with the average travel distance of the tracer).

In fractured media, the increase in the dispersion coefficient may- or may not - reach an asymptotic value depending on the interconnection of the fractures. It is important to determine if an asymptomatic value will be reached, and to measure the value of the dispersion coefficient at a distance where the asymptomatic value has been reached. Alternatively, it would be of great interest to be able to predict this asymptomatic value from measurements of geometrical properties of the fractures.

To study this phenomenon theoretically, an equivalent continuous medium approach is now used. Monte Carlo simulations of the velocity field in a random medium are generated, and the convective movement of particles (representing radionuclides) is studied, as a function of time.

2.2.1.5.6. Study of Physico-Chemical Behaviour of Actinides

Contractor : CEA, Fontenay-aux-Roses (210-81-7 WAS.F)

The work was centred on the fixation of transuranium elements, especially Np-V, on argillaceous barriers : bentonite, illite, attapulgite. For the ionic species NpO_2, PuO_{2++}, PuO_2^{++} , PuO_2OH^+, the Kd was measured, for hydroxilised species, $Pu(OH)_4$, $Pu_n(OH)$, the Ka was measured. Sorption equilibria were obtained after 24 hours ; desorption remains to be done.

Particular attention has been given to the concentration of the transuranium elements and its influence on the experimental results.

A bibliography on the subject has been completed and was followed by an experimental study of neptunium-carbonate complexes.

Experimental part

Retention measurements have been done by a static method. Clay is brought into contact with active water. The suspension is agitated for 24 hours. After decantation, the solution is filtered on millipore 0.22 um and analysed. Distilled water is used. Retention is measured at pH 5-9. The results are expressed in curves K_D = f (pH). Complementary measurements are made at pH = 11 for water-bentonite. Adding the transuranium element is done at acid conditions to compensate for the basicity of the clay. Calculations show the ionic strength of the solution to be about 10^{-3}M. Used quantities of neptunium are small from a thermodynamic point of view, but very high as compared to real leachates. The advantage of spectro-photometric detection is to be precise about the nature of the species in the solution. Equilibrium concentrations are around or below 10^{-6}M. Plutonium is dosed by liquid scintillation. For the results, see Figs. 37-40.

Experimental study of complexation of Np-V, by carbonates is done by optic absorption spectrophotometry - using a spectrophotometre CARY 17D with its modified pressure compartment inside a glove box.

Absorption for solutions at various carbonate concentrations is given as a function of pH in the spectral band 900-1000 nm. At least one Np-carbonate at pH above 7 has been detected ; its nature remains to be verified. With the exception of U-VI carbonate complexes, the other transuranium elements are very badly known in this respect.

It is the first time, to our knowledge, that a systematic study is undertaken about the retention of actinides, taking into account their chemical species. The results obtained confirm the difference in behaviour between two differnet oxidation states of plutonium. The existance of carbonate complexes - shown for Np V - is indicative for the importance of the role of this complexing anion in the chemistry of actinides as applied to their interactions with artificial barriers.

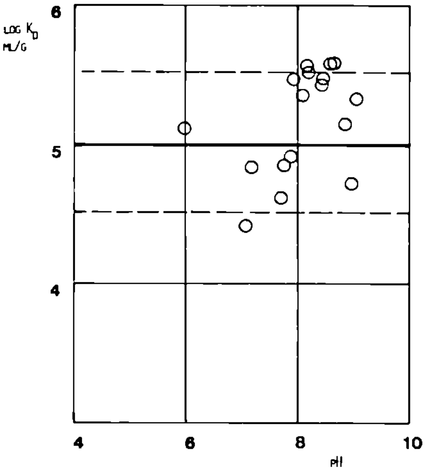

FIG. 1 Pu (IV) BENTONITE

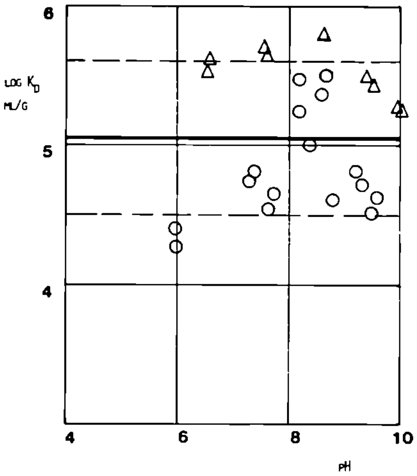

FIG. 2 P_u(IV) ATTAPULGITE

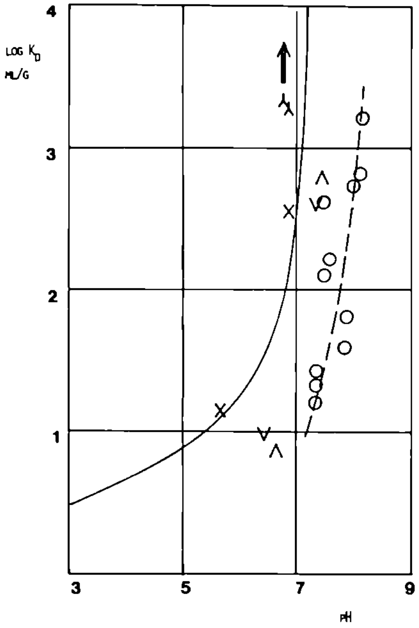

FIG. 4
Np(V) BENTONITE

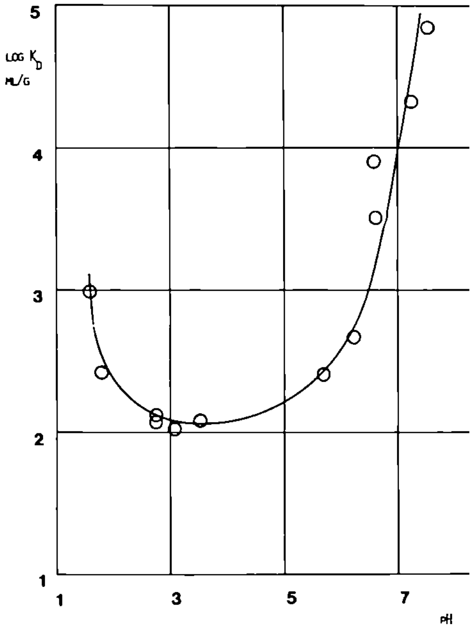

FIG. 3 P_u VI BENTONITE

2.2.1.5.7. Research on the Retention of Radio-elements on the Fracture Surfaces of Granite

Contractor : CEA, Fontenay-aux-Roses (203-81-7 WAS.F)

A special apparatus has been developed to measure sorption - desorption equilibria on the surface planes of rock samples taken from drillings of Auriat granite. To the crystalline water (Volvic water) the radioelements under study (at a pH 7 and as a cationic solution) are added : Sr^{++}, Cs^{+}, NpO_2^{+}, or as hydrolized suspension (hydroxide or insoluble polymer) Pu and Am. This active water then circulates in a closed circuit system over the fracture surface. Regular sampling of the water permits to verify the evolution of the concentration of the radioelements until an equilibrium is attained between these elements in the water phase and on the fracture surface. This equilibrium takes about twenty days for granite, whereas for more permeable rock samples (chalk) it takes longer. When this equilibrium is attained, a desorption cycle follows by circulating inactive water over the fracture surface and measuring the uptake of activity in the water. In all these cases, proper account is made of the possible attachment of radioactivity to the walls of the experimental set-up.

For different samples of granite which have been studied, the retention decreases in the following sens Pu, Am > Cs >Sr >Np.

For Pu and Am one measures Ka (surface adsorption coefficient) variable from 200 to 300 cm^3/cm^2. This retention is not markedly influenced by the state of the granite surface. Although the major part of these elements is adsorbed on the surface, there remains a non-negligible part in the water phase.

For Sr and Cs one obtains Ka (strontium) 0-65 cm and Ka (cesium) 30-200 cm. This retention is sensitive to the state of the surface. On an inaltered surface from a spontaneous crack, one measures a quasi zero retention (Ka≈0) for strontium and a weak retention for cesium (Ka ≈30). Fissures or cracks covered with alteration products of sericite or chlorite type retain these elements better with Ka (strontium) 13-65 cm and Ka (cesium) 50-200 cm.

The equilibrium concentration is very sensitive to the initial isotope concentration in the water : a concentration of 0.1mg/ litre of stable Cs diminishes Ka measured on tracer isotope Cs-137 with a factor 5

The retention of Np is characterised by weaker Ka coefficients, varying from 5 to 10 cm according to the type of fracture. As regards reversibility and Cs adsorption is only partly reversible. Reversibility of the actinides is still under study, although preliminary results are available, and only show partial reversibility.

2.2.1.6. MATHEMATICAL MODELLING

The aim of this task is to develop computer codes with which the behaviour of the stored waste products and its impact on the various barriers can be simulated. It concerns in particular codes on :

- Temperature distribution
- Geomechanical stress
- Groundwater movement
- Radionuclide migration, etc...

The coupling of the models should allow to improve the degree of certitude of the simulation and the sensibility to various parameters.

In 1982 a review study, to be performed by Atkins R&D (UK), will be carried out on the various codes available.

2.2.1.6.1. A Review of Mathematical Models for Characterisation of Geological Formations in View of Disposal of Radioactive Waste

Contractor : Atkins R&D, Epsom (219-81-7 WAS.UK)

This review study, to be started in 1982, will establish a directory of existing computer programmes on the various aspects of characterisation of geological formation in view of disposal of radioactive waste.

It is also anticipated that a need may be shown for the modification of certain programmes and for the development of new programmes to cover aspects for which the existing programmes are inadequate.

2.2.1.6.2. Improvement of the Computer Codes for the Calculation of Temperature Fields after Disposal of High-level Radioactive Waste

Contractor : KfK, Karlsruhe (266-80-7 WAS.D)

Several activities were pursued concerning the further improvement and application of the set of programmes available for near-field and far-field temperature calculations.

Work was performed in close collaboration with the "Institut für Elektrische Anlagen und Energiewirtschaft" of the Rheinisch-Westfälische Technische Hochschule (RWTH) Aachen.

Development of a flexible 2-dimensional programme for axisymmetric geometry

The coarse mesh-computer code FAST-CUBSIT which works in cartesian x, y, z-geometry has been modified for the calculation of axisymmetric temperature fields. The modifications which also enclose the temperature-dependent behaviour for the thermal properties of salt were theoretically derived and have been recently implemented. Algorithms for boundary conditions of first and second kind were adapted. To take into account air filled gaps, for instance around a waste canister column, the code was extended to consider radiative and convective heat exchange.

Testing calculations for simple configurations show good conformity of the results of the new programme FAST-RZ in comparison with other analytical or numerical calculations. The latter were done by the formerly developed finite difference code TEFELD.

Development of a programme system for the consideration of realistic storage field configurations

This system will be based on the complex use of special versions of the FAST-BEST 3D-code (for near-field calculations) and the FAST-STEP code (originally developed to describe a test emplacement of some few waste blocks). The necessary modifications of these codes were performed to be able to handle space - and time-dependent boundary conditions (delivered by RWTH Aachen).

As a further preparation for the assemblage of the programme system, several versions of a subroutine NETZPL were developed and tested for the generation of computer plots of the geometric models and spacial discretizations used in the programmes CUBSIT,FAST-BEST, CUB 3DG, GERA and ASYTE/KA, which are being used on the IBM-computer at KfK.

2.2.1.6.3. Development of Methods of Computation for Thermomechanical Interactions of the Salt and Borehole casings or Waste Canisters

Contractor : KfK, Karlsruhe (266-80-7 WAS.D)

The MAUS computer code

As a meaningful enlargement of the programme MAUS (Mechanical Analysis of Underground Storage) a plane stress version (MAUS-ES) was written.

In opposition to the current definition of plane stress, with zero stress in the direction z (normal to the plane under consideration), it is possible with MAUS-ES to take account of any proper stress in that direction. By that means a better modelling, for example of underground tunnels, can be achieved.

Founded upon the theoretical background that was obtained during the development of MAUS-ES, a further development of the former plane strain version MAUS-ED was carried out. It is possible now, to take account not only of the strain "zero" in the direction normal to the plane, but also of any meaningful strain in that direction. By that way any proper stress in direction z can be indirectly modelled.

By modelling an underground tunnel the hydrostatic state of stress can be herewith more accurately represented than with the former MAUS-ED version. This hydrostatic state of stress is usually the startpoint for the stress relaxation that occurs as an effect of the underground cavity excavation.

Thermomechanical model calculation for the HLW-borehole

Thermomechanical model calculations for in-situ heating experiments with the standard probe and for a HLW-borehole were performed using the ADINA finite element code.

This work was carried out at KfK in collaboration with the Institut für Elektrische Anlagen and Energiewirtschaft of the RWTH Aachen.

The rock salt was assumed to be homogeneous with temperature dependent material properties.

The overall results of the finite element model agree satisfactorily with the experimental data. In figure 41 the calculated and measured radial displacements are shown.

Typical results of the calculations were the borehole closure and the stress field, in particular the pressure loading on the waste canisters and the buffer material.

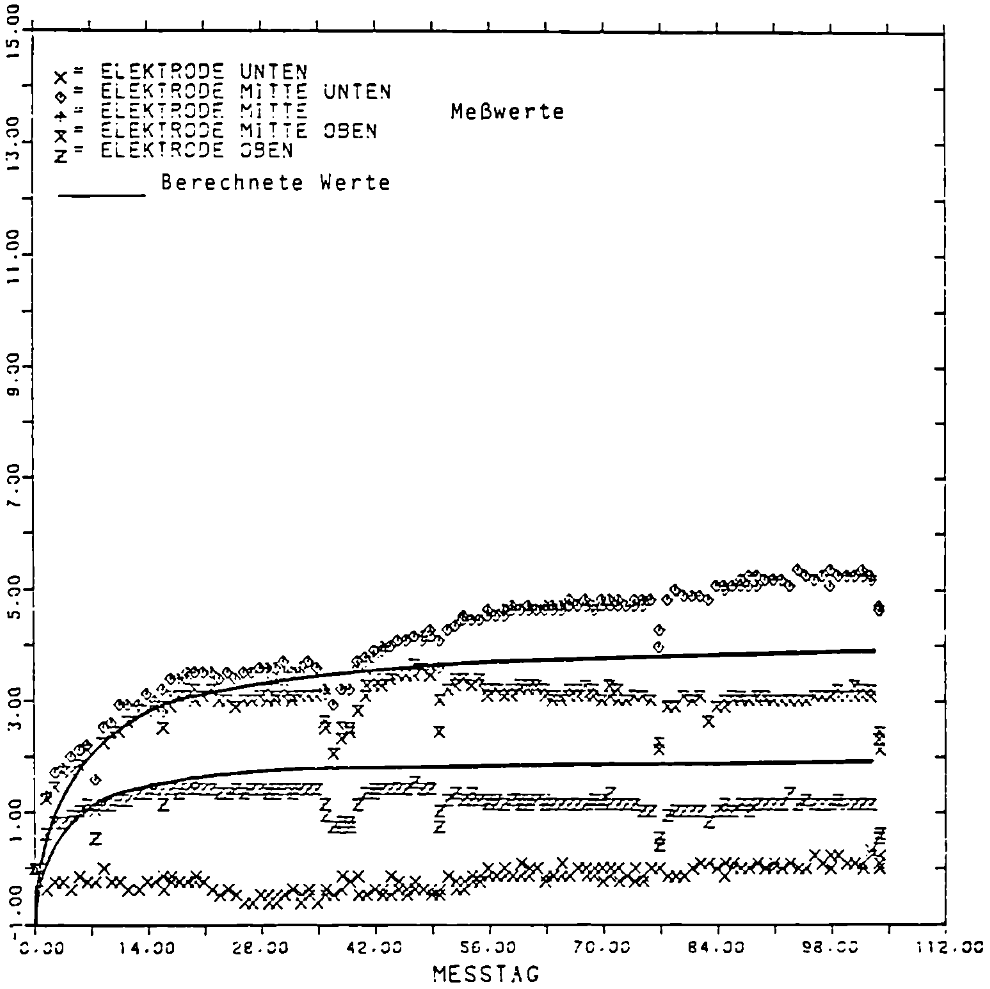

Fig. 41: Borehole diameter change with time (standard probe)

2.2.1.6.4. Development and Application of Rock-mechanical Computer Codes on the strength Behaviour of heated and unheated Rocksalt

Contractor : GSF, Clausthal-Zellerfeld (130-80-7 WAS.D)

The temperature programme CUBSIT and the rock-mechanical programme MAUS were developed further and possibilities of treating fracture formation in rock numerically were investigated by the working group from the Rheinisch-Westfälische Technische Hochschule Aachen.

A material law is being developed by this working group for salt fines which are under consideration as backfill material.

In an applied calculation of the programme CUBSIT and MAUS, the required safety distance between a storage area for high-level waste and neighbouring carnallite rocks was specified.

The results should serve as a tool to fix the position of a disposal area within a salt dome.

Trial calculations of the stress and displacement fields resulting from borehole convergence have shown that varied choises of the boundary conditions for the disc model exert a strong influence upon the calculated convergence velocity. Since this may possibly explain why previous calculations by other authors reproduced the measured rates of convergence rather inaccurately, the problem is to be further investigated. The stress field shows only negligible deviations for varied boundary conditions. In the well-known manner, following sinking of the borehole, a tangential-stress maximum moves into the rock in the radial direction where it flattens out.

Preliminary calculations which describe the behaviour of the borehole during a frac-test, have shown high tensile stresses in the tangential direction at the edge of the borehole. This clearly shows that the creep law and fracturing criteria also have to be investigated for stresses in the tensile region.

2.2.1.6.5. Development of Computer Programmes for Temperature Analysis and Deformation Behaviour in a Salt Dome

Contractor : ECN, Petten (226-81-7 WAS.NL)

Temperature analysis programme (TASTE)

The application of finite element or finite difference programmes for detailed 3D thermal analyses encounters considerable problems due to the complex geometry of the burial area and the relatively small areas in which significant local temperature effects occur. Therefore the computer programme TASTE (Three-Dimensional Analysis of Saltdome Temperatures) has been developed. TASTE is based on an analytical model of a continuous time dependent point heat source in an infinite solid of homogenuous isotropic material with temperature independent properties.

Deformation analysis programme (GOLIA)

It is generally accepted that the creep of the salt is a dominating mechanism in the deformation behaviour of a salt dome. Therefore creep deformations have to be taken into account in the deformation analyses. To perform creep-deformation analyses several large computer programmes are available. One of the disadvantages of these large computer programmes is the long running time needed for creep analyses. Therefore ECN has selected a very simple special purpose computer programme to perform the creep analyses in salt domes. This special purpose computer programme is GOLIA-RK. In order to use this programme for salt dome analyses a number of extensions are necessary. Until now the following modifications have been made :

- Implementation of volume forces.
- Extension of the number of different materials.
- Implementation of the geothermal gradient.

2.2.1.6.6. Mathematical Modelling of Groundwater Flow and Radio-nuclide Migration in Crystalline Rock

Contractor : UKAEA, Harwell (209-81-7 WAS.UK)

Grounwater flow and heat transfer

The finite element computer programme NAMMU for modelling fluid flow in porous media, with heat sources, has reached an advanced stage of development. A user's guide has been written to enable new users to tackle their own modelling problems.

NAMMU has been used for regional groundwater studies in two-dimensional sections of granitic intrusions and of the rock/clay strata which underlie Harwell.

Radionuclide migration

The computer programme NAMTAR is being developed to model the migration of radionuclides released from a waste repository in rock. All the important physical processes affecting migration will eventually be included and are being added one by one to the programme. They include not only advection by the water flowing through the rock and radioactive decay, but also molecular diffusion, hydrodynamic dispersion by the flow, complex chemical effects (collectively known as sorption) and diffusion into the non-flowing water in the microscopic rock pores. To date only advection and dispersion are included in the programme.

NAMTAR is based on methods different from conventional finite element or finite difference schemes. The approach is quite new, though it has been used recently with some succes in oil reservoir studies. It is a relative of the Particles-in-Cell and Discrete-Parcel-Random-Walk methods but with some apparent advantages. Its basic feature is the use of concentration contours of marked particles which are cracked in time.

Flow in fracture networks

In many rock masses the groundwater flow occurs predominantly through a system of interconnected fractures. If the interconnections are sufficiently numerous and the length scale of the flow is large compared to the fracture dimensions then a porous medium description can be used. However, if either of these conditions does not hold then a description in terms of laminar flow down a network of discrete fractures is appropriate.

Work has started on the application of percolation theory to the flow of water through networks of fractures. Using these techniques it is possible to look at connectivity, flow and dispersion in fracture systems.

So far the connectivity of fracture networks in two-dimensions has been examined for some idealised probability distributions for the fracture lengths and orientations. Also some progress has been made towards estimating the length scale at which a porous medium description holds.

2.2.1.7. REPOSITORY DESIGN

Within the previous R&D programme (1975-1979) various conceptua design studies were carried out for waste repositories in the geological formations considered, respectively clay, salt and granite.

Under the current programme two of such design studies are continuing. It concerns a study performed by CNEN(I) for a waste repository in a clay formation in which two concepts will be considered, respectively the mine gallery and the deep borehole concept.

The other study, carried out by ECN (NL), concerns a further optimisation of the one level mine concept in a salt dome.

2.2.1.7.1. Engineering Studies for a Waste Disposal Facility in Clay

Contractor : CNEN, Casaccia (225-81-7 WAS.I)

The study is to determine :

- Parameters for site selection ;
- Subsoil engineering problems ;
- Costs of constructing and running of the repository ;
- Suitable geometry of the repository and evaluation of its technical feasibility.

The basic hypotheses for the repository have been defined up to date, in particular :

- The general characteristics of clay formation ;
- The net of boreholes to be drilled from surface or from a subsurface cavity, according to two types of repositories. Furthermore thermal numerical codes have been adjusted in order to predict temperature distribution in the repository, according to a different geometry of borehole nets.

2.2.1.7.2. Design Work on a Repository in a Salt Dome

Contractor : ECN, Petten (226-81-7 WAS.NL)

The work is focussed on a one layer mine concept with a capacity sufficiently large to hold the high level waste resulting from 25 PWR's of 1.000 Mwe during 40 years of operation .

For a given geometry, consisting of galleries with deep boreholes (300m) for disposal of high level waste and a separate bulk disposal of low and intermediate level waste, a workplan, as well a cost estimate for the shaft sinking and subsequent mining operations, have been worked out.

The global temperature analyses for the updated repository design shows in the case of an instantaneous disposal a gradual temperature rise in the centre of the repository for the first 100 years. The maximal global temperature reaches 85°C. Later on, this max. global temperature slowly decreases. After 1.000 Years the max. global temperature is about 65°C. Local temperature analyses have been performed to investigate the influence of the canister diameter on the max. local temperature. The reference analysis was performed on a continuous stack of 300 m length with a diameter of 20 cm ; and an initial heat production of 277 W/m. The larger diameter canisters were spaced in such a way that the average initial heat production was also 277W/m borehole length. Compared with the reference analysis the 43 cm canister shows an increase of the maximum local temperature of about 32°C.

Detailed global deformation analyses are at this moment being performed. Before starting these analyses a simplified global displacement analysis has been done. In this analysis the total heat source of the disposed high level waste is concentrated in a point source. The heat input into the salt dome resulting from this point source minus the dissipated energy at the surface, was used to calculate the maximum thermal expansion volume. The expansion results in surface displacements. These surface displacements can easily be analysed assuming stress free expansion. An upper limit for the surface rise was calculated to be about 1.8 m.

Local displacement analyses have been performed for the stresses and strains around the pressure probe of the heated experiments in the dry drilled 300 m deep borehole .

These analyses in a maximum pressure on the probe of about 40MPa. Furthermore a sensitivity analysis was done to investigate the influence on this max. pressure of some of the important parameters.

2.2.1.8. SAFETY ANALYSIS

The safety analysis on the Mol site (clay), started under the previous programme, is being continued.

Furthermore, BRGM (F) has started a geoprospective study on a repository site (granite) and the HMI (D) is going to examine the safety of disposal in a salt dome. Apart from these limited projects the Commission has launched by the end of 1981 a coordinated action on the evaluation of the performances of geological formations in view of disposal of high level wastes. (See following paragraph 2.2.1.9.)

2.2.1.8.1. Safety Analysis on Waste Disposal at the Mol Site

Contractor : SCK/CEN, Mol (144-80-7 WAS.B)

Because of their relative importance, assessed by Fault Tree Analysis, three geological containment failure modes were selected to be worked out more in detail and to be considered for consequence analysis : 1) faulting, 2) glacial erosion, 3) human actions.
Possible scenarios for a hypothetical faulting through the repository, initiating a release of radionuclides into the groundwater were described.

The pathway to man taken into account were : a) ingestion of well water by pumping from aquifers under - and overlying the Boom clay and b) inhalation of resuspended dust of well water used for irrigation purposes. In the case of release to the neogene (upper) aquifer, maximum radiation doses would be in the range of 10^{-2} - 10^{-4} rem/yr. It must be borne in mind that these doses so far calculated result from conservative assumptions and in the context of the scenario could effect only a very small number of individuals. In the case of release to the aquifer underlying the Boom clay doses so far calculated for the ingestion pathway are in the order of 10^{-3} - 10^{-4}rem/yr for a maximum exposed individual. For the inhalation pathway doses are about 3 orders of magnitude lower.

A possible glaciation caused release scenario has been worked out in more detail and special attention has been drawn on the evolutionary aspects of such scenario.

The repository Evolution Analysis, based on a deterministic scenario caused by natural degradation of the repository elements has been launched. The system analysis is now underway and first assessments of the importance of the assumptions for the geometry of the release source have been made.

The study is performed in close collaboration with the JRC Ispra.

2.2.1.8.2. Geoprospective Study of a Repository Site

Contractor : BRGM, Orléans (222-81-7 WAS.F)

This study aims at developing a methodology for the definition of natural evolution scenarios for a selected landscape. Such an evolution results from two kinds of factors :

(a) Those related to "internal geodynamics" (deformations, faults, block displacements), and ;
(b) Those related to "external geodynamics" (erosion, sedimentation sensu lato).

In a further step, such a methodology could be applied to specific potential disposal sites.

The work carried out during 1981, concerned the identification and qualitative analysis of the factors to be taken into account and the quantification of these factors with the understanding of their functioning.

Volcanic activity

French volcanic areas of the Neogene age were screened and attention was focussed on three of them :

- The Mont-Dore volcano : eruptions occured at a relatively recent date (0.2 million years) ;
- The "Chaîne des Puys", which is linked to a recent graben structure, the Limagne graben ;
- The Bas-Vivarais volcanoes, which were active at a very recent date (35,000 to 10,000 years B.C.) and which are linked to thermal springs.

All three systems can be considered as potential risk zones for the time spans selected.

Faulting and fracturing

Work is in progress to assess the possible influence of tectonic movements upon the permeability of a fractured rock formation.

The study of these phenomena includes a collection of data on recent tectonic history, and implementation of geodynamics and rock mechanics (fracture behaviour) models.

Specific applications to selected geologic area (Armoric Massif and Parisian Basin) will be performed later.

Seismic activity

A collection of literature data allowed the characterisation of :

- Earthquake occurrence probability for different time spans, using statistical distributions for historically recorded

earthquakes (time-span 0-3,000 years), geological data and geodynamic models for longer periods (time-spans 0- 10,000 years or 0-100,000 years).
- Effects of earthquakes at great depth (1 000 m deep).

Field results being scarce, a collection of literature data and theoretical considerations lead to the conclusion that the amplitude of seismic movements decreases and that there is a noticeable change in the frequency spectrum with depth.

Diapirism

It was decided to include this kind of phenomena, given its importance for salt dome disposal. The aim is to get estimates of the uplift velocity of the formations.

Climatology

Literature data collection is in progress, with special attention paid to "glacial cycles".

Sedimentary erosion

Determination of erosion velocity of a given rock mass is performed taking into account :

- The transportation of the eroded materials
- The places of re-sedimentation
- The eroded site itself

The velocity of the erosion seems to be correlated with the topography, to a greater extent than with the lithologic nature of the outcropping rock, and even with the climate. Erosion velocity can be estimated at 50 mm per 1.000 years for hills and plains, and at 500 mm/1.000 years for mountains.

2.2.1.8.3. Safety Study for the Disposal of Radioactive Waste in the Salt Dome at Gorleben

Contractor : Hahn Meitner Institut , Berlin (270-81-7 WAS.D)

The objective of the study is to prove the feasibility of site specific and comprehensive safety study for the disposal of radioactive waste in a salt dome. "Site specific" means, that the analysis relates to the German disposal concept for the site Gorleben and is using as many basic data from that site as possible. This does not reduce the significance of the results also for other sites of similar geological formations. "Comprehensive" means, that the analysis includes the whole field of site evolution and release scenario development up to assessment of the consequences on man. The study will be focussed on the identification of release scenarios and calculation of corresponding consequences for the post-sealing period of the repository.

2.2.2. DISPOSAL INTO THE SEABED

Ocean sediments have also been considered as a possible option for waste disposal. In reference to the division of the activities in this field by the NEA Seabed Working Group, the following research areas are covered in the CEC programme:

- Sediment and rock : Behaviour of sediments under elevated temperature and sorption characteristics of the sediments are examined.

- Physical oceanography : Research is centered on deep sea currents important for sediment disposal, radionuclide transport and dispersion processes excluding the biological chain.
- Engineering studies : Main aspects of the feasibility of waste disposal operations are dealt with from port facilities through to final in-sediment position and verification of the waste package isolation system.

The research projects have in most cases started only at the end of 1981, so that little report can be given on results of the work. In the following the different research programmes are therefore mainly presented.

2.2.2.1. Properties of Ocean Sediments in relation to the Disposal of Radioactive Waste

Contractor : NERC/IOS, Wormley (257-81-7 WAS.UK)

The objective is to assess the properties of sediments at possible ocean disposal sites in relation to the emplacement of canisters of radioactive waste and the retention within the sediment of radionuclides eventually released from the canisters. In particular the aim is :

- To discover which sedimentary conditions offer the optimum physical situation for emplacement ;
- To investigate the chemistry of sediments to establish both their chemical and mineralogical character and their capacity to restrain or enhance the migration of radionuclides released within them ;
- To study the behaviour of natural actinides in pelagic sediments as a model for the phenomena to be expected with the eventual release of transuranic elements from the waste.

Research on physical properties of sediments

On Discovery Cruise 118 (February - March 1981) a range of different sediment types was collected using a Kastenlot corer (15 X 15 cm square section) with a 2 m barrel. About one hundred 75 mm and 50 mm diameter samples were taken for consolidation and permeability tests. Results from these tests have demonstrated the wide range of permeability and void ratio depth-profiles that may exist in pelagic sediments ranging in lithology from nearly pure carbonate oozes to deep-sea clays. The permeability has been found to range from 10^{-7}m s^{-1} to 9 X 10^{-9}m s^{-1} with no overburden and from 10^{-8}m s^{-1} to 10^{-10}m s^{-1} at a void ratio equivalent to a depth of approximately 130 m. Some horizontal permeabilities exceed vertical permeabilities by a factor of four in adjacent samples. Such a large anisotropy is expected to have a profound effect on porewater speeds and directions of flow.

Geochemistry of sediments and chemistry of natural actinides in sediments

Work to date has concentrated on Atlantic red clay sediments. Accumulation rates determined on material collected from the Nares Abyssal Plain, close to the areas favoured by the NEA Site Selection Task Group to demonstrate that lateral sediment redistribution, including turbidity current flow, has been an active process in this region over the past few hundred thousand years and has given rise to considerable vertical or horizontal variability on a local scale in the areas studied. This interpretation is supported by corresponding fluctuations in the solid phase composition and pore water data from the same cores. It should be emphasized, however, that operational constraints prevented sampling in areas which have been identified as most suitable on the basis of geophysical measurements. Examination of uranium and thorium isotopes in four size fractions separated from bulk samples of Nares

Abyssal Plain red clay of an inferred pelagic character revealed no clear relationship between specific activity and particle size (surface area). Work has also commenced on a core from the Cape Verde Abyssal Plain area. This core shows a red clay overlying two separate carbonate-rich turbidites which are difficult to account for at the depth (6 km) and from the perceived topography. Besides determination of clay accumulation rate, work has concentrated on the possible geochemical controls operative on a marked (X8) uranium enrichment at the interface between the upper and lower turbidites.

2.2.2.2. Migration Processes in Marine Sediments caused by Heat Sources

Contractor : Delta Institute for Hydrobiological Research,
Yerseke (258-81-7 WAS.NL)

The investigations concern simulation experiments with the objectives to study diffusion and convection processes in point-source heated sediments and the changes of sorption capacities of marine sediments due to elevated temperatures.

Investigations on small scale were started to determine the heat transfer factors in sediments as being dependent on different sedimentary composition (grain-size and sedimentological composition). The first experiment was executed with "model" sand of one grain-size (150-210 μm) and free of impurities. This water saturated sediment was heated with a 50 Watt point-source, automatically regulated at 50°C.

The first experiments do not yet allow conclusions on heat-transfer coefficients, but they are used to test the proper equipment relative to model calculations.

The model calculations for the experiments and the interpretation of the results to the sea-bed situations have also started

2.2.2.3. Sorption and Migration of Radio-nuclides through Ocean Sediments

Contractor : CEA, Cadarache (259-81-7 WAS.F)

To provide data required to assess the feasibility of emplacing radioactive wastes in sub-seafloor geologic formations, the sorption of radionuclides by and the migration of radionuclides through samples of Abyssal sediments collected in the North Atlantic will be investigated. The three following works are foreseen :

- Determination of distribution or sorption coefficients (Kd) for several Abyssal sediments of various physical and chemical characteristics, and for Pu, Np, and Am.
- Detailed study of long-lived beta emitting radio-nuclides sorption and desorption to and form typical sediments selected fro the previous study. Influence of experimental conditions.
- Study of the biological availability of radionuclides. The possibility of radionuclide transfer to man through benthic species will be investigated.

2.2.2.4. Lagrangian Current Measurements and large-scale long-term Dispersion Rates

Contractor : Ministry of Agriculture, Fisheries and Food,
Lowestoft(255-81-7 WAS.UK)

For several years now oceanographers from the Woods Hole Oceanographic Institution (WHOI) and University of Rhode Island (URI) in the United States have been deploying long-range neutrally buoyant acoustic floats called SOFAR floats. To date most exercises have taken place in relatively small areas of the western half of the North Atlantic and the tracking stations have been land-based. The unique features of the system are the long-ranges (1,000-1,500 miles) and the long-life (12-18 months) that have been possible.

In the last two years listening-stations have been developed which can be moored at sea and left to record signals from the free-drifting floats. A suitable spaced array of such autonomous listening stations (ALS's) would make it possible to track flocks of SOFAR floats as they drift anywhere in the North Atlantic. Thus far only prototype ALS's have been used but WHOI is now about to embark on a full-scale research programme using "production-line" equipment that will last for 5 years at least.

The results of the first phase of the WHOI/URI programme have demonstrated that the SOFAR floats are the only means presently available of building-up population statistics relating to large-scale, long-term dispersion processes.

An extension of these experiments to the eastern basin of the North Atlantic is of real scientific interest. The Institute of Oceanographic Sciences, Wormly, UK, will build its own

ALS's of WHOI design and deploy its own SOFAR floats. The outcome of this work will be much improved long-period, large scale dispersion rates for the North Atlantic ; such information is essential to any meaningful prediction of long term dispersion of radioactive polluants in the ocean.

By contributing to this project through equipment and ships time, the Lowestoft Laboratory wishes to share in creating a better understanding of mixing in the ocean.

The construction of SOFAR floats will start in 1982.

2.2.2.5. Feasibility Study of the Offshore Disposal of Radioactive Waste

Contractor : Taylor-Woodrow Constr. Ltd., Southall
(256-81-7 WAS.UK)

The study concerns the disposal of vitrified waste in drilled holes in ocean sediments. It is envisaged that the disposal operation would be carried out from a floating platform anchored in water depths in excess of 4.000 m. The structure may be of the semi-submersible variety, having a low water plane area and with most of its bulk beneath the waves, in orde to provide a stable working platform. Such a structure may provide both drilling and disposal capabilities as well as temporary shielded storage for the radioactive material within its massive hull.

The aim of the study is to carry out an engineering appraisal of the disposal of radioactive waste in this manner with a view to establish technical and operational feasibility and giving guidance on the economics of installing and running the scheme.

Special consideration will be given to the following aspects of the scheme :

- Environmental and geotechnical constraints ;
- The safe movement of radioactive material from the land and onto the platform ;
- The problems of forming suitable holes in the sea bed sediments
- Lowering and emplacement of waste canisters ;
- Backfilling ;
- Station keeping ;
- Operational logistics ;
- Safety of personnel and recovery of dropped or damaged canisters in the event of system malfunctions.

The programme of work is broken into three phases. The first phase, due for completing in mid-1982, will encompass the establishment of reference criteria and preliminary evaluation of alternative schemes. The second phase will involve a state of the art study of the various techniques involved in the disposal operation and will lead to the selection of preferred options. The third phase will develop outline designs to enable cost estimates to be made.

The first phase is well under way and progress has been made in identifying the environmental and geological conditions for the possible sites and also in identifying the critical areas for subsequent study. The initial work has confirmed that with regard to most aspects, the logistics of disposing of the required quantity of vitrified waste economically and safely are of greater significance than the technical problems.

2.2.3. DEVELOPMENT OF SITE ASSESSMENT TECHNIQUES

Under this chapter are listed the projects concerning the development, improvement and/or testing of exploring technique or methodologies for the assessment of the suitability of possible sites for the disposal of radioactive waste.

2.2.3.1. Development of Techniques for the Assessment of Geological Formations

Contractor :NERC /IGS, Harwell (128-80-7 WAS.UK)

A thermal infra-red linescan survey of part of the Altnabreac research area was undertaken early in the year to assess the technique for detecting springs and groundwater seepages. It is necessary to fly the survey in the winter or early spring in order to benefit from the maximum temperature contrasts between groundwater discharges and surface waters. Because non-ideal weather conditions prevailed at the time of the survey, only a part of the total proposed area was flown. Despite the difficulties, over one hundred and forty springs and seepages were identified and many of these were visited for the collection of hydrogeochemical samples. It is intended to complete the survey of the Altnabreac area early 1982.

Hydraulic testing, using the sinusoidal pressure wave method, has been carried out in the UKAEA operated test quarry in the Carmenellis granite, Cornwall. The method entails generating a sinusoidal variation of pressure in a small packered off section of a borehole and detecting the signal in a packered off section of another borehole. By moving both the source zone and receiver zone various orientations can be measured. The trials were successful in showing that measurements can be carried out using comparatively simple equipment and provide the opportunity to measure directional hydrogeological properties over considerable distances. Detailed analyses of the results is still being carried out.

An operational geochemical probe is being developed by the Electrochemistry Research Laboratories of Newcastle University. The objectives of this work are to further develop the NERC prototype probe to meet the parameter specifications and to construct operational probes and associated electronic control equipment in the field. It is hoped that the probe will meet the following parameter specifications in deep borehole conditions :

pH	3.5 to 9.5 ± 0.05
Chloride	0 to 1000 mg/l ± 1 mg/l or 1000 to 30.000 mg/l ± 50mg/l
Conductivity	0 to 1000µ ± 1 µs or 1000 to 50.000 µ ± 50 µ s
Eh	- 400 to + 600 mv ± 10mv
Dissolved O_2	0 to 12ppm ± 0.01 ppm
Temperature	5 to 50°C ± 0.05°C

In addition it is expected that the probe will be able to function for up to a week of continuous readings without the need for recalibration. To date work was concentrated on the suitability and reliability of the alternative pH sensors , the stability with time of the oxygen and chloride ion sensors and on the overall design of the probe and associated electronics.

2.2.3.2. Remote Assessment of Permeability/Thermal Diffusivity of Ocean Sediments

Contractor : University College of North Wales, Menai Bridge
(263-81-7 WAS.UK)

The aim of the proposed research to be started early 1982 is t examine the possibility, via series of laboratory experiments, that the permeability - thermal diffusivity of marine sediments can be acquired by geophysical means.

The consolidation behaviour of marine silts and clays will be studied in association with observations of P - and S- wave velocities, electrical resistivity, and thermal conductivity.

A range of sediment samples will be collected from land and marine sites, as well as being prepared artificially, so as to provide a representative collection of initial void ratios consolidation states, and sediment grades. From these observations correlative master curves can be drawn relating the static elastic moduli and permeability to the geophysical quantities.

2.2.3.3. Deduction of Characteristics of large deep Clay Deposits by low Cost Surface Survey

Contractor : CNEN, Casaccia (239-80-7 WAS.I)

The CNEN has started methodological research to verify the possibility to coarsely and quickly foresee some features, especially mineralogy, of deep clay deposits.

The problem was to find possible correlations between lithologically different feeding basins and mineralogy of clay sediments originated from them and accumulated in the near corresponding basins.

A systematic sampling of the existing outcropping inlandward extention of deep clay deposits has been carried out. A total amount of 75 samples have been collected on the whole Italian territory. Because of the complexity of Italian lithology (Fig.42) a production of different clay minerals as well as their clear different geographic distribution can be expected. On the other hand a non negligeable byproduct of this research could be a connection between rock types and kind of derived clay minerals. Through these expected results some predictions can be carried on sedimentary and mineralogic characteristics of deep deposits.

Granulometric data of collected samples are at the moment available.

It has been observed that silty fraction is the most frequent grain size among the analyzed clay samples. This accords with the inlandward location of the sampling position compared with the deeper parts of the clay deposits, presumably displaying thinner grain size.

Furthermore it has been found that :

- Monomineralic clay formations were absent in the explored areas. Generally the clays are characterized by mineralogical association ;
- The mineralogical associations show an evident regional distribution ;
- A certain relation exists between the lithological nature of old feeder basins and the distribution of different types of clay minerals ;

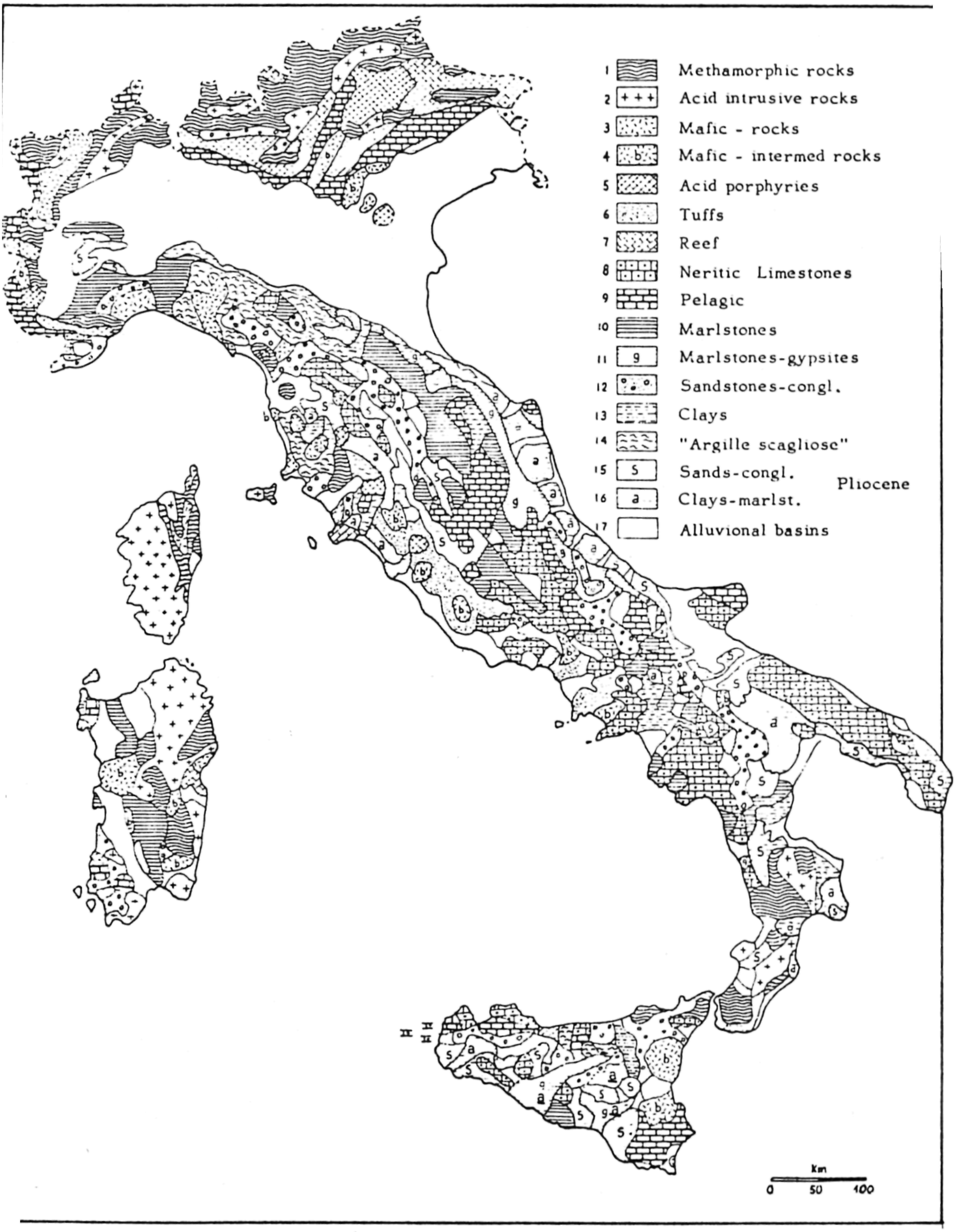

Fig. 42: Simplified lithological map of Italy.

- A set of various types of clay basins have been spotted.

As a result, it is probably possible to foresee some mineralogical and sedimentological characteristics of deep clay deposits from surface surveys.

For sitologic selection of clay basins it is possible, by means of the surface surveys, to apply exclusion criteria, complementary, to those for compiling the "Catalogo delle formazioni geologiche favorevoli allo smaltimento dei rifiuti radioattivi in Italia".

2.2.3.4. Development of a geophysical Methodology for Site Assesment

Contractor : BRGM, Orléans (264-81-7 WAS.F)

The work covered under this project is to develop a borehole probe for deep fracture location, both in the vicinity of and at great distance from a borehole in which the probe will be inserted.

The two deep boreholes previously drilled in the granite formation at Auriat, France may serve as test site.

So far a collection of available literature data has been done concerning the two boreholes. From the obtained result a relationship between fracturing and electrical properties of the rock at depth could be shown to exist.

Progress was also made concerning the data acquisition by the electric and electro-magnetic probes. The improvements were obtained using a computerized data acquisition system set up in the laboratory van. This system will allow digital data acquisition, field graphic data display and data storage on tape.

The schedule for field investigations foresees the following measurements :

- Neutron/gamma logging, electric, sonic logging
- Electromagnetic logging using induction probe for investigations in the range 1-10m from the borehole ("Romolus" and "Eric" probes)
- Implementation of a "dipole-dipole" type electric probe (currently under construction) with a view to investigate the range of some tens of meters from the borehole.
- Measurements between the ground surface and an electrode at 1.000 m depth in the borehole, the surface area having a diameter of 1.5 km centered on the borehole head.

2.2.3.5. Development of a Borehole Probe for Measurement of Permeability and hydraulic Pressure in Rocks with low Permeability

Contractor : CEA, Fontenay-aux-Roses (262-81-7 WAS.F)

When determining the permeability of a crystalline formation, by ordinary borehole tests (pumping tests), several difficulties are encountered due to :

- The presence of the boreholes which perturbates the original groundwater conditions.
- The problem of obtaining a continuous hydrogeological logging of the hole ;

- The low values of permeability, comparable with the uncertainties of the measurements
- The hardly possible re-use of the same borehole for several measurements at regular time intervals.

In 1968, the French consulting company "COYNE & BELLIER" developed and patented the PIEZOFOR device, which allows (in limited cases) the determination of the true hydraulic pressure distribution in a rock formation. However, the maximum depth for use is limited to 150m, and the determination of permeability is at present only qualitative.

The present works intend to improve the capabilities of the PIEZOFOR device. The work programme covers the development of a special borehole equipment with probe to allow the measurement of permeability and water-head without altering the initial groundwater regime and the completion of a 250m borehole followed by a 500 m one for receiving the described equipment.

2.2.4. EVALUATION OF WASTE ISOLATION PERFORMANCES "PAGIS" PROJECT

The Community "Plan of action on radioactive waste"* foresees the examination, at Community level, of measures which could ensure the long term or permanent storage of waste under optimum conditions.

In order to implement this task in the framework of a concerted action, the Commission proposed a project on the iterative safety assessment in step with the advance of the scientific and technical knowledge. The proposal was discussed with the Advisory Committee for the Programme Management of the Plan of Action, which, on the 30th of September 1981, expressed a positive advice on this project. The project (called PAGIS = Performance Assessment of Geological Isolation Systems), covers the disposal of high level waste in four geological formations, namely : clay, salt, granite and sub-seabed. It is to be performed in three successive phases. The objectives of the three phases can be summarized by the following headings:

I. Definition of the components of the repository system,
II. Validation and utilization of the knowledge,
III. Assessment of the risk.

The Commission decided to start the first phase and to launch the action after a short preparatory period. For each of the four geological formations considered, secretary bodies were identified (CEN/SCK for clay, HMI for salt, ANDRA for granite and NRPB for sub-seabed).

The first phase includes : the characterisation of the various components of the multi-barrier disposal system (site, disposal installations, waste form, radionuclide inventory and barriers), the selection of a general methodology, the description of possible release scenarios and the modelling of the various

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* Ref. O.J. no. C51, 18.2.1980.

phenomena responsable for the retention or migration of the radio nuclides through the geosphere and the biospere up to man.

The action will be based on the knowledge acquired so far through both the R&D programme of the Community and the national activities in the field of high level waste disposal ; no development or research work is foreseen.

The secretary bodies will, therefore, collect all available data and models and, with the help of experts select appropriate reference and variant parameters. They also will assist the Commission in obtaining the best possible homogeinity of the approaches and of the analysis for the four geological formations The selected references together with the justification of the choices will be described in a final report. The negotiations of the contracts for implementing the 1st phase are due to start beginning of 1982.

APPENDIX

BUDGET FIGURES°

Total funding allocated to the R & D programme 1980-1984	43.000
Contractual Commitments taken as to 31/12/1981	18.800

° In thousands of European Currency Units (ECU)

LIST OF RESEARCH CONTRACTS

Contract n°	Contractor	Chapter of report
121-80-53 WAS.UK	UKAEA	1.5.2.
122-80-53 WAS.D	HMI	1.5.2.
123-80-55 WAS.F	CEA	1.5.1.
124-80-55 WAS.B	SCK/CEN	1.5.2.
125-80-55 WAS.NL	Univ. Leiden	1.5.2.
128-80-7 WAS.UK	NERC/IGS	2.2.1.1.-2.2.1.4.
129-80-7 WAS.B	SCK/CEN	2.2.1.2.
130-80-7 WAS.D	GSF	2.2.1.2. 2.2.1.6.
140-80-7 WAS.I	CNEN	2.2.1.2.
142-80-7 WAS.NL	ECN	2.2.1.2.
144-80-7 WAS.B	SCK/CEN	2.2.1.1. 2.2.1.3. 2.2.1.4. 2.2.1.5. 2.2.1.6. 2.2.1.8.
145-80-7 WAS.D	GSF	2.2.1.4.
146-80-7 WAS.F	CEA	2.2.1.4.
147-80-7 WAS.F	CEA	2.2.1.5.
148-80-7 WAS.F	BRGM	2.2.1.4.
150-80-7 WAS.F	CEA	2.2.1.4.
151-80-7 WAS.I	CNEN	2.2.1.4.
152-80-7 WAS.I	CNEN	2.2.1.4.
153-80-7 WAS.NL	Un.Utrecht	2.2.1.4.
154-80-7 WAS.UK	UKAEA	2.2.1.4.
157-80-8 WAS.UK	UKAEA	1.6.1.
158-80-8 WAS.D	KfK	1.6.1.
159-80-8 WAS.B	SCK/CEN	1.6.2.
161-80-8 WAS.UK	NRPB	1.6.3.
162-80-8 WAS.D	KfK	1.6.4.

163-80-8	WAS.UK	UKAEA	1.6.3.
166-81-15	WAS.UK	UKAEA	1.3.4.
167-81-2	WAS.B	SCK/CEN	1.2.1.
168-81-2	WAS.D	KfK	1.2.3.
169-81-2	WAS.D	NUKEM	1.2.1.
170-81-2	WAS.F	CEA	1.2.2.
171-81-2	WAS.UK	UKAEA	1.2.2.
172-81-2	WAS.F	CEA	1.2.1.
173-81-2	WAS.D	KfK	1.2.1.
176-81-31	WAS.UK	UKAEA	1.3.3.
177-81-31	WAS.D	KfK	1.3.1.
178-81-33	WAS.D	KFA	1.3.3.
179-81-31	WAS.UK	UKAEA	1.3.1.
180-81-33	WAS.D	KFA	1.3.3.
181-81-31	WAS.F	SENA	1.3.1.
182-81-42	WAS.F	CEA	1.4.3.
184-81-43	WAS.D	KfK	1.4.3.
185-81-43	WAS.UK	UKAEA	1.4.1.
186-81-44	WAS.UK	UKAEA	1.4.1.
187-81-45	WAS.D	KfK	1.4.2.
188-81-44	WAS.D	ALKEM	1.4.2.
190-81-42	WAS.B	SCK/CEN	1.4.1.
191-81-44	WAS.I	AGIP Nucleare	1.4.1.
192-81-44	WAS.F	CEA	1.4.1.
193-81-6	WAS.NL	SML Delft	2.1.3.
194-81-6	WAS.DK	NL RISφ	2.1.3.
195-81-6	WAS.DK	NL RISφ	2.1.2.
196-81-6	WAS.F	CEA	2.1.2.
199-81-7	WAS.I	CNEN	2.2.1.3.
200-81-7	WAS.F	CEA	2.2.1.4.
202-81-7	WAS.F	CEA	2.2.1.3.
203-81-7	WAS.F	CEA	2.2.1.5.
204-81-7	WAS.UK	Mott-Hay Anderson	2.2.1.3.
206-81-7	WAS.I	CNEN	2.2.1.4. 2.2.1.6.

208-81-7	WAS.I	CNEN	2.2.1.5.
209-81-7	WAS.UK	UKAEA	2.2.1.5. 2.2.1.6.
210-81-7	WAS.F	CEA	2.2.1.5.
219-81-7	WAS.UK	ATKINS	2.2.1.6.
222-81-7	WAS.F	BRGM	2.2.1.8.
225-81-7	WAS.I	CNEN	2.2.1.7.
226-81-7	WAS.NL	ECN	2.2.1.3. 2.2.1.6. 2.2.1.7.
227-81-8	WAS.UK	UKAEA	1.6.1.
228-81-8	WAS.B	SCK/CEN	1.6.4.
230-81-35	WAS.D	NUKEM	1.3.1.
231-81-31	WAS.I	AGIP Nucleare	1.3.2.
232-81-53	WAS.D	Fraunhofer Inst.	1.5.1.
233-81-6	WAS.UK	NRPB	2.1.3.
234-81-8	WAS.B	Un.Antwerpen	1.6.1.
235-81-13	WAS.DK	NL RISØ	1.1.
236-81-7	WAS.F	Un. Clermont-Ferrand	2.2.1.3.
238-81-7	WAS.I	CNEN	2.2.1.4.
239-81-7	WAS.I	CNEN	2.2.3.
240-81-13	WAS.D	KfK	1.1.
242-81-13	WAS.UK	Un.Aberdeen	1.1.
243-81-13	WAS.UK	UKAEA	1.1.
244-81-15	WAS.F	CEA	1.1.
245-81-15	WAS.B	SCK/CEN	1.1.
246-81-2	WAS.D	KfK	1.2.3.
248-81-7	WAS.UK	UKAEA	2.2.1.3.
249-81-7	WAS.F	CNRS	2.2.1.3.
250-81-7	WAS.D.	KfK	2.2.1.3.
252-81-7	WAS.F.	CEA	2.2.1.3.
253-81-7	WAS.UK	UKAEA	2.2.1.5.
255-81-7	WAS.UK	MAFF	2.2.2.
256-81-7	WAS.UK	Taylor-Woodraw	2.2.2.
257-81-7	WAS.UK	NERC/IOS	2.2.2.
258-81-7	WAS.NL	Delta Inst.	2.2.2.
259-81-7	WAS.F	CEA	2.2.2.

262-81-7	WAS.F	CEA	2.2.3.
263-81-7	WAS.UK	Un.N-Wales	2.2.3.
264-81-7	WAS.F	BRGM	2.2.3.
266-80-7	WAS.D	KfK	2.2.1.2. 2.2.1.6.
267-81-6	WAS.F	CEA	2.1.1.
268-81-55	WAS.F	CEA	1.5.1.
270-81-7	WAS.D	HMI	2.2.1.8.

LIST OF PUBLICATIONS

1) G. BRAMBILLA et al (AGIP NUCLEARE) : Incineration in molten salts of alpha-contaminated solid waste- (contracts 036-76-7 and 063-78-1 WASI) - EUR 7336 (1981).

2) J.R. GROVER et al (AERE HARWELL) : An overview of PCM management - (contracts 100-WASUK, 101 WASF and 102 WASB 79-5) - EUR 7335 (1981).

3) J.W. ISAACS et al (AERE HARWELL) : An experimental study on critical factors in the plutonium recovery from wastes by incineration and pyrolysis - (contract 049-77-11 WASUK) - EUR 7337 (1981).

4) S. AHNER et al (BELGONUCLEAIRE & NUKEM) : Storage of HLW in engineered structures, air cooled and water cooled concepts (contracts 041 and 042-77-10 WAS B)- EUR 7340 (1981).

5) B. LONGSTAFF, P.W. CAINS, M.N. ELLIOT, R.F. TAYLOR (AERE HARWELL) : Investigation of a process for the pyrolysis of plutonium contaminated combustible solid waste - (contract 050-77-11 WASUK) - EUR 7339 (1981).

6) N. JACQUOT-FRANCILLON, F. LANDE, A. MAGNIER, E. VERNAZ : Elaboration de verres hautement actifs dans les cellules Vulcain et Piver (contract 040-77-11 and 070-78-9 WASF) - EUR 7371.

7) M. POULAIN (UNIVERSITE DE RENNES) : Incorporation de l'iode dans les verres à bas point de fusion - (contract 054-78-1 WASF bis) - EUR 7372 (1981).

8) N.J. FRANCILLON, J.P. MONCOUYOUX and G. RABOT (CEA, MARCOULE): Conditionnement et décontamination des déchets de gaine

d'éléments combustibles nucléaires par fusion - (contract 033-77-8 WASF) - EUR 7369 (1981).

9) W. KRISCHER and F. LANZA (CEC) :
Activities in the frame of the E.C. programme on radioactive waste management in the field of testing and evaluation of solidified HLW products - Paper ORA 19993 - (Third, IAEA research coordination meeting on evaluation of solidified HLW products - Bombay 23-27 February 1981).

10) P. BO, L. CARLSEN (Risø) :
Ion exchange properties of soil fines (contract 038-78-1 WASDK) - EUR 7355 (1981).

11) D. TAILLAR, J. CLAES, D. HENNART (CEA CADARACHE - CEN-SCK MOL):
Traitement et conditionnement des déchets solides de faible et moyenne activité - (contracts 092 WASF et 093 WASB) - EUR 7374.

12) P. HENRION (SCK-CEN) :
Interpretation and prospective study of diffusion phenomena in the context of a geological repository for high level waste in a clay formation (contract 084-78-7 WASB) - EUR 7416 (1982).

13) B. STOJANIK-WIECZOREK, G. DAMERAU, G. KEMMLER, G. MUHR (KfK):
Nassverbrennung plutoniumhaltiger Abfälle - (contract 044-77-7 WASD) - EUR 7441 (1981).

14) M. PIGNOL, P. HALLER (SENA) :
Poste pilote d'enrobage des déchets radioactifs dans les résines thermodurcissables - (contract 039-77-6 and 077-78-11 WAS FB) - EUR 7415.

15) P. BO (Risø)
In situ determination of permeability and solute retention (contract 065-78-1 WAS DK) - EUR 7651 (1981).

16) R. HEREMANS et al. (SCK-CEN) :
Programme R&D relatif au rejet de déchets radioactifs en formations géologiques profondes. Etude d'une formation ar-

gileuse en Belgique (contracts 013-76-1 and 060-78-1 WASB) - EUR 7650 (1982).

17) Etude de conception générale d'une installation permettant l'évacuation de déchets radioactifs dans une formation granitique - (contract 107-79-2 WASF) - EUR 7620F (1982).

18) The Community's research and development programme 1980-84 on radioactive waste management and storage. Annual progress report 1980 - EUR 7652 (1982).

19) I.L. JENKINS et al. (UKAEA, HARWELL) :
The characterization of activities associated with irradiated fuel element claddings - (contracts 037-76-11 and 072-78-4 WASUK) - EUR 7671.

20) A. VAN DALEN et al. (ECN, PETTEN) :
Sea disposal of radioactive Krypton - (contract 052-78-5 WAS NL) EUR 7670 (1982).

21) H. KARTES, U. RIEGE, A. GORTZEN and TH. DIPPEL (KfK, KARLSRUHE) :
Verfestigung alphahaltiger Abfälle in keramischer Matrix. (contract 082-78-7 WASD) - EUR 7654.

22) N.J. BRIGDER and C.V. DE FREITAS (UKAEA, HARWELL) :
Tritium immobilization by incorporation in inorganic solids (contract 075-79-7 WASUK) - EUR 7653 (1981).

23) A. BRUGGEMAN et al. (CEN/SCK, MOL) :
Separation of tritium from aqueous effluents (contract 090-78-7 WASB) - EUR 7669 (1982).

24) High temperature incineration and densification of granules from high temperature incineration (contracts 017-76-7 and 064-77-7 WASB) - EUR 7675.

25) B. SKYTTE JENSEN (Risø) :
Critical review of available information on migration phenomena of radionuclides into the geosphere (contract 066-78-1 WASDK) - EUR 7676 (1982).

26) O. NOMMENSEN, D. NIEPHAUS and H. BRÜCHER (KFA, Jülich) : Versenkung von Krypton-85 in das Meer - (contract 051-78-1 WASD) - EUR 7731.

27) D. NIEPHAUS, O. NOMMENSEN and H. BRÜCHER (KFA, Jülich) : Langfristige Lagerung von radioktivem Krypton in Druckbehältern - (contract 047-77-7 WAD) - EUR 7771.

28) H. FROTSCHER, H. GRABNER AND H. KAPULLA - (KfK, Karlsruhe) : Konditionierung von Hüllen bestrahlter Brennelemente durch Walzen und Einbetten in Beton - (contract 035-76-11 WASD) - EUR 7728 (1982).

29) General conceptual design study for a high level radioactive waste repository in a granite formation - (contract 107-79-2 WASF) - EUR 7620 E (1982).

30) R.D. PENZHORN, P. SCHUSTER and H.E. NOPPEL (KfK, Karlsruhe) : Die Langzeitlagerung von Kr-85 durch Fixierung in Zeolith 5A - (contract 074-78-1 WASD) - EUR 7829.

31) J. BROOTHAERTS et al. (CEN/SCK, MOL) : Axial distribution of tritium in Zircaloy clad fuel rods - (contract 071-78-9 WAS B) - EUR 7830.

32) A. LEUDET and A. LESEUR (CEA, FONTENAY-AUX ROSES) : Comportement des radionucléides gazeux dégagés pendant les opérations initiales du retraitement - (contract 089-78-7 WASF) - EUR 7822.

33) J. BROOTHAERTS et al. (CEN/SCK, MOL) : Conditioning of cladding waste by press compaction and encapsultation in low-melting metal alloys - (contract 030-76-11 WASB) - EUR 7876.

34) J.A.C. MARPLES et al. (UKAEA HARWELL) : Testing and evaluation of the properties of various potential materials for immobilising high activity waste. - EUR 7138 (1981).

35) W. HEBEL, G. COTTONE, ed. (CEC) :
Management modes for Iodine - 129, Proceedings of Specialists meeting, Brussels, 25 Sept. 1981 - EUR 7953 (1982).

36) J. HAMSTRA, B. VERKERK (ECN) :
The Dutch geologic radioactive waste disposal project (contract 062-78-1 WASN) EUR 7151 (1981).

37) Programme of research into the disposal of radioactive waste into geological formations (studies on crystalline rock) (contract 059-78-1 WASUK) EUR 7976 (1982).

38) K. NILSSON and B. SKYTTE JENSEN (Risø) :
Determination of the stability constants of bicarbonate and carbonate complexes using Strontium-85 (contract 038-78-1 WASDK) - EUR 7901 (1982).

MEETINGS AND SEMINARS

During 1981 the following ordinary progress meetings took place with regard to the implementation of the R&D programme :

January, 29,	Working group on storage and disposal in geological formations.
February, 18,	19th meeting of the Advisory Committee on Programme Management (ACPM).
April, 28,	Working group on fuel claddings and dissolution residues.
30,	Working group on gaseous wastes.
May, 26,	Working group on waste product characterisation (LLW and MLW).
June, 11,	20th meeting of the Advisory Committee on Programme Management (ACPM).
23,	Working group on storage and disposal in geological formations.
September 24,	Working group on testing and evaluation of HLW products.
October 1,	Working group on liquid, medium-level waste.
6,	Working group on the disposal into the sea bed.
13/14,	Working group on fuel claddings and dissolution residues.
15/16,	Working group on combustible alpha bearing waste.
22/23,	Working group on waste product characterisation (LLW and MLW).
26/27,	Working group on gaseous wastes.
28,	21st meeting of the Advisory Committee on Programme Management (ACPM).

November, 5/6, Working group on shallow land burial of solid low-activity waste.

December, 2, Working group on storage and disposal in geological formations.

On September 25, 1981, a topical seminar was organized for specialists of the Community member states on the subject of "Management Modes for Iodine-129".

MEMBERS OF THE ADVISORY COMMITTEE ON PROGRAMME MANAGEMENT
(in 1981)

Belgium	L. Baetslé
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ABBREVIATIONS

ACPM	Advisory Committee on Programme Management
AERE	Atomic Energy Research Establishment
BAM	Bundesanstalt für Materialprüfung
BRGM	Bureau de Recherche Géologique et Minière
CEA	Commissariat à L'Energie Atomique
CEC	Commission of the European Communities
CNEN	Comitato Nazionale per l'Energia Nucleare
CNRS	Centre National de la Recherche Scientifique
EC	European Communities
ECN	Energieonderzoek Centrum Nederland
ECU	European Currency Unit
GSF	Gesellschaft für Strahlen - und Umweltforschung
HLW (HAW)	High Level Waste
HMI	Hahn-Meitner-Institut
IGS	Institute of Geological Sciences
IOS	Institute of Oceanographic Sciences
JRC	Joint Research Centre
KFA	Kernforschungsanlage Jülich
KfK	Kernforschungszentrum Karlsruhe
LLW (LAW)	Low Level Waste
MAFF	Ministry of Agriculture, Fisheries and Food
MLW (MAW)	Intermediate Level Waste
NERC	Natural Environment Research Council
NRPB	National Radiological Protection Board
OJ	Official Journal of the European Communities
SCK-CEN	Studiecentrum voor Kernenergie - Centre d'Etudes Nucléaires

SENA	Société d'Energie Nucléaire Franco-Belge des Ardennes
UKAEA	United Kingdom Atomic Energy Authority

related titles of interest

RADIOACTIVE WASTE MANAGEMENT
An International Journal

Edited by D. R. Anderson, Sandia Laboratories — A. M. Platt, Battelle Pacific Northwest Laboratories — Francesco Girardi, Joint Research Centre, Ispra

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RADIOACTIVE WASTE MANAGEMENT AND DISPOSAL 1980

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RADIOACTIVE WASTE : ADVANCED MANAGEMENT METHODS FOR MEDIUM ACTIVE LIQUID WASTE

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(First Annual Progress Report of the European Community Programme 1980-1984)

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