Commission of the European Communities

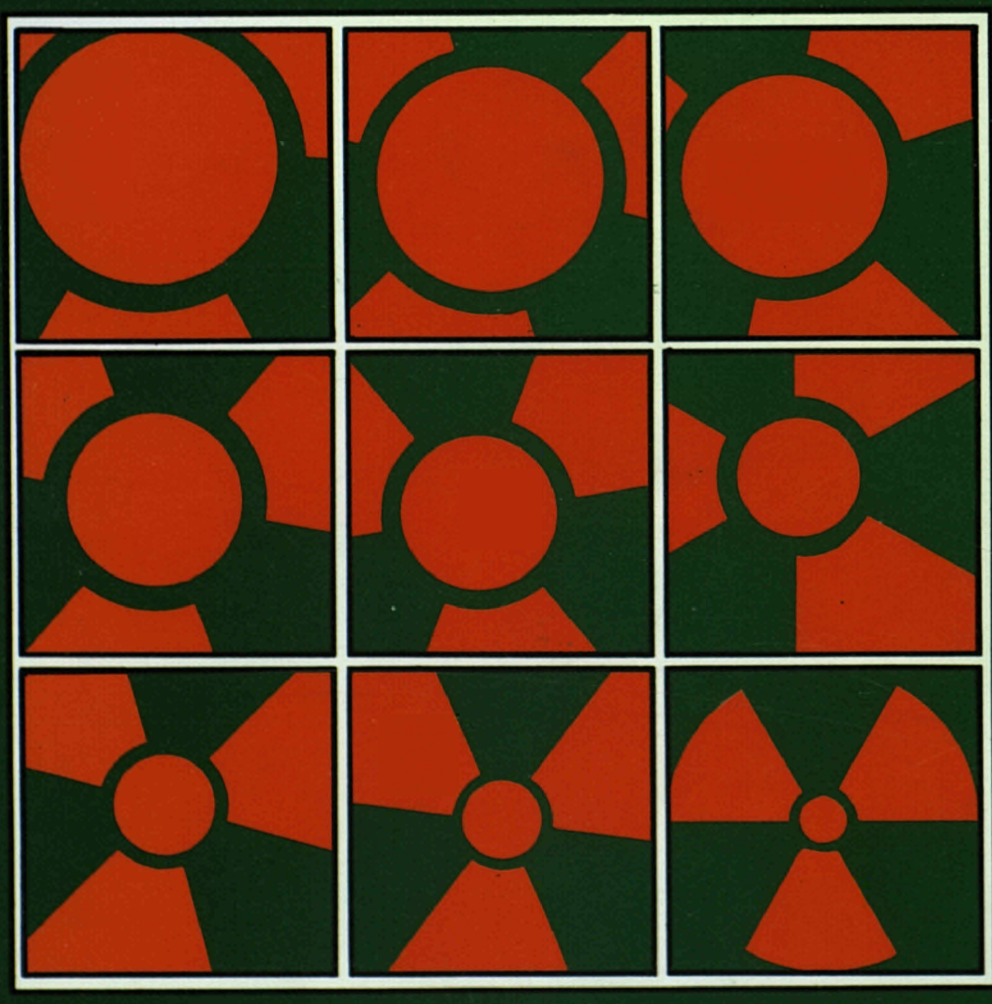
nuclear science and technology

The Community's research and development programme on radioactive waste management and storage

Shared cost action

Annual progress report 1989

Volume 2



Report

EUR 12761/2 EN

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Directorate-General
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Foreword

The Council of the Ministers of the European Communities adopted through its decision on March 12, 1985 (1) a five year R&D programme on "Management and Storage of Radioactive Waste" for the period 1985-1989. This R&D programme is being carried out on the competent public organisations and private firms within the Member States. At the end of 1989 over 256 contracts had been concluded with some 70 bodies within the European Community (43 % national bodies and research centres, 36 % private industries, 21 % universities and institutes). The Commission's contribution to the cost of the programme amounts to 62 million ECU (for the five year period).

In the management of the programme, the Commission is being assisted by the Management and coordination advisory committee "Nuclear fission energy - Fuel cycle/processing and storage of waste" (see for list of members pageVII, Vol. 2).

The launching of specific coordinated research projects within the programme has promoted the co-operation between various teams within the Member States. Half of the contracts concluded are dedicated to such projects. The exchange of fresh scientific information on the progress and results of the work carried out in the framework of the programme is assured at operational level by periodical progress meetings of working groups for the various projects. The final results of the research contracts are published in the EUR series which publication are e.g. listed in the six monthly news letter, EC FOCUS. In addition the Commission is publishing annual progress on the overall programme. This report, covering the year 1989, is the fourth of this type, the previous being published under no EUR 11089 for the year 1986, no EUR 11482, for the year 1987 and EUR 12141 covering the year 1988. For each contract the objectives, working programme and a summary of progress and results obtained have been given as prepared by the contractor, under the responsibility of the project leader. The Commission wishes to express its gratitude to all scientists who have contributed to this report. The overall results of the five year programme will be presented, as usual, to the worldwide scientific community at the third European Conference on Radioactive Waste Management to be held September 17-21, 1990 in Luxembourg.

S. ORLOWSKI
Head, Nuclear Fuel Cycle Division

1) O.J. No. L83, March 25, 1985, page 20

VOLUME 2

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(1) This Committee was established by the Council Decision of 29 June 1984 dealing with structures and procedures for the management and coordination of Community research, development and demonstration activities (OJ No L 177, 4. 7. 1984, p. 25).

PART A

WASTE MANAGEMENT STUDIES AND ASSOCIATED R & D ACTIONS

T A S K N o 4

RESEARCH IN SUPPORT OF THE DEVELOPMENT OF DISPOSAL FACILITIES; SHALLOW BURIAL AND GEOLOGICAL DISPOSAL STUDIES

TASK No. 4 : RESEARCH IN SUPPORT OF THE DEVELOPMENT OF DISPOSAL FACILITIES; SHALLOW LAND BURIAL AND GEOLOGICAL DISPOSAL STUDIES

A. Objective

Evaluation and modelling of the long-term behaviour of the geological barrier

Development of disposal facilities.

B. Research topics dealt with under the 1980-1984 programme

a) Work related to sites and their characterization

- General survey of geological formations and development of measuring techniques with a view to develop large scale in-situ characterization of the geological formations by direct or indirect methods

- Geoprospective studies : development of an operational method for the prospective analysis of the characteristics of geological containment

- Rock mechanics studies.

b) Work related to geological repositories and barriers

- Improvement of the designs and technologies required for the setting up of repositories in geological formations (salt, granite, clay)

- Development of long-lived containers for vitrified waste and of methods for the backfilling sealing of openings in geological repositories.

c) Work on radionuclide migration in the geosphere

- The work mainly comprised integral experiments on migration simulation, laboratory studies concerning the properties of materials from specific sites, hydrogeological investigations, research on natural geological migration systems and the role of micro-organisms, and, finally, the development of calculation tools and the intercomparison of codes regarding transport and geochemistry.

d) Shallow land burial

- Studies dealt with migration phenomena, improvement of barriers and radiological assessments.

C. 1985-1989 programme

The work is mainly a continuation of the research started during the 1980-1984 programme; however, special emphasis is being put on calculation tools and their intercomparison, on investigations attached to specific sites as opposed to laboratory work of general nature, on the role of colloids and complexes in radionuclide migration, on studies of natural analogues, and on the development and assessment of various backfilling materials and concepts.
Coordination is ensured by a structure of projects or working groups :

COSA	:	Comparison of Rock Mechanics Codes for Salt
COMPAS	:	Container Mechanical Performance Assessment
B & S	:	Backfilling and Sealing
MIRAGE	:	Migration of Radionuclides in the Geosphere
COCO	:	Colloids and Complexes
CHEMVAL	:	Geochemical Benchmark for Mirage
NAWG	:	Natural Analogue Working Group

D. Programme implementation

The available information on the contracts signed is listed hereafter.

4.1 RESEARCH RELATING TO SITES AND THEIR CHARACTERIZATION

4.1.A. General survey of geological formations and development of measuring techniques

The 600 m borehole project: "Development of a surveillance method during dry-drilling of a 600 m deep borehole in salt and performance of geotechnical measurements in the 600 m hole"

Contractor : Netherlands Energy Research Foundation (ECN)
Petten, The Netherlands
Contract No. : FI-1W/0084
Working Period: August 1986 - December 1990
Project Leader: J.R. van Seuren

A. OBJECTIVES AND SCOPE

The experiments performed in the Asse II salt mine in the FRG under the contract with the CEC during the previous programme (1980 - 1984) were carried out in a drilled hole of 30 cm in diameter and 300 m in depth; since then a dry drilling technique was developed for larger diameter holes and greater depths. In this project this technique will be tested by drilling a borehole with a diameter of 60 cm, typical for a disposal hole, and a depth of 600 m.
An alternative for the reconnaissance drilling which takes place before the actual drilling, will be developed with GSF.
The free convergence measurements of the salt as a function of depth of the hole, will be carried out. At the bottom of the hole convergence measurements with variable back pressure (isothermal lithostatic measurements) will be performed. Because of the complexity of the total construction the measurements will be done in two boreholes (Figure 1). All these results will be used for the validation of analytical techniques and computer codes.

B. WORK PROGRAMME

B.1 Drilling of the borehole.
B.2 Surveillance method.
B.3.1 Isothermal convergence measurements.
B.3.2 Isothermal iithostatic pressure measurement.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Drilling of the borehole

Although not in the contract with the CEC, the drilling of the 600 m holes with a diameter of 60 cm is a vital part of the experiment. This drilling is carried out by GSF (Gesellschaft für Strahlen- und UmweltForschung) under contract with BMFT.
The drilling has resulted in the following:
1. A borehole with a depth of 101,5 m.
2. A borehole with a depth of 300,5 m.
3. A borehole with a depth of 347,4 m.
The drilling of hole 1 and 2 will not continue. The drilling of hole 3 is temporarily stopped. The drilling rig needs major modifications and repair. It is expected that in 1990 the drilling of hole 3 can continue.

It was decided to use the second hole for convergence measurements first and later, when the lithostatic pressure measurement device was designed and tested, use this hole for those measurements.
This forced change in measuring strategy, which means measurements in a not optimal drilled 300 m hole instead of in an expected straight 600 m hole, resulted in extra effort.
The hole had to be measured for straightness and the lithostatic pressure measurement device has to be surrounded by cement at the 300 m level in order to make experiments possible.

Surveillance method

The design of the system has been described in the previous reports. With this system the flushing air during drilling of borehole 3 has been analyzed. The results are summarized in table 1.

Isothermal convergence measurement

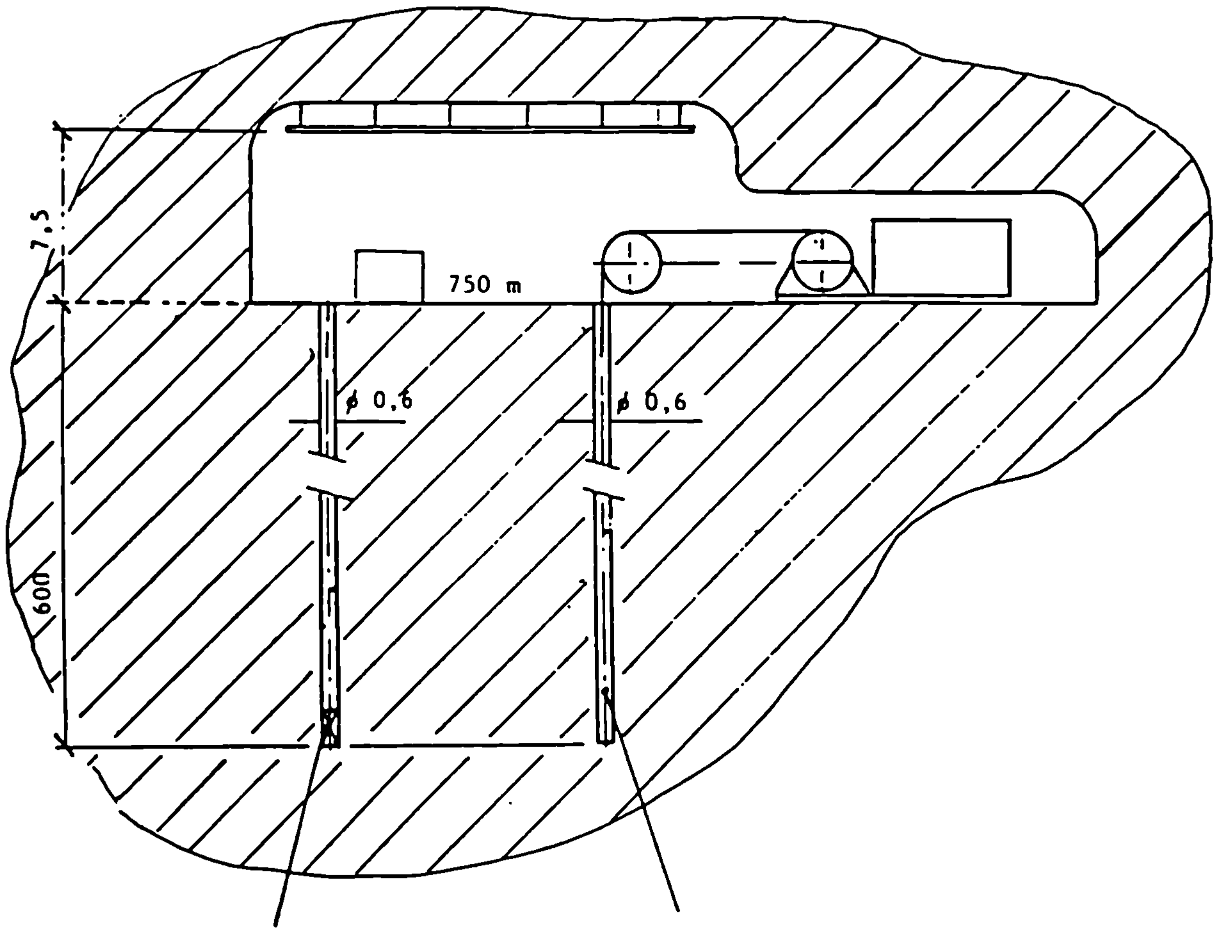
These measurements at the 300 m level in the second borehole have been started in November 1988. The free convergence at this depths is a relatively slow process. It has been calculated for a depth of 300 m and is shown in Figure 2 as part of an experimental program. It can be seen that the maximum displacement to be expected for a wall with an age of about 1 year is about 2 mm for a ½ year period. For a drilled hole with the same conditions this means double the amount. The hole diameter will therefore be decreased with about 4 mm.
A data report with experimental results will be available in 1990. The free convergence will in addition be measured at 200 m and 100 m in the 300 m hole. The unit which will measure this is in its testing fase and will be installed in the beginning of 1990.

Lithostatic pressure measurement

The design of the measuring construction has been finalized together with the pressure control station.
The design consists of a flexible rubber balloon in the form of a torus, surrounded by a rubber skirt. The torus is supported by a steel structure.
This construction has to be lowered, in the 300 m borehole, to the bottom location. To make measurements possible, it has to be surrounded by salt cement, which has to be poured from the 750 m floor. This is done via tubing, which is lowered, together with the construction, to 300 m level.
An amount of about 2 m^3 cement will, by gravitation, be transported in liquid form. It has a cure time of about 24 hours and will then act as an intermediate between the rubber and the salt wall, thereby assuring that measurement can start immediately.
The experimental program, which will be carried out, is shown in Figure 3. This program will start in 1990.

Table 1. Maximum values of the gas content in the flushing area of borehole 109/750/89

gas component	depth	drilling speed	concentration in the flushing air	concentration in the mine air	amount of flushing air	liberation
CO_2	180-181 m	0,125 m/min 0,035 m^3/min	1220 vpm	400 vpm	40 m^3/min	33 Nl/min 942 Nl/m^3 salt
hydrocarbons	258-261 m	0,11 m/min 0,031 m^3/min	40 vpm	3 vpm	40 m^3/min	1,5 Nl/min 48 Nl/m^3 salt
H_2O	79- 82 m	0,08 m/min 0,023 m^3/min	24% relatively humidity	13% relative humidity	40 m^3/min	180 g/min 7,8 l/m^3 salt



DYNAMIC ISOTHERMAL LITHOSTATIC PRESSURE MEASUREMENT

FREE CONVERGENCE MEASUREMENT OVER THE LENGTH OF THE HOLE

FIG. 1: SKETCH OF THE 600 m BOREHOLE MEASUREMENT SET-UP

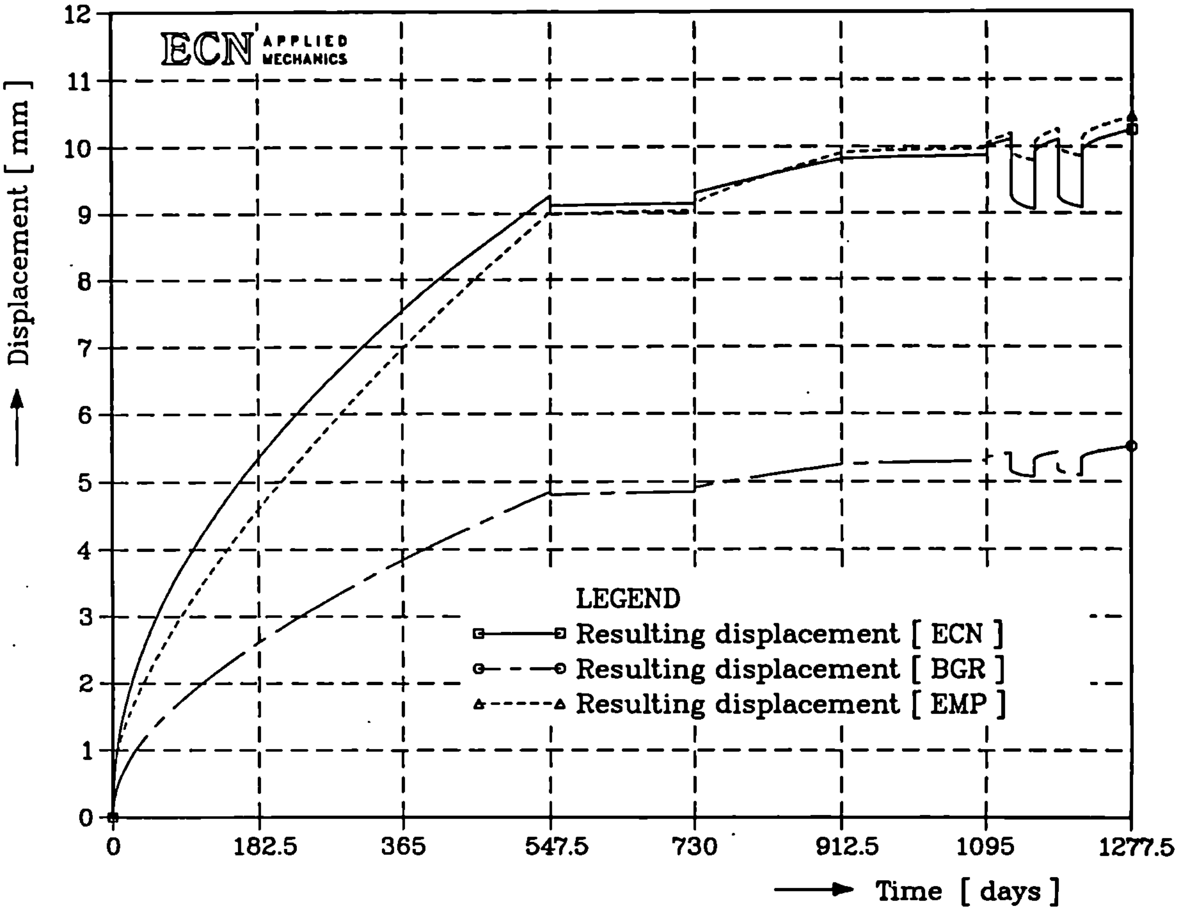


Fig. 2. Displacement of a saltwall in a 300 m deep borehole.

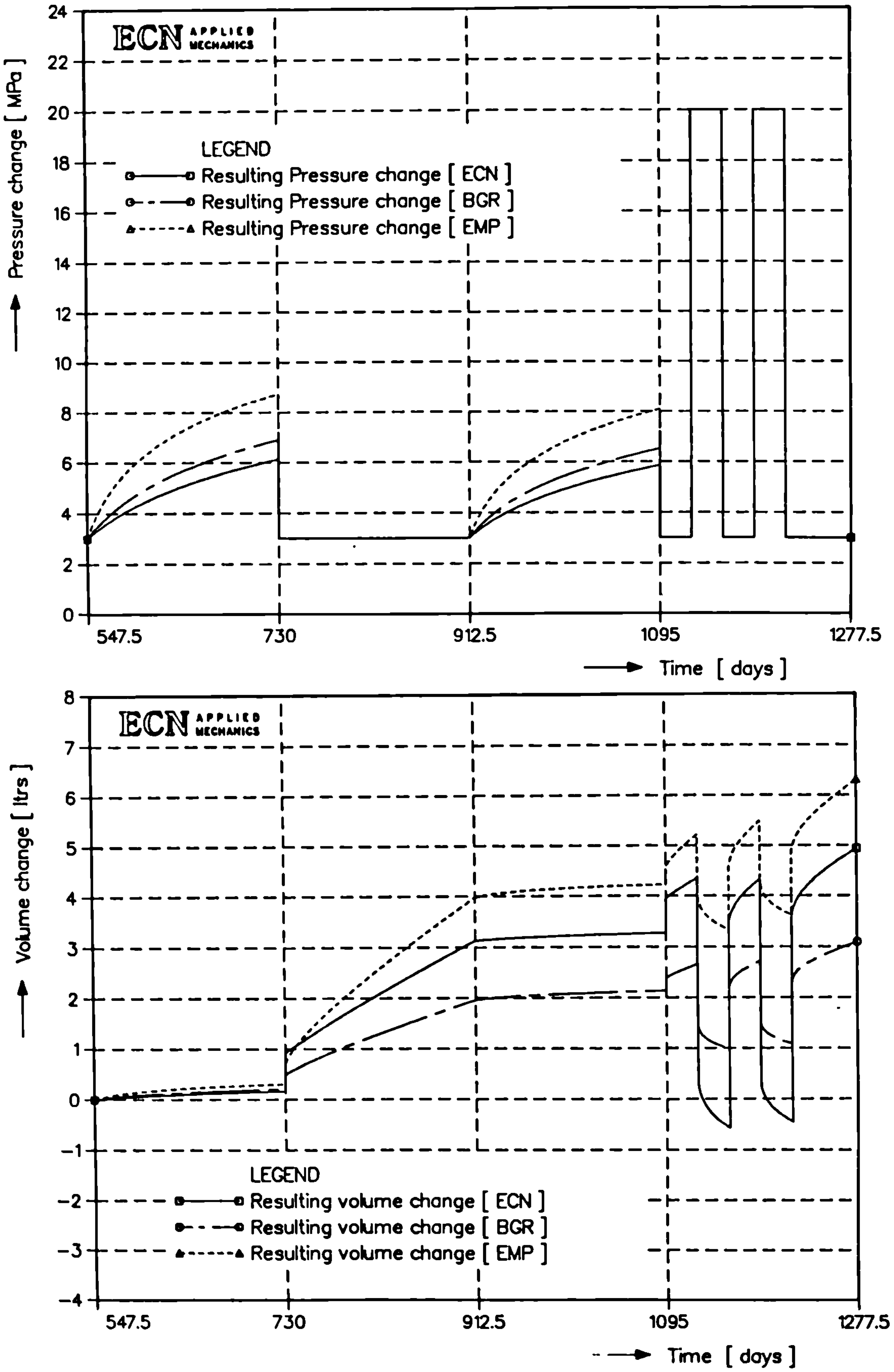


Fig. 3. Experimental program with the lithographic pressure measurement station in a 300 m deep borehole.

Faults in clays: their detection and properties

Contractor: BGS, Keyworth, Nottingham, UK

Contract No.: FI1W/0085/UK

Contract Period: October 1986-October 1989

Project Leader: M.A.Brightman

A. Objectives and Scope

Faults occur in most mixed sedimentary environments but their effects on regional groundwater flow patterns are poorly understood. The hydrogeological significance of faulted clay layers is of particular relevance where mudrocks are potential host formations for radioactive waste repositories.

In cooperation with ISMES of Italy two faults through clay layers will be studied (one site in the UK and one in Italy). The project has three objectives :-

1. To develop suitable geophysical techniques to detect water bearing faults in clays. These techniques will aim to differentiate between hydraulically active faults and those which are either sealed or non-water bearing.
2. To measure the hydrogeological properties of faults in sequences of mudrocks and aquifers. This will be achieved by measuring the hydraulic and chemical properties of the fault directly and by measuring the effect of the fault on the underlying aquifers.
3. To define suitable techniques for use in site investigations and methods for assimilating faulted boundaries into flow and transport models in clays and mixed sediments.

B. Work Programme

B.1. Desk study to evaluate a number of potential UK study sites; selection of two preliminary sites.

B.2. Initial geological and geophysical investigations of the preliminary UK sites; selection of the final study site.

B.3. Development of geophysical techniques for fault identification.

B.4. Detailed geophysical survey of the study site.

B.5. Borehole drilling.

B.6. Wireline geophysical logging of the boreholes.

B.7. Hydrogeological testing of the boreholes.

B.8. Synthesis of the results to evaluate the significance of the fault.

C. Progress of work and obtained results

State of advance

Seismic cross-hole tomographic surveys of the fault have been completed. Hydrogeological testing and groundwater sampling have been completed. Data interpretation and reporting have been completed.

B.3.,B.4.,B.7. and B.8. have been completed.

Progress and results

1. Detailed geophysical characterisation of the fault zone (B.3.,B.4.)

Cross-hole seismic tomography surveys have been completed in the borehole array at Down Ampney (Jackson et al, 1989b) with the twin aims of imaging the fault plane to examine its geological structure, and to determine changes in physical properties of the clay caused by the faulting. The measurement borehole array was designed to allow both the gross lithological changes on the upthrown side, and the fault zone within the clay on the downthrown side, to be investigated by the cross-hole seismics.

The environment at Down Ampney was favourable for cross-borehole transmission seismic tomography, with no background noise, and very low attenuation of the seismic waves. A high resolution borehole seismic source and a multi element hydrophone array was used for the surveys. This propagated energy of 1.5m wavelength in the Oxford Clay, representing a frequency of 1.2KHz which is far higher than that used in standard site investigation practice. The noise levels were low enough to enable whole waveforms to be acquired which often exhibited both reflections and refractions from the thin Cornbrash layer, indicating the benefits of underground seismic surveys that are not degraded by thin attenuating surface layers.

The effects on the velocity re-construction of using different arrivals were studied, and the removal of those events which were critically refracted along the Cornbrash improved the velocity inversion. Even though the geological setting was very favourable, it was concluded that the simplest straight ray interpretation procedure produced the best results with the real field data. The finite wavelength and the uncertainty in the exact position and shape of the refracting zones affected by the fault reduced the benefits of bent ray modelling. This is an unusual situation as the majority of reported case histories show that bent ray modelling produces results that identify geological structures more clearly.

The results from Down Ampney show a zone of 5% lower velocity within the clay coincident with the fault zone observed by the geological logging of the core. This lower velocity suggests the physical properties of the clay within the fault zone are altered, having a higher porosity and lower strength. This implies that the hydraulic conductivity of the fault zone is likely to be greater than the surrounding clay. Furthermore the reconnaissance resistivity surveys showed fault responses thought to be due to altered zones within the clay, that were of far greater magnitude than at the site of the borehole array. This may suggest that the fault zone detected by the seismic tomography is atypical of the Down Ampney site on a larger scale, and that the changes in physical properties may be greater in other parts of the fault.

2. Hydrogeological and hydrochemical characterisation (B.7.)

Groundwater heads and hydraulic conductivities have been measured at Down Ampney to investigate the local groundwater flow regime and are listed in Table 1. The array of measurement boreholes was designed to provide three vertical profiles of groundwater head, one through the upthrow side, one through the downthrow side and one through the fault zone.

The groundwater levels measured in the boreholes with screened sections through the Great Oolite and the Cornbrash, on both sides of the fault, were lower than had been expected, being depressed by groundwater abstraction for public water supply by two pumping stations a few kilometres distant. The groundwater heads measured in the piezometers did not show any consistent trends supporting either upward or downward groundwater flow. The most likely explanation seemed to be that the groundwater heads in the low hydraulic conductivity had not reached equilibrium with the heads in the aquifer which were generated by relatively recent pumping effects.

This hypothesis to explain the head distribution was tested by mathematical modelling using values of the hydraulic conductivities and storativities measured at the site. A 2–D vertical section about 2km long and 100m deep was modelled using the finite element code FEMWATER. The clay and other impermeable layers were assigned conductivities in the range 10^{-10}–10^{-11} ms^{-1} and the limestone layers 10^{-5}–10^{-6} ms^{-1}. The fault elements were given various properties in order to simulate the measured head pattern. The top surface at ground level was given a fixed head boundary condition of 0m and the lower surface was taken to be a no–flow boundary. The boundary conditions at each end of the model were altered for various simulations. The water abstraction history was modelled as a simple step drawdown from heads of +10m in the White Limestone to the present -11m around 30 years ago. This simplification was as accurate as was justified by the available data. A number of simulations showed that the measured heads were consistent with this model if the fault was assigned a low conductivity of approximately 10^{-10} ms^{-1} except between the downthrown Cornbrash and upthrown White Limestone where it was given an intermediate value of 10^{-8} ms^{-1}.

		Upthrow side	
Depth	Formation	Groundwater head	Hydraulic conductivity
(m below gl)		(m above gl)	(ms^{-1})
14.1-16.2	Oxford Clay	-0.2	$9x10^{-11}$
28.2-30.3	Kellaways	1.7	$9x10^{-12}$
32.1-37.0	Cornbrash	-6.0	$1x10^{-5}$
48.9-85.0	Great Oolite	-11.0	$5x10^{-5}$
		Fault zone	
Depth	Formation	Groundwater head	Hydraulic conductivity
(m below gl)		(m above gl)	(ms^{-1})
14.3-16.3	Fault zone	4.9	---
22.7-25.2	Fault zone	3.0	$2x10^{-10}$
28.8-30.8	Fault zone	0.8	$2x10^{-8}$
42.2-44.9	Fault zone	-1.53	$4x10^{-11}$
		Downthrow side	
Depth	Formation	Groundwater head	Hydraulic conductivity
(m below gl)		(m above gl)	(ms^{-1})
23.0-25.1	Oxford Clay	3.5	$4x10^{-12}$
49.0-51.4	Oxford Clay	5.2	$6x10^{-12}$
67.6-69.9	Kellaways	2.4	$3x10^{-11}$
82.8-88.1	Cornbrash	-10.0	$4x10^{-6}$

Table 1 Groundwater heads and hydraulic conductivities measured at the Down Ampney Fault Research Site.

Hydraulic conductivity was measured in the aquifer units by constant rate abstraction pumping tests and in the clays by pulse tests. The results show that the hydraulic conductivity of the fault zone tends to be between one and two orders of magnitude greater than the adjacent unfaulted clay.

Pore water has been squeezed from clay core samples and groundwater samples have been collected during pumping tests and from piezometers. These samples have been analysed for major elements and stable isotope compositions and profiles of variations in pore water and groundwater chemistry on both sides of the fault and through the fault itself have been constructed. These profiles suggest that the fault has had a significant effect on solute transport.

3. Evaluation of the fault (B.8.)

The following technical reports have been produced to evaluate the significance of the fault :

/1/ Alexander, J. 1989. Faulting in mudrocks : the selection of potential research sites. Report of the British Geological Survey WE/89/12

/2/ Brightman, M.A., Hallam, J.R. and Ambrose, K. 1989a. The boreholes and geology of the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in press)

/3/ Brightman, M.A., Abbott, M.A.W. and Sen M.A. 1989b. Hydrogeological investigation of the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in press)

/4/ Hallam, J.R., Jackson, P.D., Raines, M.G., and Rainsbury, M.P. 1989. High resolution resitivity measurements at the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in press)

/5/ Horton, A., Ambrose, K., and Cox, B.M. 1989. Geological sequence at the Down Ampney Fault Research Site, Gloucestershire, England. Report of the British Geological Survey WE/89/7

/6/ Jackson, P.D., Hallam, J.R., Raines, M.G., Rainsbury, M.P., Greenwood, P.G., and Busby J.P. 1989a. Reconnaissance geophysics to locate major faults in clays. Report of the British Geological Survey WE/89/32

/7/ Jackson, P.D., Greenwood, P.G, Raines, M.G., Rainsbury, M.P., and Hallam, J.R. 1989b. Seismic tomography investigation of the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in press)

/8/ Raines M.G., Jackson, P.D., Evans, C.J., Meldrum, P., and Rainsbury, M. 1989. An evaluation of the anisotropy of the clays of the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/48

/9/ Ross, C.A.M. 1989 The chemistry of the clay pore fluids at the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in prep)

/10/ Sen M.A. 1989. Hydrogeological modelling of the Down Ampney Fault Research Site. Report of the British Geological Survey WE/89/ (in prep)

METHODOLOGY FOR APPLICATION OF ELECTRIC AND ELECTROMAGNETIC BOREHOLE TECHNIQUES FOR DETAILED EXPLORATION OF FRACTURED ROCKS

Contractor: BRGM, Geophisics Dpt., ORLEANS (F)
Contract No: FI1W/0086

Duration of contract: July 1986 - June 1989
Period covered: January 1989 - June 1989
Project leader: G. Pottecher, P. Valla

Progress report not yet available.

Title : DEVELOPMENT OF A SELF-CONTAINED DRILL-HOLE CHROMATOGRAPHIC PROBE

Contractor : CEA - CEN CADARACHE F
13108 - Saint Paul lez Durance

Contract n° : FI1W/0087
Working period : November '86 - November '89
Project Leader : J.M. Vinson

A. OBJECTIVES AND SCOPES

The study of the transuranian nuclide migration from radio-active waste storage places is based on the knowledge of the natural environment and, in particular, of the chemical composition of the water : the transfer vector.

In addition to major elements, the water also contains trace elements which play a prominent part not only on the general equilibrium, but also on the radionuclide migration possibility.

This is in particular the case of the Lanthanides, present in the granitic and argillaceous environments, whose role of sorption competitors with respect to the Actinides has been exhibited throughout numerous experiments.

The scope of this contract is to manufacture and operate a chromatographic probe to be used in a deep drill-hole, so designed as to acquire a representative sample under conditions of equilibrium of the natural environment, to preconcentrate it by elimination of the saline content and to store it for ulterior analysis at the surface.

B. WORK PROGRAM

B.1 Process Development

B.1.1 Principle laying-down and delineation. In laboratory model.
B.1.2 Definition of final probe design after study on model. Realization and adjustment on CADARACHE site.

B.2 Probe Qualification in the Deep Drill-hole of AURIAT

B.3 Application to 3 reference sites

C. WORK PROGRESS AND RESULTS OBTAINED

Summary

The analysis of AURIAT samples shows that the very low level of the lanthanides concentrations -i.e. 1 ppb- so another technic, ICP/MS, has been tested.

The probe is now manufactured and has been qualified on AURIAT site. The study of AURIAT deep bore-hole is undergoing.

Some anionic columns are also used to study anionic species in the bore-hole water.

C.1 Progress and results

C.1.1 Analytical studies

A lot of tests driven with several analytical methods show the difficulties of working at very low concentration level.

The observed AURIAT water concentrations are about 10 ppb for Ce, 1 ppb for La and lower for other lanthanides (10^{-1} to 10^{-2} ppb).

These concentrations do not allow a reliable analysis classical spectrophotometry after chromatographic separation. They make the use of the most achieved methods necessary. ICP/MS technic needs more development but could be the most interesting in correlation with the neutronic activation, heavier but more accurate.

Varied kind of stresses limit the analytical abilities. They are : saline charge, reconcentration and more generally all operations intended to surface separations. The probe target is the limitation of stresses by on-line saline charge elution and in situ studied species concentration.

The realisation of an acidic buffer medium of pH2 appeared to allow both elimination of monovalent cations and better concentration of lanthanides.

C.1.2 Manufacture and development of the equipment

During this year, all the tool manufacturing has been realised. The logistical support and the electronical surface device have been defined and built.

All workshop tests have been made even with drastic thermal conditions in order to check good making and durability of mechanical, electrical and electronical components.

The probe has a length of 14,5 meters and a weight of 162 kg.

C.2 Qualification on AURIAT site

C.2.1 The probe

Site experimentations have been realised in order to check the ability of the probe in bore-hole with hydrostatic pressure and 2000 m of wire-line.

Some defects depending of length of imersed wire-line appeared and were corrected. After some 6 tests the probe was able to work correctly.

C.2.2 Experimental process

During these experiences, the analytical methods were tested. The concentration factor has to be adjusted in order to avoid acidic impurities effect even for suprapure acid, because of lanthanide occurence in it at the same level than in water.

This disturbance occurs especially when lanthanides are carried away from columns by concentrated acid for analysis.

The procedure depends of the level of the elements the pH is adjusted at 2 by alternative sequences of 90 % water and 10 % nitric acid 0.1 N. 100 or 200 ml of water are percolated for Ce and La analysis. Volume of 1 to 2.5 l are needed for other lanthanides with a concentration factor of 200 to 500.

Fracture mapping in clays

Contractor: University of Exeter, Exeter, UK.
Contract No: FI1W/0088
Duration of Contract: September 1986 - August 1989
Period covered: January 1989 - August 1989
Project Leader: Dr. E.M. Durrance

A. OBJECTIVES AND SCOPE

If faults occur in the rock mass surrounding a nuclear waste repository, there is a risk that the return of hazardous radionuclides to the biosphere will take place by migration along these zones of higher permeability. However, the detection and characterisation of faults is difficult, especially in soft rocks such as clay, and little development of techniques has taken place. The objective of this programme is to develop techniques that will be suitable for routine use in both the preliminary and detailed stages of site investigation. The approach used is based on the observation that faults act as zones of preferential migration in the natural degassing of the Earth. Soil gas exploration methods are applied to detect zones of anomalous gas geochemistry. The procedure followed is based upon samples obtained from a depth of about 0.5m along a series of traverse lines. Once a fault has been located, spiking of the high permeability zone from a borehole drilled to intersect the fault plane, will take place with specific gases of different compositions. The ground will then be resurveyed to determine the migration characteristics of the gas within the fault. Test sites in the UK and Italy are to be investigated in co-operation with BGS (Keyworth) and ISMES (Rome). BGS and ISMES are responsible for site selection and the drilling programme, but some trials will be conducted at sites near Exeter.

B. WORK PROGRAMME

B.1. Equipment development.
B.2. Site selection
B.3. Soil gas geochemistry
B.3.1. Reconnaissance soil gas surveys measuring ^{4}He, ^{220}Rn, ^{222}Rn, O_2 CO_2 and some organic gases.
B.3.2. Detailed soil gas surveys of anomalous zones identified in the reconnaissance surveys.
B.3.3. Spiking of vertical boreholes and resurvey of soil gases.
B.3.4. Spiking of inclined boreholes and resurvey of soil gases.
B.4. Modelling and interpretation of results.

C. PROGRESS OF WORK AND OBTAINED RESULTS

The main experiment on gas flux through faulted clay ground was conducted at Down Ampney, Gloucestershire (UK), using a gas injection techniques via a borehole. Enhanced concentrations of the injected gases (CO_2 and He) were successfully detected later in the soil around the borehole. A second visit was made to the Narni site in Italy for a more detailed soil gas survey over the suspected extension of a fault in the adjacent quarry, this time including Rn with CO_2,O_2 and He measurements.

The contract ended on 31 August 1989. A final report has been produced summarising the results from the three-years' work.

Soil gas geochemistry (B.3.2.)

A five-day programme of soil gas surveying at the clay quarry site at Narni (Italy) enabled 85 sample points to be sampled and analysed for CO_2, O_2, He and Rn. A ten-metre sample spacing was used on three parallel traverses (10m apart) along each of two traverse lines at 028° and 053° directions from north. These 120m long lines were those used by ISMES (Roma) for geophysical measurements. The use of three parallel sets of readings enabled radial averaging techniques to be applied to the results, to eliminate random spurious values. Overall statistics for each gas analysed show log-normal distributions for Rn and CO_2, making them potentially more useful for fraction detection than He which showed low anomalies and a normal distribution. The two main anomalies detected,one on each traverse line, suggest a 180° trend of a major fracture. Consideration of all peaks above background, however, suggests linkage along a parallel series of lineations at a 125° trend. Intersection of both trends may be the most likely explanation of the highest anomalies.

Spiking of boreholes (B.3.3.)

A borehole (No. 10) was made available for gas injection work by BGS at the Down Ampney (UK) site. This vertical borehole is located just north of the outcrop of the northerly dipping normal fault plane in Oxford Clay. Gas was introduced to the Cornbrash aquifer horizon at a point close to the fault, upward passage of gas being prevented by the clay above and a rubber packer inflated in the borehole at the base of the clay. Gases reaching the surface via the fault, a distance of about 35m, were sampled by means of soil probes.

For CO_2, probe sites were position along and to either side of the fault outcrop 12.5m apart in the form of a grid. For He more closely spaced sites on a traverse orthogonal to the fault was adopted. This was necessary to avoid ground which had become waterlogged. Gas injection was controlled by the gas cylinder regulator and monitored with flow meters and pressure gauges.

About 1000ℓ of CO_2 and 1800ℓ of He were injected into the aquifer. Values gained at the surface appeared erratic for both gases and this is attributed to waterlogging of the soil profile in addition to vacuum problems common in clay ground experiments. Nevertheless the mean results prove valuable in showing distinct changes over time. Carbon dioxide values gave an initial mean of 1.01%, comparable to that of previous work at 1.05%. At 24 hours it gave a value of 1.04% rising to 1.24% at 120 and 2.69% at 262 hours. Similarly initial mean helium concentration at 55ΔHe ppb, slightly higher than previous work, increased to 130ΔHe ppb at 21 hours and with no more supply a plateau 2 to 3 times the original background level was established. A massive ΔHe value 50 times background close to the borehole was considered to be leakage via the borehole itself and was discounted from the mean results. The first appearance of He and CO_2 above background levels suggests travel times in the order of 11 and 100 hours respectively.

Sample sites responsible for the increased means do not show clear correlation with the predicted fault outcrop. However, CO_2 results to the south are generally higher than those to the north both for sites penetrating the concrete runway and those in the adjacent field. Increased He appears to be concentrated near the fault outcrop.

The results show that the technique can be successful and that a statistical element is needed for evaluation and consequently an extensive programme of data collection is necessary.

FAULTS IN CLAYS: THEIR DETECTION AND PROPERTIES

Contractor : ISMES S.p.A. - Bergamo
Contract No. : FI1W/0103
Duration of contract: from January 1987 to January 1990
Period covered : January 1989 - December 1989
Project leader : Ferruccio Gera

A. OBJECTIVES AND SCOPE

The research is coordinated with a companion contract to BGS to perform similar work in UK clays and, for some aspects, with a research carried out by the Exeter University (UK) on soil gases.

Faults and fractures are known to intersect argillaceous formations that may be considered suitable as host rocks for a radioactive waste repository. In some cases these structural discontinuities have been considered responsible for the enhanced hydraulic conductivity of mudrocks. Consequently, any argillaceous formation considered for the location of a waste repository should be characterised thoroughly from the points of view of occurrence of structural discontinuities and their hydraulic significance.

If geophysical techniques with the capability of revealing the existence of structural discontinuities, particularly water bearing ones, in mudrocks, could be developed, then site investigation studies would rely more on geophysics and less on deep exploration (mainly drillings) with significant advantages from the viewpoints of time, money and disturbance of the formation, etc...

The objectives of the contract are:
- to develop suitable geophysical techniques to detect water-bearing faults in clays. The main intended application is for surface investigations, but possible application in tunnels and boreholes should be considered;
- to carry out hydraulic testing across deep faults, if clearly detected, and to relate the in situ measured characteristics of the faulted clay with the results of laboratory testings;
- to define suitable techniques to be used in site investigation campaigns.

B. WORK PROGRAMME

The project consists of the following activities:
1) a survey within the Italian territory to identify sites where faults intersect clay formations;
2) choice of the most suitable sites for carrying out field work;
3) drilling of two boreholes in such a way that a fault plane is intersected at a depth of some tens of meters;
4) performing gephysical investigations of the fault zone using both surface and down-hole techniques;
5) hydraulic testing of the fault zone, if considered useful (carried out by BGS);
6) geotechnical measurements of samples obtained from the cores;
7) geochemical measurements (ISMES on clay and BGS on pore water);
8) possible hydraulic modeling of groundwater flow in the fault zone.

C. PROGRESS OF WORK AND RESULTS

State of advancement

All field and laboratory activities related to the Faults in Clays Project have been completed by the end of 1989 and the final report is being assembled.
Work carried out in 1989 by ISMES can be summarized as follows:
- completion of geotechnical laboratory testing of clay samples from the Orte quarry;
- geochemical determinations on Orte clay samples;
- laboratory testing programme on British clay samples;
- choice of a new site for field geophysics;
- geophysical surveys of the Narni site;
- structural geology survey of the Narni site.

Progress and results

Geotechnical laboratory testing of Orte clay samples
Laboratory testing has been carried out on samples collected in borehole OS2 at the Orte site. The main results, relative to classification, oedometric characteristics, P and S waves velocity and permeability, indicate that the Orte clay is a strongly overconsolidated sediment (OCR > 5).

Geochemical determinations on Orte clay samples
An experimental study on the disequilibrium of the U and Th series in the Orte clay samples provides information on the migration of natural occurring radionuclides. The aim is to measure the radioactive disequilibria of uranium and thorium which could indicate the existence of a migration process. After an acid attack and a spiking with a known ratio of U/Th, alpha-spectrometer analyses give parameters indicating the possibilities of radionuclide migration with respect to local redox conditions. An interesting feature of this research is that it allows to estimate the time, from a radiometric point of view, that has been necessary for the system to reach the disequilibrium conditions. The processes occur at the scale of nuclear interactions between clay minerals and interstitial fluids. In this way, it is possible to use equations giving quantitative estimates of the diffusion parameters which control the migration process. Data elaboration is presently being completed.
BGS has analysed water squeezed from Orte clay samples collected at different depths in borehole OS2; plotting the obtained data on bidimensional diagrams show the existence of an anomalous peak for almost all measured parameters at a depth of about 51 meters.

Laboratory testing programme on British clay samples
A number of sealed clay samples coming from the study area of Down Ampney in Britain have been analysed in the ISMES geotechnical laboratory of Seriate. The geotechnical determinations carried out on BGS samples consist of:
- classification (granulometry, density, Atterberg limits, calcimetry);
- history of the geological formation (Edo Ko);
- P waves and S waves velocity (for different frequency waves);
- triaxial cell tests;
- permeability.

Choice of a new site for field geophysics

After completion of the activities at the Orte site, it was decided to move to another area, in which only a programme of geophysical surveys would be carried out. The new site, selected from the list produced in the first phase of the Faults in Clays Project, is a hill at Narni (Umbria region, central Italy). At the site, a hill of pliopleistocenic age is cut by a clay quarry in which a fault plane was clearly visible up to the mid 80s, when it was obliterated by the collapse of the quarry front.

Geophysical surveys of the Narni site

The geophysical programme carried out at Narni has been somewhat similar to that of Orte, excluding all tests performed in boreholes. Two high resolution reflection seismic profiles have been performed: they both had a linear extention of about 100 meters (lines NAR-01-89 and NAR-02-89, see the Figure). Two geoelectrical profiles (HEL - horizontal electrical lines) have been performed to evaluate the electrical resistivity of the clay formation. The first profile covers exactly the seismic line NAR-02-89; on the same alignment, four vertical electrical soundings (VES) have been carried out (see Figure). Elaboration of results is presently being completed.

Structural geology survey of the Narni site

To better define the geostructural setting of the Narni hill a structural geology survey has been performed. Even if data elaboration is presently under way, it can be anticipated that the many dislocations encountered follow the main structural regional patterns.

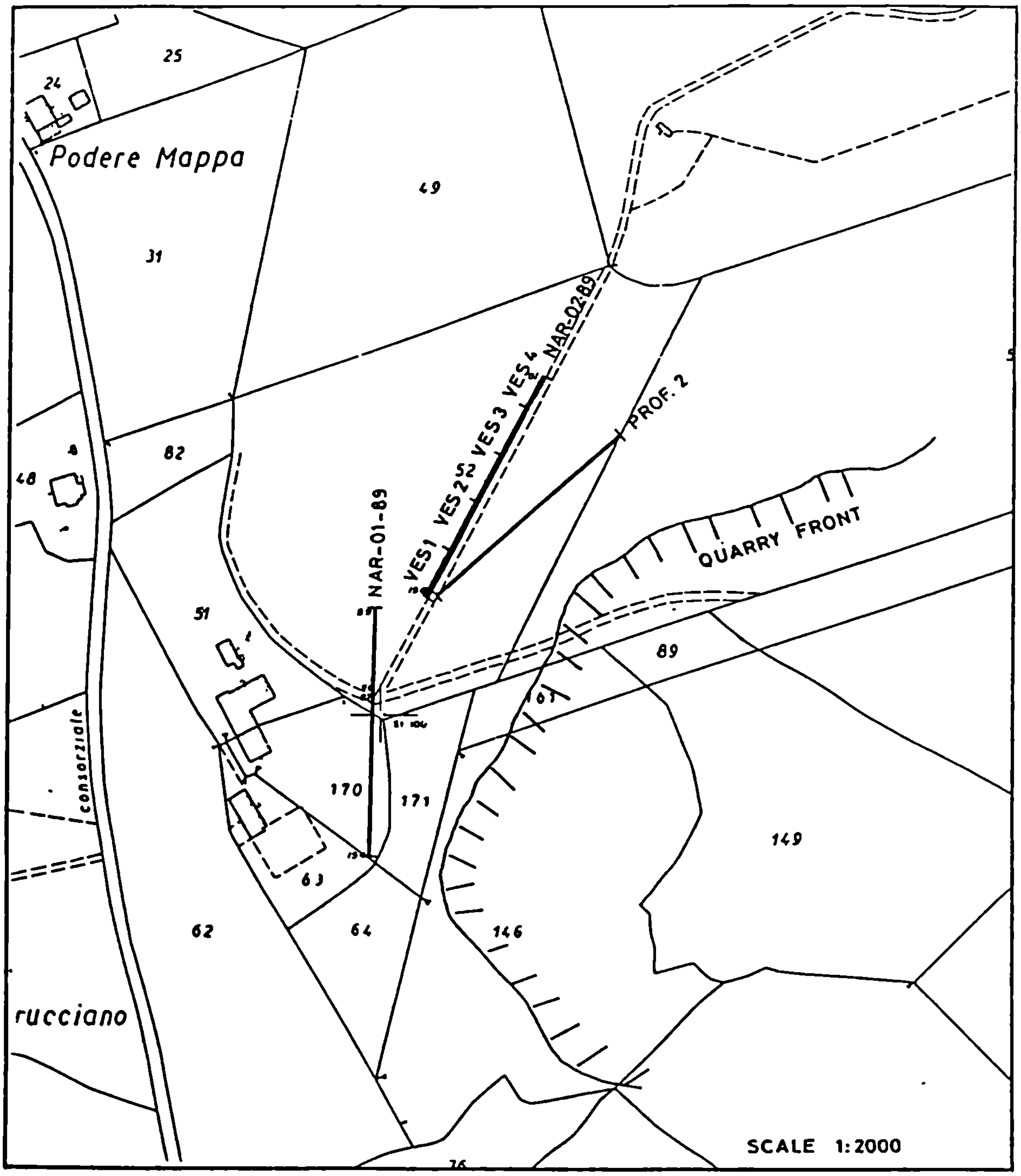


Figure - Geophysical survey of the Narni site.

EVALUATION AND DEVELOPMENT OF GEOHYDROLOGICAL SURVEYING METHODS IN AREAS WITH SALINE GROUNDWATER

Contractor : RIVM, Bilthoven, the Netherlands
Contract No : FI1W/0160
Duration of Contract : from November 1987 to May 1989
Period covered : January 1989 to December 1989
Project Leader : P. Glasbergen / G.C. Wijland

A. OBJECTIVES AND SCOPE

Both deep and shallow sedimentary rocks have been explored by oil- and watercompanies respectively. A lack of knowledge about the geohydrological parameters and a limited research-experience exist with respect to Oligocene, Eocene an upper Cretaceous sediments in the subsurface of the Netherlands.

The aims of the project are:

- to investigate, evaluate and indicate necessary developments of in-situ measuring techniques, which obtain geohydrological parameters, in boreholes;
- to give recommendations for the set-up of deep boreholes near salt structures in phase 2 of the Dutch Nuclear Waste Disposal Research Programme;
- to test and compare measuring techniques in several existing boreholes.
- to test and compare the effect of different watersampling techniques on water quality and natural isotope data.

B. WORK PROGRAMME

1.1 Investigation and evaluation of well-test methods of wireline logging, in-situ measurements of pH, Eh and temperature, sampling and isotope dating techniques of both undisturbed unconsolidated rock and pore water.

1.2 Recommendations for the construction of boreholes.

1.3 Recommendations for an optimized borehole- and survey programme.

2.1 Selection of existing deep boreholes in The Netherlands accessible for well-tests.

2.2-2.8 Performing of well-tests and sampling of formation water. Evaluation of the results of the different methods used and comparison with results of different laboratory measurements.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Current techniques and methods, which are considered to be of practical use for the determination of the geohydrological parameters of sediments around and above salt formations, have been investigated and evaluated. The results of this study have been reported in the first interim report no. 728515001, which covers item 1.1 of the work programme described previously /1/.

Uptil now no salt structure has been designated for a geohydrological survey programme and it is unlikely that a descission is made before the termination of the present research project. Therefore item 1.2 and 1.3 of the work programme will be restricted to some general recommendations, which will be included in the final report.

Using the existing wells, it is not possible to perform tests, which will lead to parameter values representative for Oligocene, Eocene and Cretaceous formations around and above salt structures in the North-East of the Netherlands. Therefore the second part of the work programme is focussed on the comparison of several existing techniques in low-permeability formations. The range of permeabilities of the investigated formation are probably in agreement with those, which can be expected in the NE Netherlands at large depths in formations overlying salt structures. Three boreholes were selected which are suitable for performing the well-tests. The results for an existing borehole in Reusel have been described in the second interim report no. 728515002, which covers part of items 2.1 to 2.8 of the work programme /2/. Furtheron well-tests on a borehole nearby Stekene (Belgium) have been performed. Results will be presented in report no. 728515004 (in preparation) /4/.

progress and results

1.1 Investigation and evaluation of geohydrological surveying methods of sediments overlying salt formations.

- Different well-test methods to obtain the permeability and the formation pressure have been evaluated including pumping tests, slug tests, packer tests drill stem tests and wireline formation tests. The investigation showed that the vertical permeability in anisotropic formations can be difficult to obtain with currently used well-test methods. Formation-pressure measurements can be subject to errors caused by the conversion of the water-head into formation-pressure. This problem cannot be solved by measuring the pressure downhole due to instrumental drift during long-duration measurements.
- Wireline logs have been considered for the determination of a large number of hydrogeological parameters and characteristics including the clay content, porosity, permeability, temperature, pressure of the geological formation and the salinity of the formation water. The determination of the permeability, pressure and salinity, using wireline logs, can be subject to large errors.
- Sample methods currently used have been reviewed. Taking corematerial samples turned out to be of importance in a geohydrological site-specific safety assessment. Chemical and mechanical alteration can be minimized using the right systems and methods. Various core-taking techniques are available for the different formation types. A low recovery may however be expected in unconsolidated sediments.
- Current water-sampling methods have been reviewed. Sampling using submergible pumps was considered for it's use in standard chemical and isotope analysis. Due to effects of degassing this method is supposed to be inaccurate in the determination of the pH , Eh and C-14 isotopes of deep groundwater. Tools which take in-situ pressurised samples were briefly reviewed.
- The french CEA downhole pH/Eh probe was compared with the swedish SKB system. Both were developed for investigation on behalf of the nuclear waste disposal programme.

1.2-1.3 Recommendations for the construction of boreholes and for an optimized borehole- and survey programme

For these items see remark under "State of advancement".

2.1 Selection of existing boreholes for well-tests.

Because it didn't seem possible to perform tests on existing wells, leading to useful parameter values representative for Oligocene, Eocene and Cretaceous formations around and above salt structures, the Berg Sand formation was selected instead.

The Berg Sand formation, which forms the base of the Oligocene Rupel formation, was chosen because of it's low permeability, it's isolation from other water-bearing layers by almost impermeable clay layers and the availability of existing accessible wellscreens.

Three wells have been selected for the field study:

a) A well in Reusel (the Netherlands), close to the dutch-belgian border, drilled in 1981.
b) A well nearby Stekene (Belgium), also close to the dutch-belgian border, drilled in 1989 by the RIVM in co-operation with the Belgian Geological Surveying Institute (BGD)
c) A well in Asten (the Netherlands), which has been drilled within the Dutch Research Programme on Geothermal Energy in 1987.

2.2-2.3 + 2.5 Fieldwork and evaluation of well-tests methodologies

- Tests are performed on an existing well in Reusel. Two types of well-tests are used.
  a) A pumping test using a pneumatic pump leading to a transmissivity of 0.18 m^2/day. The recovery data of the pumping test are illustrated in figure 1 (following the Theis recovery method. The possibilities for a pumping test in a 2-inch borehole are limited because the maximum flow-rate and head of the submergible pumps, which fit in these wells, are low. Large pumping time with a pneumatic pump with a capacity of 120 l/hr were needed to obtain a reliable set of residual drawdown data. The similarity between the calculated transmissivity from the recovery test for air-lifting and pumping are noteworthy.
  b) Then a slugtest using different techniques, leading to a transmissivity varying between 0.10 and 0.15 m^2/day was used. Adding water to the well has the disadvantage that it is practically impossible to give large changes in the length of the water column instantaneously. Using compressed air has improved the slugtest technique which is reflected in a more clear-cut curve-fitting procedure.

  In respective to the savings on time and costs the small difference between the two types would plead in favour of the slugtest method in the investigated sediment.
- Tests are performed on a newly-drilled well nearby Stekene (Belgium). Here again the two types of well-tests were used to determine the permeability. Furthermore some flow-measurements in combination with a pumping test with a short duration were performed by TNO-Delft. Results on these test are still preliminary. The obtained permeability of the Berg Sand formation is ca. 7.8e-6 m/s.

2.4 Water-sampling from deep aquifers.

More deep boreholes with highly mineralized groundwater have been sampled and analysed this year for their stable isotopes. Resulting data are given in table I. The stable isotopes indicate a contribution of meteoric water for all samples. The salinity has no unique source but is attributed partially to mixing with sea-water and partially to dissolution of evaporites. The oxygen-18 and sulphur-34 contents of aqueous sulphate are useful tools in identifying the origin of the dissolved salts /3/. The age of the deep groundwater is still in discussion. The hydrologically derived age exceeds the limits of carbon-14 dating. In co-operation with the Université de Paris-Sud samples will be analysed on Cl-36 which can function as a possible dating technique.

2.6 Selection of quarries for collection of rock-samples.

A first indication of the permeabilty determined on samples from quarries in outcrop areas pointed to values strongly differing from those expected in the NE Netherlands. Therefore this item was not worked out in more detail.

2.7-2.8 Laboratory measurements and comparison with field data

Core samples have been taken along a depth-profile during drilling of the Stekene well. A laboratorium was chosen which performed measurements on the permeability of the taken samples. It occured that the obtained permeability-values were significantly lower than obtained by the well-tests performed in the field.

References

/1/. Langemeijer, H.D., Evaluation of geohydrological surveying methods of sediments overlying salt formations. First Interim Report no. 728515001, RIVM, Bilthoven (1989).

/2/. Langemeijer, H.D., Evaluation of permeability tests on the Berg Sand. Test results of the Reusel well. Second Interim Report no. 728515002, RIVM, Bilthoven (1989).

/3/. Glasbergen, P., J.L.Michelot, D.Poutakis, G.C.Wijland, Investigation and evaluation of hydrogeological surveying methods of sediments overlying salt formations. Third Interim Report no. 728515003, RIVM, Bilthoven (in press).

/4/. Wijland, G.C., Evaluation of permeability tests and borehole constructions. Results for the Stekene well Berg Sand member. Fourth Interim Report no. 728515004, RIVM, Bilthoven (in preparation).

Location	Formation		Depth (m)	$\delta^{18}O$ ‰SMOW	$\delta^{2}H$ ‰SMOW	$\delta^{18}O(SO_4)$ ‰SMOW	$\delta^{34}S(SO_4)$ ‰CD
Kastanjelaan	M1	Dinantian	235	-8.90	-58.00	14.07	13.20
	M2	Devonian	470	-8.20	-56.20	13.60	17.10
Broekhuizenvorst	B	Oligocene	535	-6.52	-44.50	18.23	51.53
Nieuweschans	N	Eocene	559	-3.23	-24.20	17.36	30.67
Oploo	OP	Paleocene	513	-5.91	-37.50	17.63	30.31
Ockenburgh	02	Pleistocene	21	-6.98	-41.97	12.17	14.93
	05	Pleistocene	87	-2.74	-19.99	17.29	26.60
	06	Pleistocene	102	-1.68	-14.66	17.74	25.95
	07	Pleistocene	143	-0.56	- 7.98	15.42	22.34
	08	Pleistocene	181	-2.73	-22.27	15.17	25.15
	09	Pleistocene	209	-2.26	-18.34	14.47	24.04
	010	Pleistocene	337	-3.75	-27.50	21.69	39.23
Rosmalen	R6		279	-6.56	---	11.79	18.06
Seawater Scheveningen	SH			---	---	9.13	20.66

Table I. Stable isotopes analysis of deep groundwater.

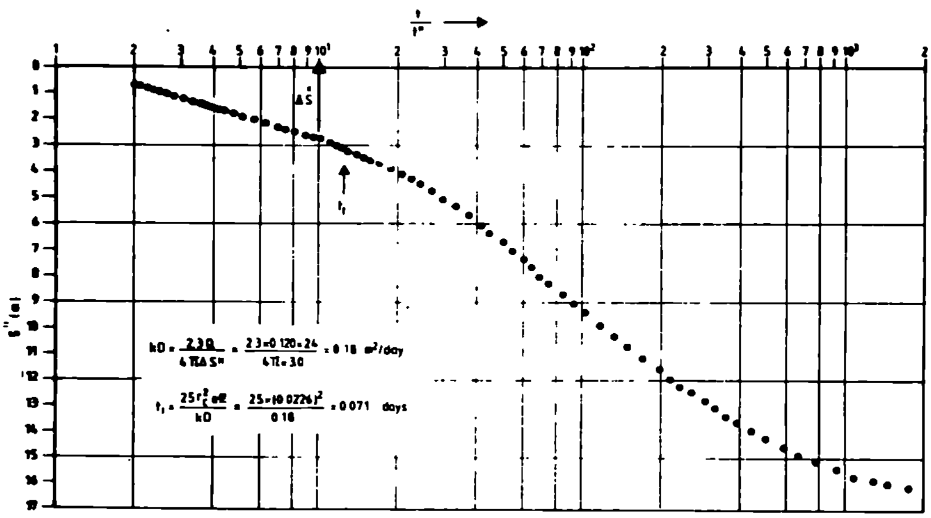


Figure 1. Theis's recovery graph of pumping test data from the Reusel well (56F7).

Acquisition of physico-chemical properties of clay. Development of methodology for taking samples and making experiments.

Contractor: CEA, CEN Fontenay-aux-Roses
Contract N°: F11W/0208
Working period: January 1989- December 1989
Project leaders: H.COULON, A. LAJUDIE

A. OBJECTIVES AND SCOPE

The decision to dispose of radioactive wastes in a deep clay formation cannot be taken without obtaining an in-depth knowledge of the phenomena involved in the transfer of water and radioelements *in situ*.

The difficulty in this approach lies in the necessity of taking samples and preserving them without modifying their intrinsic properties.

The general objective of this study is, therefore, to develop a methodology that allows us to specify the techniques for obtaining the *in situ* mechanical and chemical properties required for material transfer evaluations. This involves making an inventory of the properties to be obtained, identifying the possible perturbations resulting from the sampling method, selection of the core drilling methods, handling and treatment of the cores, sample preparation and analytical procedures.

The bases for this are, in particular, the experience gained by the CEN/SCK at Mol in the work in using the Boom clay, the observations made by ANDRA on the core drilling tests with deep clays and the methods developed by the CEA for the studies of clays (contracts N° 031 and 061).

The second stage is to test the methodology by taking, using the specifications reached above, a test core on the site at Mol, then comparing the results of the core analyses with the tests *in situ* in the underground research facility. This will be done following a protocol that is yet to be determined.

B. WORK PROGRAM

B.1 Inventory of the physicochemical parameters to be obtained both in order to evaluate the transfers in a clay formation and the perturbations induced in the clay by the sampling method.

B.2 Selection of the methods of sampling, of the treatment and of the preservation of the samples. Specification of the experimental procedures. Installation on the site at Mol (Belgium).

B.3 Comparison of the *in situ* and laboratory properties. Verification of the methodology.

C. PROGRESS OF WORK AND OBTAINED RESULTS

The preliminary study of the literature carried out in the B1 phase made it possible to list the ensemble of the phenomena that may modify the physicochemical characteristics of deep clays during and after sampling by core drilling.

- contamination by the drilling fluids;
- expansion of the cores, resulting in fractures and the migration of the drilling fluids into the core, and changes in the density, porosity, water content;
- oxidation of organic matter changing the chemical conditions (Eh - pH) and, thus, the diffusion transfer parameters.

The use of core drilling machines developed for soil engineering measurements and equipment for reapplying the pressure and maintaining anaerobic conditions should make it possible to avoid or limit the ensemble of the potential perturbations listed.

These techniques have just been tested on the site at Mol, in the Boom clay at a depth from 180 to 222 m.

The drilling was done by the FORAKY Company (Brussels) through arrangement with INTRAMINES (Paris).

The 0-180 m phase was carried out by destructive drilling with recuperation of the cuttings every 50 cm. These cuttings were made available to CEN/SCK.

From 180 to 222 m, the soil was continuously cored. In addition to the 8 m of undisturbed material requested by the CEA in pure clay, the CEN/SCK undertook the sampling of 21 m of undisturbed material in the sand, the transition zone and the pure clay.

Two types of core drilling machines were tested:

- a Longyear PQ3 Wireline system mine core drill (cable core-barrel) which can take 3 m long, 85 mm diameter cores. This triple core-barrel with an internal casing formed by two semicircular metal shells, was equipped with a frontal discharge bit, limiting the contamination by the drilling fluids
- a Mazier soil engineering cable core-barrel, allowing the recuperation of 1.30 m long, 84.5 mm diameter cores. This machine, with an internal PVC casing, is equipped with a cutting sleeve that is independent of the external tube, which penetrates the earth ahead of the bit (frontal discharge) decreasing the risks of contamination by the drilling fluids.

After comparing the characteristics of the cores obtained by these two methods, the Mazier core-barrel was chosen for all of the work.

The first tests carried out on the clay seams of the transition zone made it possible to optimize the drilling parameters:

- use of a modified extractor with steel blades;
- a load of 500 kg on the cutter, so as to always have the cutting sleeve well inserted in the soil during the advance of the bit;
- rotation speed of 40 rpm;
- penetration rate of of 2 cm/mn;
- a the end of a pass, an increase in the pressure of the fluid simultaneously with the decrease in the load on the cutter, so that the drilling fluid can begin to cut the base of the core, thus decreasing the stress on the extractor.

These procedures have made it possible to obtain apparently good quality pure clay cores. The cores containing more sand taken for the CEA/SCK seem to be of lower quality (presence of drilling fluids on the core surface, rotation marks on some cores,...).

Several treatment systems have been used:

- sealed galvanized iron storage containers (food can), provided by the CEN/SCK, especially suited to anaerobic storage:
- equipment developed by ANDRA for recuperating the samples under pressure:
  * pneumatic cells, allowing the injection of argon at 1 MPa;
  * hydraulic cells, the isostatic pressure of 1,5 MPa is ensured by pressurized silicone oil;
  * mechanical cells, the containment is ensured by an expanding-type cement and the normal pressure by a compressing spring (1 Mpa).

The permeability and diffusion measurements made on these samples in the B3 phase will allow us to compare these different systems.

The instruments for measuring the permeability *in situ* foreseen in the B3 phase are being studied:

- the first procedure, developed in collaboration with BRGM (Bureau de Recherches Géologiques et Minières), in using their experience in this field, makes it possible to measure low permeability in a transition regime by the following principle: a cylindrical sample where there are no radial or axial deformations, is saturated with degassed water. When the pressures are in equilibrium in the sample and in the circuit the upstream face of the sample is subjected to an increase in pressure H, and the injection valve is immediately closed. From the changes in the upstream and downstream pressures with time we can determine, from a nomograph, the coefficients of permeability, k, and the specific accumulation Ss of the sample.

The essential problem is the necessity of avoiding any deformations of the volume being studied, this is why the construction of shutters especially adapted to clay (in the form of a screw) is envisaged.

This technique should be developed and tested in the CEN/SCK underground research facility in the summer of 1990 and adapted in the laboratory to allow direct comparisons.

The second technique envisaged will be at the feasibility study level. It is based on the experience obtained in the use of the piezocone and on its possible transfer to the underground research facility. As in the above case, the principle is the measurement of the attenuation of the interstitial pressure generated, this time, by the insertion of a penetrometer. A calibration with values obtained in laboratory studies and other *in situ* techniques should make it possible to extend the measurement of low permeabilities in clays.

DEVELOPMENT AND EVALUATION OF AN X-RAY-ANALYTICAL TECHNIQUE FOR CORES FROM EXPLORATION HOLES DRILLED ON A POSSIBLE WASTE-DISPOSAL SITE

Contractor : Bureau de Recherches Géologiques et Minières
B.P. 6009 45060 ORLEANS Cédex - France

Contract : n° FI 1W/0209

Duration of contract : from january 89 to july 90

Period covered : 1989

Project leaders : Ph. Massal, S. Gentier

A. OBJECTIVES AND SCOPE

The exploration of site for the disposal of radioactive waste in various types of deep geological formations, and of possible locations for underground laboratories, is mainly based on the use of cored boreholes. It is desirable to study the resulting cores in a non-destructive manner.

X-Ray photography is widely used in the mechanical and mettalurgical industries.

In geology, only a few stationary X-ray machines are used for limited applications.

The aim of the research is thus, for different environments:

- to adapt radioscopic techniques to the study of geological materials that are available as drill core;
- to create a mobile unit that can be transported to the sites where cores are available and must be studied, such as a drill site, an underground laboratoy or a vessel engaged in sediment studies;
- to allow non-destructive testing and study of the cores that are taken within a barrel and require immediate confinement after being pulled from the hole, such as is the case for clay and salt-bearing materials that are destined for mechanical and geochemical tests;
- to have access, before any destructive operations, to the internal structure of cores, as a further aid to mechanical tests and to geological and geochemical studies;
- to adapt and develop image-processing software for the geological study of radiocopic images.

The equipment that is the subject of research differs from existing X-ray apparatus in the following points:

- an image-analyser tube is fitted in order to be able to make video pictures available; picture quality can be controlled and adapted to the geological material being studied, by varying the feed voltage of the emitting tube;
- a diaphragm allows adaptation to the size and shape of the samples to be analysed;
- the video picture allows real-time observation, as opposed to classical radiography where static images are obtained after an exposure of long duration. It will thus be possible to give a movement to the core and to obtain an analog-type log that can be correlated with logs obtained by other techniques;
- the video images can be digitally processed, for graphic reproduction and the recognition of geological subjects.

B. WORK PROGRAMME

The contract will comprise the following stages:

a. Drawing up a detailed pilot study of the apparatus (consulting suppliers of components, accuratety defining the apparatus and its operating environment).
b. Constructing the apparatus with the assistance of specialized suppliers.
c. Testing the apparatus on various cores of geological materials that might be suitable for the disposal of radioactive waste, such a clay (in a sleeve), salt, granite and schist or shale.
d. Adapting an developing image-processing software with the following objectives:
  - geological examination and identification (lithology, structure, texture) without damage to the structural integrity of the sample;
  - testing the suitability of this technique for identifying homogeneous zones and for localizing mechanical and geochemical samples in particular in the case of cores covered by a sleeve;
  - improving the clarity of the pictures so that better use can be made of them, using image-processing software developed for image-analysis purposes;
  - correlating the X-ray logs with other borehole or geological logs.
E. Preparing documents tha present the results obtained for various types of geological materials, and for different structures and textures being studied.

C. PROGRESS AND RESULTS

1. Progress to date

The beginning of the year was devoted to the construction of the apparatus, which was completed in May, 1989. It was immediately put to work on the X-ray analysis of clay cores recovered from deep boreholes (around 300 m of cores). All cores were recorded on video tape. Each 15 cm length of core gave rise to a digitized image, which was stored on magnetic tape.

As a follow-up to drilling, a software was developed for utilizing the drill-core images. It comprises an recombination and integration of the data derived from the 15 cm samples and those provided by the enhanced-image algorithms. Use of the processing software has, among other things, brought to light additional data concerning changes in sedimentation, fissures and inclusions.

In addition to the preliminary campaign referred to above, a number of other tests were carried out on various rocks, either in the form of cores (salt, schist, marine sediments) or in the form of sections cut perpendicular to the axes of cores (schist, granite, breccias, cherts).

2. Progress and results

In its final form, the apparatus comprises:

- a bank of X-ray machines, incorporating a device that enables the core to be rotated or otherwise manipulated;
- an assemblage that allows the video signals to be reproduced in real time, recorded on video cassette or presented as hard copy, and also permits the video signals to be digitized.

The digitized images can be stored in a microcomputer or a streamer, and be processed in the microcomputer by means of an image card linked with image-processing software.

This equipment is installed in a movable container, measuring 6 X 2.56 X 2.64 m and weighing about 9 tonnes, which can be transported on a lorry so that it can be used on site, immediately after the cores have been pulled.

The analytical technique employed is non-destructive, and can be applied over the protective sleeve, of PVC or other plastic material, in which the core is wrapped.

The information furnished relates to the entire volume of the core. The image obtained corresponds to the projection of the volume on a plane surface.

In the context of the investigations undertaken into potential storage sites for nuclear waste, the following materials were tested:

. Clays: Study of the clays has revealed a number of significant details:

- fine variations in lithology of the order of 1 cm (limestone, marl, clay, sandy partings)
- decompression fissures;
- pyritic inclusions;
- signs of wear on the core, linked to the drilling; the amount of attrition varies, depending on the rock type involved. In fossiliferous horizons, only those fossils rich in pyrite, with a density markedly different from that of the matrix, are well preserved.

The diameter of the cores examined was 15 cm, so it was not possible to study the ferruginous oolitic limestone horizons satisfactorily; this would have required more power for the X-Ray beam. It seems that the larger the diameter of the core, the stronger the density contrasts have to be if the fine structures are to be clearly visible.

. Salt Study of the 8.5-cm cores disclosed the presence of:

- marly bands;
- anhydrite inclusions;
- fissures and discontinuities.

Where the salt occurs as large crystals a centimetre or more in size, grain boundaries can be clearly seen.

. Schist Both 9-cm cores of schist and discs 10 to 20 mm thick were investigated. Among the various types of schist tested, the alternations arising from the sedimentary characteristics of the rock were exposed by so rotating the cores as to bring the bedding-plane into a position parallel to the X-Ray beam; in other positions, these horizons are blurred or even, at times, invisible.

In the case of slate, the long and short axes in the plane of schistosity could be detected with the aid of the pyritic inclusions. Rotation of the cores reveals the size of these inclusions; by measuring them regularly, the favourable directions can be determined, if they exist. These observations are confirmed by the studies of sections parallel to the plane of schistosity.

The slates display two types of discontinuity:

- closed fractures containing pyrite inclusions. They show up when the core is rotated with the fracture plane parallel to the X-Ray beam;
- open fissures within the schistosity.

. Granite Textural study of this type of rock is difficult. As granite is a polymineralic rock, composed of mineral grains that are almost randomly arranged, the X-Ray beams become diffused, producing a grey, indistinct image. Textural studies can be made, however, by using sections of a thickness suited to the granulometric characteristics of the rock.

With the aim of fully comprehending the potential and limitations of the apparatus, other substances were studied; these included cores of concrete and marine sediments, and sections of breccias and chert.

Study of the marine sediments, which were made available from a drilling programme being carried out at sea, showed up:

- the laminations;
- the change in density accompanying the compaction of soft sediments;
- the bioturbation;
- the sea-shells.

The tests on the chert sections revealed the progressive silicification of the nodules they were derived from. The breccia studies disclosed the mineralized zones in those rocks.

For the clays and marine sediments we tested, gamma-gamma density logs are available in parallel with our X-Ray logs: a systematic comparison of the two sets of logs is an imperative future committment.

CARROTTES DE SEL (Varangeville-France)

Ø 85 mm

Image brute

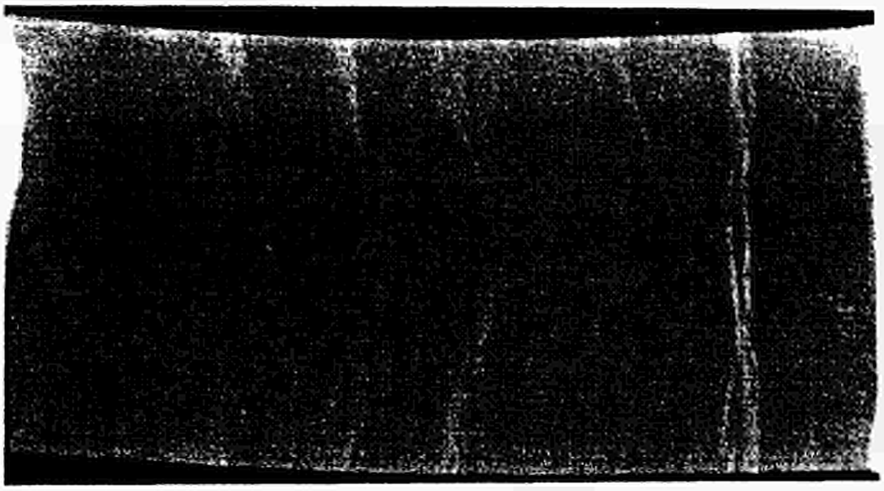


Image inverse

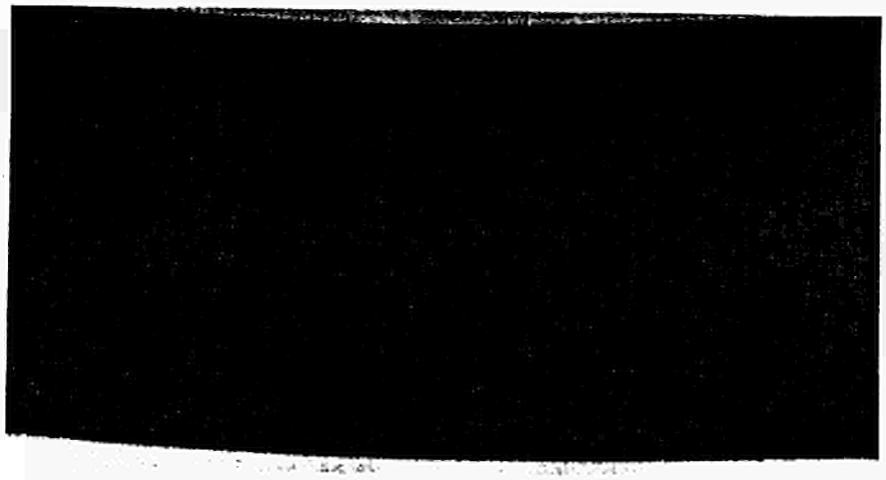
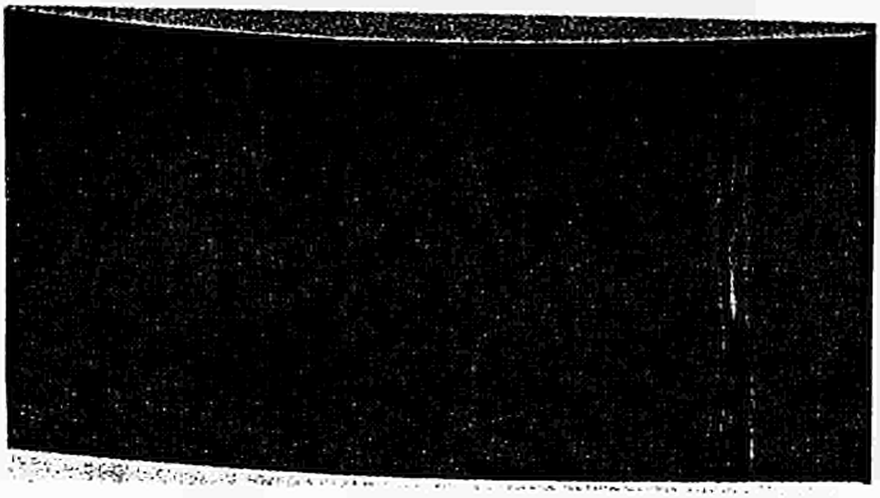


Image traitee

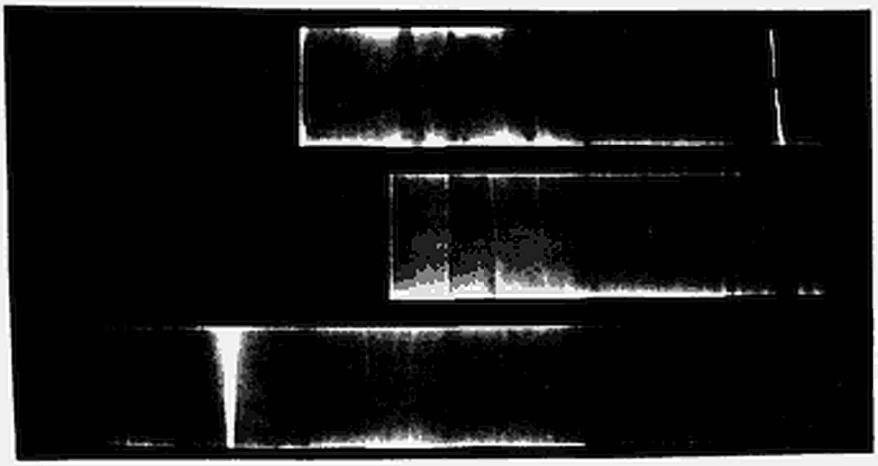


CAROTTES D'ARGILE (Aisne)

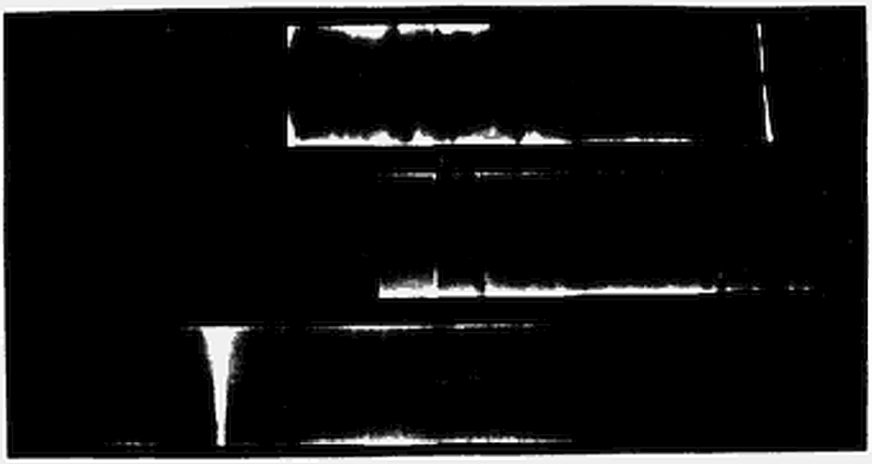
Reconstitution d'une passe

(Ø 149 mm - L = 2 m)

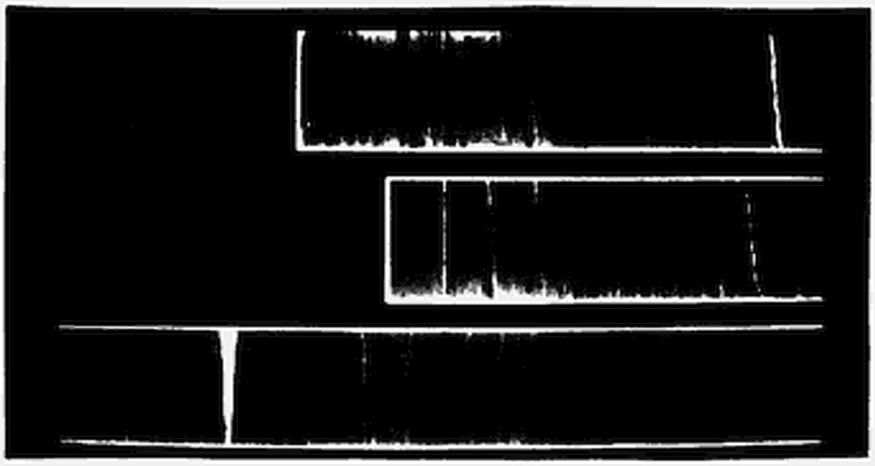
Images brutes



Images avec soustraction d'un niveau de gris constant



Application d'un operateur Laplacien



4.1.B. Geo-forecasting studies

GEOPROSPECTIVE MODELLING

Contractor : Bureau de Recherches Géologiques et Minières
B.P. 6009 45060 ORLEANS Cédex - France

Contract : n° FI 1W/0048

Duration of contract : from 1986 August 1st to 1989 March 31st

Period covered : 1989 January 1st to March 31st

Project leaders : P. Peaudecerf - J. Fourniguet

A. OBJECTIVES AND SCOPE

Since 1981, the BRGM has been working on the development of a method for systematically studying all the factors which might influence the evolution of a waste storage site and their interactions. One of the work phases consisted in a relative quantifying of the links between the factors and in modelling them so as to complete realistic scenarios These operations are carried out with a simulator called CASTOR ("Construction automatique de scénarios d'évolution d'un site de stockage de radionucléides" /Automatic design of scenarios evolution of a radionuclide storage site).

The first simulations showed that a few main mechanisms governing site evolution should be represented more realistically.

They are essentially those concerning climate variations, weathering and erosion processes and relationships between stress and hydraulic parameters. The simulation programme will have to be modified accordingly to take these mechanisms and their interrelationships into account simultaneously.

This work should make the CASTOR code operational when applying the methodology to specific sites.

B. WORK PROGRAMME

Work programme will revolve around two main aspects:

1. Increasing knowledge and modelling of mechanisms which appeared essential in previous phases.
   a. Climatology:as well mathematical expression of fluctuations in the near past and future according astronomical data, as search for present climatic equivalents to past climates.
   b. Weathering erosion: better quantifying of rates according to lithology, slopes, vegetal cover, rainfall, temperature, and a tentative modelling of erosion processes.
   c. Relationships between stress and hydraulic parameters mainly in the case of the occurence of an ice-cap covering the site area.
2. Improving the modelling of the phenomena and the representation of the results obtained.

C. PROGRESS OF WORK AND OBTAINED RESULTS

1. State of advancement

Item 1.a has been treated and mainly completed in 1987 (see annual report). The work on weathering and erosion (item 1.b.) began in 1987 by a bibliographical analysis which produced a review of processes (nature, rates, ...). A new and specific model of erosion was designed in 1988 since no available model was able to fit precisely with our requirements. Item 1.C was also treated during the past years; the resulting model has been completed in 1989.

Item 2 has been performed in 1989 after the development of the new models.

The set of the four final reports were sent to the Commission in September 1989.

2. Progress and results

2.1. Erosion and wasthering processes

Most of the existing models concern the short term modelling (human scale of some tens of years). They require the supplying of abundant quantitative data on numerous parameters, obtained by field instrumentation during several years. These results are difficult if not impossible to extrapolate at the geologic scale (several tens of thousands years).

Some models deal with more global approaches of erosion but do not provide the quantitative data we need for a geoprospective modelling.

The newly created model allows the simulation of various erosional processes in one or several catchment basins, each of them defined by their topographic values in a rectangular grid.

It also takes into account the weathering of one or several geologic layers whatever its dip. Erosion and weathering are two coupled phenomena in the model.

The driving parameters of the model are as follows:

- erodability of soils (submittted to spatial variations),
- susceptibility of rocks to weathering (with spatial variations),
- climatic action, susceptible of variation as a function of time.

The progression of erosion/weathering front can be followed on screen by the evolution of a longitudinal profile in one or several basins.

2.2. Stress and hydraulic parameters under an ice-cap

The finite element method was used for the evaluation of the hydro-mechanic behaviour of the geologic formations under an ice-cap. The phenomenon is supposed to be isothermic since the arrival of the next glaciation is foreseen after several tens of thousands years, when the thermic activity of the storage is largely if not totally weakened.

A parametric survey was conducted mainly on the limit hydraulic conditions and on the progression rate of the ice-cap.

A quick progression of the ice stops the removal of interstitial water and is considered as the undrained condition. A speed of 100 m/year was adopted as a minimum. On the downstream part of the fictive site, an imposed pressure seems valuable since it corresponds to the initial stage, after the creation of a permafrost.

On the upstream part, two imposed conditions were tested: pressure and flow.

The permeability of the faults on the fictive site was considered as far greater than the one of geologic layers. This is supposed to be the worst possible case.

The permafrost is considered as impermeable thus stopping any water movement toward the surface.

The progression of the ice-cap induces a sub-horizontal movement of underground waters downstream. But the rapidity of the ice movement stops this flow rather quickly.

3. LIST OF PUBLICATIONS

- International Association of Hydrogéologists - International Symposium hold in Orléans (France) in June 1988: *A quantitative forecast of the long term evolution of the hydrogeolgical characteristics of deep storage sites* by Ch. Filippi & al.

- ISPRA courses: Advanced Seminar on Risk Analysis in Nuclear Waste Management, May-June 1988:
  Predictive geology in the analysis of the repository evolution; description of the relevant natural phenomena, by J. Fourniguet.

  Modelling combined effects and making scenarios, by G. Aubertin.

- International Symposium fo the Safety Assessment of Radioactive Waste Repositories organised by CEC, IAEA and DECD/NEA in Paris - 9-13 October 1989: *Long term evolution of wastes disposal sites: scenario selection and methods* by P. Peaudecerf and P.L. Blanc.

4.1.C. Rock mechanics

IN-SITU CHARACTERISATION OF THE BEHAVIOUR OF DEEP CLAY LAYERS

Contractor : ANDRA, PARIS, FRANCE
Contract n° : F I1W/0049
Duration of contract : August 86 - December 89
Period covered : January 89 - December 89
Project leader : M. RAYNAL

A - OBJECTIVES AND SCOPE

The objectives of this project are :

- To complete geomechanical investigations in the Boom deep clay formation, under natural conditions and after heating,
- To develop laboratory and in-situ methods to study deep clay layers, and to compare the results obtained by these two approachs.

This work forms a basis for dimensionning the storage facilities in deep clays. The interesting point is to study the time-lag behaviour of this material, at ambient and high temperature.

The sub-contractors of ANDRA for this project are :

- LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique) for the laboratory tests, for the conception of borehole probes in cooperation with SEDITECH-MAZIER and with the BRGM, and for the in-situ test interpretation,
- BRGM (Bureau de Recherches Géologiques et Minières) for the set up, and monitoring of in-situ test,
- SEDITECH-MAZIER for the design and fabrication of borehole probes.

Experiments are performed in the U.R.L. of CEN/SCK at MOL (Belgium).

B - WORK PROGRAMME

1. Experiments at ambient temperature
1.1. Long term borehole dilatometric tests
Three tests are considered, two in horizontal holes and one in a vertical hole. Surveys of experiments are still carried out.
1.1.1. Measurements
1.1.2. Interpretation
1.2. Laboratory tests on thick tube samples of clay
1.2.1. Experiments
1.2.2. Interpretation

2. Experiments at high temperature
Design of apparatus led to construction of the firts probe in 1988.

2.1. Apparatus fabrication and test in laboratory
Preliminary tests on components of the probe are still undertaken.

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In-situ studies : the two first dilatometers are used for creep tests since December 1986 for the horizontal borehole and June 1987 for the vertical one. The third device is used for a relaxation test since December 1987 in horizontal borehole. Their loading states have been increased by stages. As for the heating dilatometer, the realization of the deformable part of the probe has brought up some difficulties relating to the adherence between the rubber body and its reinforcement. New apparatus pieces are fabricated.

Laboratory test : a set of different experiments (creep, relaxation and internal unloading tests) has been performed on Boom clay samples at varying moisture contents.

Progress and results

1. Experiments at ambient temperature
1.1. Borehole dilatometer long term test
1.1.1. Measurements

The detail of the boreholes loading is reported in tables I and II. The creep test principle is to apply a fixed pressure on the hole wall, smaller than the lithostatic stress, and to record the borehole closure. The relaxation test proceeds from the opposite principle : the borehole closure is imposed and the pressure variation is observed. The rough measurement of pressure must be corrected in order to consider the apparatus inertia pressure (about 0,4 MPa). Respectively, the rough convergence measurement must be added to the initial closure value, which occurs during the time-lag between drilling and setting of the probe, to give real convergence value. The initial closure is estimated from previous test in unlined boreholes in Boom clay.

After two previous stages the same pressure has been maintained by the first dilatometer for 1,5 year. Then the pressure has been decreased until 1,6 MPa but closure has increased very quickly (figure 1)

For the second test, the decreasing stages of pressure were chosen smaller to multiply the points on the long term closure-confinement diagramm of the material and to better border its limit pressure of instability. However the closure is rather moderate in this vertical borehole (figure 2), although final pressure is smaller than for the first test.

The last stage of the relaxation dilatometer shows that the increase of pressure stabilizes slowly and reaches a final value very near of the previous one (figure 3).

1.1.2. Interpretation

A rheological model of Boom clay has been established from numerous laboratory tests performed by another way. It reproduces in a satisfactory way the results of dilatometer tests, which confirms that scale effects are moderate for Boom clay /4/.

1.2. Thick tube test

1.2.1. Experiments

All the tests begin with an isotropic loading to the lithostatic pressure value of the sampling level. Then three types of experiment are carried out :

- regular internal unloading, with different rates (figure 4),
- creep (figure 5),
- relaxation : contrary to in-situ test, the initial pressure is not zero.

1.2.2. Interpretation

These tests give the short and long term closure-confinement curves of the material, either directly and continuously for the short term one through rapid internal unloading tests, or point by point from the stabilized couples of parameters (Pi, Ui) obtained in the long term tests. The results are coherent with each other. They show the difference between short and long term behaviour, that is the effect of ultimate capacities of Boom clay, and the influence of the moisture content (figure 6).

2. Experiments at high temperature

2.1. Apparatus fabrication and tests in laboratory

The probe body has been made. Several specimens of the deformable tube which surrounds the central probe have been realised. They failed during preliminary inflation tests probably because a bad adherence between the rubber material and its reinforcement. Since this events cause considerable delay in the project it has been decided to postpone the in situ test.

REFERENCES

/1/ - ANDRE JEHAN, R. and al, OCDE/AEN workshop on excavation responses in deep radioactive waste repositories. Winnipeg, Canada, 26-28 April 1988.

/2/ - ANDRE JEHAN, R. and al, International Symposium on Rock Mechanics "Rock mechanics and Power Plants". Madrid, Spain, 13-16 September 1988.

/3/ - ROUSSET, G. and al, CEC contractor's meeting on "Geomechanics of clays for radioactive waste disposal", Brussels, Belgium, 1-2 December 1988.

/4/ - ROUSSET, G. "Comportement mécanique des argiles profondes - application au stockage des déchets radioactifs", Thesis E.N.P.C., July 1988.

TABLE I / DETAIL OF LOADING OF BOREHOLES CREEP DILATOMETRIC TESTS

TEST	stage	rough pressure (MPa)	corrected pressure (MPa)	duration (days)
TEST 1 (horizontal)	1 2 3 4	3.65 3.10 2.40 2.00	3.25 2.70 2.00 1.60	35 130 515 >365
TEST 2 (vertical)	1 2 3 4 5 6 7	3.60 3.10 2.70 2.35 2.10 1.85 1.65	3.20 2.70 2.30 1.95 1.70 1.45 1.25	120 130 90 120 175 150 >145

TABLE II / DETAIL OF LOADING OF BOREHOLE RELAXATION DILATOMETRIC TEST

TEST	stage	corrected closure (%)	duration (days)
TEST 3 (horizontal)	1 2 3 4	1.0 2.5 #4.5 #6.5	100 265 210 >150

(stages 3 & 4 : corrected closures are not yet processed)

Figure 1 :

Rough measurements of dilatometer n° 1 (horizontal)

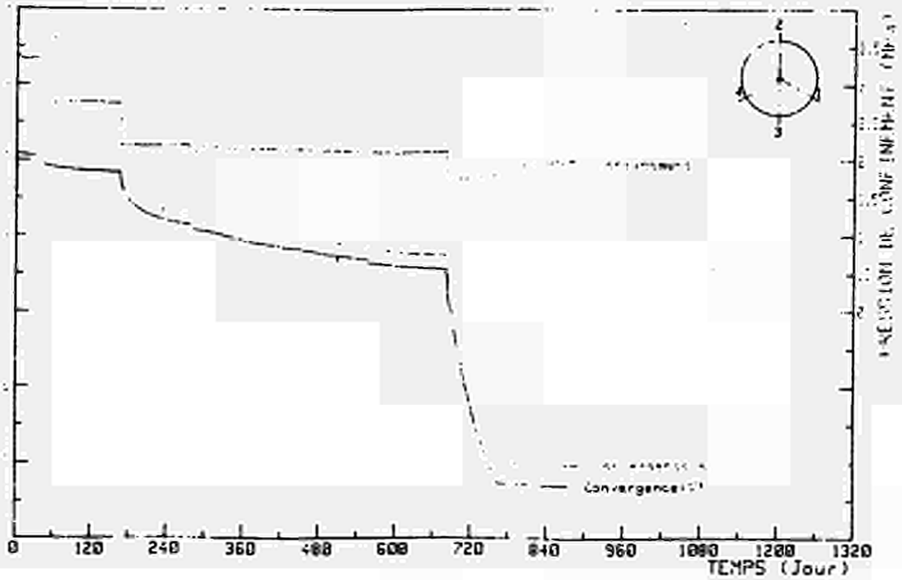


Figure 2 :
Rough measurements of
dilatometer n° 2 (vertical)

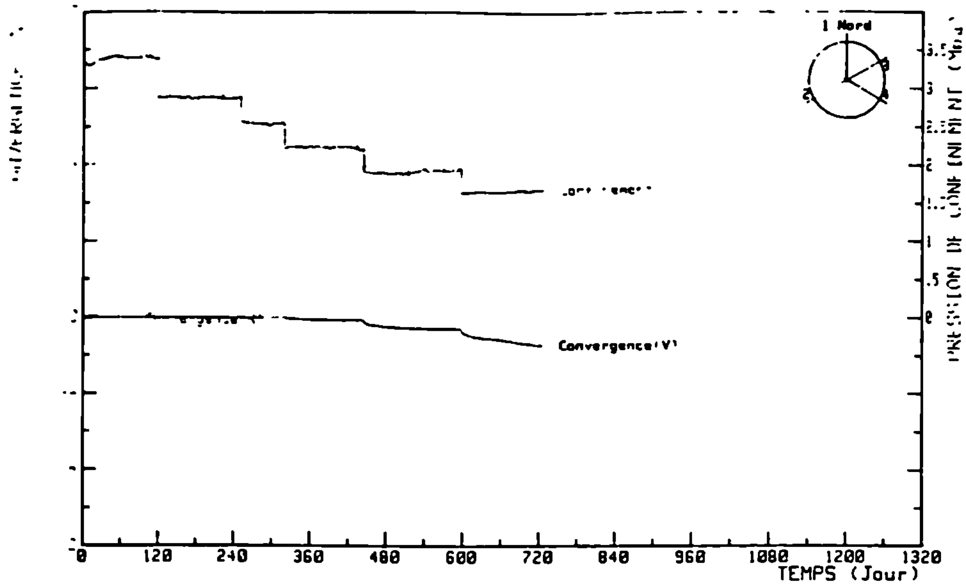
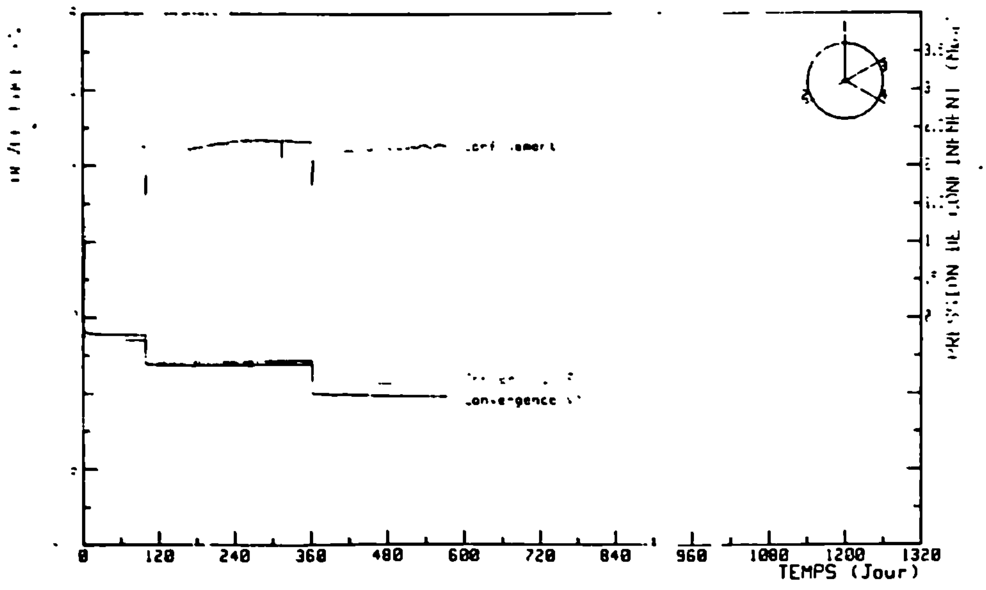


Figure 3 :
Rough measurements of
dilatometer n° 3 (horizontal)



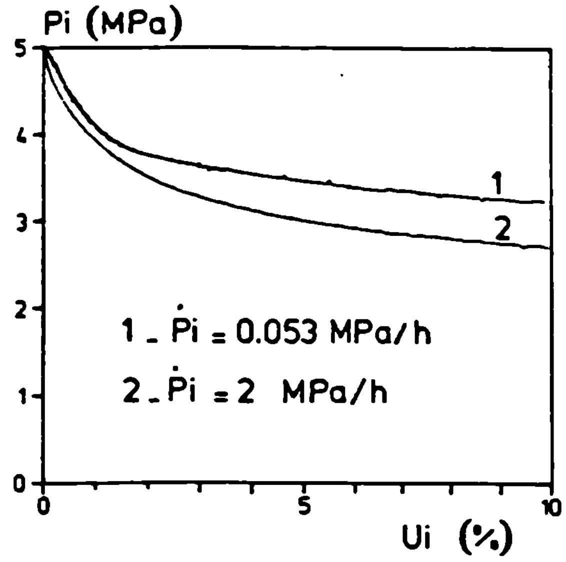
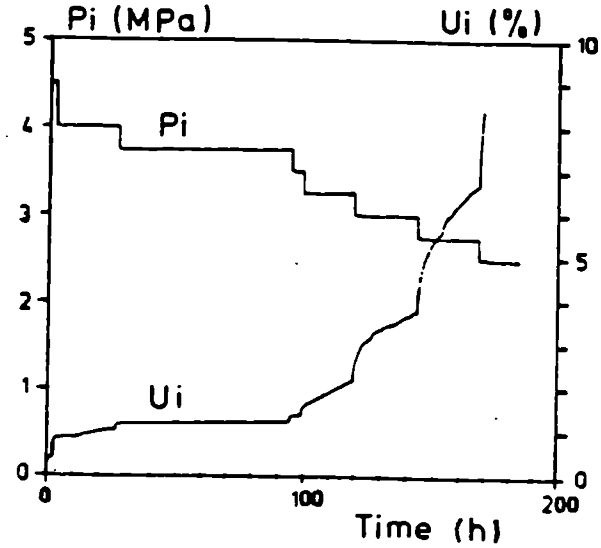
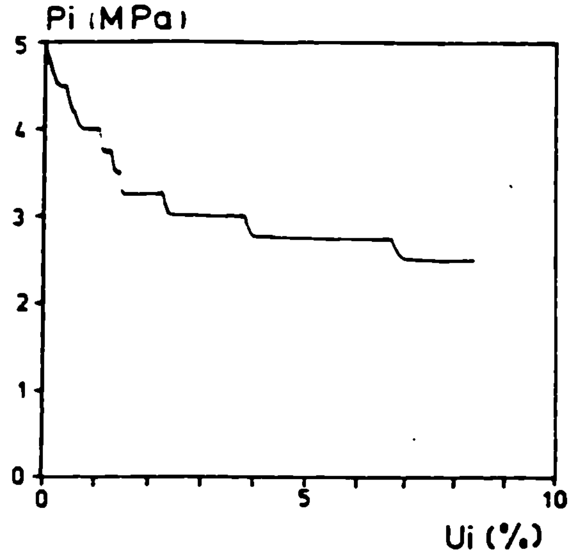


Figure 4 :
Internal unloading tests on thickwalled tube sample closure confinement curve





Figures 5 : creep test on thickwalled tube sample

5a : Pressure and closure versus time

5b : Closure - confinement curve

moisture content	w=25%		w=27%		w=29%	
time dependent behaviour	short term	long term	short term	long term	short term	long term
limit strain at failure(%)	7	7.5	14.5		11	5.5
max. deviatoric stress (MPa)	3	2.25	2.3	1.3	1	0.5

Figure 6 : ultimate capacities of Boom clay

CENTRIFUGE MODELING OF SALT DOMES

Contractor: ANDRA, PARIS (F)
Contract No: FI1W/050

Duration of contract: October 1986 - September 1989
Period covered: January 1989 - September 1989
Project leader: M. Raynal

Progress report not yet available.

Study on fracturing and microfissuration of granite

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract n° : FI.1W.0053 F
Duration of contract : from 01.12.1986 to 01.06.1989
Period covered : December 1986 - June 1989
Project Leader : S. DERLICH

A. OBJECTIVES AND SCOPE

Digging of drifts or tunnels with explosives or tunnelling machines and boring holes induce in massive rocks (granite) two kinds of stresses : the first one are stress waves and the second one are variations of the natural stresses induced by the underground openings.

Explosives loads induce cracks of tens of centimetres, or microcracks with thicknesses of some microns. Depending on the value of natural stresses and also on the shape of openings, slabing of walls and boreholes or spalling of cores may occur. Such fractures modify the mechanical characteristics of the medium and create new flow paths for the underground waters.

Few studies have been made in order to evaluate these possible effects.

Our purpose is to determine the extension of fractures around mine openings and to try to quantify fracturing or microfracturing of cores versus lithostatic pressures.

B. WORK PROGRAMME

1. Sampling

Fractures and microfractures will be studied on samples of granite.

1.1. Samples from 100 m depth

Study of explosive induced fractures in drift walls and of stress release in zones far from explosive mining ; release of stress will be studied by overcoring.

1.2. Samples from surface to 1000 meter depth (Auriat borehole).

2. Preparation of rock samples

Successive impregnations will be made with different dyes in order to identify the fractures induced by each operation.

3. Study of fractures

Different sizes will be considered :

. thicknesses from 10^{-3} to 10^{-6} m.

. " or pores less than 10^{-6} m

Several methods will be tested and the best ones used for the study.

3.1. Volume study

. Optical examens on thin sections cut following three directions.
. Electron microscope
. Castaing micro probe
. Porosimetry tests

3.2. Surface study

Fissures will be opened and the two surfaces limiting the voids will be studied (rugosity).

4. Interpretation

It will be a tentative to find a relation between the variation of stresses applied to the sample and the observed mechanical effects of fracturing

C. PROGRESS OF WORK AND OBTAINED RESULTS

Sampling

In large diameter cores (1 m diameter)

Parallel to the core axis and along a diameter, 4 smaller cores have been sampled in order to study the distribution of microfracturing between the center of the large core and his external limit.

Studies

Auriat samples

1) Geometrical characteristics of small fractures in thin sections and the void distribution at depth between 300 m and 999 m were determined.

2) Oriented petrographic studies are now going on.

Results will be compared with those obtained on granite samples coming from the URL (Canada).

PROGRESS AND RESULTS

Porosity and quantitative analysis of fractures and microfractures - Final results

The first selection criterium was the aspect of the samples : fresh, sligtly weathered, highly weathered.

Porosimetry results allowed to determined the pore (or microfracturing) size distribution.

In a second phase of work, samples were choosen at depth increasing from 300 m to total depth in the Auriat borehole (1000 m).

Considering previous results (see APR 1988) on the pore volume variations versus stresses, figure 1 shows that the compressibility of the granitic matrix, of the small fractures (the largest pores) and of the microscopic fissures has a weak influence compared to the weathering induced porosity.

The porosimetry samples are cylinders, 24 mm diameter, 26 mm height. They are diamond-cutted and they need massive pieces of core to allow a good cylinder shape.

Same remark must be made upon the thin sections prepared for the quantitative microscopic studies.

Figure 2 shows the distribution of the number of micro-fissures per cm^2. This value shows no correlation with the depth of the samples. These two methods are very good to evaluate the ability of the matrix to absorbe underground fluids but not to determine mechanical fissures induced by stresses.

The programe on microstructural analysis has begum in the two last months of 1989.

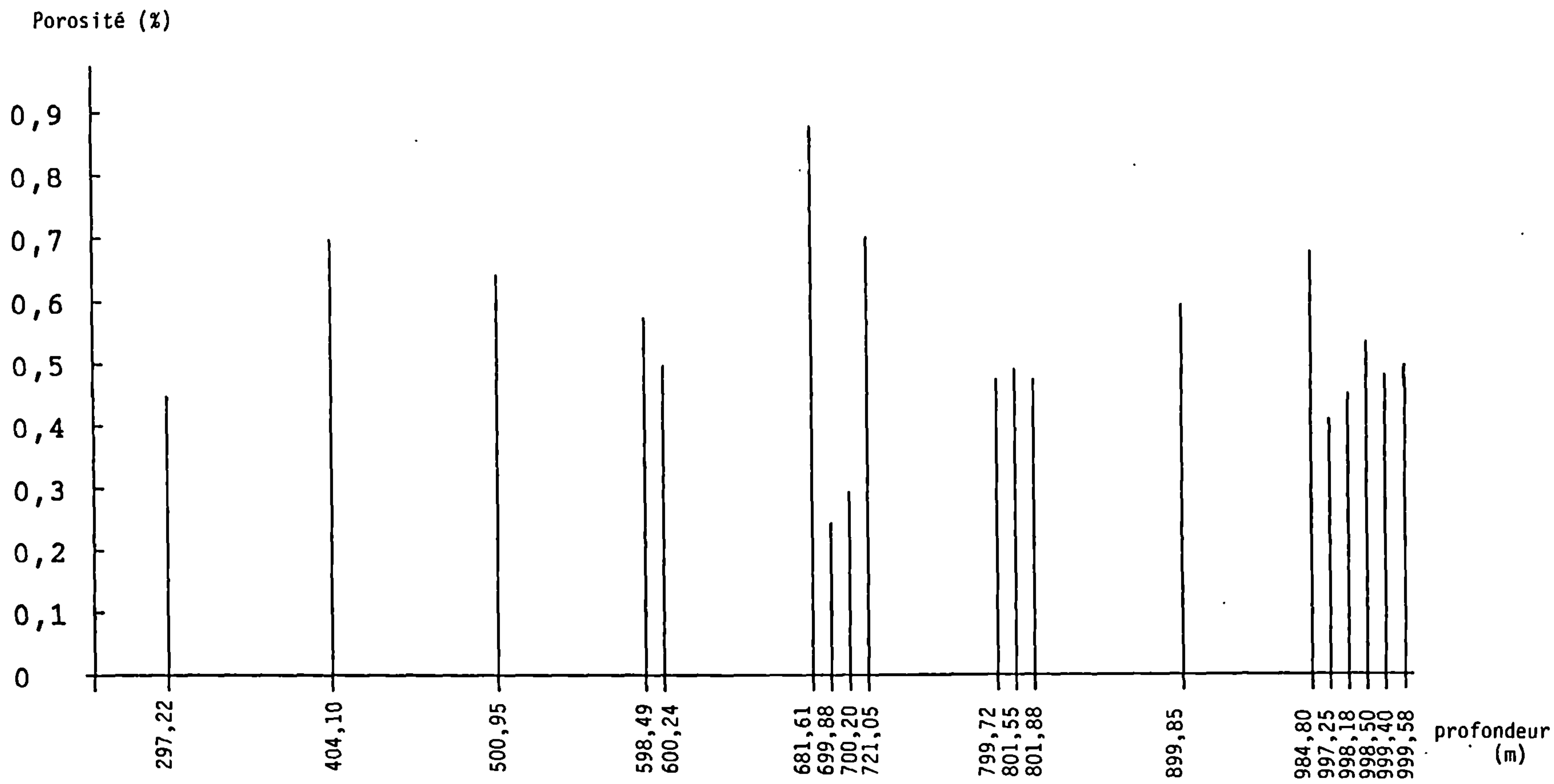

Porosité (%)
0,9
0,8
0,7
0,6
0,5
0,4
0,3
0,2
0,1
0
297,22
404,10
500,95
598,49
600,24
681,61
699,88
700,20
721,05
799,72
801,55
801,88
899,85
984,80
997,25
998,18
998,50
999,40
999,58
profondeur
(m)

FIGURE 1

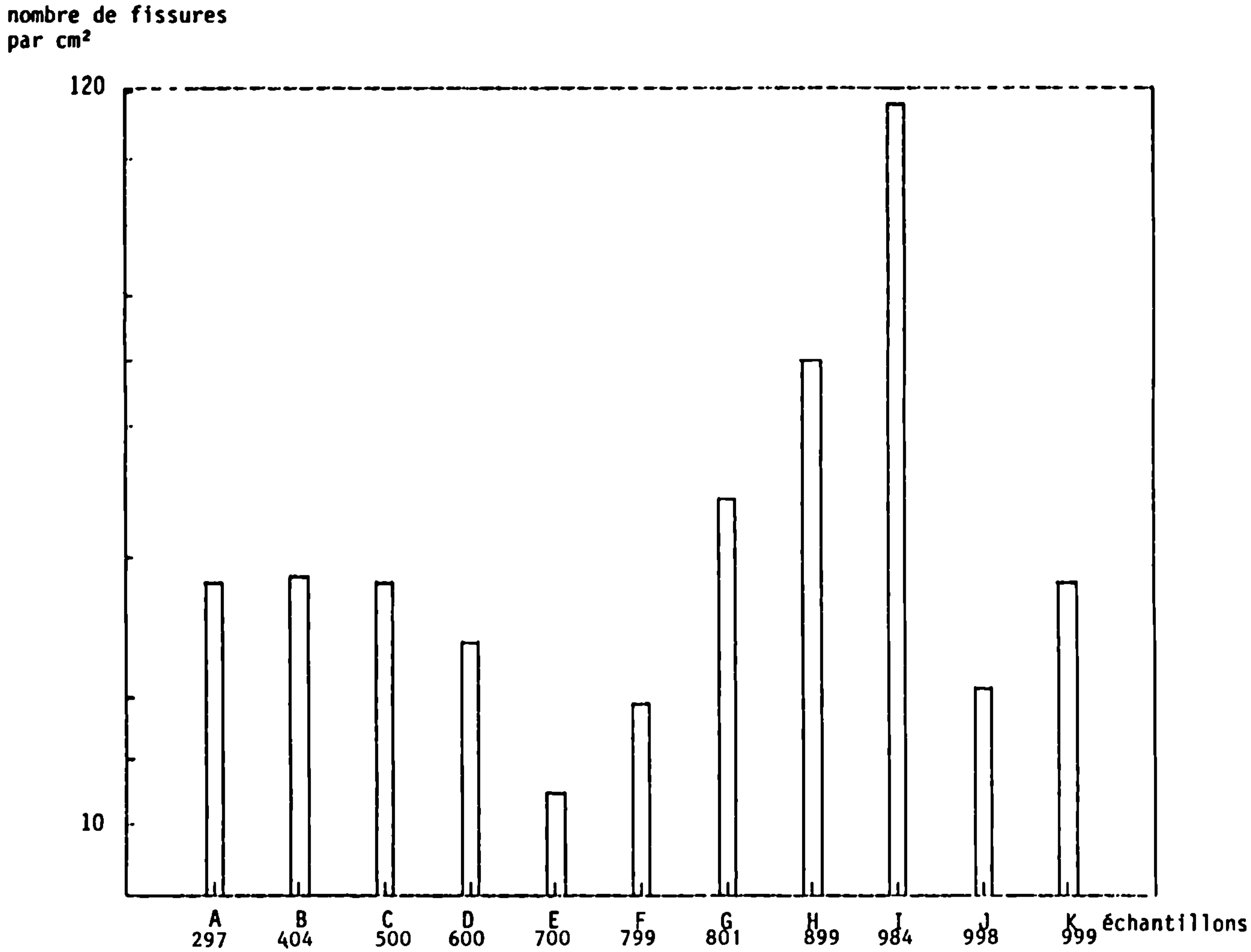

nombre de fissures
par cm²
120
10
A
297
B
404
C
500
D
600
E
700
F
799
G
801
H
899
I
984
J
998
K
999
échantillons

FIGURE 2

FURTHER BENCHMARK EXERCISES TO COMPARE GEOMECHANICAL COMPUTER CODES FOR SALT (COSA II)

Contractor : WS Atkins Engineering Sciences, Epsom UK
Contract No : FI1W/0054
Duration of Contract : November 1986 - January 1989
Period Covered : January 1989
Project Leader : N C Knowles

A. OBJECTIVES AND SCOPE

Research into geomechanical aspects of HAW repositories in salt formations has been active in the European Community for nearly two decades, with particular interest being placed on problems of heat producing waste. Central to this work is the prediction of stresses and deformations in the host strata, for which a number of computer codes have been used /1/. A preliminary exercise ("COSA 1") to compare the ability of the different codes /2/3/ provided a limited "snapshot" of the European capability to predict the behaviour of rock salt under well defined conditions. The purpose of the present contract which was completed in January 1989, was to extend the comparison to more complex but realistic situations.

Comparison problems in COSA I were relatively simple, and a number of difficulties to do with modelling the in-situ behaviour of rock salt were deliberately avoided. The present exercise is directed at comparisons of realistic, albeit relatively short-term, in-situ behaviour. Emphasis was placed on the requirement to predict (rather than replicate) real-life behaviour and individual participants were allowed considerable freedom to characterise the physical situation and material behaviour according to the dictates of their experience. A key feature was that, as far as possible, calculations were performed 'blind' (i.e. without any knowledge of the observed behaviour), using the best information available a priori.

There were 10 participants in the exercise, each acting as a sub-contractor to the coordinator. In addition two independent experts provide advice as necessary on aspects of salt rheology (Table I).

B. WORK PROGRAMME

The Work Programme was as follows:

- B.1 Participants jointly, at plenary meetings, agree programme of suitable in-situ benchmark problems.
- B.2 Coordinator prepares discussion documents as necessary and circulates to participants.
- B.3 Coordinator prepares and circulates detailed specifications of agreed problems.
- B.4 Participants solve benchmark problems as specified by coordinator, to the best of their ability using appropriate codes.
- B.5 Coordinator collects and collates results and other data from participants.
- B.6 Coordinator prepares draft reports for discussion at plenary meetings to be held approximately every 6 months.
- B.7 Coordinator prepares and issues final reports taking due account of participants comments.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

Three benchmark problems were identified at the start of the exercise. They are based on the series of experiments performed by ECN in the 300m dry-drilled borehole at the Asse research facility.

The first problem was the prediction of the isothermal free convergence (IFC) at the bottom of the borehole over a period of some 800 days. This was largely completed in 1987 but a number of additional results were submitted later.

The second problem was to predict the behaviour (including pressure build-up) as the borehole wall converged onto a heated probe (HPP1). Preliminary results for this benchmark were collated and subsequently discussed at a plenary meeting of participants at Ismes, Bergamo in February 1988.

The third problem was to predict the free convergence of the borehole during a heater test (HFC1). The specification of this benchmark was discussed at the plenary meeting in Bergamo, it was then finalised and calculations were performed, prior to comparison and discussion of the results at a plenary meeting in Madrid, September 1988. The predictions again agreeed qualitatively with the experimentally measured behaviour, but quantitatively there were considerable differences, which can be attributed to the various constitutive models used.

A plenary meeting of participants was convened in Palaiseau in April 1989 following formal completion of the project in January 1990.

Progress and Results

1. IFC Benchmark

In this benchmark participants were asked to predict the convergence of the bottom of the borehole under the isothermal conditions of lithostatic stress. The results of the preliminary calculations by the various participants contains wide variations. These can be attributed to the differing assumptions about the initial state of lithostatic stress and the material constitutive law, since when normalised with respect to stress and creep law constants very good agreement was obtained. This is in contrast to benchmarks of COSA I in which analyst error and differing discretisation techniques caused large variations.

2. HPPI Benchmark

The experimental test on which this benchmark was based, involved monitoring the variation with time of pressure and temperature on a heated probe fixed in the borehole at a depth of 262m over a period of 60 days. Participants were asked to predict this behaviour. As with the IFC benchmark, the specification gave no quantitative details of the measured behaviour and defined only the physical arrangements for the test. It was left for each participant to decide how best to model the test, especially with regard to the material constitutive behaviour. There was also some discussion about the importance of the inevitable small gaps between the probe and the borehole wall, but nevertheless perfect contact was specified in order to confine the model variations to reasonable bounds. Subsequently the effect of gaps was investigated by a number of participants and was shown to be relatively minor compared to other influences.

In general there was good agreement among the predictions of the temperature field but the prediction of the temperature at the borehole were all higher than measured. The reasons are attributed to a variety of sources, including uncertainty about thermal properties of the salt, heat loss in the cables and experimental error.

Predictions of the geo-mechanical behaviour agreed qualitatively but differed substantially in absolute terms (eg predicted peak pressures differed by a factor of approximately 2.5). The behaviour is dominated by the early thermo-elastic response to a steadily increasing temperature and accordingly is sensitive to spatial temperature gradient and to the values used for Elastic modulus and coefficient of expansion.

3. HPCP Benchmark

The heater test on which this benchmark was based, followed chronologically the HPP test and was conducted at a depth of 237m down the borehole. The benchmark required the prediction of the temporal and spatial temperature and stress variations in the salt mass adjacent to the borehole over a period of 20 days due to a heat source of approximate 5KW in the borehole.

As before, participants were, quite deliberately, allowed complete freedom in modelling the test. In practice the key issues again centred on the material constitutive model.

The temperature predictions for the salt generally agreed well with each other. Moreover, given the difficulties encountered in obtaining reliable temperature measurements during the test, they appear to be consistent with reality.

The stress and convergence predictions exhibited wider variation (maximum displacements varied by a factor of approximately 4) but were qualitatively similar. In contrast to the HPP benchmark the behaviour at the borehole wall exhibits a complex interaction between the visco-elastic response to a time varying thermal straining and the thermally accelerated creep observed in the IFC. The predictions bounded the measured behaviour and the variations can be attributed to the different models used to account for the material behaviour.

The 3 benchmarks provide good insight into the role of material models and highlight the difficulty in establishing reliable ones for "blind" predictions. It appears that most models can be "tuned" to replicate measured behaviour satisfactorily, but the reliability of such models to predict unknown behaviour is unproved.

List of Publications

1. LOWE M.J.S., KNOWLES N.C. 'Project COSA - A Benchmark of Computer Codes for the Geomechanical Behaviour of Rocksalt'. Proc. Int. Conf. Reliability of Methods for Engineering Analysis, Swansea, 1986.
2. KNOWLES N.C., LOWE M.J.S. 'A Benchmark Exercise on a Thermal Elasto-Creep Problem'. Structural Analysis Systems World Conference, Paris, 1986.
3. KNOWLES N.C. 'Project COSA - A Benchmark of Computer Codes for the Thermal-Mechanical Behaviour of Rock Salt'. Workshop on Mathematical Modelling for Radioactive Waste Repositories, Madrid, 1986.
4. KNOWLES N.C., LOWE M.J.S. 'Some Experiences of Finite Element Calculations in a European Benchmark Exercise'. NAFEMS Int. Conf. on Quality Assurance and Standards in Finite Element Analysis, Brighton, 1987.

5. KNOWLES N.C., LOWE M.J.S., PIPER D. 'An Update on Project COSA' Trans. 9th Int. Conf. on Structural Mechanics in Reactor Technology, Lausanne, 1987.
6. COME B. 'Benchmarking Rock Mechanics Computer Codes : The Community Project COSA' Proc. 6th ISRM Congress on Rock Mechanics, Montreal, 1987.
7. COME B. 'Le Projet Communautaire COSA : Un Exemple d'Intercomparaison de Codes de Calcul Geomecaniques pour le Sel' Revue Francaise de Geotechnique, 1987.
8. PIPER D., KNOWLES N.C. 'Some Computational Experiences of a Geomechanical Benchmark in Rock Salt'. Proc. 6th Int. Conf. on Numerical methods in Geomechanics, Innsbruck, 1988.
9. KNOWLES N.C., COME B. 'A Progress Report on Project COSA'. Workshop on Excavation Response in Deep Radioactive Waste Repositories, Winnipeg, 1988.
10. LOWE, M.J.S., KNOWLES N.C. 'COSA II Further Benchmark Exercises to Compare Computer Codes for Salt' Draft CEC EUR Report 12135 EN
11. KNOWLES, N.C., LOWE, M.J.S. and COME, B, 'On the Reliability of Predictions of Geomechanical Response - Project COSA in Perspective', Proc. Int. Symp. on Radioactive Waste Repositories OECD, Paris, 1989.

References

/1/ BROYD, T.W., et al. CEC EUR Report 8669 (1985)
/2/ LOWE, M.J.S.L., KNOWLES, N.C., CEC EUR Report 10760 EN
/3/ PIPER, D., LOWE, M.J.S., VOUILLE, G., CEC EUR Report 12134 EN

Table I : List of Organisations involved in COSA II

WS ATKINS ENGINEERING SCIENCES - Epsom (UK)	Co-ordinator
FORAKY - Brussels (B)	Calculation Team
LGC - Louvain-la-Neuve (B)	Calculation Team
KfK - Karlsruhe (D)	Calculation Team
RWTH - Aachen (D)	Calculation Team
CEA-DEMT - Saclay (F)	Calculation Team
EMP - Ecole des Mines - Fontainebleau (F)	Calculation Team
LMS - Ecole Polytechnique - Palaiseau (F)	Calculation Team
ISMES - Bergamo (I)	Calculation Team
ECN - Petten (NL)	Calculation Team
ENRESA/ETSIM - Madrid	Calculation Team
GSF - Braunschweig (D)	Salt Specialist
Technical University Delft (NL)	Salt Specialist

MECHANICAL BEHAVIOUR OF CLAY UNDER AMBIENT AND ELEVATED TEMPERATURE CONDITIONS

Contractor : CEN/SCK, Mol (B)
Contract No : FI1W/0055/B
Duration of contract : from October 1986 through June 1990
Period covered : January 1989 to December 1989
Project leader : A.A. Bonne
Report by : B. Neerdael

A. OBJECTIVES AND SCOPE

In 1974 CEN/SCK launched a R&D programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site.

Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hosting radioactive wastes as well from engineering point of view as for long term safety and performance assessments.

A particular area of interest is the one of (thermo-) mechanical behaviour of clay. Several in situ experiments and tests are developed and performed jointly with ANDRA in this field.

B. WORK PROGRAMME

B.1.1 Stress measurements in non-frozen clay
B.1.2 Fracturing in clay
B.1.3 Long term dilatometric tests

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

On the short term few information will be available concerning the behaviour of Boom clay under elevated temperature conditions; this is due to the low temperature level obtained in the vicinity of heating elements (e.g corrosion tests) implanted in the Boom clay at Mol before some of them definitively failed.

The design and calibration of the "heated" dilatometers developed in France and to be experienced in Mol for ANDRA does not progress according to schedule and cannot be tested in situ anymore in the frame of this research contract.

This year was essentially devoted to the follow-up of the hydraulic pressure gauges installed in clay to get information on the stress field. Geophysical investigations were performed regularly from the underground research laboratory for the detection of natural or induced fractures in clay.

Further, the SCK/CEN-team contributed to the thermo-mechanical modelling studies on clay, made by ISMES; complementary mineralogical analyses could be needed to improve the interpretation of the laboratory tests.

Progress - Main achievements

Stress field

The measurements at the three Gloetzl stress monitoring stations are going on; the set of available data has been recently increased since that measurements are also performed on two complementary devices located in the same portion of the underground laboratory around the experimental set-up of the "Bacchus" test.

The data available now are sufficient and allow to go more in detail with the interpretation, trying to make the correlation with previous results.

Progress was made in the placement techniques.

Clay fracturation

Due to the very slow temperature evolution and low temperature level at the place of the test, only three measuring campaigns were made this year.

All the information available has been synthetized with the experimenters themselves and presented at the recent colloquium organized by the Belgian Committee for engineering geology /1/.

More attention is now devoted to the correlation of the actual survey with the clay stratigraphy at this location.

One laboratory experiment was started to investigate at small scale and using seismic sensors the response of a clay sample submitted to various confining and heating conditions; these conditions also cover the in situ experimental conditions.

With regard to the consolidation state and process, the heating (80°C) seems to be of negligible influence on the propagation velocity through the sample.

In the framework of the ME2I research programme for ANDRA concerning the study of clay behaviour around a heat source by way of spectral analysis, three measuring campaigns have been made between December 1988 and December 1989, all of them before starting the heating phase. The next campaign will bring some more information about heating influence on clay (scheduled for February 1990).

Mechanical behaviour

The three long term dilatometer tests, considered in the characterization programme of ANDRA (application of the convergence/confinement theory), were running until now but will be disconnected begin next year. The very consistent results are commented by ANDRA in their corresponding contribution.

List of publications

/1/ JONGMANS.D and al. Etude géophysique en champ proche à partir d'une galerie dans l'argile, Colloq. Nat. "Application des méthodes de prospection géophysique à la géologie de l'ingénieur", Liège, Sart Tilman, 17-19 oct., 1989

A THERMO-MECHANICAL BEHAVIOUR OF BOOM CLAY.

Contractor: Ismes S.p.A.- Viale Giulio Cesare, 29 -24100 BERGAMO ITALY
Contract n.: FI1W/0150
Duration of contract: 29 months, from 1.11.1988 to 1.2.1990
Period covered: 1.1.1989 to 1.1.1990
Project Leaders: Dr. A. Peano, Ing. G.Baldi.

A. OBJECTIVES AND SCOPE

The understanding of thermal effects due to decay heat like pore pressure rise, change in mechanical properties and in the hydraulic field under high lithostatic pressures is fundamental in the evaluation of safety and in the design of nuclear waste repositories in clay.

For this purpose a thermo hydro-mechanical model has been developed in ISMES. Its suitability in reproducing the thermo mechanical response of the samples tested under different loading conditions has been positively verified with experimental tests performed on the HITEP triaxial cell operating in ISMES laboratories on various types of stiff clays, either reconstituted and undisturbed.

Furthemore, the interpretation procedures of thermal tests showed that for some of the clays tested the Campanella an Mitchell procedure can lead to inconsistent results.

In order to study possible reasons for this inconsistency, first thermal expansion coefficient values will be studied referring to the conditions of water in stiff clays, which are suspected to affect significantly its thermal-behaviour. This study will be conducted with AECL of Canada.

The results of this research could affect also the predicted undrained response of clay in terms of pore pressure rise.

In the meantime a better understanding of thermo-mechanical behaviour of Boom clay is foreseen in order to improve the model.

Thermo-mechanical effects on clay due to realistic disposal geometries will be then studied with this model with the cooperation of SCK/CEN.

Similar studies are planned also for sand/betonite-mixtures, widely adopted for backfilling.

B.WORK PROGRAMME

1. Microstructural studies
2. Sampling of the Boom clay in the Mol site.
3. Experimental tests in the HITEP apparatus on natural and artificial clay soils.
4. Identification of material parameters on the basis of experiments on Boom clay.
5. Introduction of the options expected from activiy 1 in the mathematical model.
6. Check on the results of the laboratory tests using the above model.
7. Definiton of the boundary value problems to be treated on the basis of prospective in-situ experiments and/or typical disposal technology at Mol site.
8. Simulation of clay mass behaviour with reference to the problems envisaged in activity 7.
9. Evaluation of simulation results.

C. PROGRESS OF WORK AND OBTAINED RESULTS.

State of advancement.

Studies on the microstructural properties of stiff clays have been undertaken in order to assess the role of adsorbed pore water layers in clay response to nuclear waste heat. A law for thermal expansion coefficient of pore water has been developed at ISMES together with a mathematical framework to account for adsorbed water presence within the theory of mixtures of porous deformable continua. Experiments aimed at supporting the modeling of thermal water expansion coefficient of pore water for Boom clay have been performed in a parallel research performed by AECL on the behaviour of stiff active clays within a cooperative research agreement.

Laboratory tests at ambient and elevated temperature are in progress at ISMES laboratories for specification of the thermo-mechanical properties of Boom clay. The results should specify the constitutive functions of the thermo-mechanical model as well as be used for calibration of its constants. An unexpected effort has been devoted to study the mechanical response of Boom clay at -223 m. The material appeared at last definitely different from that studied by ISMES and BGS in a previous test campaign referred to as Boom -240m level. Due to the relevance of this level for the thermo-mechanical in situ tests in the Mol underground laboratory, it has been judged more appropriate to turn back to investigate it. Further mechanical and thermo mechanical tests has been selected then. This supplementary test program started in July will end by January 1990.

The analysis of the results of the laboratory campaign at ambient temperature lead to a revision of the constitutive model. The mathematical framework has been settled. The dependence of the revised constitutive functions with temperature will be analyzed on the basis of the thermomechanical tests.

PROGRESS AND RESULTS

1. Microstructural studies

Boom clay has been included as test material in a research performed by the WNRE of Atomic Energy Board of Canada Ltd. on microstructural properties of stiff clays.

The 1-D prototype cell was ready for tests in the early spring of 1989. Calibration tests with de-ionized water gave α_w values in satisfactory agreement with those published in technical literature. The test program involves heating and cooling cycles from 25°C to 100°C with clays with different mineralogic properties. The material selected were: Sealbond (smectitic), Avonlea clay (Illitic), kaolin, Silica Sand (quartz), Boom clay. It resulted that inert clay minerals as well as quartz minerals show no deviation in α_w values respect to those of pure water. Clay with active minerals show a remarkable decrease of α_w value with temperature respect to that of pure water. This was also the case of Boom clay. Experimental and processing inaccuracies are not judged capable of altering the observed trend (Fig.1.1).

2. Sampling of Boom Clay in the Mol Site.

Two additional samples, n.17 and n. 30, have been provided by SCK/CEN, for the additional test campaign. They have been taken during 1983 from elevation -241m and -243m and kept in wooden boxes.

3. Tests on Boom Clay

Tests on Boom clay are in progress at ISMES using conventional geotechnical devices and the HITEP apparatus.

Classification, mineralogic tests have been performed on Boom clay samples R116-W112, R116-W110, R118/W112, R66/W42 and R96/W66. Results confirm that with some reasonable scatter, Boom clay at -223 m has different properties respect to that present at -240 m level. X-ray diffractometry on random powder and on the $\leq 2\ \mu m$ fraction has been performed together with DTA,TG and DTG analyses.

Measured clay content and plasticity index are greater (Fig. 3.1.1). Clay and interlayer minerals (chlorite, smectite) found in Boom clay lead to consider this material as less expansive respect to Boom - 240 m (Fig. 3.1.2).

Considerable scatter in compressibilities has been found from stress and strain controlled oedometer tests; higher compressibility has been found with respect to the clay at 240m depth. Preliminary evaluation of the results seems to confirm that maximum consolidation vertical stress is around 5MPa (Fig. 3.1.3).

Two triaxial drained test and one undrained in the range of high OCR have been performed (0.8 MPa 1.7 MPa, 2.5 MPa initial isotropic stress). The new material revealed more compressible and with a lesser residual resistance (ϕ'=15.8° respect to meanly 23° of ultimate resistance in Boom level -240).

It resulted that more tests than those foreseen were necessary. Six triaxial tests on HITEP apparatus on R116/W110 samples have been then performed. Three triaxial drained compressions tests at increasing initial stress (p'=2, 3, 4 MPa respectively) and one undrained at 4.75 MPa have been performed as to judge about yield onset, elastic shear compliance, dilatancy, ultimate stress values.

From the shearing phase, it resulted that shear response is characterized mainly by softening-compactive response. The ultimate friction angle is again 15.8° in each test (Fig. 3.1.4). Elastic secant shear modulus is increasing with initial isotropic stress and is comparable to that of Boom clay at -240 m level.

Particle size and mineralogy tests on new samples taken from -240 m level have been performed. The clay tested revealed some scatter in sand and clay content. Clay from level -240 has a greater sand content respect to that from -223 level, and lower water content. In the case of the two samples tested the sand content is remarkably high (28% sample n. 30) and water content low (16%), probably because of the long storage time. This is confirmed by the observation that samples were initially not saturated: saturation has been achieved at approximately 18.5% water content. Mineralogic tests reveal greater amount of active clay minerals (smectite 10%).

Two compression triaxial tests have been performed on HITEP apparatus, one in undrained conditions (p'= 4.75 MPa) and one in drained conditions (p'=2.00 MPa) with sample n.30. The material showed ultimate shear resistance very similar to that typical for Boom -240 m. The material seems however more rigid also respect to Boom -240 m, may be because of the high sand content.

Three thermomechanical tests have been then performed on clays taken from sample n. 30. They consist in heating-cooling cycles at different isotropic stress values and loading at different temperatures. An heating test in undrained conditions under sustained shear stress. The results are under evaluation.

4. Mathematical modeling

An elastic plastic model has been developed with the following characteristics. In the elastic domain, non linear elastic behaviour has been described where shear modulus is dependent on the isotropic state of stress.

The yielding locus is described by two functions that merge without discontinuity: n the highly overconsolidated states,a hyperbolic - linear in the stress invariants space; in the slightly overconsolidated to normally conslidated states an ellipsis. The yield surface is flatter in the overconsolidated domain respect to what predicted by the Mod Cam Clay at the same preconsolidation pressure.

The flow rule is non associated. The plastic potential has been defined as an ellipsis in the stress invariants space. In order to meet the variety of the dilative response shown by Boom clay, the locus of potential separating the compactive from dilative response may not be coincident with the critical state line, to which it should collapse only at terminal part of the loading process.

The evolution of the yield surface depends on the accumulation of plastic volumetric and deviatoric strains (double hardening).

A phenomenological relationship for thermal expansion coefficient of interstitial water based on the results obtained in isotropic heating experiments on the HITEP apparatus on reconstituted Pontida silty clay at high stresses and temperature has been developed at ISMES.

It considers not only temperature dependence, as usual for pure water, but also bonding pressure dependence. Predicted values of thermal expansion coefficient are considerably lesser at high pressures. The lower water expansion coefficient may be due to the different physical state of the interstitial water in stiff clays. Strains calculated following this relationship agreed well with the experimental strains obtained from the axial measures.

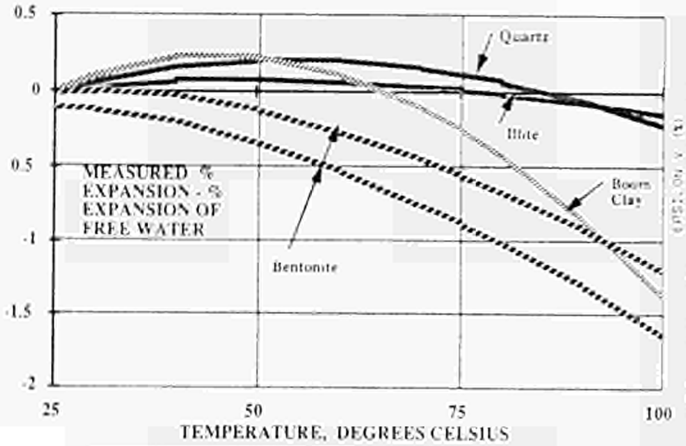
A sensitivity analysis of the model has been performed as to see if the mid-space bonding force can be considered equal to the effective stress. The simulations carried out indicate that while this approximation was acceptable in the range of pressures considered in the normally consolidated ranges of the clays studied, where the mid-space values for the adsorbtion force were of the order of few to ten MPa, it can lead to erroneous expansion data when used for real low pressure values.

5. List of publications

/1/ PELLEGRINI R., PEANO A., BALDI G. CEC Technical Contractors Meeting "Geomechanics of clays for radioactive waste disposal", Bruxelles 1-2 December (1988).

/2/ PELLEGRINI R. 9TH EURATOM - AECL Exchange Meeting Pinawa, 19-21 September (1989).

Fig. 1.1 Difference between measured expansion of soil water and expansion of free water as a function of temperature for four minerals



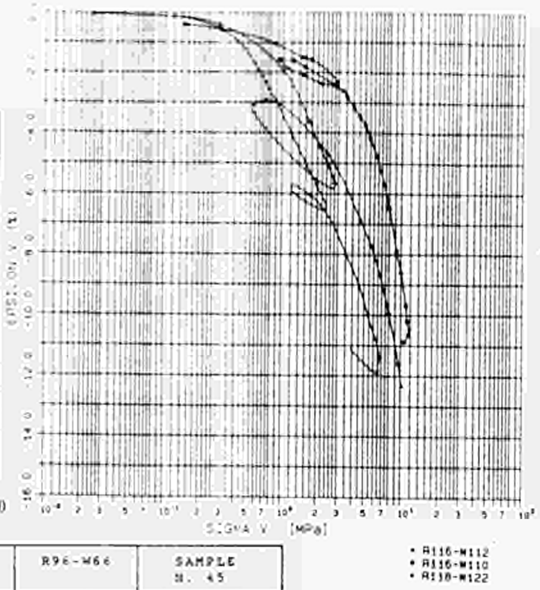


Fig. 3.1.3

Tab. 3.1.1

	R118-W122	R116-W122	R116-W110	R66-W42	R96-W66	SAMPLE N. 45
SAND	5%	2.0%	3.0%	3.0%	7.0%	12.3%
SILT	49%	37.0%	49.0%	38.0%	48.0%	47.2%
CLAY	46%	61.0%	47.0%	58.0%	45.0%	40.5%
LL	77.9%	61.0%	72.0%	68.1%	60.8%	56.6%
PI	55.9	50.0	46.0	45.4	40.5	29.7
SL	16.3%	/	19.5%	13.1%	13.0%	
GS	2.69	2.71	2.70	2.69	2.68	2.69
W	/	27.5%	26.0%	30.3%	25.2%	20.0%

Tab. 3.1.2

	RANDOM POWDER	RANDOM POWDER	RANDOM POWDER	RANDOM POWDER	RANDOM POWDER
	Z=223 M (R116-W112)	Z=223 M (R116-W110)	Z=223M (R118-W122)	Z=223M (R66-W42)*	Z=223M (R96-W66)
SMECTITE	< 5%	< 5%	< 5%	5-10%	5-10%
CHLORITE	5-10	< 5	< 5	TRACES	TRACES
MIXED-LAYER INTERGRADE *	< 5	10-15	15-20	10-15	10-15
ILLITE	20-25	15-20	< 20	< 20	< 20
KAOLINITE	20-25	15-20	< 20	15-20	15-20
QUARTZ	20-25	15-20	< 20	15-20	15-20
FELDSPAR	> 5 (4 8)	< 5	<< 5	<< 5	TRACES
ORGANIC	5-10%	5-10	5-10	5-10	5-10

(*) ALSO PRESENT CALCITE <=5%

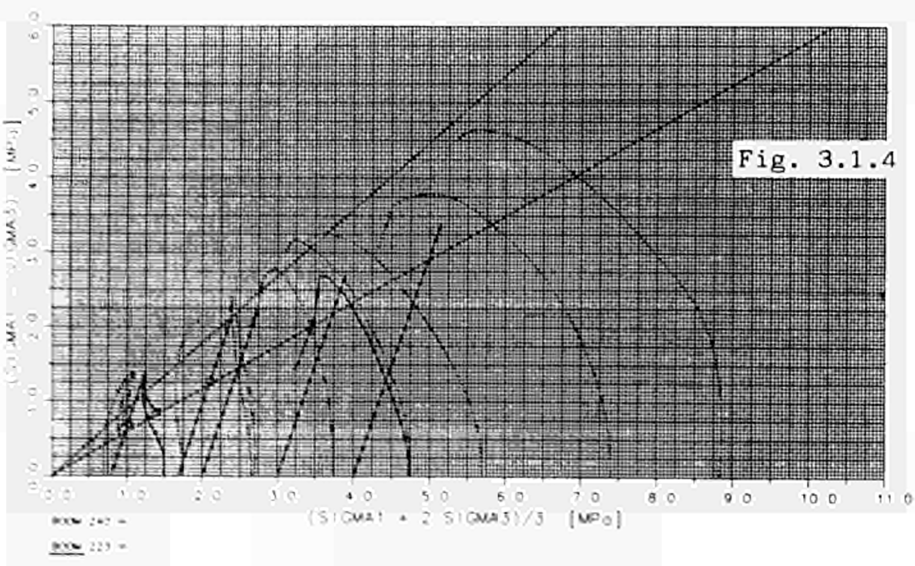


Fig. 3.1.4

STUDY OF THE CLAY BEHAVIOUR AROUND A HEAT SOURCE WITH FREQUENTIAL SEISMIC

Contractor : ME2i, 38 PLACE DE LA LOIRE, SILIC 451, F-94593 RUNGIS
Contract n°: FI1W/0152.
Duration of the contract : from 01.12.87 to 31.12.89
Period covered : 1989
Project leader : P. De Sloovere

A. OBJECTIVES AND SCOPE

Wave propagation into soft rock is not completely described by purely linear elastic theory. Through spectrum analysis of a wave propagated into such a medium, one can see that several frequencies are selected by the ground. This behaviour is not forecast by linear elasticity, but is related to mechanical characteristics of the medium. Any change in the mechanical characteristics modifies the frequency content of a signal passing through the medium. This agrees with the behaviour of a wave propagated into a BIOT medium or a viscoelastic medium. Therefore, one can detect by spectrum analysis of propagated wave any change in mechanical characteristics of a medium. Around an underground storage in clay, displacement or swelling will provoke a shift in main frequencies.

We use the general concept of cross-hole method in order to produce or receive the signal. Previous experiments confirmed that an improvement of the soil characteristics is easily shown by this method. One can detect the lack of grouting in alluvium, sands, a.s.o. one can detect a cavity and its filling after grouting. Cracks on piles have been dtected by this method.

One can imagine that such a method is well adapted to detect any change around a radioactive waste deposit, mainly in clay or in soft rock. Around a bore-hole heated by a corrosion tube, we will set up a cross-hole system and follow the frequency content during the heating of the clay.

B. WORK PROGRAM

1. Construction of a new shock transmitter for horizontal bore-holes and three receivers;
2. Boring of four holes on the MOL site (CEN/SCK);
3. Setting-up transmitters in the bore-holes (taken in charge by CEN/SCK);
4. First measurements;
5. Realization of the big bore-holes for corrosion tube (CEN/SCK);
6. Measurement of the effect of the excavation;
7. Heating of the clay;
8. Measurement of the heating of the clay effect;
9. Interprétation.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The new shock transmitter for horizontal bore-hole developed and built in 1987-1988 was used on the MOL site, for its first field test in January 1989. A second field test occurred in March, and a third one in June.
Only subhorizontal bore-holes are available for frequential seismic tests at MOL site and no shock transmitter is able to work properly in such bore-holes . This is a general problem and it was necessary to build a hammer able to work in any bore-hole : vertical - up and down - horizontal and dipping.

The fully pneumatic transmitter is named " MARGOT " for : "MARteau Générateur d'Ondes Transversales" (Shear Wave Hammer Generator). It has two parts - see figure 1 - the hammer and the anvil clamped at the casing by a packer.
The receiver is a triaxial probe with three high sensitivity accelerometers clamped the same way as the transmitter.

The tests occured in the usual MOL clay. During all these tests, the clay remains at ambiant temperature. The heting of the clay was forecast for the end of 1989. So the work performed this year, main ly shows the feasability of the method, the MARGOT'S ability to produce good shock and the stability of the frequency during the first months of the tests. The next tests on heated clay will give the most interesting results on the frequential seismic method.

Progress and results

1./3. FEASABILITY OF MARGOT

As one can see on figure 2 the shock provocked by MARGOT is brief and powerful; so its spectrum is wide and appears like a "white noise" decreasing as the frequency increases. MARGOT is able to produce 200 hits a day. The shocks are repeatable and the dipping of the bore-hole has no influence on the shape or size of the signal. So the feasability of the shock-hammer is proven.

The remaining problems are :
- rebound of the hammer
- impermeability of the tool : the connection between the hammer itself and the probe has to be flexible. It is difficult to assure impermeability during the shock with a flexible joint.
- length of the bore-hole. the high pressure is out of the tool, so the pressure goes to the hammer through tubes; their length can not be infinite; so it will be necessary to embark the high pressure system on the probe.

The main results are :
- ability to work in any oriented hole
- very good shock
- approach by mechanical shock seems better than by piezo-electric quartz.

4./5./6 FREQUENTIAL SEISMIC AT AMBIANT TEMPERATURE

Figure 3 shows the diagram of the cross-hole method; test plants at MOL are depicted figures 4 and 5; figure 6 shows the signal received by the axial accelerometer fixed on the receiver probe in a bore-hole two meter away of the emitting hole.
Two terms of frequential seismic are defined on this figure :
- Main Frequency is the frequency of the most important ray;
- Maximal frequency is the highest frequencywith an amplitude of the ray higher or equal to 20% of the Main Frequency.

At Mol site, general main frequency is 320 Hertz. Usually in subsurface clay, the main frequency does not exceed 220 Hertz.
One observes an increase of 8 Hertz between the results of January and those of March, possibly due to the enclosure of the clay around the bore-hole.

A more detailed study, ray-path by ray-path shows the following main frequencies :
- panel A : 334 Hertz, shift : -17 Hertz
- panel B : 330 Hertz, shift : +38 Hertz
- panel C : 336 Hertz, shift : +19 Hertz

The main frequency lessens on panel A close to the bore-hole and rises on the panels B and C. This effect is probably due to the more cracked zone around the bore-hole.

The maximal frequency is 516 Hertz for the two tests. The maximal frequency seems to be more sensitive than the main frequency at the evolution of the clay. The global shift between the two tests reaches 25 Hertz.

The P-Wave frequency seems to be 1400 Hertz and increases of 100 Hertz between January and March.

These results show that frequential seismic is accurate, the main frequency is function of the behaviour of the clay, and little changes in it shift the main and maximal frequencies.
It remains now to test the heated clay in the same conditions.

Figure 1: Shear wave hammer MARGOT

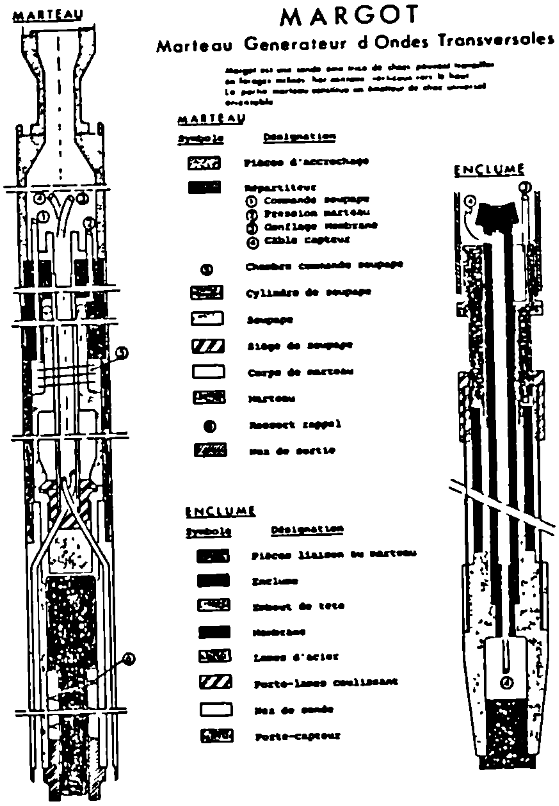
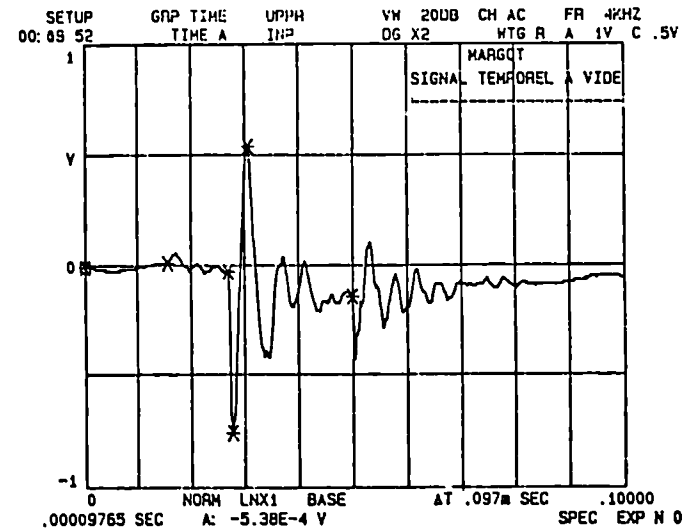


Figure 2: Time history of the emitting signal



	Temps (sec.)	
	(Amplitudes)	(Volts)
0	---	---
1	.015430	.0156	Ouverture soupape
2	.026758	-.0225	Choc
3	.027539	-.752	
4	.030469	.542	
5	.049706	-.140	2ème choc (rebond)
6	---	---	
7	---	---	
8	---	---	
9	---	---	

Figure 3: Cross-hole scheme

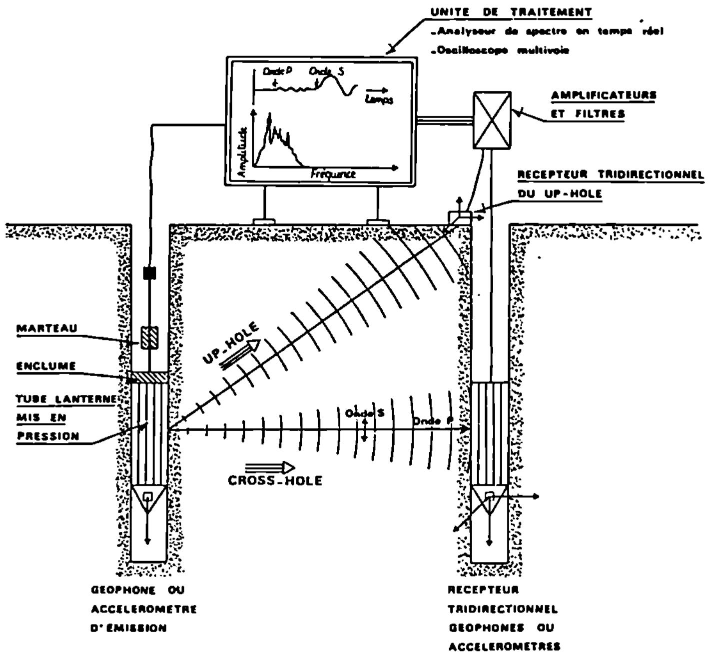


Figure 4: Test plan at MOL site

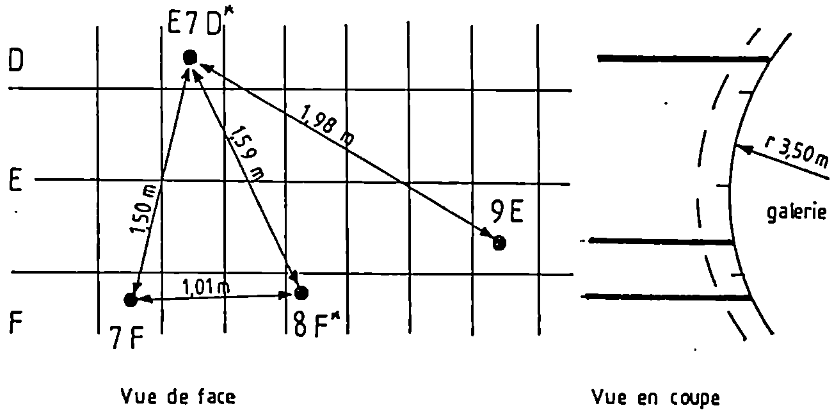
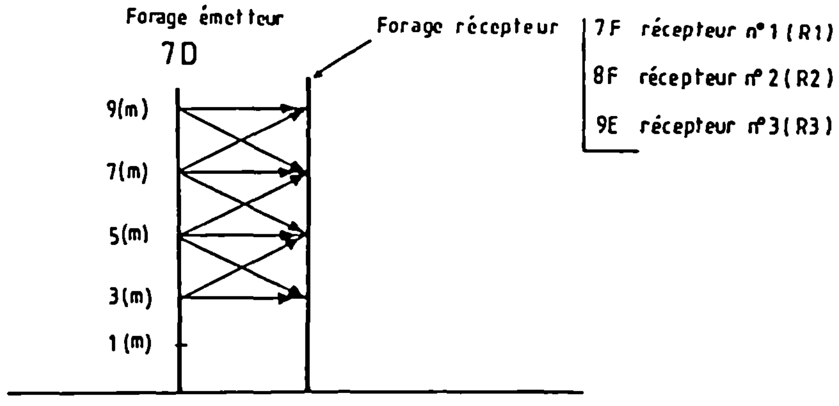
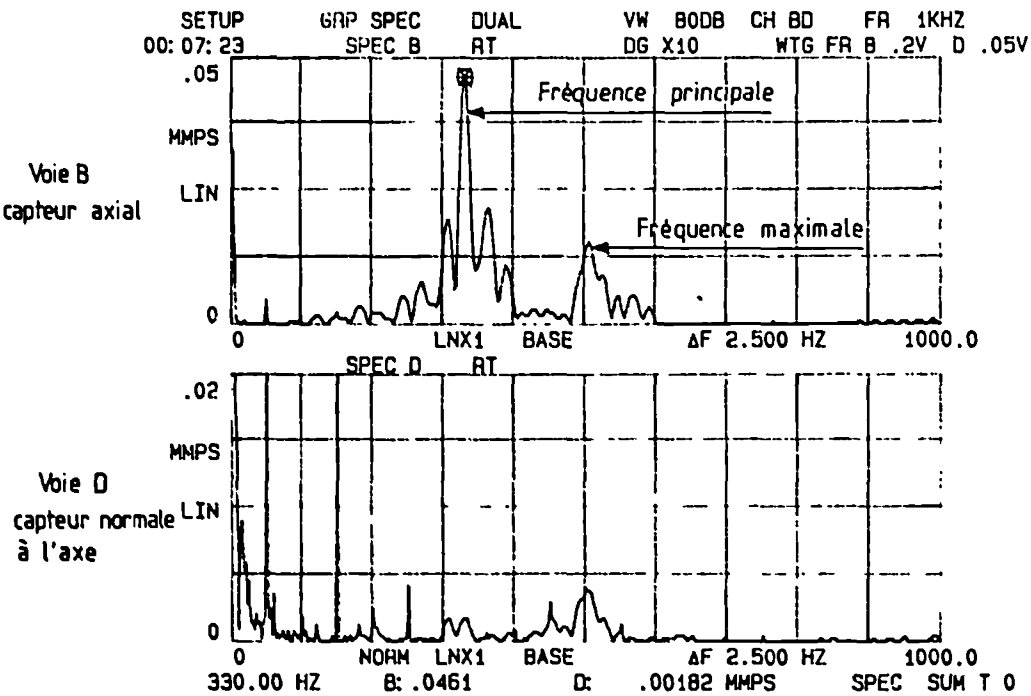


Figure 5: Seismic pannel with ray paths



Vue en plan ou projection dans le plan horizontal
d un couple forage émetteur forage récepteur

Figure 6: Characteristic receiving signal



Research project: Fracture mechanics for hard rock

Contractor: J. Gramberg

Contract No.: FI IW/0153

Duration of contract: From 01-07-1987 to ultimo 02-03-1990

Period covered: 32 - 33 months

Project Leader: J. Gramberg

FRACTURE MECHANICS FOR HARD ROCK

Sub-title: Fracture Mechanisms.

The characteristic of this report is the tendency to apply observations, phenomenology and simple mathematical (graphical) representations with Mohr circles in order to frame, sometimes, far reaching conclusions. These conclusions are usually very much adapted to the virtual circumstances and, often, in detail better than sophisticated mathematical calculations. This might be called the "naturalistic" approach.

The report comprises 4 parts

First Part	:	Orientation
Second Part	:	Structural changes in rock
Third Part	:	Application of the Principal Law - Constructions
Fourth Part	:	Destressing and diverse analyses

The report contains 96 figures, whereas one particular figure stands for 84 "items", figures as well.

In the "First Part" diverse degrees of brittleness in rock are discussed. The axial cleavage- or extension fracture is in focus, being the most occurring fracture phenomenon in underground mining and in tectonics. The "Principal Law" for primary brittle fracturing is framed.

In the "Second Part" diverse mechanisms of structural changes in underground rock when a gallery or bore hole approaches are discussed. For this study rock salt specimen and the result of acoustic measurements are applied, both from the Asse II rock salt mine. the phenomenon of "stress peak shift" is discussed. Our "naturalistic" analysis leads to the same result as the mathematical analysis by Santarelli et al. based on the theory of bifurcation. An expert has commented this similarity as follows: "In conclusion we

may expect that we are comming gradually into very good agreement between "naturalistic" observations and our mathematical modelling".
In the "Third Part" the Principal Law is applied to long wall mining as well in coal exploitation as in the 2000 - 3000 m deep well known aurifirous quartzitic reefs in South Africa. The "caterpillar-looper" effect of the advancing S-shaped deformation is analyzed resulting in the definition of the "stress-strain-energy pump" through which high horizontal stresses are generated.
Application to geology covers a theory on vertical and sub-vertical joints, a mechanism of domal uplifting and the generation of three types of schistosity.
The "Fourth Part" is dedicated to destressing a.o. by means of the "Self Supporting Rock Ring" (S.S.R.R.; J.P.A. Roest); diverse analyses are discussed.
Our "fracture-and-fracture-plane analyses" is applied to the long wall mining in the very deep gold bearing quartzites of South Africa.
3 Different cyclic processes and 5 different stress regimes were recognized.
Our "naturalistic" analyses lead to the insight that diverse cyclic processes in the carboniferous hanging wall of the Western European coal mining must belong to the same family of phenomena: cyclic processes as a result of the "stress-strain-energy-pump" mechanism appear generally inherent to long wall mining in tabularly structured rock.

We think that this conclusion forms a topic of our research. Therefore we will explain the principles in the next figures. Possibly they might be useful for the software engineer.

FIGURE 1 The cyclic process in long wall mining: the "caterpillar-looper" effect

a: The advancing coal face of a Western European long wall coal exploitation; the hanging wall forms an S-shaped advancing deformation;
A is the position of a particle U; the horizontal stress is small; the original horizontal length is L

b: The advancing face causes U to pass through the positions B,C and D; the horizontal length has first increased to L + ΔL, then decreased to L + dL; dL stands for the permanent enclosed secondary porosity; in the position E the particle has regained its original length L;
the excess-length dL has vanished as a result of freshly generated horizontal stress σ_h; the advancing deformation reminds of a continuously moving caterpillar

c: The elevated horizontal stress causes the hanging wall strata to stiffen and to behave like a brittle beam; they break off causing a violent "bump", in this stage the behaviour resembles a "looper", a special kind of caterpillar; the caterpillar-looper behaviour forms together one cycle which can be repeated with various intervals

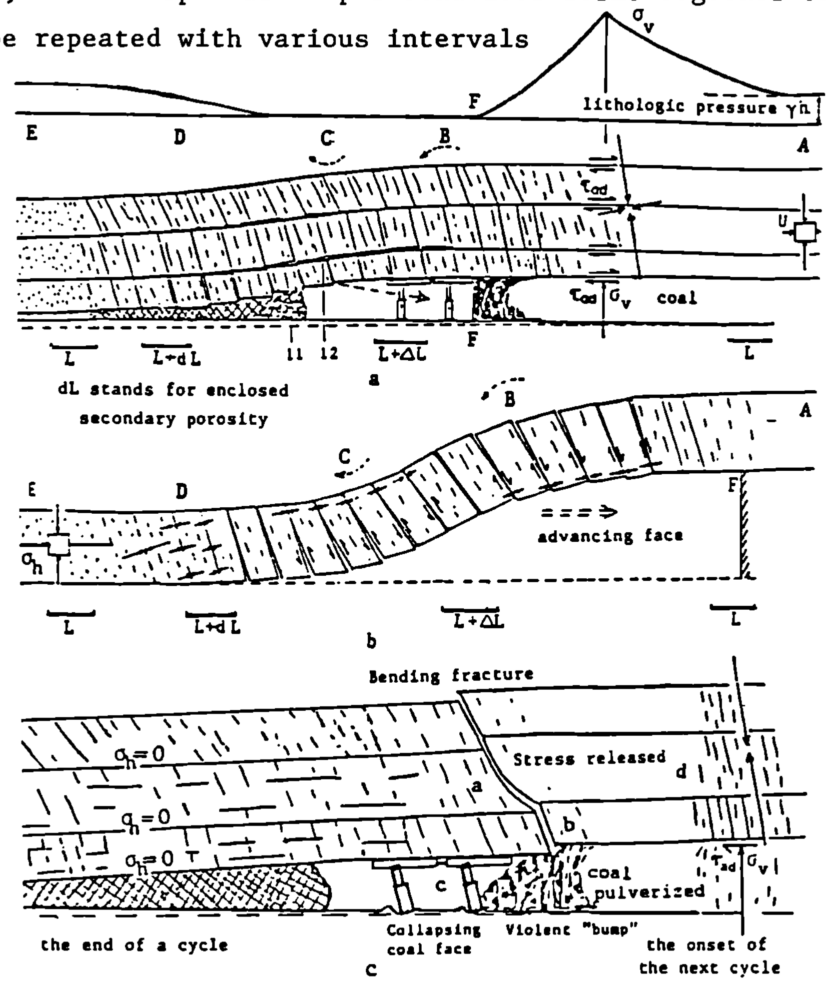


FIGURE 2 The "stress-strain-energy-pump, being the mechanism of the generation of the freshly added horizontal stress

I : A replica of the vertical stress distribution from figure 1

II : The particle U in its diverse position a .... g

III : The advancing S-shaped deformation showing the structural changes in the particle U between 3 and 6; in the position E = 6 the excess length dL is shown as a structural element: secondary porosity

IV : The same series, but now the particle U is kept in place whereas the stress peak and the deformations are passing the particle; in the end, in the position E = g the secondary porosity has caused the generation of the fresh horizontal stress σ_h; the energy content of U is increased

V : The freshly generated stress σ_h is transferred, through the tabular structure of the strata, to the particle U' in the position b'; the continuity of the process brings about a pumping effect with respect to the horizontal stress and the energy content

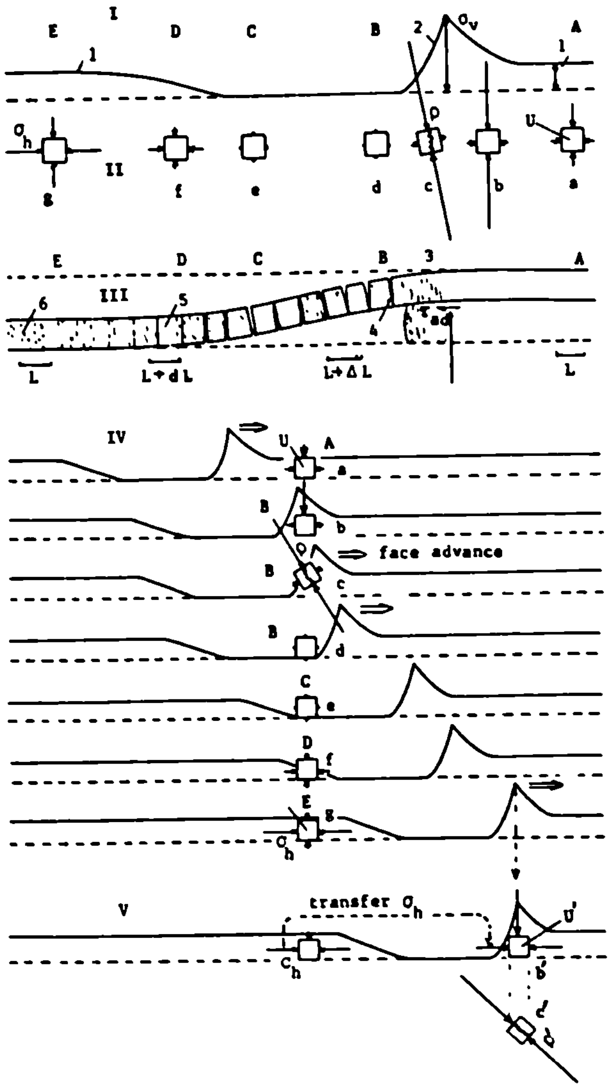


FIGURE 3 Schematic picture of the possible cyclic processes at the 2000 - 3000 m deep long wall gold exploitation in Africa, based on the fracture-and-fracture-plane analysis applied to the ample descriptions and diagrams from litterature

a: The higher strata +/- 45-60m must harbour an unknown high triaxial pressure system (1); they form the unfinished megacycle

b: The macrocycle with a periodicity between about 5m and 8m on the +/- 7m level

c: The stope- or minicycle with a periodicity of about 1.80 m

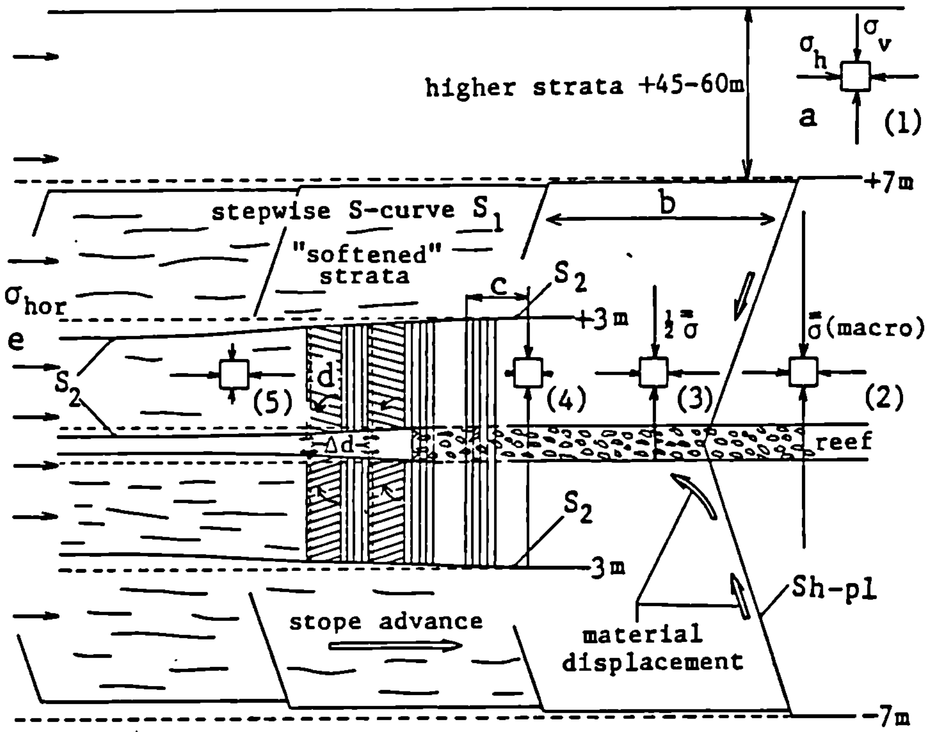
Sh-pl.: The shear plane or shear zone which shields the area (at the right) with a variable high severe stress regime (2) from the zone (left) with an about constant mild stress regime (3);S_1: stepwise S-curve, responsible for the pattern of the shear zones

S_2: Stepwise S-curve, responsible for the alternating multifractured and intact blocks; it causes the very local "reduced mild stress regime (4) within the +/- 3m zone; it causes the closure of the stope (at the left)

d: The wedging action which causes extra-large horizontal dilatation Δ_d and clamping

(5): The stope-stress regime, characterized by very high horizontal stresses and small vertical stress; horizontal stresses of 20 MPa are measured

e: The horizontal stresses, inherited from the former exploitation over more than thousend meters by the "caterpillar-looper" effect: 20 MPa.



Experimental study of the mechanical behaviour of argillaceous rock

Contractor: Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract N°: FI1W/0163
Duration of Contract: October 1987 - December 1989
Period covered: January 1989 to December 1989
Project Leader: J.Y. BOISSON

A. OBJECTIVES AND SCOPE

Changes in the mechanical behaviour of clays according to the temperature are not well known yet. If, for the short term behaviour studies, one can admit that there is no drainage effect and, that, a temperature rise leads, at the short term, to a clay when increase, this cannot be stated concerning the long term, where the drainage effect must be taken into account.

Recent studies have shown that, owing to their texture rearrangement, saturated remolded porous soils, replaced in a normal consolidated state, exhibit, in the long term, a volume decrease, with, as consequences, settling effects and crackings.

It is possible that the long term response of the clays at such thermal prompting is a fonction of their overconsolidation degree : normal consolidated clays will decrease, in fact, in volume under thermal solicitation . but this could be different for the over consolidated clays. The aim of this study is to bring clear and quantitative experimental answers to these questions both with a theoretical interpretation of these phenomena.

This research will be performed with the scientific support of the Centre de Géologie de l'Ingénieur (ARMINES-Ecole des Mines de Paris).

B. WORK PROGRAMME

2.1. Choice of the sites, and sampling of clay

The selection will be made first considering that the sites answer to some textural granulometric criteria (clays silts) and mechanical criteria (normally or over consolidated clays).

After that, we will take these materials under predetermined conditions to avoid remolding and overconsolidation due to surface dessication.

We will determine then, in a precise way, their initial overconsolidation degree and their texture, considering that these two points are essential for this study of the behaviour of clays towards thermal sollicitations.

2.2. Responses study of the selected material towards the thermal mechanical solicitations

These responses are mainly textural rearrangements which will be studied in the laboratory by precise texture identifications and by comparisons between initial and final state.

2.2.1. Basic petrographic, mineralogic and textural identification

Different procedures will be used : X ray diffractometry analysis, adsorption tests with methylene blue, Atterberg limits, porosimetry, observations with scanning electron microscope, and permeabilities.

2.2.2. Determination of the overconsolidation degree.

2.2.3. Control owing to blank tests of the thermal behaviour of the experimental apparatus used.

The aim will be to well separate, in the experimental results, the information concerning the clay geomechanical behaviour, from artefacts due to the thermal behaviour of the tests apparatus itself.

2.2.4. Creep tests in oedometer cells and permeability measurements

An axial backpressure will be applied so as to be able to scan the temperature range between 20°C and 110°C (instead of the 120°C previously envisaged) without pore water loss. The stress will be equivalent to the one in situ, and will not be less than 0.01 MPa.

In case of swelling during the sample saturation, the applied stress will be equivalent to the swelling pressure. The thermal prompting applied to each selected clay sample will correspond to the 4 following temperatures : 20, 50, 80 and 110°C. Different temperature rises will be applied, favouring the exploration of parameters linked to the thermal prompting and to the creep :

a) direct temperature rise (creep study at mid term during 2 months, (instead of the 3 months initially envisaged) tests at the 4 mentionned temperatures).
b) direct temperature rise (creep study at long term during 6 months, tests at 20° and 80°C) (instead of the 9 months initially envisaged)
c) progressive temperature rise from 20° to 110°C (for each temperature level, creep study at mid term for 2 months).
d) progressive temperature rise and then progressive decrease, creep study during 2 months (for each temperature level) : one test from 20° to 50°C and then from 50°C to 20°C ; one test from 20°C to 80°C and then from 80°C to 20°C ; one test from 20°C to 110°C and then from 110°C to 20°C.

A certain number of these tests will be repeated (two or three times) so as to ascertain the reproductibility of the results.

2.2.5 Textural study

This study will be done by applying the mercury porosimetry tests, scanning electron microscope, permeability tests on each sample (initial and final state). The compared analysis of the whole results will certainly give a contribution to the thermal mechanical behaviour knowledge related to the texture and overconsolidation degree.

The basic equipement for the textural study and for the mineralogical, petrological identifications and classical geotechnical tests is available. Nevertheless, for the thermomechanical tests it will be necessary to adapt existing devices.

C. PROGRESS OF WORK AND OBTAINED RESULTS

2.1. Choice of the sites, and sampling of clay

The choice of the studied clays has been slightly modified from the initial choice previously presented ; the black clay from Limay (LIMN) has been tested only for the direct temperature rise tests. Then, the complete study deals with three clays, from which, one is tested both in an undisturbed and disturbed state (after swelling) : St Genest "normal" (STG-N) and St Genest "disturbed" (STG-G), Lagny (LAG), Limay red (LIM-R).

The development of the study during this year has been perturbed by a technical problem which has been the cause of an interruption of a certain number of long term tests (6 months). It appeared that the heating of the experimental oedometer cells at 110°C led to a premature ageing of the "Bellofram" membrane. To solve this problem, it has been necessary to design a special new membrane and, consequently, this has been the reason of delay in our tests planning. Nevertheless, it has been possible to minimise this delay, by reducing the time of each temperature level from 3 months to 2 months. In fact, it appeared that, as shown by the first obtained results, this decision will not be detrimental to the scope of the study. Table I shows the state of progress for these tests.

2.2. Responses study of the selected clays, towards thermo-mechanical solicitations.

2.2.1. Basic identifications choice

2.2.2. Overconsolidation ratio

2.2.3. Control tests of the experimental apparatus. cf. A.P.R. 1988

2.2.4. Presentation of the different parameters measured during the creep tests

The study of the primary consolidation is done from the experimental compression curve, from which the coefficient of primary consolidation C_v is determined (Casagrande's construction).

The creep phenomena is analysed owing to two approaches :

- from the rate of secondary compression C_α (slope of the curve $dh/h_o = f\ (\log t)$), and the creep index $C_{\alpha e} = C_\alpha\ (1 + e_o)$; (e_o = initial void ratio) ;
- from the total theoritical strain (h_{tt}) due to the creep extrapolled after 100 years, determined from the consolidation curve according to Felix (1980) using the equation :

$$C(t) = h_f\ (1 - e^{\alpha t^\beta})$$

Permeability measurements are made, before temperature rise, after the primary consolidation phase, and at the end of the creep tests, with calculation of the correspondant void ratio.

2.2.5. Obtained results

Results reported there, deal essentially with parameters from the theoritical creep curves (according to Felix for tests at direct temperature rises (see table II), and with $C_{\alpha e}$ variations towards temperature for each clays (fig. n°1).

Examination of the results given table II shows that, in the general case, the total theoritical strain (h_{tt}) due to the creep increasesslightly with temperature

Analysis of the curves $C_{\alpha e} = f$ (T°) shows too an increase of these $C_{\alpha e}$ values with higher temperatures ; this tendency seems to be more important for the Lagny and LimayR clays than for the St GenestN and St GenestG in the case of temperatures equal to 50°C and 80°C.

To illustrate these remarks, the curves of consolidation with temperature rises at different levels for St GenestN and St GenestG are reported fig.2 to 4, indicating the values of the different measured parameters.

Permeability and void ratio variations during the tests are very low and their values are in the same order of magnitude than the measurement accuracy.

The observation of the clay texture with Scanning Electron Microscopy does not reveal at the present time significant and systematic modifications, in agreement with what could be predicted from the void ratio calculations.

However, a decrease of the macroporosity spectrum seems to become apparent from the first mercury porosimetric tests. This last point could be verified and confirmed by a simultaneous decrease of the microporosity in the next tests.

REFERENCES

1. B. Felix, (1980)
   Le fluage et la consolidation unidimensionnelle des sols argileux. Rapport de recherche des Laboratoires des Ponts et Chaussées n°94 (1980), 176 p.

	T°C	STG-N	STG-G	LIMR	LAG	LIMN
Direct T°C rise - Creep = 2 months	20	- - - -		———	———	
" "	50	———	———	———	———	———
" "	80	———	———	———	———	———
" "	110	———	———	———	———	———
T°C rise at different levels Creep = 2 months each level	20 50 80 110	——— ——— - - - -	——— ——— ———	- - -	——— ——— - - -	
T°C rise and decrease by levels Maximum T° level = 50°C Creep : 2 months each level	20 50 20	——— ——— - - -	——— ——— ———	- - -		
T°C rise and decrease by levels Maximum T°C level = 80°C Creep = 2 months each level	20 50 80 50 20	——— ——— ——— ——— ———	——— ——— ——— ——— - - -	——— ——— ——— ——— ———	——— ——— ——— ——— ———	
T°C rise and decrease by levels Maximum T°C level = 110°C Creep = 2 months each level	20 50 80 110 80 50 20	——— ——— ——— ——— ——— - - -	——— ——— ——— ——— ——— - - -	——— ——— ——— ——— ——— - - -	——— ——— ——— ——— ——— - - -	
Direct T°C rise Creep = 6 months	80	- - -	- - -	- - -	- - -	

Table n°I : Progress of the tests

		20°C			50°C	80°C	110°C
STG N	α	0,570	0,820		0,374	0,504	0,275
	β	0,073	0,105		0,105	0,080	0,105
	hf	0,194	0,407		0,407	0,359	0,413
	hftt	0,110	0,280		0,280	0,217	0,307
STG G	α	1,965	1,590	2,110	1,615	1,674	0,983
	β	0,034	0,038	0,031	0,044	0,039	0,056
	hf	1,708	1,989	1,951	1,840	1,971	2,429
	hftt	0,239	0,405	0,239	0,320	0,369	0,909

		20°C			50°C	80°C (1)	80°C (2)	110°C
LAG	α	0,412	0,440	0,366	0,443	0,417	0,276	0,408
	β	0,089	0,082	0,093	0,087	0,088	0,105	0,086
	hf	0,231	0,198	0,248	0,230	0,691	0,285	0,717
	hftt	0,152	0,127	0,168	0,147	0,455	0,217	0,477

h_f and h_{ftt} in mm

Table II : Coefficients of the Felix' equation for clays from St Genest undisturbed (STG-N) St Genest disturbed by free swelling (STG-G) and Lagny (LAG)

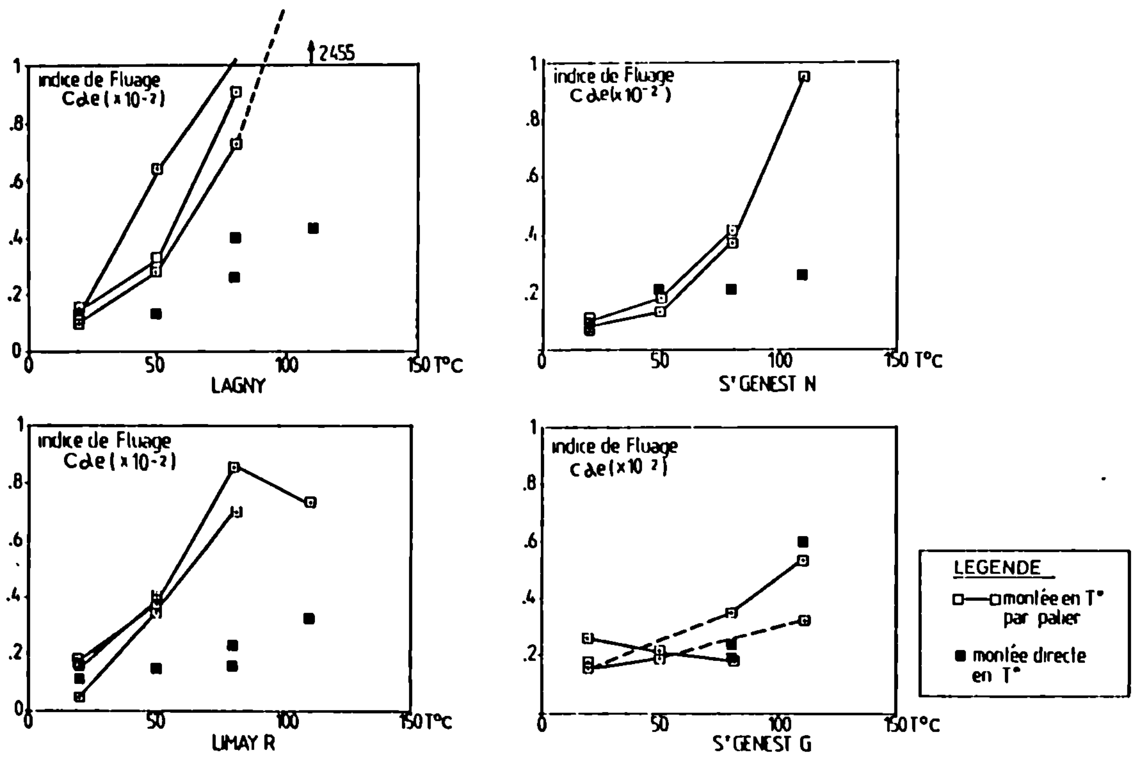


Fig. n°1 : Variation of the creep index $C_{\alpha e}$ versus temperature

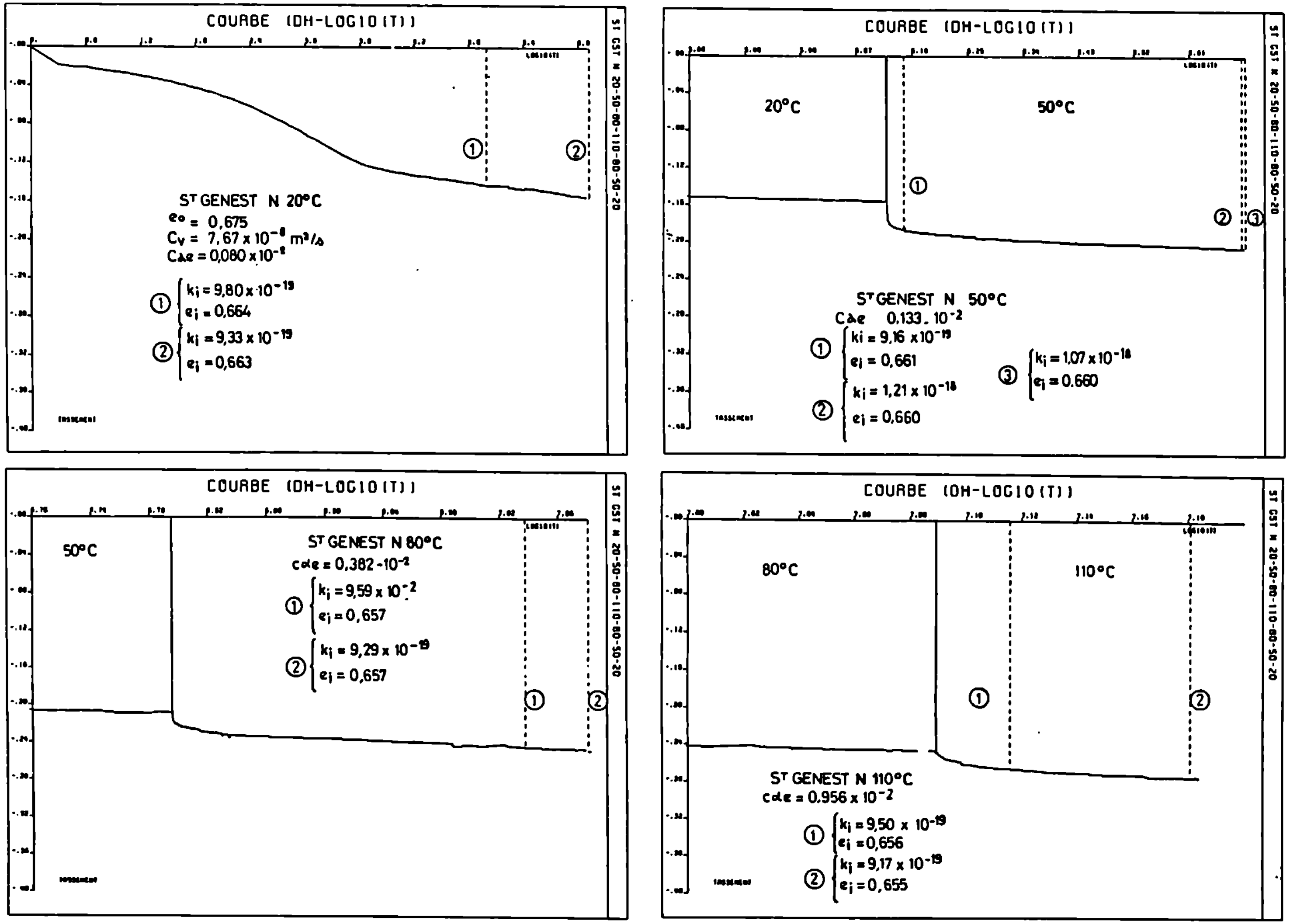


Fig. n°2 : Creep tests with St Genest undisturbed clay (direct temperature rises)

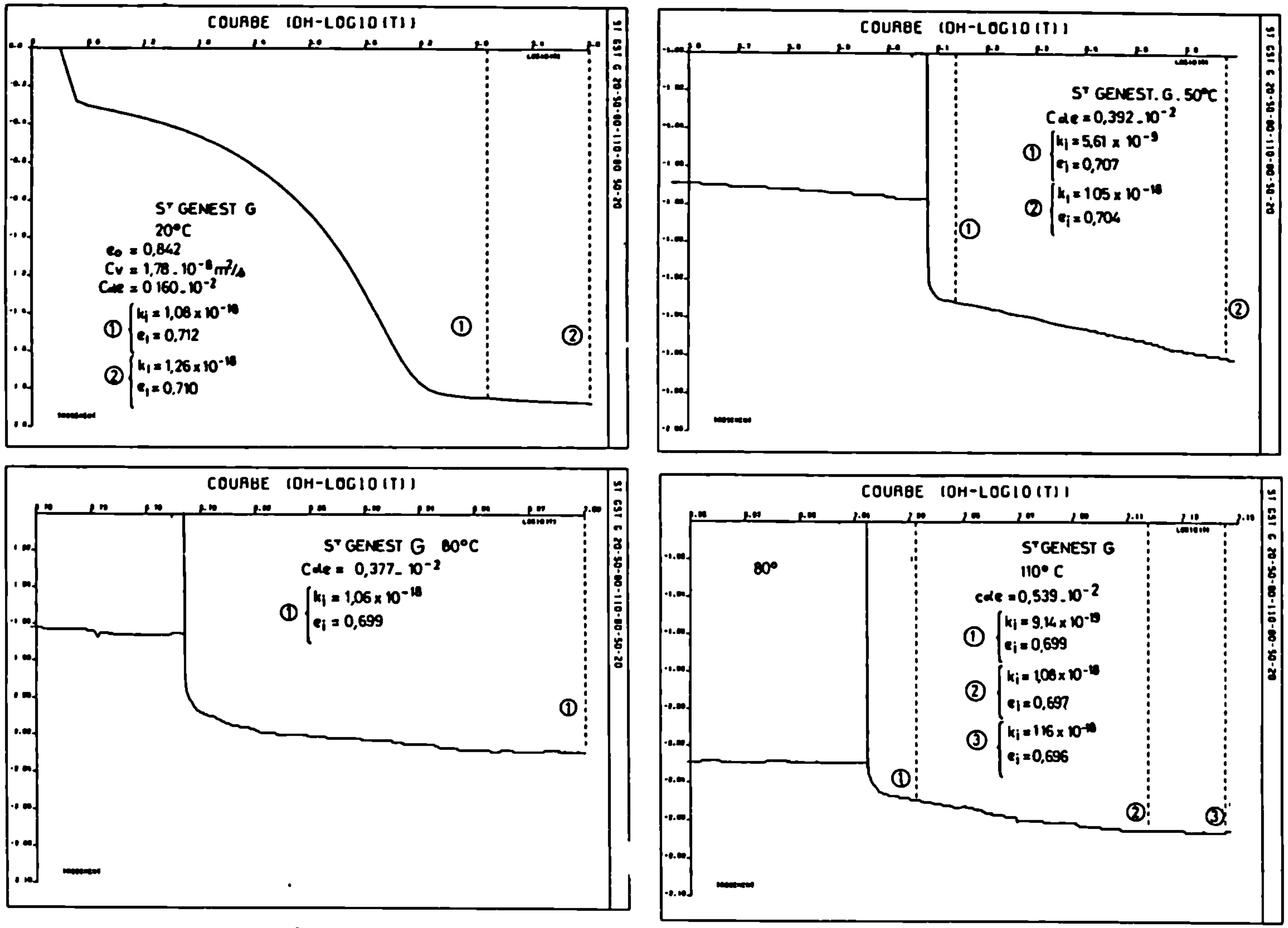


Fig. n°3 : Creep test with St Genest clay disturbed by free swelling (direct temperature rises)

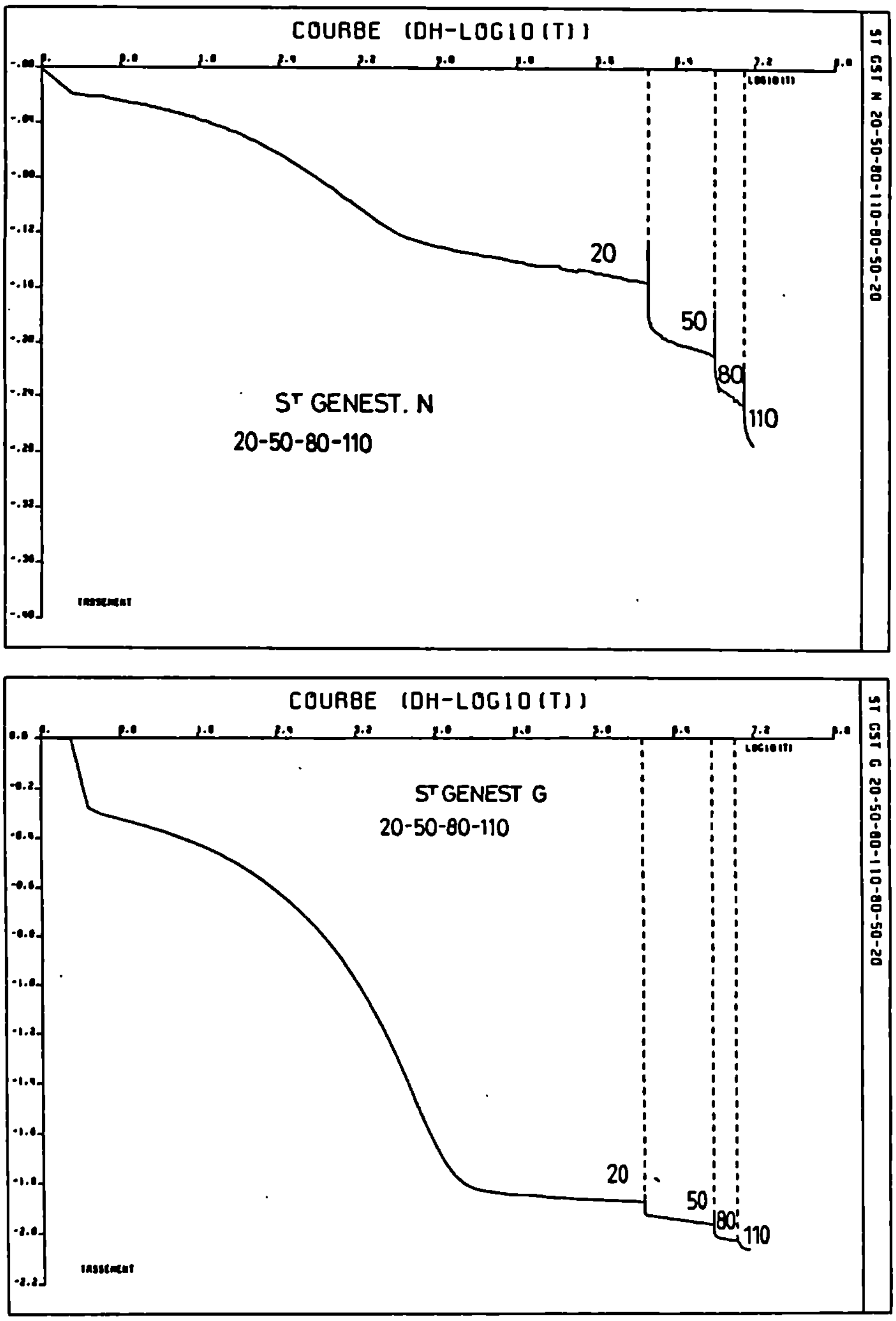


Fig.n°4 : Creep tests at four temperature levels (3 months) with the St Genest clay (undisturbed (N) and disturbed (G))

Title: Deformability and failure behaviour of a saliferous formation near cavities.

Contractors: BRGM-ARMINES

Contract n°: FI 1 W/0206

Duration of contract: From August 88 to July 90

Period covered: 18 month

Project Leader: Ph. MASSAL, G. VOUILLE
Tél. : (33) 38.64.34.34
FAX: (33) 38.64.33.90

A - OBJECTIVES AND SCOPE

Rock salt, because of its very low permeability and its capacity for absorbing large amounts of deformation by creep, is a material favoured for the disposal of radioactive waste.

Nevertheless, in situ observation of the salt's behaviour shows that under certain conditions, rock salt may fracture. It is therefore important to confirm the behaviour established in the laboratory by in situ mechanical tests, and in particular to define the boundary between brittle and ductile behaviour, in terms of average pressure, rate of loading and temperature. Nor must the risk of fracturing during cooling be ignored; similarly the influence of past temperature on its behaviour should be investigated.

The purpose of this contract is to propose a Lemaitre model for the interpretation of laboratory mechanical tests and to validate it by appropriate in situ tests. The site is the Varengeville mine, near Nancy (France).

B - WORK PROGRAMME

The contract comprises three closely related main parts:

1 - A series of laboratory tests of salt samples from Varangeville:

The purpose of this test campaign is to use the test results to construct a model that reproduces the behaviour of the Varangeville salt in order to provide answers to questions relating to deformation and failure such as:

- strain-hardening by deformation;
- the influence of the intermediate stress;
- the viscoplastic potential;
- the influence of temperature.

Adjustment of a rheological model:

Use of code AJUST to adjust rheological law coupled with an algorithm for integrating differential equations governing the variation of the stress (relaxation test).

2 - In situ tests:

The tests envisaged will require the drilling of two holes about 25 m deep.

- Deformability test: "short-term" dilatometer

This will be a standard deformability test enabling the in situ determination of young's modulus for the rock salt.

- Creep test: "long-term" dilatometer

A special dilatometer will be developed for measuring creep in a drill hole in the salt over a long period, at least twelve months and, if possible, longer.

3 - Simulation of in situ tests and modelling

This will be done using the VIPLEF code, introducing the rheologic model derived from the results of all the tests and simulating the loadings during the in situ tests.

Using the results of these computations, the closure and possible failure of an idealized cavity for radioactive-waste disposal will be calculated, taking into account an increase followed by a decrease in temperature.

C - PROGRESS AND RESULTS

1 - State of progress

Implementation of the initial programme was somewhat delayed by the difficulties encountered in derising and constructing the prototype of the long-term dilatometer.

Furthermore, the boreholes cored in the Varangeville mine were only completed between the 20th November and the 8th December, 1989. The salt cores were analysed by gamma-gamma logging and X-ray radiography before being sent to the laboratory for the mechanical tests, which began in January, 1990. The in situ trials were completed in December 1989. They comprised:

a - Deformation trials with the short-term dilatometer. Four tests were carried out, each comprising three loading-unloading cycles, at different positions in the borehole.

b - Installation of the long-term creep dilatometers:

The specially constructed prototype was installed in a boreholes sited in the middle of a pillar covering some 30 m × 30 m in cross-section. Measurements should continue to be taken over at least one year. The installation of the prototype was preceded by a simulation of this procedure, using the VIPLEF package, to ascertain the stresses and displacements likely to be encountered as time passes.

2 - Results

a - Construction of the borehole dilatometer

This apparatus for measuring salt creep over long periods has to be able to measure both variations in the diameter of the borehole and changes in the volume of that part of the borehole in which it is sited. To achieve this, it must be possible to apply, and control, an internal pressure. Bearing in mind the simulation calculations carried out, it amounted to the measurement of extremely small desplacements, measurable in micro-metres, but with an internal pressure always lower than 4 MPa.

In its finished form, the prototype consists of three main components (fig.): the probe, the device for measuring volume and controlling pressure, and the measurement recorder.

- The probe consists of an inflatable tube, 1.50 metres long, containing four displacement gauges orientated across the diameter of the tube at intervals of 45°, and each provided with a sensitivity of about 1 micrometre. An inflating pipe and an electric cable connect the probe with the measuring and controlling device on the one hand and the measurement recorder on the other.

- The measuring and pressure-control device comprises an air-oil interface which enables pressure to be applied to the dilatometer. Pressure is supplied to the air circuit by a bottle of compressed nitrogen, and is adjusted to a level determined by a regulator valve. The internal pressure of the dilatometer is measured by a pressure detector in the oil circuit, and the variations in volume are established by measurement of the displacement of the air-oil interface.

- The measurement recorder contains data processors for the various gauges and a measurement processor controlled by a micro-computer. The device can be interrogated remotely via a Modem and telephone link.

The prototype as a whole was tested and calibrated in the laboratory.

b - Results to date

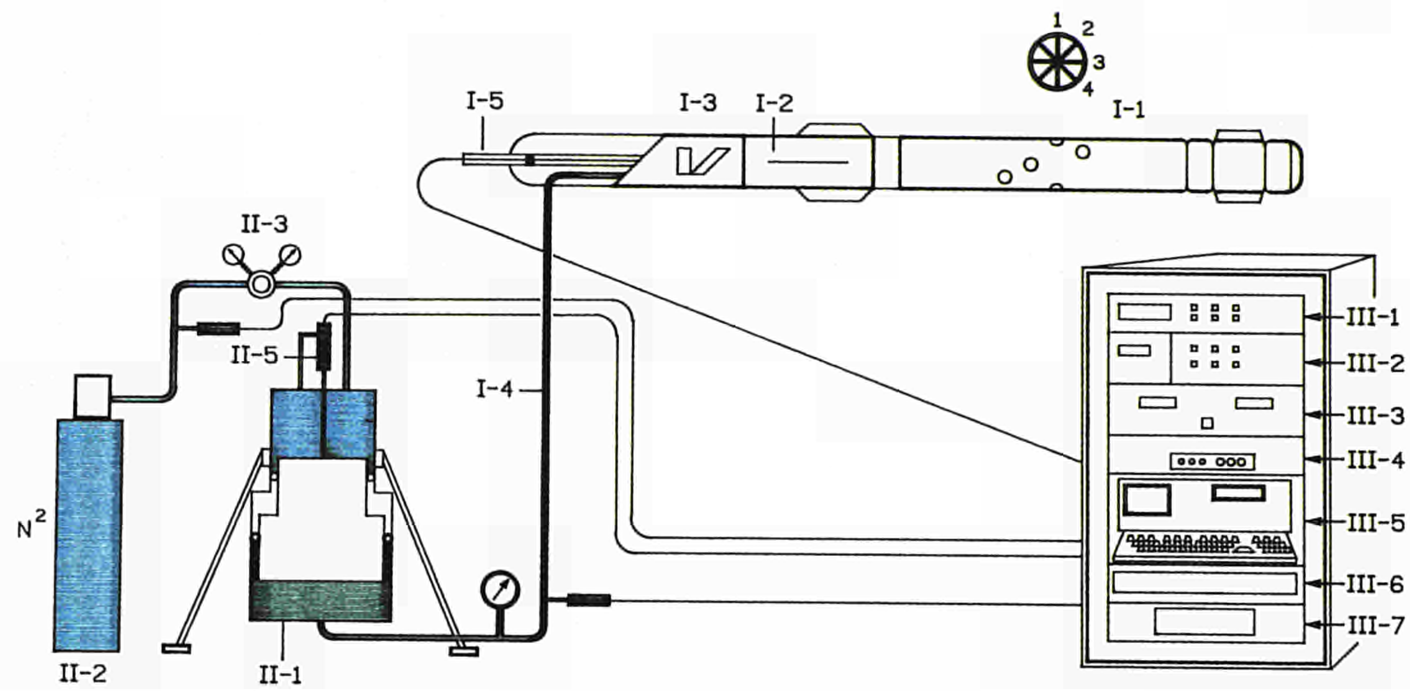
- The apparatus was installed in December 1989, in a horizontal borehole drilled into a salt support pillar in the Varangeville mine, about 250 m below the surface. It was orientated with the aid of two inclinometer sensors, enabling one of the displacement gauges to be exactly aligned on the vertical axis. The dilatometer sleeve was attached to the side of the borehole and inflated to a pressure of 2.5 MPa.

  This pressure will be kept constant for at least three months, during which the deformation of the borehole wall will be closely monitored.

- Four other deformation tests were carried out in another horizontal borehole, at varying distances from the face. For each tests, three loading-unloading cycles were applied, at 1 MPa, 1.5 MPa and 2 to 3 MPa respectively, with a loading rate of around 0.12 MPa per minute. Based on the stress-strain curves obtained, the deformation moduli are in the range of 2.700 MPa and 8.300 MPa.

DISPOSITIF DE MESURE DE FLUAGE EN FORAGE DANS LE SEL

BRGM



I DILATOMETRE

I-1 4 capteurs diametraux
I-2 électronique de conditionnement
I-3 inclinomètres d'orientation
I-4 tubing de gonflage
I-5 câbles de mesures

II DISPOSITIF DE MISE EN PRESSION

II-1 pot multiplicateur de pression
II-2 réserve d'azote
II-3 détenteur
II-4 capteur pression air
II-5 capteur de variation de volume

III ARMOIRE DE MESURES

III-1 fréquencemètre
III-2 centrale de mesure
III-3 afficheur de pression
III-4 modem de transmission
de donnée
III-5 microordinateur
III-6 onduleur
III-7 imprimante

Design of the tests.

In order to forecast the order of magnitude of the deformations of the borehole, a numerical modelisation has been carried out with the VIPLEF F.E.M.code, assuming a non linear viscoplastic behavior of the rock salt with parameters already known from a former study . The mesh which has been used represents 1/4 of a pillar (fig.1) in which, in the central part, the hole itself and its neighbourhood are divided in finer elements (fig.2).

The calculus was performed in the following way:

- at time 0 the pillar is created in an isotropic compressive stress field of 4.5 MPa,
- at time 4000 days, in the center of the pillar, the compressive vertical and horizontal stresses are respectively equal to 9.6 and 6.2 MPa. At this time the borehole is created and immediately its convergences in the vertical and horizontal directions are respectively equal to 64 and 22.10-6 m,
- during the two following hours the borehole is left free and the evolution of its convergence versus time is plotted on fig.3,
- at the end of this period, the measuring device is put in the borehole and begins to measure the convergences either with a 2MPa radiat pressure or with no pressure : the corresponding curves are plotted on the fig.4.

The general conclusion of this preliminary study is that the maximum variation of the diameter of the hole during the first year of measurement should not be more than 100 or 200.10-6 m.

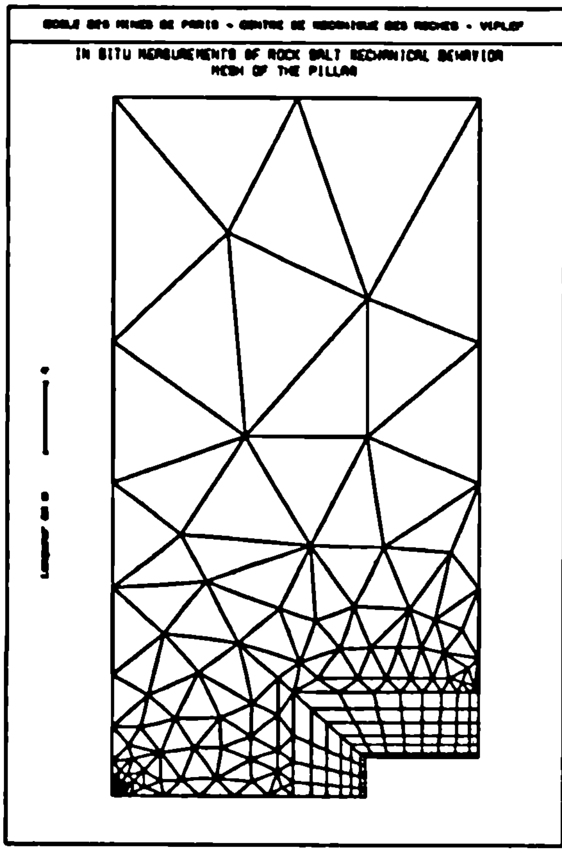


Figure 1

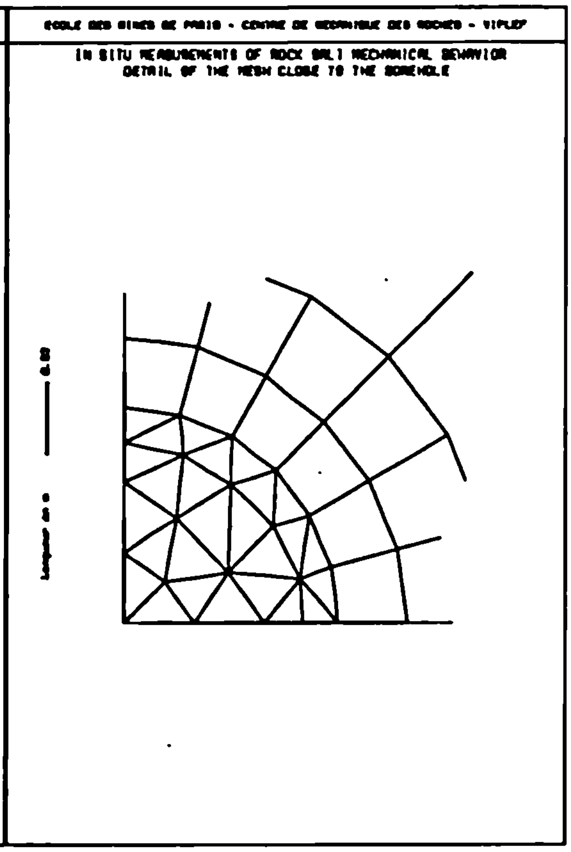


Figure 2

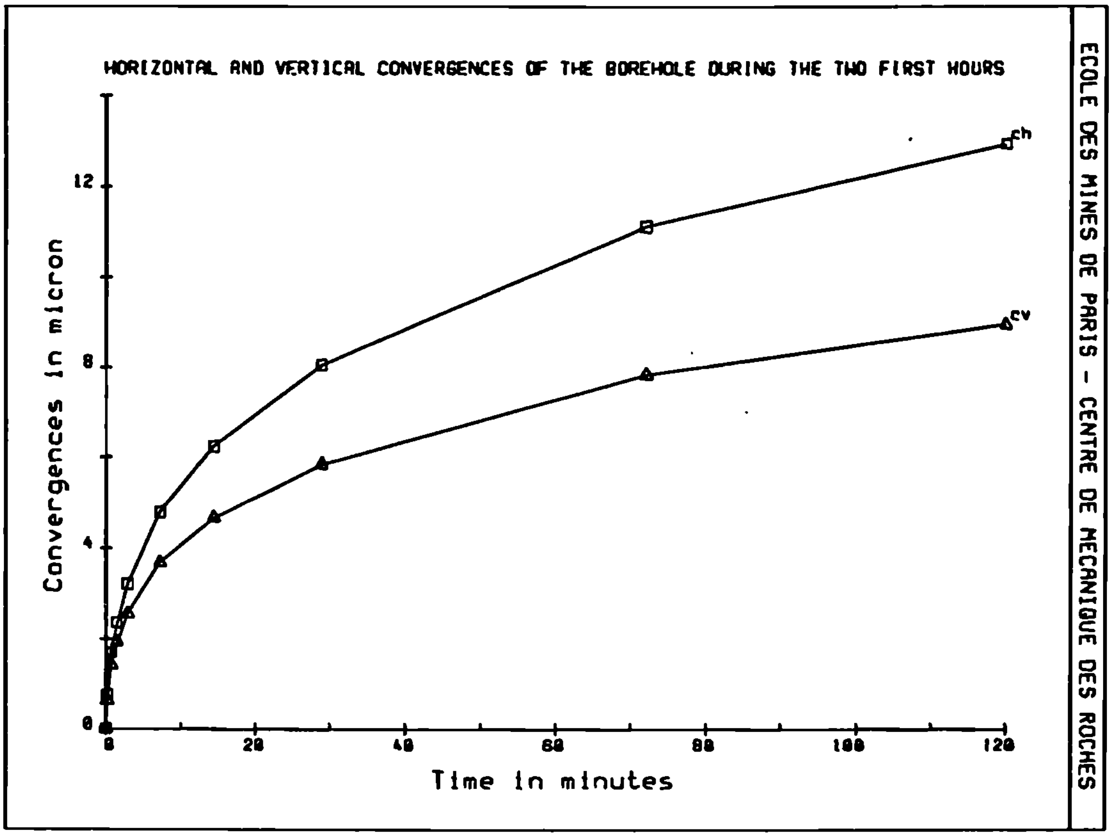


Figure 3

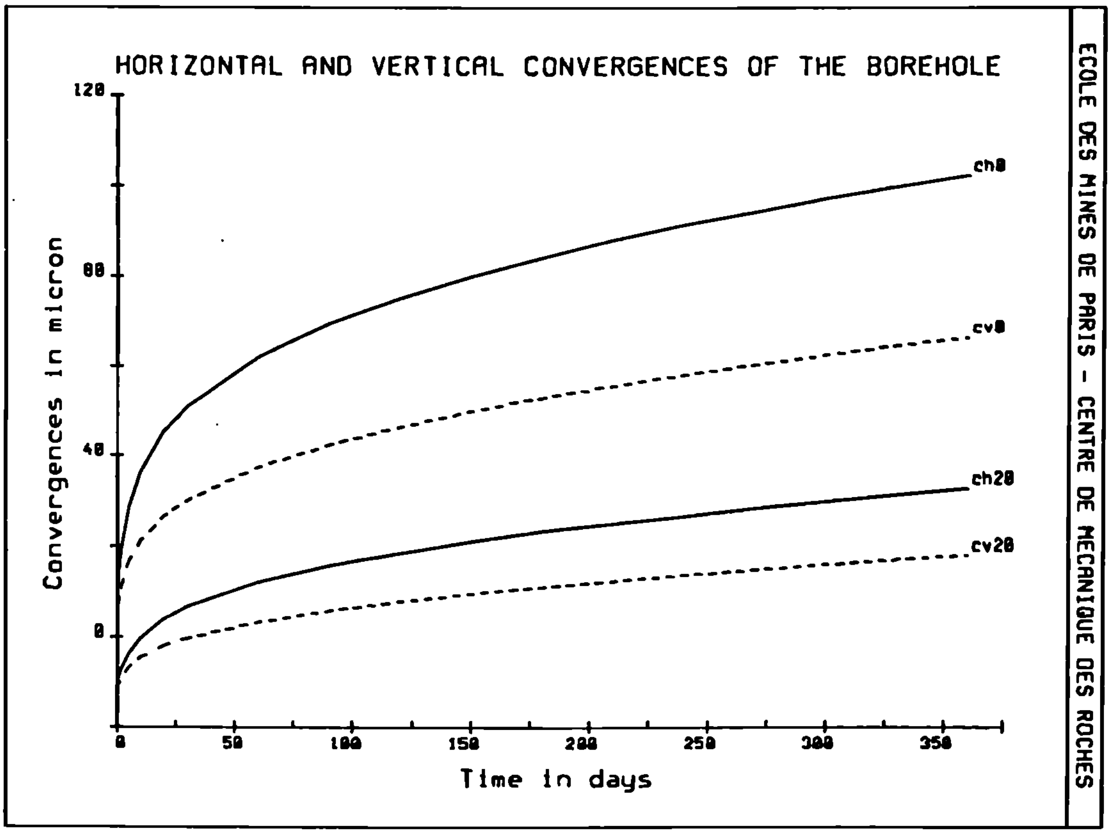


Figure 4

Verification and validation of coupled thermo-hydro-mechanical

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, FRANCE
Contract n° : FI.1W.0246
Duration of Contract : July 1989 - January 1991
Period covered : July 1989 - December 1989
Project leader : S. DERLICH

A. OBJECTIVES AND SCOPE

An underground experiment on hydromechanical effects of massive granite under thermal loading has been previously realised in granitic botholite partly fractured.

The purpose of the experiment was a simulation both on space and time of phenomena related to thermal release of a waste repository.

The lows of similitude give relations between a natural site and a reduced experimental model, which are strictly valid only for thermal release in continuous medium.

The first evaluation was made with this hypothesis.

In the experimental conditions, the rock mass is affected by tridimensional set of fractures in unsaturated conditions.

Our study will concern three kinds of parameters generally considered independently :

. thermal phenomena
. mechanical phenomena
. hydraulic phenomena

An interpretation of experimental results will be realised in two parts :

1. Determining experimental values of the physical parameters (strain or displacement, temperature), and interpretation taking into account discontinuities or anomalies of the medium.

Comparison between experimental results and computational results starting with the same initial conditions.

DESCRIPTION OF WORK

Thermal phenomena

Previously realised experiments show that heat transfert obeys Fourier low and can be correctly obtained by using a model based on the assumption of a continuous medium. Major disturbance should be introduced in the results by an active water bearing fracture system. This possibility will be also examined.

Mechanical phenomena

Differences introduced by discontinuities in the medium obeying Hooke low should be brought into evidence by the interpretation of strain or displacement of measurements.

Thermo mechanical characteristics of granitic matrix have been carefully measured.

Computational simulations will be made as carefully as possible, as well for the volume geometry as for the thermal evolution.

Analysis of differences between the computed results (temperatures, displacements, stresses) and the experimental results will show influence of discontinuities and allow to quantify it.

Fitting a model according to the experimental results will be achieved by the research of an equivalent continuous medium including anisotropy, non linearity, elastoplasticity, no traction material...

3D description of the geometry should lead to complicated work. So it will be better to use 2D models with various mechacial lows. Nevertheless, major discontinuities such as main fractures of the granite will be considered as jointed elements and will need 3D evaluations.

Hydraulic phenomena

Variations of the fractures thicknesses may be introduced by the heat effects and give variations of hydraulic properties.

Different permeability measurements have been realised (injection test, low flow in watertight boreholes).

Interpretation objective is to determine the hydraulic behavior of some identified fractures by two methods :

- continuous medium model used to identify permeability tensor and the influence of mechanical stresses due to heating.
- discontinuous medium model : opening of fractures will be related to applied stresses and to measured hydraulic conductivity.

In the latest phase, possible effects of temperature on density of water and on the flow (viscosity) or effect of flow on intersticial pressure on the stress distribution will be examined.

2) Models validation

Experimental results will be compared to computed results obtained with the preceeding models.

Differences between the two sets of results will be identified and related to geological discontinuities.

Equivalent continuous media will be improved by using new limits conditions.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Compilation and interpretation of the experimental results (temperatures, displacements ans strains) have been made.

Preliminary results on thermal-elastoplastic models have also been made.

Progress and results

Temperature distribution

The results of temperature measurements show a very good agreement with the predictions and with corrected limits conditions mainly at the surface of the experimental room.

The heat transfert in fractured granite obeys the Faurier low. The 2D revolution model gives satisfactory results and allows good correlations with temperature measurements.

Measurements of mechanical effects

Surface and volume measurements give quantitative data and thermomechanical effects of the heat sources.

Interpretation shows important effects of fissures on the stresses in granite.

In the future, three directions of investigation will allow :

- to use a 2D model of revolution with a "no tension" condition at fracture surfaces ;
- to use a 3D model with the same "no tension" condition ;
- to study some fractures by 3D model.

4.2. REPOSITORIES AND ENGINEERED BARRIERS

4.2.A. Repository design and disposal techniques

.B. : No particular research contracts have been concluded on this subject

4.2.B. Engineered barriers

4.2.B.1 HLW container development

ASSESSMENT OF STRUCTURAL PERFORMANCE OF HLW CONTAINERS (COMPAS)

Contractor:	Ove Arup & Partners, London, England
Contract No:	FI1W/0111
Duration of Contract:	April '87 - Dec '89
Period Covered:	Jan '89 - Dec '89
Project Leader:	J Miles

A. OBJECTIVES AND SCOPE

The COMPAS project has been designed to look at the mechanical performance of those containers which will be used for overpacking and disposal of high level radioactive waste. The following partners are also participating in the project: CEA (F), Equipos Nucleares (E), NAGRA (CH), PSI (CH), SCK/CEN (B) and STEAG (D). By agreement of the partners it is restricted to the examination of containers for vitrified waste rather than containers for the direct disposal of spent fuel. The project is not concerned with the production of a specific design for licensing purpose; it is only intended to investigate the characteristics of representative designs.

The objectives of the COMPAS project are to look at the mechanical performance of these containers and to develop an understanding of how they will behave when subject to the most extreme conditions which can be foreseen in realistic disposal scenarios.

In order to predict the ultimate mechanical performance of the disposal containers it is necessary to use computer aided modelling techniques. The early part of the COMPAS project therefore includes a considerable amount of computational work which is aimed at developing confidence in the use of these techniques.

B. WORK PROGRAMME

During the previous year (1988) the contract was extended to include a more comprehensive programme of testwork and associated computing. The revised project plan now comprises the following activities:-

(i) Directory of Computer Codes
(ii) Containment Concepts
(iii) First Benchmark Exercise
(iv) Preliminary Ring Tests
(v) Intermediate Testwork
(vi) Advanced Testwork
(vii) Prediction of Ultimate Performance.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

The third and final year of the COMPAS project has seen the completion of all experimental and analytical activities. Preparation of the final report is in progress.

Progress of Work

(i) Directory of Computer Codes

During 1987, various finite element (and finite difference) codes were investigated to establish a directory of codes suitable for the stress analysis of containers for high level nuclear waste.

The directory included information on each code's supplier, its distribution and availability, technical background and pre- and post-processing capabilities. This directory has been published by the CEC as report EUR 12052 EN.

(ii) Containment Concepts

This work, performed during 1987, confirmed the state of development of repository plans for the project partners, and was used to determine feasible repository loading conditions to be used in the prediction of ultimate container behaviour (see section vii).

(iii) First Benchmark Exercise

The purpose of this study was to set up a series of problems against which the partners could test their favoured codes. Two different container concepts were investigated during this exercise - a thick walled 'corrosion tolerant' container and a thin-walled 'corrosion resistant' container. For both containers, the calculations investigated the effect of an external pressure on an annular slice through the container. Both geometrical and material non-linearities were considered.

All of the partners took part in the exercise, although not all of them attempted every problem. There was good agreement in prediction of yield pressures but further assessment was inhibited by the lack of experimental data available. The report on this work was published by the CEC under the title "Stress Analysis of HLW Containers" as EUR 12053 EN.

(iv) Preliminary Ring Tests

The First Benchmark Exercise confirmed that several codes were capable of analysing problems involving non-linear behaviour of HLW containers. However no experimental information was available to assess the accuracy of the codes' predictions. To address this shortcoming the Preliminary Ring Test programme was commissioned. In these tests ring specimens, representative of sections through a thick-walled HLW overpack, were loaded radially between parallel plattens. A number of tests were carried out on rings with representative defects. These included rings with internal or external notches and rings with an external layer of weld deposit.

The project partners were invited to model these tests, and most of the partners attempted all the problems. This exercise showed the partners' ability to produce acceptable results for a simply-defined but nevertheless still quite complicated problem. The comparison of experiment and analysis was most useful. The report of this phase was published by the CEC as report EUR 12401 EN.

(v) Intermediate Testwork

The Intermediate and Advanced Testwork programmes involved isostatic pressure tests on scale models of HLW containers. The Intermediate tests looked at simplified one-third scale models of thick-walled containers. By including several variations to the standard container (reduced wall thickness, non-uniform corrosion and different welding methods), these tests gave an opportunity to examine the effect of these variations on the performance of the container.

Eight containers were tested at the Schlumberger test facility in Paris between February and April 1989. The containers were subjected to a uniform external pressure of up to 100 MPa, and were extensively instrumented to allow post-test comparisons with results from the partners' calculations.

The analytical predictions were less accurate than in previous phases, reflecting the increased complexity of the problem. Some partners were unable to model the non-axisymmetric containers, and the ability to allow buckling in the calculations emerged as necessary for accurate predictions. This work has been submitted to the CEC for publication.

(vi) Advanced Testwork

Following on from the Intermediate tests there were a smaller number of tests on more realistic scale models of actual HLW containers. These containers were as near as possible to an existing design, but there were restrictions on the cost, size and strength of the models.

Three Advanced tests took place in August 1989. They were extensively instrumented and gave increased information concerning the likely behaviour of real containers under disposal conditions.

The partners' predictions were of a similar level of accuracy as for the previous Intermediate phase, but increased experience allowed the more complicated geometries to be considered. A report will be published by the CEC.

(vii) Prediction of Ultimate Performance

This study was intended to predict the ultimate performance of the containers under extreme disposal conditions. It drew on the conclusions from the experimental programme, especially the Advanced Testwork. A typical HLW overpack container was subjected to severe but conceivable loadcases and the analysis predicted that the containers would not suffer loss of integrity under load. Although the very high and anisotropic pressures meant that no testwork was possible, the experience of the previous phases lends confidence to the analyses and conclusions.

4.2.B.2. Backfilling and sealing of radioactive waste repositories

IN SITU REDUCED SCALE BACKFILL AND HEATER EXPERIMENT

Contractor : CEN/SCK, Mol (B)
Contract No : FI1W/0055/B
Duration of contract : from October 1986 through June 1990
Period covered : January 1989 to December 1989
Project leader : A.A. Bonne
Report by : B. Neerdael

A. OBJECTIVES AND SCOPE

In 1974 CEN/SCK launched a R&D programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site.

Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hoisting radioactive wastes as well from engineering point of view as for long term safety and performance evaluations.

In support of these also further modelling efforts are required in order tc improve and confirm our prediction capability.

This particular item aims at investigating in situ the performances of an argillaceous seal and its immediate surrounding clay in a thermal field.

The experiment is developed jointly by the CEA/DRDD and the CEN/SCK where the field test is taking place in the HADES-URL (Underground Research Laboratory).

B. WORK PROGRAMME

B.2.1 Investigations about materials and instrumentation
B.2.2 Integral reduced scale in situ experiment
B.2.3 Validation of heat transfer and thermo-mechanical codes

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The Bacchus experiment ("BACkfilling Control for High level wastes Underground Storage") has been designed in 1987 as a thermal loading experiment where a heater was providing temperature conditions in the clay mass likely to take place around a HLW canister after a cooling period of 50 to 60 years (surface interim storage).

The backfill aspect was integrated in the design of the test by considering the in situ behaviour of potential clay-based materials placed around the heater (prefabricated blocks). The ability of Boom clay to be re-compacted was also investigated.

The composition of a compacted buffer material was defined by the two parties involved (CEA/DRDD and CEN/SCK) in order to meet the experimental conditions prevailing at Mol.

The CEA provided blocks of compacted buffer material (engineered barrier around the heater) and of recompacted Boom clay (seal plug at the top) and designed the heating elements (D=60 mm,H=1500 mm) for the heater test at constant heat power of 500W.

Thermal probes and temperature sensors were placed by CEA in the experimental set-up. The CEN/SCK instrumented the clay mass in the near field (earth and pore water pressure, humidity, temperature).

The experiment has been run from March 1989 to Augustus 1989; the different phases of the experiment which are partly illustrated by the diagram of figure 1 (temperature evolution at the interface buffer material/clay host) are the following :

23/11/88 : the experimental set-up is placed in the access hole
- heater and buffer from 12.5m to 14m
- seal plug from 11 to 12.5m.
(instrumentation in host clay already operational)

15/12/88 : the access hole cased until 11m is concreted.

16/03/89 : the heating phase is started with a 100W thermal power (local ground conditions not yet restored).

23/03/89 : the power is increased to 500W.

16/06/89 : the power is off for a few minutes due to an external perturbation on the electrical circuit.

12/08/89 : the thermal power fallen to 0; no satisfying reason has been found up to now to explain this event.

Progress and results

2.1. Investigations about materials and instrumentation

The natural clay selected by CEA/DRDD (Ca-smectite) is mixed in a proportion of 50% with sand (45%) and graphite (5%) in order to match with the swelling pressure (5 MPa) and the thermal conductivity (1.7 W/m°C) of the host rock.

The characterization of the backfill material and the re-used Boom clay has been made by CEA. The isostatic compaction level of 20 MPa finally chosen for fabrication does not compromise the mechanical behaviour of large cylindrical cores (D=300mm).

The clay host instrumentation, in charge of CEN/SCK, has been installed by way of access drillholes prior to the installation of the test cell (temperature sensors, earth and pore water pressure cells and humidity probes). The last ones are based on the time domain reflectometry (TDR) technique or are using a neutron source.

Hydraulic pressure cells were also foreseen in contact with the buffer and the Boom clay to measure the swelling pressures which develop when the material becomes hydrated.

2.2. Integral reduced scale in situ experiment

Figure 2 gives the location and dimensions of the different components of the test cell.

The compacted backfill material is composed of 5 annular blocks, 0.30m high and the upper plug consists in 4 annular blocks of Boom clay, 0.36m high. The cables connecting the command and measuring units in the URL with the instrumented and heated lower part are introduced in the central hoisting pipe.

From a 760mm diameter access hole provided in the cast-iron lining, a 11m long vertical hole was drilled in 450mm diameter and lined by a steel casing. From this level, the hole was deepened in smaller diameter (390mm) on a distance of 3m and the test cell, 330 mm in diameter, lowered down the hole.

2.3. Validation of heat transfer and thermo-mechanical codes

Simulation of temperature and pressure evolution curves is reproduced using the TEMPPRES code. This computer code has been written for calculating temperature, pore pressure and stress distribution 3-dimensionally as a function of time around a heat source (of parallelepiped geometry) in an infinite medium.

In spite of assumptions which had to be made (values of thermal, physical and hydraulical parameters for clay and buffer) and not optimized time steps in the calculation, a comparison between experimental and numerical temperature values gave satisfying good agreement (figure 3 gives an example for one sensor located 28cm higher than the mid-plane of the heater in a hole located 52cm from axis to axis with regard to the test cell).

For humidity sensors, very encouraging results have been recorded by TDR prototype sondes ; further developments of this technique seem to be of interest for the future.

The reproduction by calculation of the thermal impact for the pore water and earth pressure cells is rather good in spite of a too great difference in magnitude; this one can be explained by the limitations of the code and/or the fact that we had to start the test at a time where the ground conditions were not restored yet.

The stress variations are ranging from 0.15 to 0.3 MPa for an initial effective stress level of 1 to 1.5 MPa and for distances from the sources of 0.5 to 1.5 meters.

The dissipation of the excess pressure takes place in the 3 weeks following the end of the thermal phase.

The swelling pressures exerted by the compacted buffer material are still increasing slightly.

List of Publications

/1/ NEERDAEL.B and al. The Bacchus experiment at the Hades underground research facility at Mol, Belgium. NEA/CEC Workshop on "sealing of radioactive waste repositories" Braunschweig, 22-25 May 1989

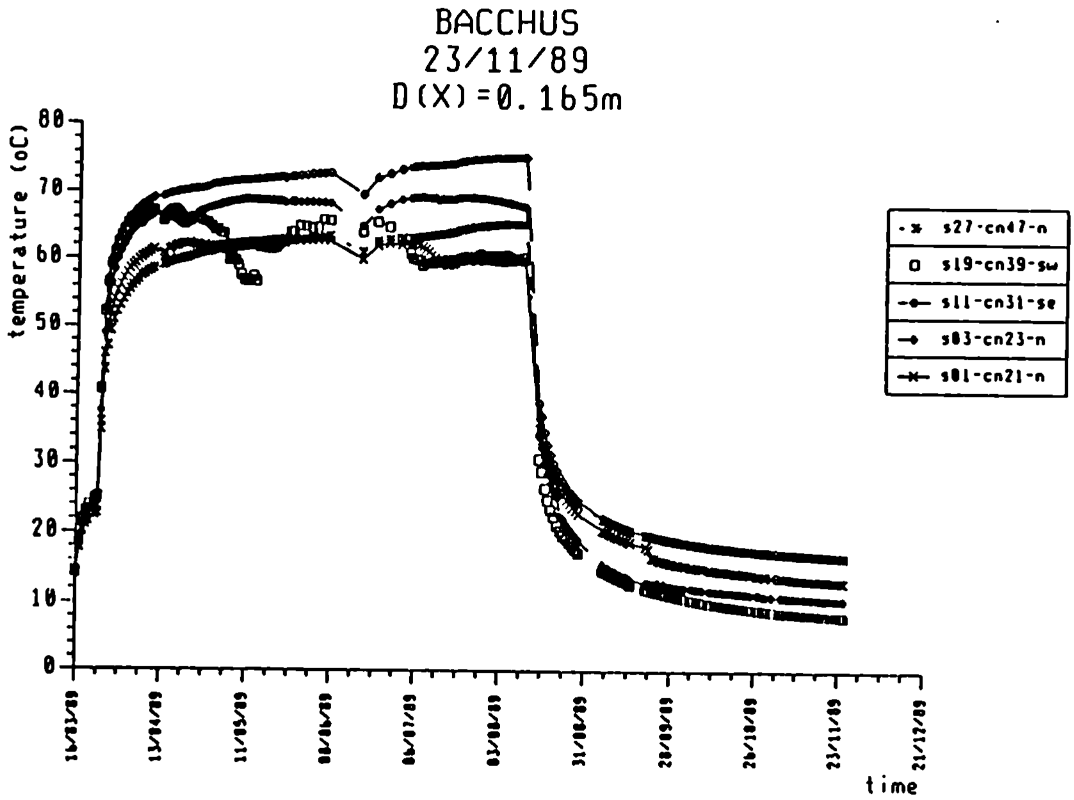


Figure 1. Temperature evolution profile.
Interface clay/buffer

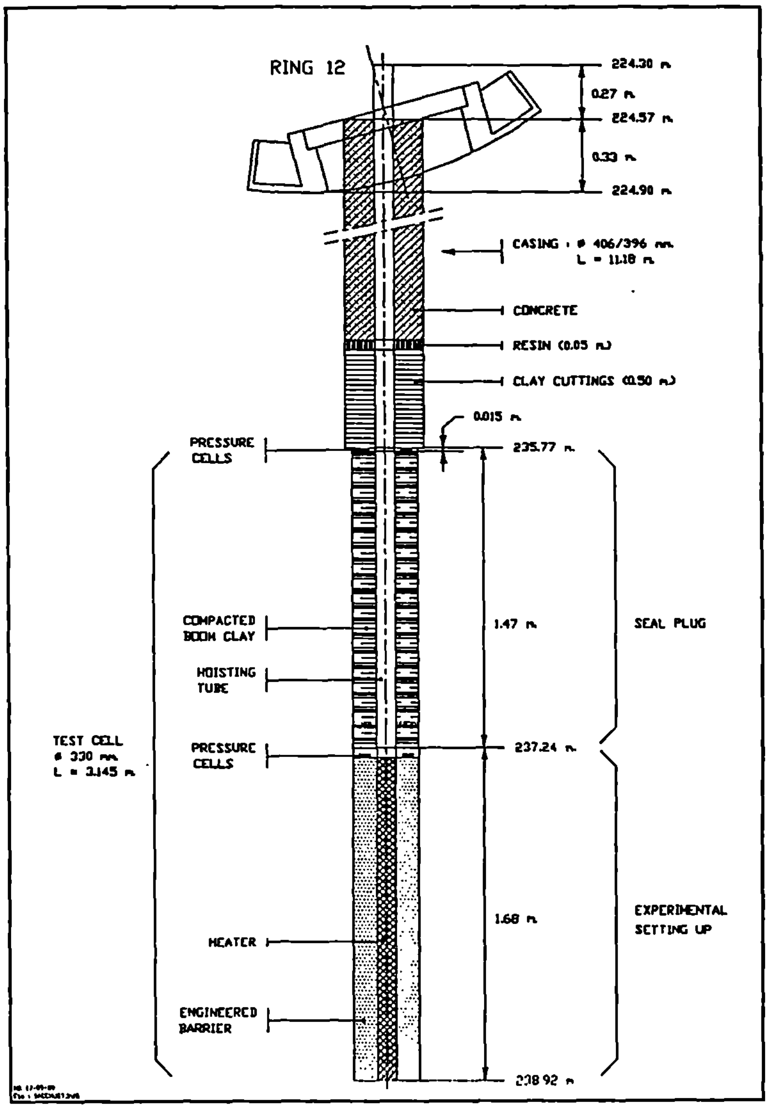


Figure 2. Design of the Bacchus test

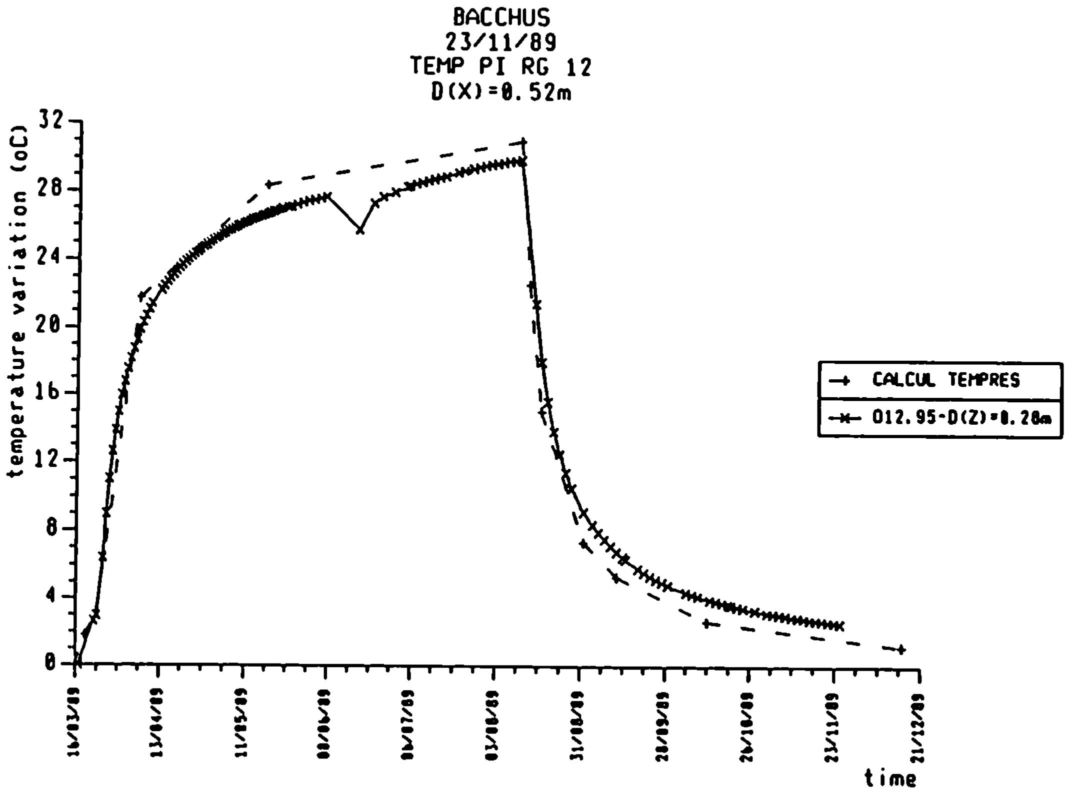


Figure 3. Temperature versus time.
Comparison experimental/numerical.

FEASIBILITY STUDY FOR HIGH LEVEL RADIOACTIVE WASTE DISPOSAL IN DEEP BOREHOLES DRILLED FROM THE SURFACE

Contractor : ENEA, CRE CASACCIA, ROME (ITALY)
Contract No. : FI1W/0056-I
Duration of contract : January 1987 - December 1989
Period covered : January 1989 - December 1989
Project leader : E. Tassoni

A. OBJECTIVES AND SCOPE

A preliminary feasibility study has been carried out in the frame of a previous contract (255-80-7 WASI) on the disposal of high level and cladding hull waste in the Pliopleistocenic blue clay of Italy. Two preliminary repository models have been assessed :

- mined repository
- deep boreholes drilled from the surface

Of the two models, assessed for the same reference power programme (10 GWe), the deep borehole facility seems preferable if compared to the mined repository from several points of view but especially as regards total cost and flexibility. The deep borehole facility is also modular, it doesn't heavily interfere with local and use; even if inacceptable geological conditions are encountered in one borehole, operations can be moved to another part of the site with little cost penalty. The main goals of this project are the following :

- elaboration of a preliminary demonstration test of high level radioactive waste disposal in deep boreholes drilled from the surface;
- determination of the in laboratory and in situ behaviour of plugging and backfilling materials for deep borehole disposal.

B. WORK PROGRAMME

1. Feasibility demonstration test of deep borehole plugging.
1.1. Laboratory permeability tests, carried out both in small triaxial cells and in big oedometric cells, on samples formed by natural clay as well as on samples formed by clay sealed with different materials (cement, bentonite, etc.).
1.2. On site plugging test of a deep borehole, sealed with the material selected under item 1.1.
2. Project of a demonstration test for high level waste disposal in deep boreholes drilled from the surface.

C.PROGRESS OF WORK AND OBTAINED RESULTS

No progress has been made

TRIALS AND CONTROL OF PLACING FILLING AND SEALING MATERIALS FOR DEPOSITS IN SCALE MODELS

Contractor : SOLETANCHE ENTREPRISE S.A.
Contract n° : FI 1W/0057
Duration of contract : February 1987 to March 1990
Period covered : January 1989 to December 1989
Project leader : D. GOUVENOT

A. OBJECTIVES AND SCOPE

The containers of radioactive wastes of high or low level activity will be disposed of in vertical shafts or horizontal galleries. In order to avoid any groundwater circulation in contact with the containers, the voids between the rock and the containers will be filled with sealing materials.

The materials, subject of this study, are cement based mortar formulations which are the results of previous studies and experiences of SOLETANCHE and C.E.A. These materials made of water, cement, clay and additives have to exhibit various mechanical characteristics as stability, imperviousness, durability, possible retention towards radioactive cations and above all easy placing in order to assure a good quality of filling and sealing.

The first objective is to evaluate the hereabove characteristics of various mortar formulations, through lab tests, in order to select four formulations best suited to the filling and sealing.

The second objective is to improve and finalize the placing method of materials in scale models. To that aim, each of four scale models will be, one after each other, filled, cured at various temperatures, tested and dismantled for examination in order to define the best material placing technique.

B. WORK PROGRAMME

1. Laboratory studies

1.1 Theoretical studies. From the previous studies and experiences, choice of filling material formulations.

1.2 Preselection tests (based on the placing criteria). The rheological properties of the hereabove formulations are measured in fluid state (viscosity, shear strength, bleeding, workability limit).

1.3 Final selection of formulations. Measurements of characteristics of hardened materials (strength, shrinkage, water content, permeability, thermal conductivity, microstructure, retention and diffusion of caesium ions) cured at various temperatures (20, 50 and 80° C).

2 Experimental studies

2.1 Vertical scale models. Filling and quality controls of three successive vertical scale models :
- study and improvements of materials placing and instrumentation,
- controls of fresh and hardened materials in lab and in situ,
- controls and tests of filling and sealing properties of materials in the scale models.

2.2 Horizontal scale models. Filling and quality controls of an horizontal scale model :
- same tests and controls as vertical models,
- finalization of materials placing technique. Particular specifications for horizontal scale models.

C. PROGRESS OF WORK AND OBTAINED RESULTS

1. State of advancement

After the first year (1987) of the research program devoted to laboratory studies, the two following years were mainly aimed at the experimental studies, i.e. at the tests on the scale models.

During these two years, three scale vertical test models, simulating a waste container placed in a shaft, were prepared, sealed and tested after curing at 80° C. The models themselves were successively slightly modified in order to improve the design and the quality of sealing. Three various sealing materials were used : the first two ones were Portland (CPA 55) cement base mortars with two different water/cement ratios and the third one was made of CLC 45 cement (clinker + fly ash + slag).

The fourth and last scale model is an horizontal one, simulating a waste container placed in an horizontal gallery ; this model is in progress.

2. Progress and results

2.1 Laboratory studies. Laboratory studies are completed except the ones concerning diffusion properties of various materials which were still going on in 1989. Diffusion tests of caesium 137 and of tritium water (HTO) on three various sealing materials, differing over the cement type, have been in progress since 1988 at the C.E.A. facilities. The cements used in the three materials are :

- CPA 55 (clinker),
- CLC 45 (clinker + fly ash + slag),
- CLK 45 (Slag + clinker).

After almost two years of test the stabilizations are not reached yet for some samples. The results are as follows :

	De, HTO	De, Cs 137
- CPA 55	$1.1.10^{-3}$	7 to 14.10^{-5}
- CLC 45	$< 3.10^{-4}$	2 to 7.10^{-6}
- CLK 45	$2.7.10^{-4}$	Not stabilized yet.

(De = diffusion coefficient in cm^2/day).

Portland cement appears to be not as good as the two other ones, since the diffusion coefficients are about 5 times greater. CLC 45 and CLK 45 exhibit very similar diffusion characteristics for tritium, whereas CLK 45 should likely be better than CLC 45 as far as caesium 137 is concerned.

2.2 Experimental studies

2.2.1 Vertical scale models

2.2.1.1 Design. Figure 1 shows the design of the vertical scale model which was carefully described in the previous progress report. Only slight modifications were made to the successive models according to the experience gained with each test. The concrete cylinders of the first two models were built by means of three cylindrical concrete elements (50 cm high and 5 cm thick) stacked and glued over each other. These elements are made of very lightly reinforced concrete so that excessive deformation was supposed to have occured during the leak tests on the first model. The second model was reinforced by means of steel ribbons surrounding the model in order to avoid deformation due to inside water pressure applied during the leak test. The concrete cylinder of the third model was a one-piece cylinder made of steel reinforced and vibrated concrete (160 cm high and 8 cm thick). On the other hand, it is supposed that a light evaporation of water of the sealing material occured through the concrete of cylinder during the curing period and mainly during the first days, leading to potential shrinkage of the material, despite the protection of the wet soil in which the model was buried. Thus, for the second model, the concrete cylinder was coated in and outside coated with epoxy resin. However, at the time of the dismantling of this model, it appeared that the binding strength between mortar and set epoxy was not as high as the one between mortar and concrete. Finally the third model was only outside coated with epoxy. The results of leak tests given hereafter show that these successive modifications, at least, participated to the improvement of the overall watertightness of the models.

2.2.1.2 Quality control equipments. These equipments were described in the previous progress report. Five various tests are performed :

- temperature variations measurement,
- ultrasonic logging,
- leak test,
- sampling by diamond coring ; physical characteristics on samples,
- visual observation.

2.2.1.3 Filling the scale model with sealing material. After heating the "waste container" at 80° C, the annular space is filled up with sealing materials. The three mortars used for the sealing of the models are made of :

model n°	1	2	3
	water	water	water
	CPA 55	CPA 55	CPA 55
	Silica fume	Silica fume	Silica fume
	Clay	Clay	Clay
	Fluidifying agent	Fluidifying agent	Fluidifying agent
	Retarder	Retarder	Retarder
	Siliceous sand	River sand	River sand
W/C ratio	0.66	0.50	0.50

The fabrication of the sealing materials is performed in 2 steps :

- preparation of a slurry with all components except the sand, in a high turbulence mixer,
- mixing of slurry and sand in a concrete batch mixer.

The placement of the materials in the annular spaces is performed, by gravity by means of a small tremie pipe.

The viscosity/rigidity (shear strength) of the material used for the first model was a bit too high (200 Pa). For the models n° 2 and 3, the rigidity was thus tuned up to lower levels (70 and 90 Pa respectivily). These lower rigidities are thought to be participating to the easier placing of materials and to the overall better quality of the set materials as well.

2.2.1.4 Tests on cured scale models

2.2.1.4.1 Temperatures. The variations of temperatures in the sealing materials during and in the hours following the placing are very similar for the three scale models. A first phase of rising temperature due to conduction of container heat, a second phase of faster rising due to the additional heat of cement hydratation, then a short stabilization between 70 and 75° C followed by a slow drop down to balanced temperatures stable between 70° C against the container and about 50° C close to the wall of the concrete cylinder.

2.2.1.4.2 Thermal conductivity. The thermal conductivity computed from the recorded temperatures after stabilization are 2.7 +/-0.5 W/mK for the first model and 2.6 +/- 0.5 for the second one.

2.2.1.4.3 Sonic logging. Sonic tests were performed before filling up the annular space with sealing materials and after the curing period. For the first model no significant differences could be noted between the two series of tests. However, some tiny adherence defects were detected between the outside steel pipes and the concrete cylinder likely due to unsufficient sealing. The same problems occurred on the two other models despite very thorough cares taken to glue properly the pipes against the concrete cylinders so that these tests did not allow any defect in the sealing materials to be detected.

2.2.1.4.4 Leak tests. The leak tests performed by water injections into the bottom of the models showed a regular improvement of the sealing quality on the successive models. For the first model, under a water pressure of 50 kPa, the water came to the surface between the sealing material and the concrete cylinder along a small split which was extending with time likely under the action of water pressure. The average flow rate was around 500 ml/min. For the second model, the pressure was limited to 20 kPa. The water came very slowly to the surface through a very small split along the wall of the concrete cylinder, but also along two thermocouples inserted in the sealing material not far from the concrete wall. The global flow rate was about 15 ml/min and the one along the concrete was estimated to only 8 to 10 ml/min. In the third model the only leak detected was along a thermocouple even when the water pressure was increased from 20 to 40 kPa. The flow rate was 4 ml/min et 20 kPa.

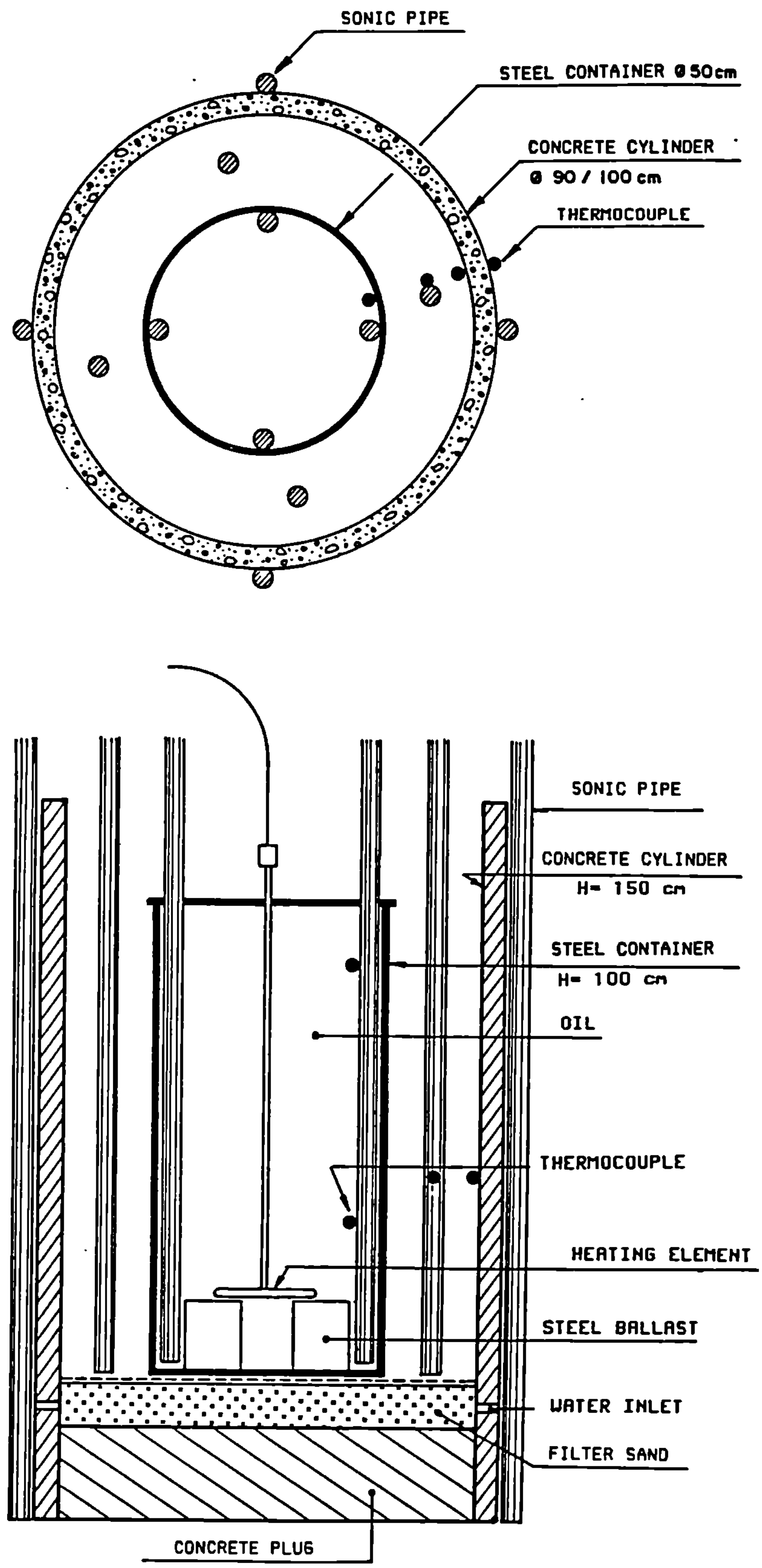
2.2.1.4.5 Tests on cored samples. Tests performed on cored samples recovered from the first two models gave the following results :

	Model n° 1		Model n° 2	
	U.C.S. (MPa)	Permeability (10-12 m/s)	U.C.S. (MPa)	Permeability (10-12 m/s)
. Top	34	5	43	25
. Bottom	36	4	52	6

2.2.1.4.6 Visual observations. Visual observations were made on both cored samples and cut models. On the first model the presence of small bubbles of air was noted as well as some tiny flat lenses of air likely trapped during the placing phase, due to the too high rigidity of the mortar. The lenses were localized along the concrete cylinder. In the second model a few very small bubbles of air were also noted at the top of the model. Along the wall of the concrete cylinder very fine "cracks" (1/10 or 2/10 mm thick and 1 to 3 cm long) were noted in two locations at the bottomm of the model.

3. Next stage

The next and last stage of this programme is the horizontal scale model which is under construction and will be sealed early 1990. The design is rather similar to the one of the vertical model with a steel cylinder placed in the middle of a concrete "gallery". The waste container will be first heated to 50° C and the void will be injected with the sealing material by means of a pump. After curing, the tests will be the same as for the vertical models.



VERTICAL SCALE MODEL

FIGURE N° 1

Sealing of fractures and boreholes

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, France
Contract No : FI1W/0058
Duration of contract: October 1986 - September 1990
Period covered: January 1989 - December 1989
Project Leader: J.Y. BOISSON

A. OBJECTIVES AND SCOPE

Radioactive wastes disposal into deep cristalline rocks requires to carry out important underground works or geological investigations by the means of boreholes which might induce direct pathway or short circuits for water, between repository and ground surface. It is essential to use specific technics and methods allowing the sealing of these potential pathways.

Among several other possibilities, clay materials have been considered to fill the boreholes and shafts. One family of clays, the montmorillonites, are widely studied because of their qualities : good adsorption towards radionuclides, swelling properties in contact with water.

The aim of the study is to determine, in laboratory and in situ, the behaviour of such clays, used as a plug in a borehole in contact with an hydraulic fracture, and to study the possible erosion of such a plug so as to be able to estimate the longevity of the sealing.

B. WORK PROGRAMME

2.1. Laboratory studies

2.1.1. Experimental erosion apparatus with bentonites are used, owing to scale models simulating a fissure. Appropriate technics are carried out for the determination of the eroded clay mass by a water flow inside the model fissure.

Tests will be performed with different types of representative clays, i.e. those which are presentely investigated and could be used as sealing material in a real repository :

- the Na bentonite "Green Bond", from Wyoming (USA)
- the Na bentonite "MX 80", also from Wyoming (USA)
- one Ca bentonite (FoCa), from Fourges-Cahaignes (France).

The waters which will be used, will be of different types, either demineralised, natural (from cristalline formations), or reconstituted with different percentages of relevant ions.

2.1.2. The behaviour study of this sealing material is completed with direct observations of the clay in the fissure, with granulometric analysis, and mineralogic analysis....

We intend to ascertain the influence of the nature and velocity of the water flow. The experimental parameters will be then ions concentrations caracteristics and water flow rates.

2.2. In situ study

A fracture with a significative water flow, in a well known direction, will be selected, in situ, in a mine or quarry. The geometric position of this fracture will be identified owing to boreholes, used themselves for hydraulic studies and sealed with bentonite at the fracture level.

The resulting water flow will be characterised by the means of tracers, water flow rates measurements at the free surface of the drift or on the face of the quarry.

After the experimentation, for a time depending upon the amount of the recovered clay mass, an over coring operation will be done, perpendicular to the fracture plane, both at the plug level, and around.

Modifications of the clay plug will be analysed in the laboratory, using the same technics as those for the scale model tests.

The clay, used for this test, will be one of those used in the laboratory.

2.3. Numerical model

The evaluation of the transported clay mass will be analysed and quantified both for the laboratory and in situ experimentations.

The results will be compared, and it is intended to establish a correlation between the two configurations.

This must lead us to provide a numerical model concerning the erosion behaviour and the durability of such bentonite plugs in boreholes in contact with water.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The first part of this research has consisted in laboratory tests, simulating the behaviour of a bentonite, sealing a fracture owing to its hydration and swelling due to the presence of water, and submitted to erosion phenomena due to the action of the same water flow.

Tests have been performed under different conditions, considering two types of geometry for the simulated fracture, different types of bentonites at different physical stages of hydration. The measurements which are made aim to caracterize and evaluate the longevity of the clay under these different conditions.

The last series of tests have been achieved this year by studying the erosion processes with highly compacted and dry Na and Ca cylindrical plugs of bentonite intersected by horizontal fracture.

In parallel, concerning the in-situ study, a suitable site was identified in a uranium mine (COGEMA-Limousin), consisting in a gallery with a main fracture hydraulically active. The complete characterization of the site was done, the borehole was drilled through the fracture, and it is ready to the emplacement of the clay plug.

PROGRESS AND RESULTS

2.1. Laboratory studies

- Experimental tests

. To simulate partially the in situ geometric conditions, we have carried out this last laboratory study to attempt to recognize and compare the behaviour of highly compacted and dry bentonite samples (Na and Ca), using clay plugs set down in scale models of boreholes including an hydraulically active fracture. Tests are performed in "Plexiglass" devices simulating a portion of vertical borehole (Ø = 58 mm) with an horizontal fracture simulated by an hollow disc inside which a water circulation is created (fig.1). Clay plugs used for these tests are cylindrical samples, (Ø = 57 mm, h = 100 mm) of highly compacted (under 1000 bars) and dry (W% ∿ 10 %) bentonites, either Na (MX 80) or Ca ("FoCa") French bentonite (2) and (3). Tests are run with demineralised water at constant flow rate (Q = 10 ml/min ; approx.10^{-4} to 10^{-3} m/s) with control of the resistivity.

. Swelling and erosion processes of the clay plugs in contact with water coming from the fracture are controlled during each tests under three aspects :

- measurements of the radial swelling volumes of the clay from the plug, in the fracture, and observations (with photos) on the aspect evolution on this bentonite filling in the fracture ;
- characterization of the outflow, both with nephelometric measurements and corresponding particles concentrations, and evolution of the particles sizes distribution of the eroded matter ;
- characterization of the clay at the end of each tests, with water contents repartition, granulometric and mineralogic differences at different levels both in the plug and in the fracture.

- Results

. The radial swelling of the two kinds of bentonites in the fracture seems in good agreement with what has been observed for previous free static swelling tests : higher swelling of the Ca bentonite FoCa at the beginning of the tests (∿ 40 days), and after this period, the Na bentonite MX80 does not show the continuous swelling developped in static conditions, and, hence, the observed swelling values appear to be in the same order of magnitude for both bentonites cf. fig.2,

. Comparative tests have been carried out, in the same conditions as previously mentioned with the cylindrical samples, but without any water circulation (Q = 0), giving us radial swelling values in "static conditions" in the same fracture. It can be observed that, in these conditions, this static swelling for the Ca bentonite FoCa, reaches values of the same order than with water flow, but, in the case of the Na bentonite MX 80, the "static" swelling values are higher that those observed with water circulation.

. This point leads us to think that erosion phenomena might be more active in the case of the Na bentonite MX 80.

. This statement is corroborated by observations made on the clay aspects in the fracture during the tests : as soon as the hydration of the plug takes place and the clay begins to migrate in the fracture, the aspect of the two kinds of clay is somewhat different. In the case of the Na bentonite MX80, as the clay migrates, there is the same development of zones as observed in the tests with highly hydrated Na bentonite. From the core to the clay/water interface, we can distinguish : a gradual lightening (4) of the clay including individual black particles radially oriented, a dark zone constitued of black particles forming a crown of approximatively 5 mm of thickness after 5 months of tests, and an interface zone having the aspect of a grey gel, easily draged along (this grey gel is almost non-existent in the case of tests with flowing water, but is well developed in the "static test", as in the case of tests with highly hydrated Na bentonite). In the case of the Ca bentonite FoCa, the migration of the clay in the fracture appears homogeneous concerning the colour, but with development of mini or micro fissures radially oriented with degradation into small blocks of particles at the front contact between water and clay.

Analysis of the outflow :

. The characterization of the outflow is made owing to a turbidity measurements technic. Samples are analysed regularly during the five months test. Raw data are expressed in "F.N.U." units. A calibration, with the same demineralised water and different concentrations of the tested clays, is made to convert FNU units into mass concentration. The comparative results between the Na bentonite and Ca bentonite expressed in FNU units are reported fig.3. The values concerning the MX80 are rather irregular, with mean values in the order of 5 to 10 FNU with peak values probably due to variations of the flow rate, or irregular transport of the clay matter in the device. However, in the same conditions, the FNU units values measured during test with Ca bentonite FoCa are quite non-existent. Refering to our calibration, the equivalent mean concentration of eroded Na bentonite particles is in the order of 25 to 50 mg/l. Concerning the Ca bentonite FoCa, the values are below the possibility of our measurement technic. This fact does not mean that there is no eroded particles in the case of the Ca bentonite FoCa, but that, as it can be seen directly through the transparent fracture model, the transported eroded matter has the aspect of small aggregates of clay with no dispersion in the water ; as consequence they are very difficult or impossible to identify using turbidity technics.

At the end of each test, the experimental device is dismantled and the plug plus the clay in the fracture are sampled in order to perform water content, grain size and mineralogic analyses of the different zones.

The resulting distribution of the water contents in the clays after 5 months are reported fig.4. Starting from the same initial water content W% $\sim$ 10 % the final hydration of the plug is obviously different between the two bentonites, with higher sensitivity towards water for the Na bentonite MX80. But this difference is widely amplified for the expanded bentonite into the fracture :

in the case of the Ca bentonite, the water content increases logically (1) from the core to the external water front from 40 % to ∿ 100 %, but, in the case of the Na bentonite MX80 we can notice a rapid increase of this value particularly at the level of the lightened zone (up to 550 % just close to the dark particles zone) (Note that the front grey zone has not been analysed).
This difference concerning the water content distribution of the clay in the fracture can be directly related to the grain size distribution of each zone. From the results of these analysis it can be noticed that there is no fundamental change in the case of the Ca bentonite FoCa between the original clay from the core and the zone close to the water front, but for the Na bentonite MX80, we can see the granulometric differenciation of the different zones specially for the dark zone with coarser particles. The consequences of this differenciation is a great heterogeneity close to the water flow, with fine particles at high water content just behind coarser particles.

The mineralogic analysis performed on samples taken from plugs and clay in the fractures underline these differences between the two bentonites. No change are observed with the FoCa7 clay, while for the MX80 it can be noticed a decrease in the initial amount of Montmorillonite and Beidellite at the edge of the clay mass in the fracture, accompanied with appearence of an interstratified clay mineral (14,25 A). In conclusion it can be said that the observations which have been made during these laboratory tests show that, with the pessimistic conditions imposed such as demineralized water, width fracture of 2 to 5 mm, flow rate up to 10 ml/min, the bentonites possibly used as plugs to seal borehole could be sensitive to erosion phenomena. It is confirmed that, as for other properties of these types of clays, this possibility of erosion is essentially governed by the chemical characteristics of the water through the development of chemical exchanges processes. It can be noticed that, in the same conditions, the behaviour of the two types of bentonites (Na and Ca) appears to be relatively different, leading to erosion processes of different nature.

2.2. In situ study

As planed, the last part of this study in the framework of this contract consists of an in situ experiment, with a clay plug in a borehole intersected by an hydraulicaly active fracture.

A suitable site was identified in an uranium mine (Silord site, Fanay-COGEMA), with favourable characteristics (cf. fig.5) :

- . the structural analysis of the site led us to select a portion of gallery including a major fracture with low pitch (20°W).
- . the borehole was drilled at the roof of the gallery, intersecting the fracture, perpendiculary (total length : 6 m, ∅ 56 mm, inclin. 75°SE).
- . the core has allowed to precise the characteristics of the fracture : 2 or 3 mm apperture, filled with clay material.
- . the borehole was equipped so as to perform tracers tests from the intersection fracture borehole : d spite the lack of water at the present time, the main role of the selected fracture was confirmed.

Furthermore, a clay plug one meter long was built. It is made up of a cylindrical (Ø 54) casing constitued with a perforated stainless steel sheet (0,5 mm thickness), in which are stacked five cylinders of dry and dense (compacted under 1000 bars) bentonite MX 80, similar to what was used in Sweden (5).

After several months of contact between the bentonite plug and the water from the fracture, it is planed to overcore the first borehole (Ø 250 mm), including the plug itself so as to analyse the behaviour of the clay and evaluate the erosion phenomena.

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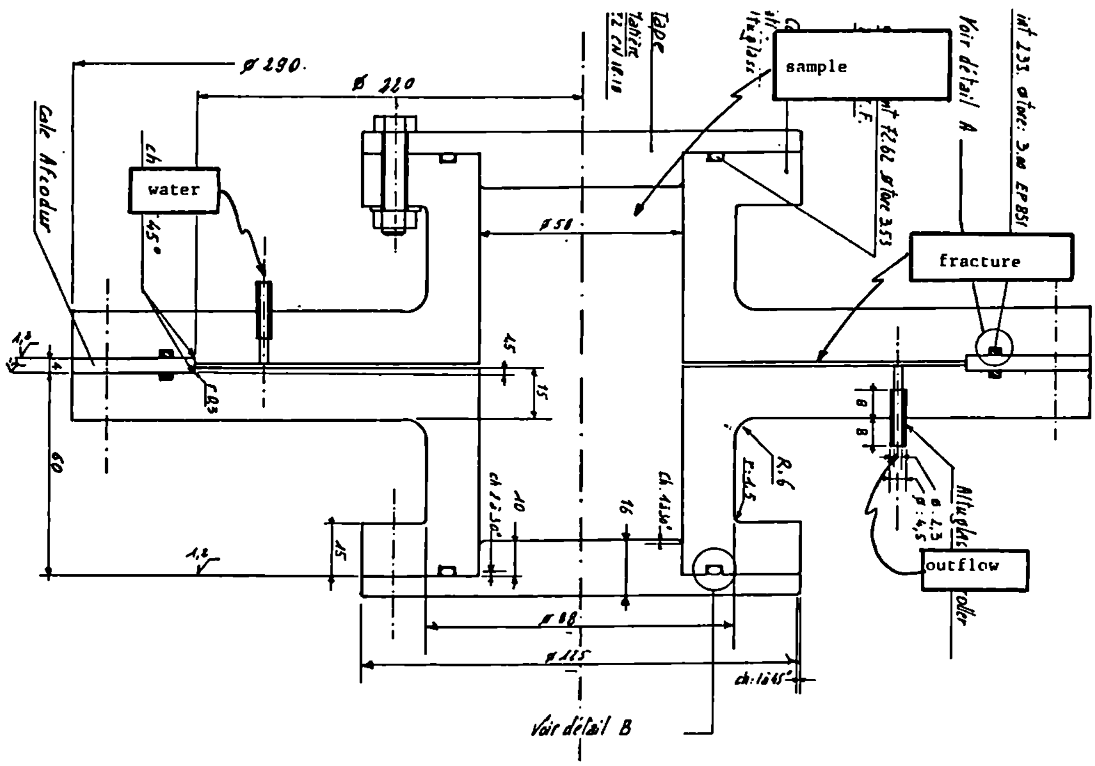


Fig. n°1 : Scale model of an horizontal fracture filled with cylindrical bentonite plugs

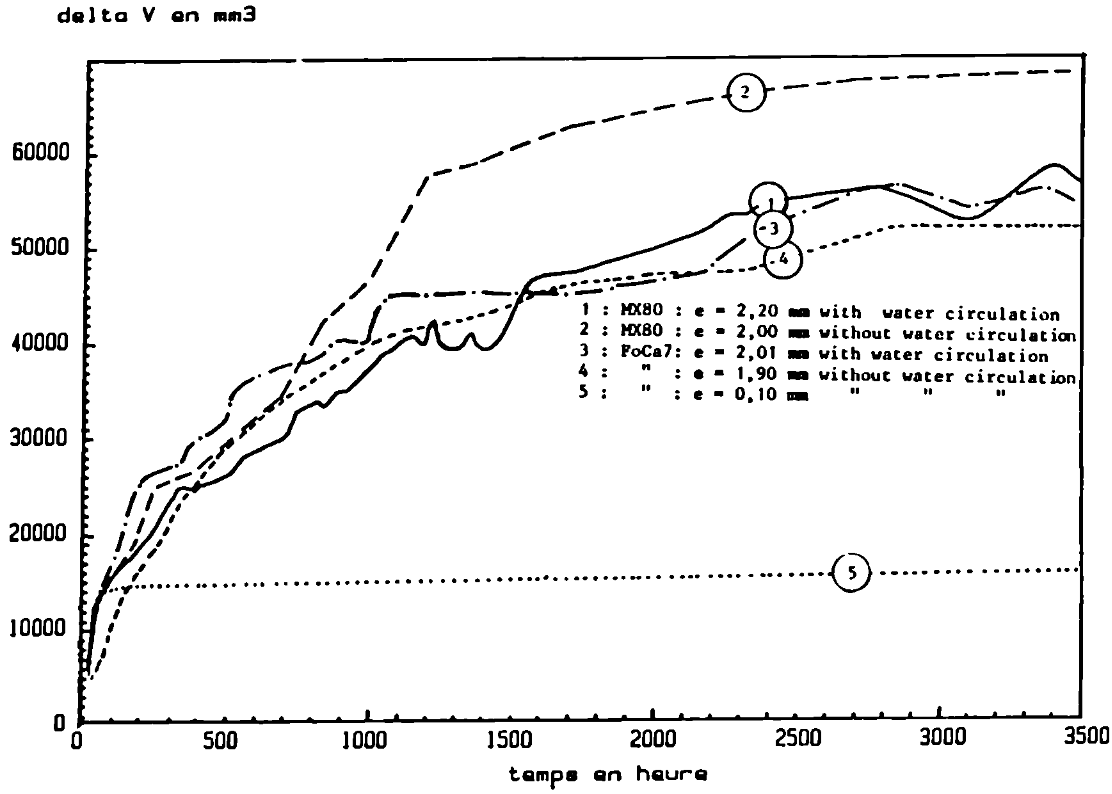


Fig. n°2 : Volumetric swelling of compacted bentonites in the fracture

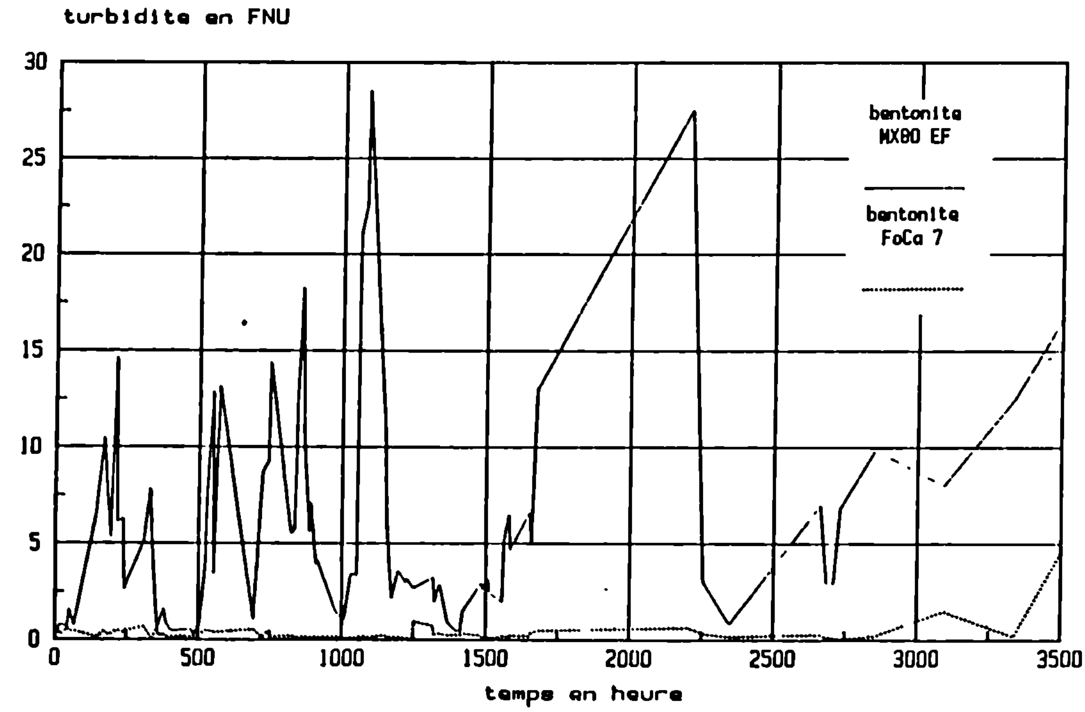


Fig. n°3 : Turbidity evolution in the out flow erosion tests in the horizontal fracture on two compacted bentonites (flow rate = 10 ml/min)

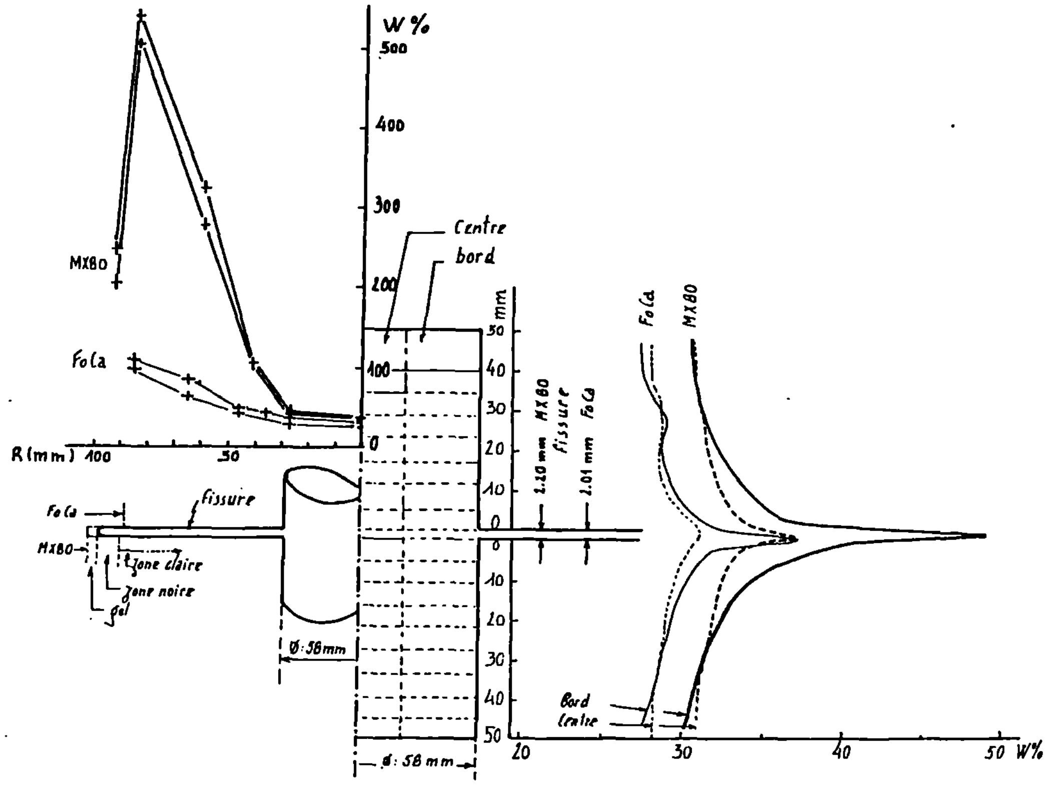


Fig.n°4 : Distribution of W% in the core and the clay in the fracture after 5 months tests for two bentonites

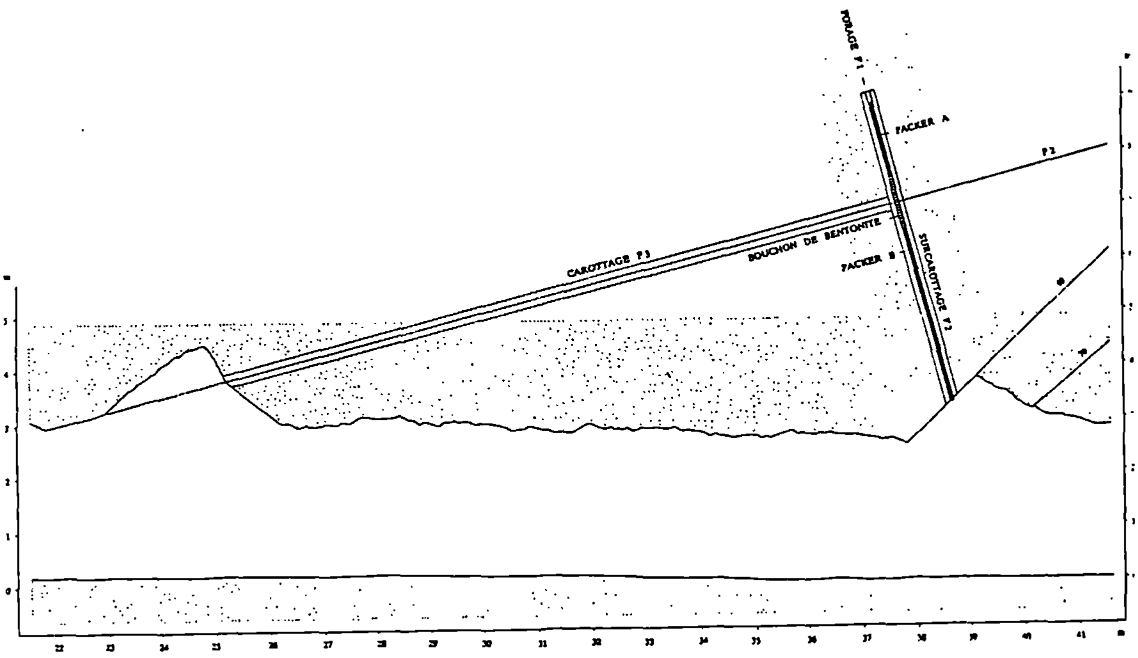


Fig.n°5 : Configuration of the in situ test with MX80 plug

Research on Backfilling and Sealing of Rooms and Galleries in a Repository in Salt

Contractor: GSF/Institut für Tieflagerung (IfT), Braunschweig, F. R. G.
Contract No.: FI 1W/0059/D
Duration of Contract: September 1986 - December 1989
Period Covered: Jan. - Dec. 1989
Project Leader: M. W. Schmidt

A. OBJECTIVES AND SCOPE

The occurrence of the hypothetical incident access of brine into the repository containing radioactive wastes must be assumed. As a consequence the transport of radionuclides by brine into the biosphere must be prevented. These objectives are to be obtained already during the operational phase by the constructional elements backfilling and sealing.

In this respect various in situ and laboratory investigations have been carried out in the Asse salt mine. The investigations serve to indicate the suitability resp. the degree of effectiveness of processes and building materials. Of importance besides the retentive properties of these components are especially those which exert a supporting and stabilizing effect on the rock since they contribute towards a minimization of fracturing and consequently permeability. The influence of the disturbed zone in the vicinity of an excavation must be analyzed with priority.

Laboratory tests on fine salt grit and precompressed salt briquettes have supplied important information on the material behaviour of selected backfill and sealing materials. The results of in situ measurements from backfilled chambers in the Asse salt mine have supplied prime knowledge regarding the long-term behaviour of backfill, which, by continuing these measurements, will provide a considerable contribution towards valuations on the stability of part of the mine.

B. WORK PROGRAMME

B.1.1 Soil mechanical laboratory research on backfill materials to investigate backfill characteristics
B.1.2 Geotechnical in situ measurements to determine the interaction between rock salt and backfill
B.1.3 Performance of geotechnical in situ measurements and rock mechanical laboratory tests on old, preconsolidated backfillings
B.1.4 Development of an evaluation matrix for selection and use of suitable backfillings in salt formations
B.2.1 Investigations in the laboratory on the mechanical and hydraulic parameters of sealing components
B.2.2 Surveying supervision of existing chamber and gallery sealing systems at the Asse salt mine
B.2.3 Development of an evaluation matrix for selection and use of suitable sealing components

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In the course of compaction of fine-grained salt grit characteristic dependencies of the density upon the compression rate and the moisture content were determined. These investigations are being continued under variation of further material parameters.

The vertical stress measured by the flat jacks positioned on the floor of chamber 8a/532 m level does not increase significantly surpassing its own weight 3 years subsequent to backfilling activities. At this time practically no indication of beginning load influence as a consequence of convergence is recognizable. This phenomenon can, however, only be defined after evaluation of the future measurements.

Three years after start of measurement registration the settlement rates of the backfill in chamber 3/658 m level are presently approaching a preliminary final rate.

Progress and results

B.1.1: With the aid of a test stand newly developed by GSF, compression tests are carried out on samples of salt powder with an average grain diameter of < 0.063 mm. This test stand enables determination of the radial and axial stress distributions. Empirical compaction functions have been evaluated at compaction velocities from 0.01 to 10.0 mm/min and at moisture contents from 0 to 4.0 wt %. With fine grained salt grit the compaction velocity had an impact only on the compaction behaviour of moisturized material. The density increases with moisture and decreases with compaction velocity at a stress range of < 100 MPa (Fig. 1 and 2).

B.1.2: In the Asse salt mine stresses and deformations are being continuously measured in the backfill as well as in the surrounding rock in two backfilled chambers since 1982.

In chamber 8a/532 m level the stepwise backfilling was completed at the beginning of 1987, which is now demonstrated by the stress curves of the flat jacks installed in the floor of the chamber (Fig. 3). Since then the measured stresses induced by the overlying backfill are practically constant. The values generally range between 0.16 MPa and 0.24 MPa, corresponding to a density of 1.1 t/m^3 to 1.6 t/m^3.

The supporting effect of the walls (skin friction) is demonstrated by the lower stresses ranging from 0.04 MPa to 0.12 MPa registered by the flat jacks positioned in the corners of the chamber resp. near the walls. Up to now the backfilling does not take over horizontal supporting effects.

Nine settlement cells have been installed in the backfilling at 5 m to 13 m height above the floor. The settlement in relation to the reference point at the northern wall actually varies between 105 mm to 250 mm (Fig. 4). The measurement values alternate with a cyclic period of 2.5 years and a range of about ± 40 mm. In order to find an explanation for this behaviour, gauging stations are presently being installed at various reference points of the deformation measurements. In conjunction with the mine leveling devices evidence for these alterations might be found.

The settlement rates of the approximately 33 m thick backfill of chamber 3 at the 658 m level increase by only 15 mm/a three years after installation to actual values of about 105 mm at a depth of 3 m. Horizontal compaction in the southern area takes place with a constancy of up to 3.3 mm/m/a fastest towards the northern area where the actual compaction rate of 1.0 mm/m/a is still increasing. This is confirmed by other measurements in the vicinity of this chamber, as is shown by deformation vectors in northerly direction.

B.1.4: Current investigations on the assessment of backfilling procedures have made clear that the choice of backfilling procedures can only be correctly evaluated by taking different waste package storage methods into consideration. In the case of an unbedded disposal of cylindrical waste packages a non-compactable void ratio of at least 30 % for uncompacted backfill must be assumed. Bedded storage can increase the compressibility of the pore volume by means of convergence up to 100 %, with the consequence of increasing the total initial void volume of the backfill and decreasing of the ratio of the waste package to the backfill. Nevertheless, by means of compaction processes the conditions can be considerably improved in the event of bedded storage.

B.2.1: Commercially available salt briquettes from Kali + Salz AG were subjected to triaxial compression and creep tests. The triaxial tests under a confining pressure of 2.5 MPa to 20 Mpa rendered compaction rates of max. 4 wt %. Breakage occurred almost immediately subsequent to maximum compaction. Under an isotropic state of stress of < 20 MPa, such as will be prevailing in seals of final repositories at a depth of approx. 1000 m, a maximum compaction rate of 2 wt % can be obtained.

Uniaxial creep tests were carried out under temperatures ranging from 25 °C to 80 °C and at a state of stress from 4 MPa to 15 MPa. This resulted in permanent consolidation rates and reduced creep rates under both changes of temperature and changes of stress.

B.2.2: The stresses within 8 gallery seals as well as in their nearer surroundings at the 750 m level are measured by hydraulic flat jacks. The measurement results differ greatly according to their position in the mine (neighbouring excavations) as well as due to influences of the interaction between the sealing construction and the surrounding rock. Presently being tested are the correlations between varyingly strong decreases of stress - partially to 0 MPa - and to reactions to activities in the mine as, for example, to the drivage of new excavations.

The pressure build-up begins in most seals shortly after their completion. In some constructions pressure build-up can be determined only after the elapse of approx. 4 years, depending upon the selected constructional method and on the spatial arrangement of the measurement gauges. Reasons for this may be that the construction is rather weak in comparison to the surrounding rock, or that the loosening zone has its effect, which must still be investigated in more detail. The maximum measured stresses reach 22 MPa.

In the multibarrier concept the assessment of the seals depends directly upon backfilling process corresponding to the selected storage method for the waste packages and on the geometry of the cavities. In case of high permeability of the backfill in a repository high quality seals are required which will have to exert their efficiency already during the operational phase of the repository.

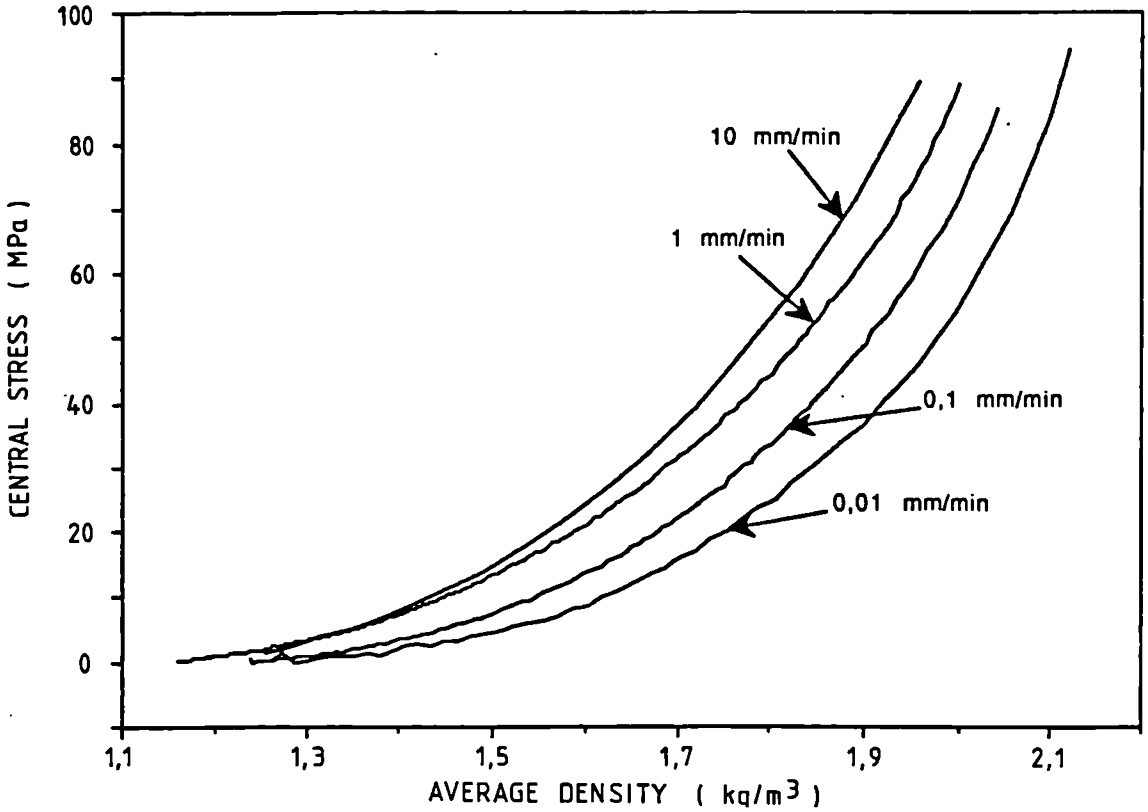


Fig. 1: Relations between central stress and average density at 1 wt % moisture at different compression velocities

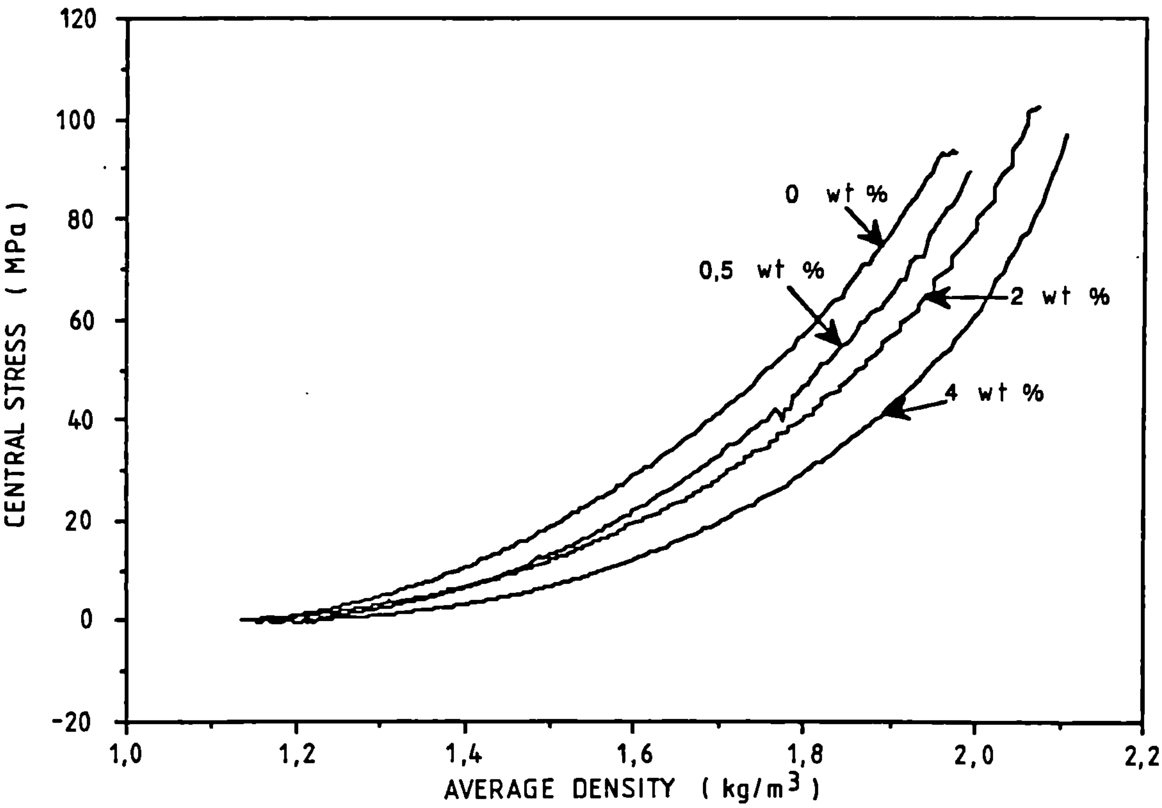


Fig. 2: Relations between central stress and average density at 1 mm/min compression velocity and different states of moisture

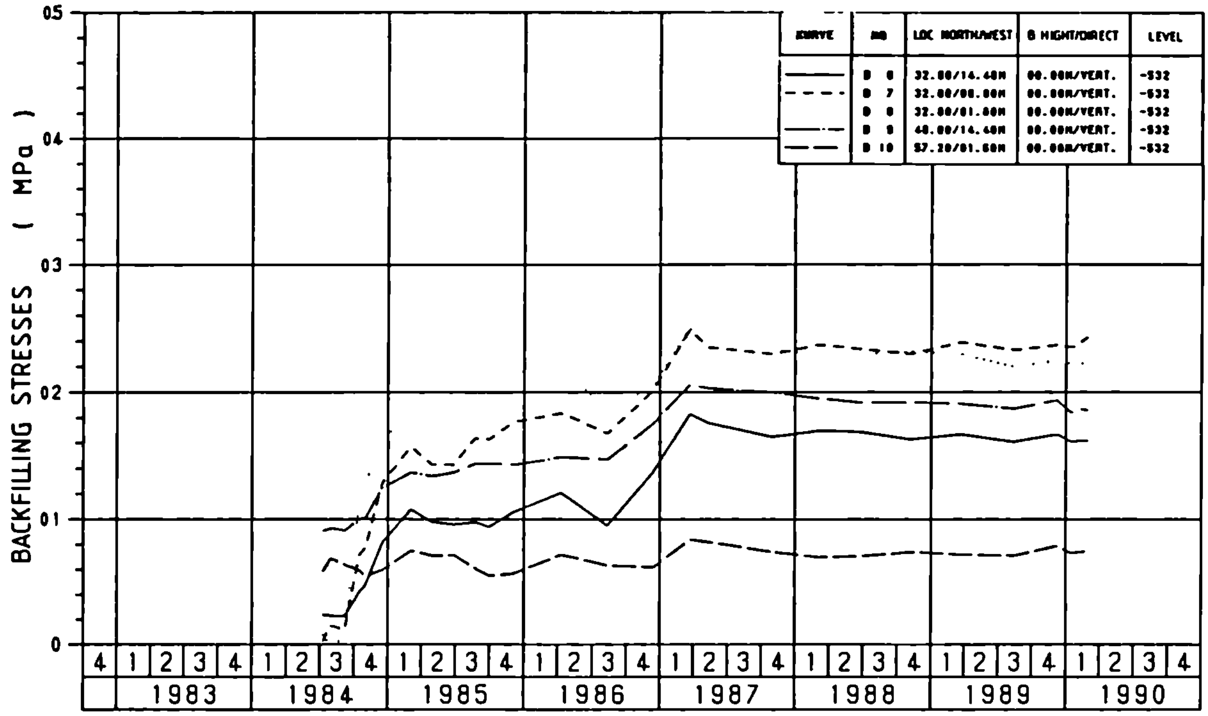


Fig. 3: Measured stresses at different points at the bottom of the backfilling

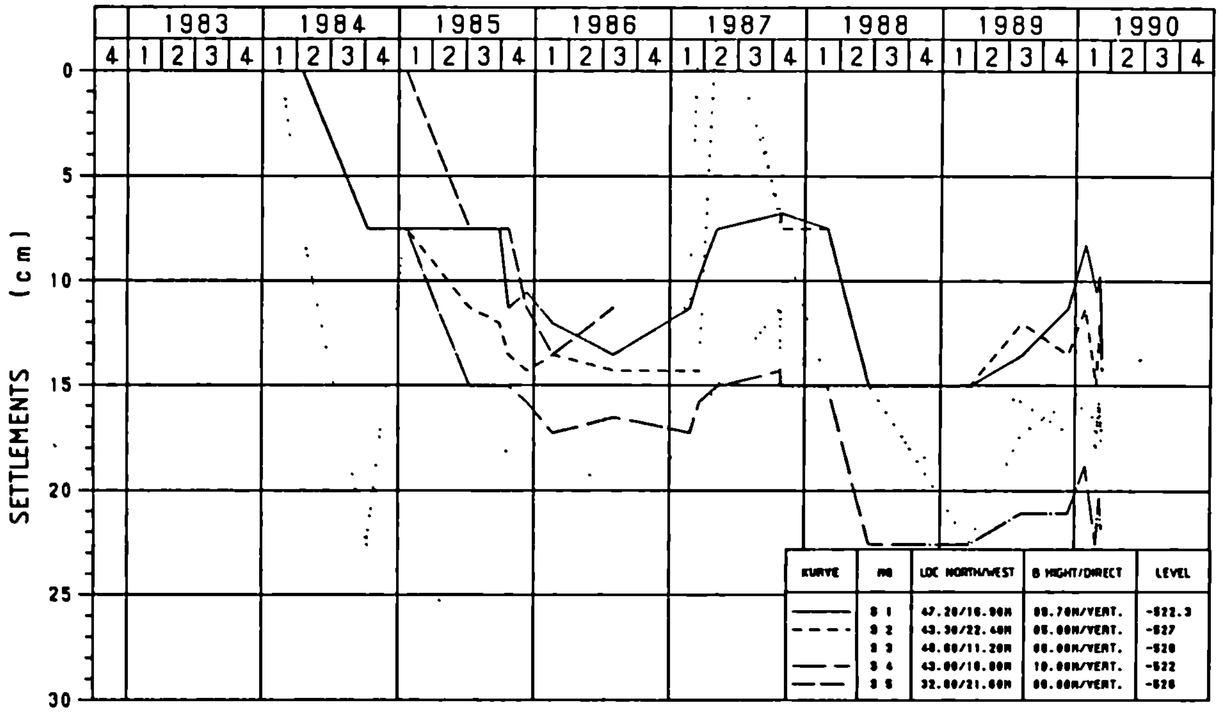


Fig. 4: Settlement of the backfilling in chamber 8a at the 532 m level at different measurement points

CRUSHED SALT BEHAVIOUR UNDER EFFECT OF A HEAT SOURCE IN BOREHOLES DRILLED IN A SALT MINE

Contractor : ANDRA, PARIS, FRANCE
Contract n° : F I1W/060
Duration of contract : January 87 - April 89
Period covered : January 89 - April 89
Project leader : M. RAYNAL

A - OBJECTIVES AND SCOPE

The study concerns the final stage of a nuclear waste disposal in deep salt formations : the partial or complete closing of a site. Sealing of drifts and shafts must be performed as tight as possible to set up a barrier as similar as possible to the natural geological barrier.
As crushed salt backfilling corresponds to one of the considered concepts a research program is actually developped on the basis of the following axis :

- *thermomechanical behaviour of crushed salt during thermal climax and during cooling,*
- *mechanical interaction between crushed salt and rocksalt,*
- *evolution of fluids, especially trapped air in crushed salt pores.*

This study led to performing an in-situ experiment by heating crushed salt in several boreholes drilled in a salt layer in the Alsace Potash Mine (MPDA).

ARMINES (Association pour la Recherche et le Développement des Méthodes et Processus Industriels) is contractor for ANDRA to support the experiment which is conducted by LMS (Laboratoire de Mécanique des Solides) and the Engineering Department of MDPA (Mines de Potasses d'Alsace).

B - WORK PROGRAMME

1. *Preparation of the test site*
   Drilling of the boreholes :
   - *six 240 mm diameter boreholes destined to be heated,*
   - *small measurement boreholes : four around each of the heated ones.*
2. *Instrumentation*
   - *Setting up of the computer controlled system developped by LMS for data acquisition and data transmission (from the test site to LMS).*
   - *Equipment of the borehole :*
     + *introduction of the framework supporting the electric wires and several transducers (for measurement of temperature, interstitial pressure of trapped air, total pressure of crushed salt, borehole closure),*
     + *backfilling of crushed salt in five of them, the sixth one leaving without backfill in order to check natural closure of heated salt and to compare its behaviour to the others'.*
3. *Follow up of the experiment and interpretation of results.*

C/ PROGRESS OF WORK AND OBTAINED RESULTS.

State of advancement.

Heating on the test site was definitively stopped at the end of 1988 ; it had been going on during 247 days in Boreholes 2 to 5, 316 days in Borehole 6 (fig. 2). However, the measurements have been performed on the whole 297 parameters until end of March 1989, and then on a representative selection of 12 parameters for three additional months.

Progress and results.

One has to notice that stabilization, both on a thermal and a thermo-mechanical plan, requires a cooling period almost equivalent to heating period. However, analysis of the most significative experimental results, hereafter presented, confirm that the most important part of the supplied energy - that is of the order of 4.10^{10} J in each heating borehole - was already evacuated at the end of the experimental cooling period of six months :

1. *Borehole wall temperature (fig. 3 : T42) shows two steady state levels corresponding to the heating supplied powers of 1.62 kW and 2.25 kW ; interruption of heating is characterized by an instantaneous decreasing response which duration is of the order of 10 hours, the characteristic time evaluated for start of heating. This response time is of course the most important in rocksalt as the measurement point is distant from the heating borehole (fig. 3 : T424, 1 metre distant). After a period of time of about 30 days, main influence on the evolution of temperature appears to be provided by the variations of ambient temperature (fig. 3 : Tam) in the gallery of the test site.*

2. *Borehole closure, as a consequence of thermal stress relaxation, has reached comparable levels in the different boreholes of the order of 5% (fig. 4) ; this results from an almost identical temperature field inside and around all boreholes, without any significant influence of the varieties and compaction degree of the crushed salts used as backfilling material. Interruption of heating period is characterized by a divergence of the boreholes, followed by a closure resumption at very low rate, showing at a large scale the elastoviscoplastic behaviour of rocksalt.*

3. *Rockmass extensions measured in boreholes parallel to the heating boreholes, are reported to the distance between the measurement points and the relative fixing points at the gallery wall. They present in a perpendicular direction (fig. 5) similar evolutions than borehole closure, but they are the least influenced by power supply changes during heating period, and divergence phases after heating interruption are the most important as the measurement points are distant from the heating borehole. Existence of extension strains confirm that modelling calculations cannot be performed with the asumption of plane strains.*

.../...

4. *The developped* <u>*pressures*</u> *within the backfilling materials appear to depend on crushed salt grainsize (fig. 6a and 6b) ant to have evolutions as a function of borehole closure of the type* $P = \left(\frac{\Delta V}{V}\right)^n$ *(fig. 7a and 7b).*

   Unfortunately, the loss of reliability of the transducers after heating interruption does not allow to give observations about cooling effects.

Nevertheless, the running of the test on an experimental ground can be considered as quite satisfying ; with the exception of the electric wires in borehole 1 and to a smaller extent the pressure transducers, the definition of the experimental conditions, the choice of the range for the various transducers, allowed to follow evolution of the parameters during cooling period too.

<u>*Future prospects.*</u>

This analysis of experimental results concerning cooling period will be completed by a modelling interpretation in the global synthesis of the test to be prepared.

On the test site, it is planned to develop a dismantling program on the following axis :

- *observation of non backfilled Borehole 6 wall (endoscopy) in order to examine its eventual fissuration state ;*

- *extracting of one of the supporting frames for the transducers and the wires, in order to examine their state with regard to corrosion ;*

- *cutting off rocksalt samples at various distances from the heating boreholes, in order to examine effect of thermal loading on chemical, petrographical and mechanical properties with comparison to those of natural non heated rocksalt ;*

- *taking off crushed salt samples of the various used backfilling materials in order to submit them to similar comparative analysis than rocksalt here above.*

List of publications.

1/ CEC/OECD Workshop on "Backfilling and Sealing of radioactive waste repositories", 3rd meeting, May 22nd - 25th, 1989 in Braunschweig - RFA.

2/ Proceeding of International Symposium "Mécanique des roches et physique des roches en condition de grande profondeur", Pau - France, Août 89.
"Comportement thermomécanique du sel broyé utilisé comme remblai dans un massif salifère". pp 545 - 552.

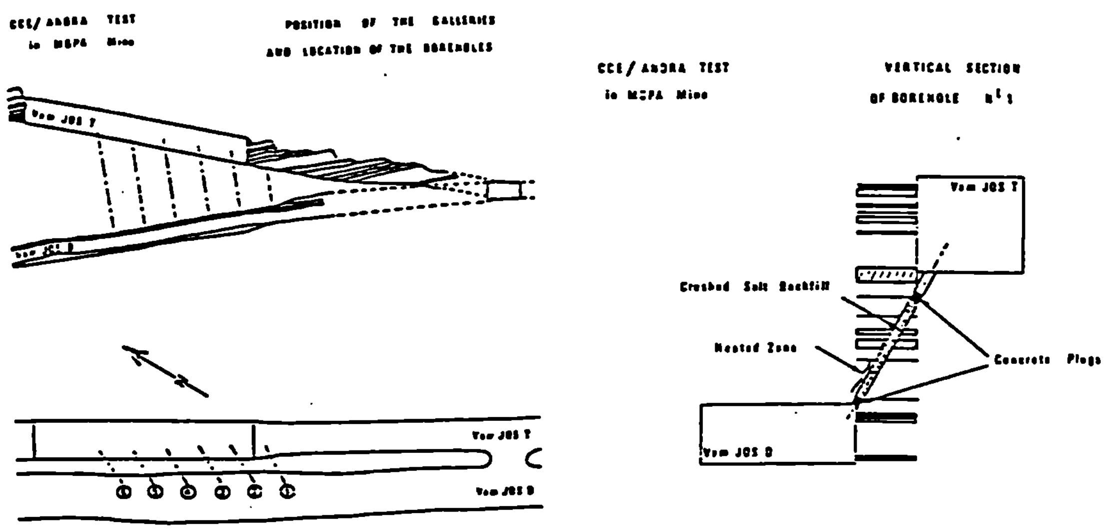


Figure 1 : Site of the test and location of the boreholes

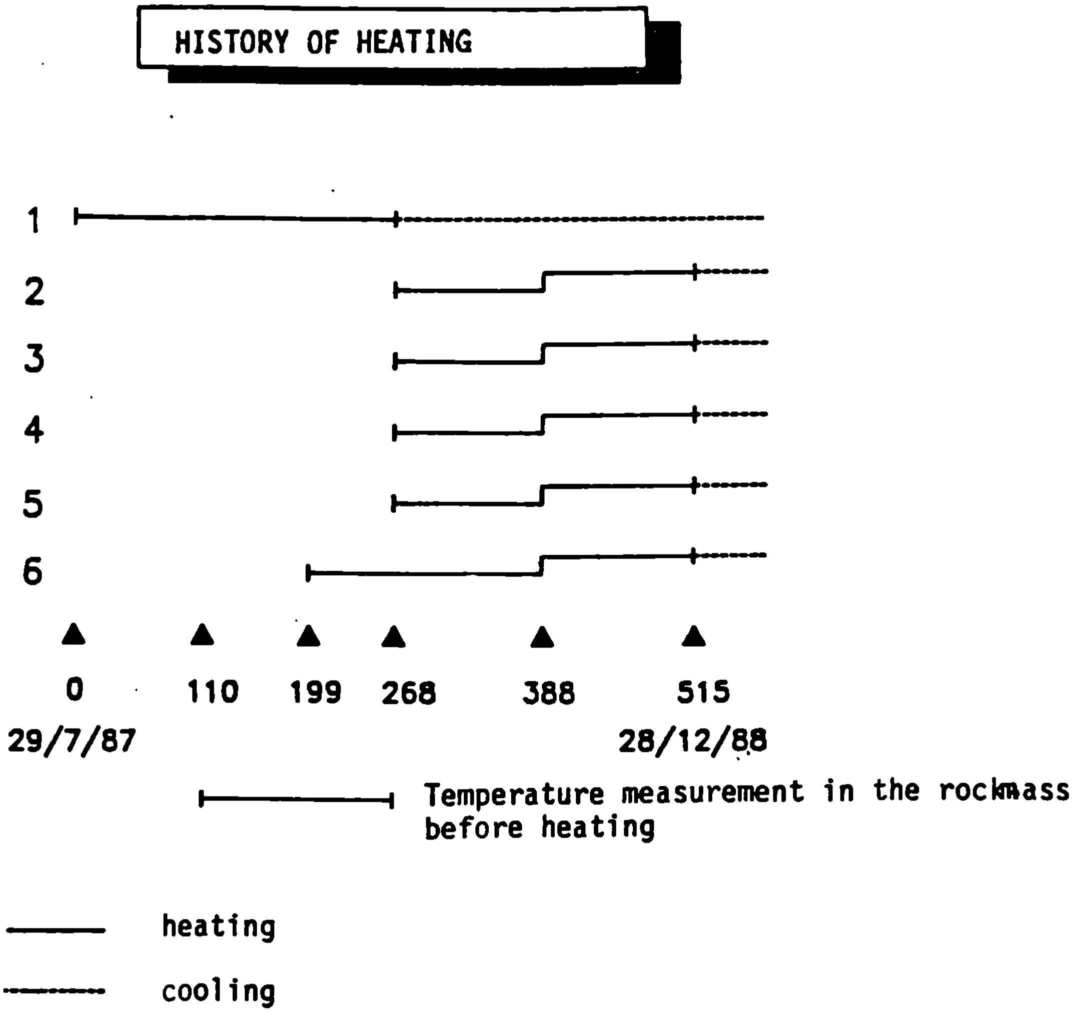


Figure 2 : History of heating in the boreholes

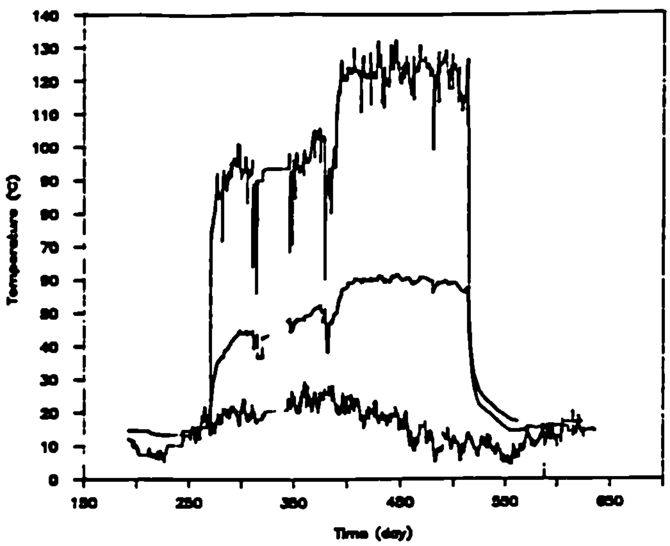


Figure 3 - Temperature evolution around borehole 4

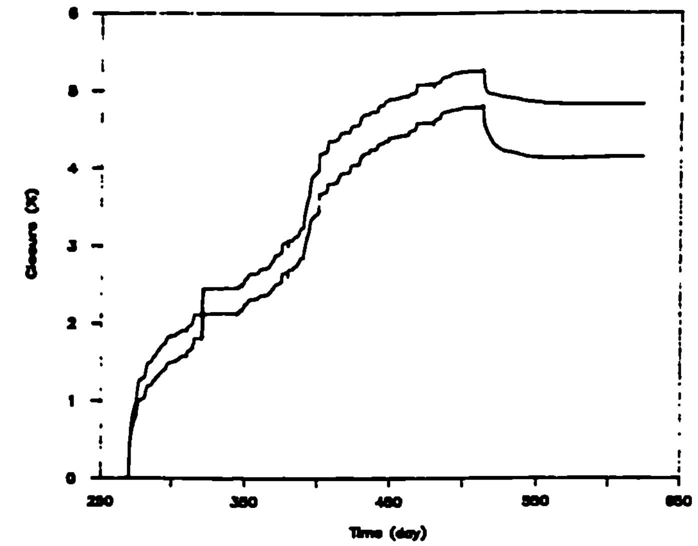


Figure 4 - Borehole closure evolution

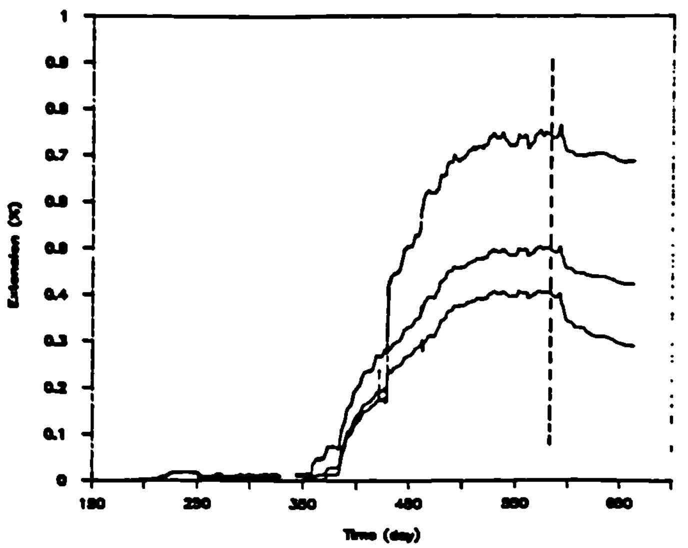


Figure 5 - Rockmass extension evolution at a distance of 1.12 m from borehole 4 and at the dephs of o.55 m, 1.24, 1.93 m

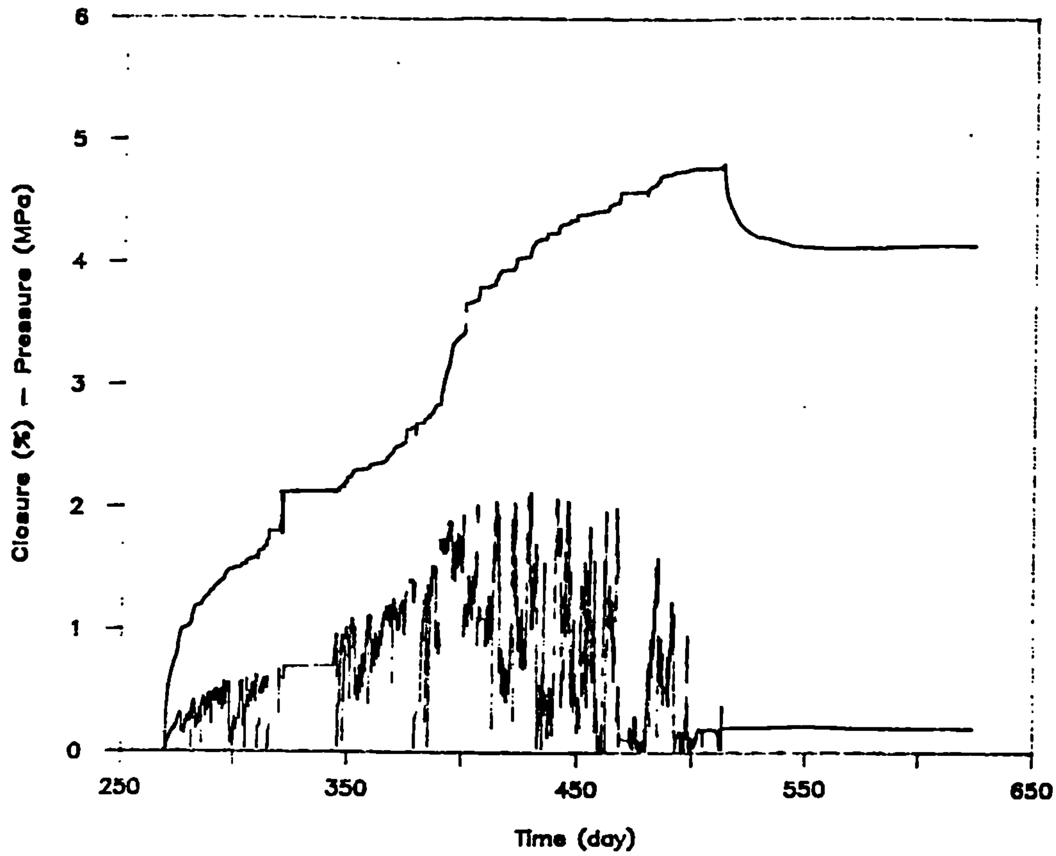


Figure 6a : Closure and pressure evolution in borehole 3 fine grained compacted crushed salt

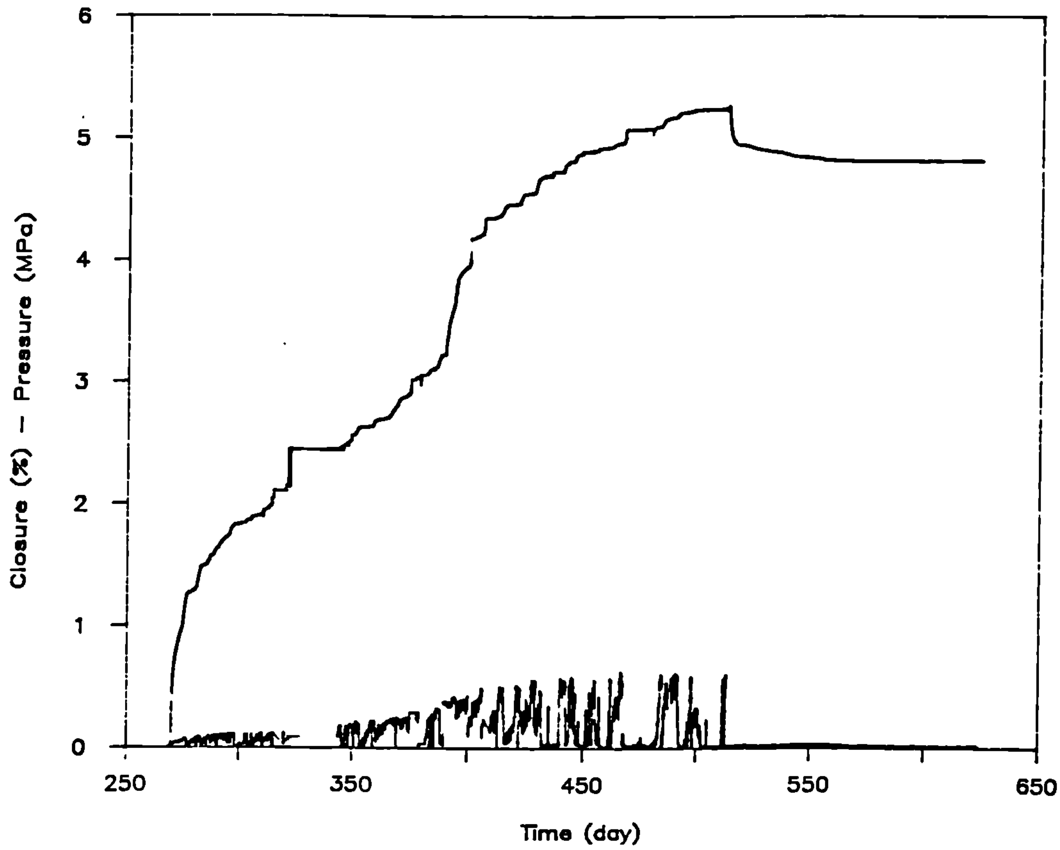


Figure 6b : Closure and pressure evolution in borehole 5 coarse grained non compacted crushed salt

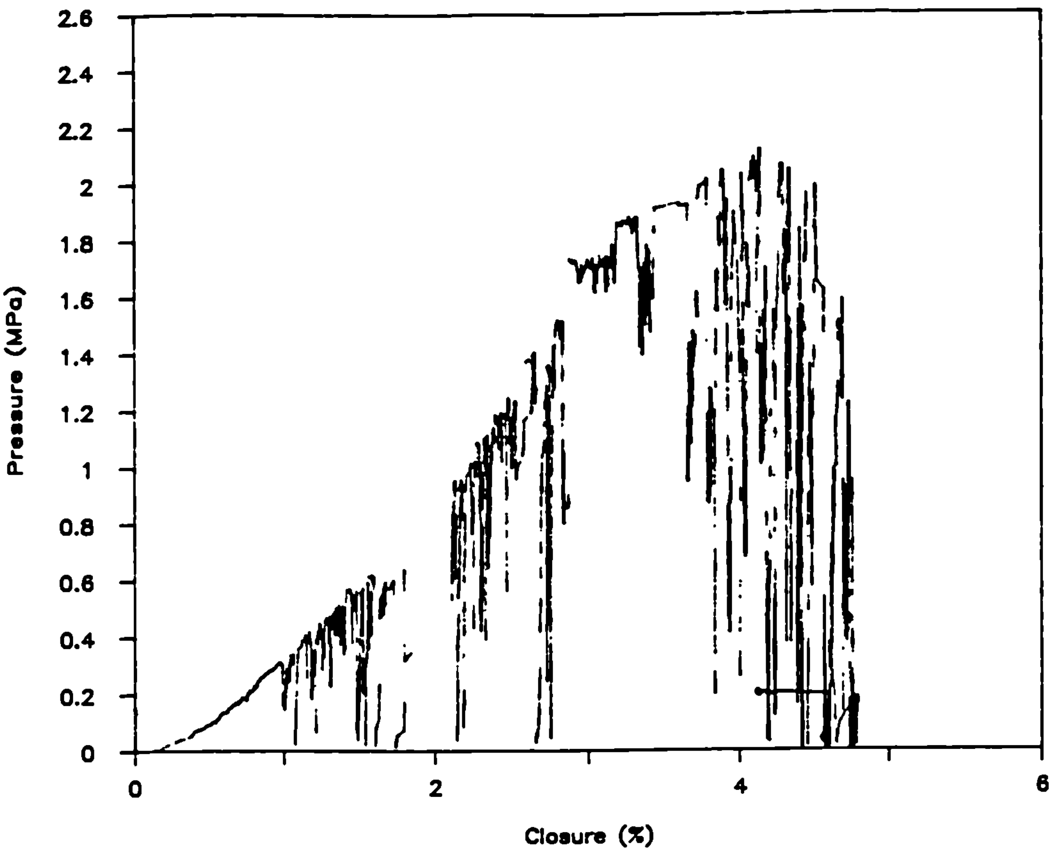


Figure 7a : Pressure evolution versus closure in borehole 3

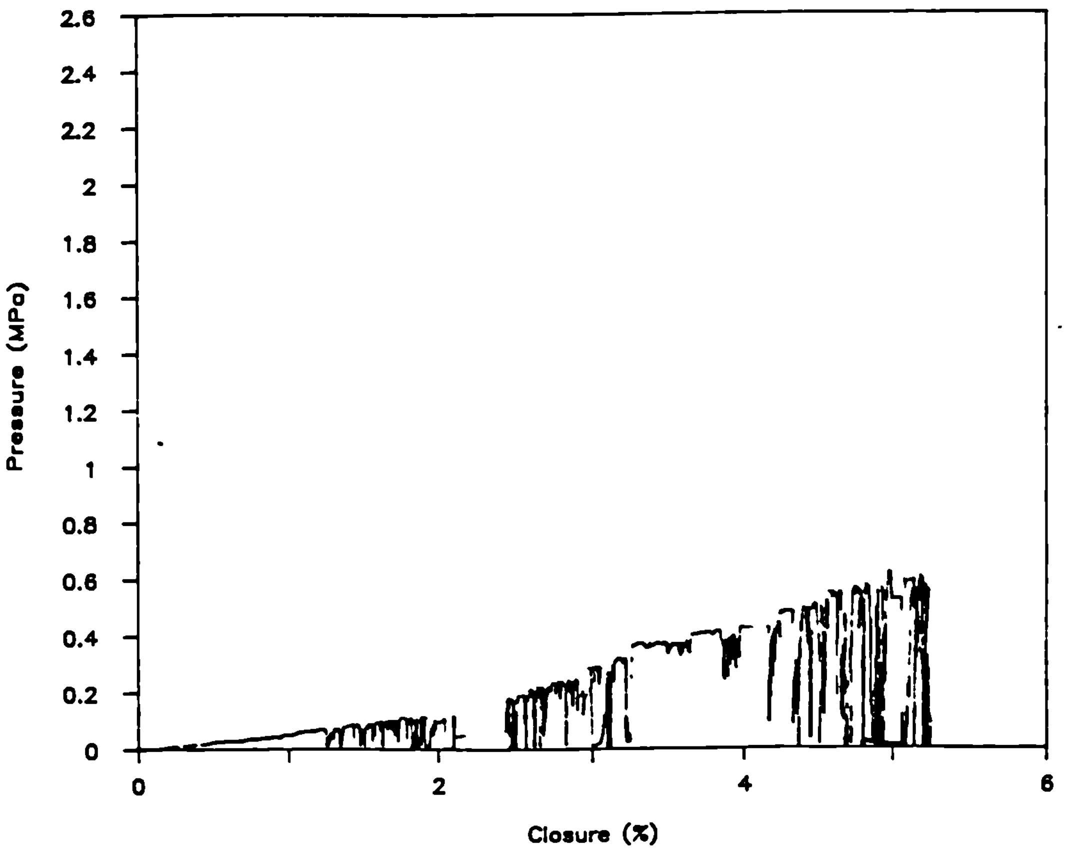


Figure 7b : Pressure evolution versus closure in borehole 5

STUDY OF THE THERMAL BEHAVIOUR OF CLAY-BASED BUFFER MATERIALS ON REDUCED SCALE MOCK-UPS AND IN AN UNDERGROUND LABORATORY

Contractor : CEA, CEN Fontenay-aux-Roses
Contract no : FI1W - 0061
Working period : July 1986 - December 1989
Period covered : January 1989 - December 1989

Project leader : M. DARDAINE

A. OBJECTIVES AND SCOPE

Clay materials could be used as components of engineered barriers placed between high level waste canisters and host rock.

The purpose of this work is to compare the behaviour of different types of materials, both homogeneous and heterogeneous, simultaneously subjected to heat and humidity gradients. Only the initial stage of storage, the so called "dried stage", is simulated.

The study, which includes instrumentation, first involves the design and construction of an experimental device to reproduce the actual physical conditions of waste disposal : temperature and water content. Subsequently, in collaboration with CEN/SCK, an in situ heat transfer experiment will be carried out in the Mol underground facility. An electrical heater will be surrounded with buffer and backfill materials. The entire system will be instrumented with temperature, moisture, and pressure sensors. The test will be supported by heat transfer modelling.

B. WORK PROGRAMME

B.1 Research and Development work on water content sensors :
- thermal conductivity sensor,
- capacitance sensor.

B.2 Design and construction of an experimental heat transfer device. Experiments and modelling.

B.3 Properties of backfill materials, determined in the underground experiment facility at Mol (Belgium). In situ experiments on heat transfer and modelling.

STATE OF PROGRESS AND OBTAINED RESULTS

State of progress

The water content determination of different engineered barrier candidates can be normally deduced from effective measurement of their thermal conductivity.

Complementary measurements at different temperatures with thermal shock probes have been carried out in laboratory, in order to compare them with the ones obtained in situ, as part of BACCHUS experiment.

The laboratory stüdy of an engineered barrier candidate, exclusively formed with French clay 4a, submitted to a thermal gradient has been completed by a close investigation of the material, after cooling down. (Water content profiles, cracking state).

Finally, most of the program consisted to follow up and exploit the BACCHUS experiment, set up at the end of 1988 in MOL underground laboratory, and whose thermal phase, which lasted 150 days, started on March 16th, 1989. This experiment is done jointly with CEN/SCK, which, more particularly, has equipped the close site with instrumentation, and taken into account the whole modelling aspect.

PROGRESS AND RESULTS

B.1 Research and Development work on water sensors.

Sensor behaviour, with respect to temperature, has been tested (endurance and measurements).
Height shock probes, identical to the ones set up in BACCHUS experiment, have been put and kept up to 120°C during three months. Values of heating resistor and of thermistor have been regularly checked. On the whole sensors, only one was damaged.

A compacted cylindrical block, with same characteristics as BACCHUS crowns (50 % 4a clay, 45 % sand, 5 % graphite) in the axis of which a resistor was introduced, has been radially instrumented with two shock probes. Conductivity measurements have been performed, from ambient temperature up to 100°C without operation malfunctions noted.

B.2 Design and construction of an experimental heat transfer device. Experiments and modelling.

The laboratory study of an engineered barrier candidate submitted to a thermal gradient ended in making a close investigation of the material, after stopping, in order to examine cracks and determine water content profiles (Figure 1, a and b).

These measurements, associated to pinpoint temperature and thermal conductivity measurements performed during the whole experiment duration, allow to result in the following conclusions :

- Hydrous transfers inside the material are slow. Water migrations from hot to cold part are hidden by an important evaporation phenomenon at heating mandrell contact.

So, in the area which is not disturbed by edge effects, thé final distribution of water content ranges within 5 and 7 % and the resulting shrinkage has for result to create large radial cracks, associated to a dry density variation.

- Taking into account the low water content variation to which is associated a density increase, it is not surprising to note no variations in thermal conductivity measurement with shock probes.

B.3 In situ experiments on heat transfer and modelling : BACCHUS experiment

Set up in MOL underground laboratory in November 1988, the BACCHUS experiment started on thermal phase on March 16th, 1989. For one week, the power supplied to the heating mandrell was 100 Watts, sufficient value to test the regulation and measurement acquisition assembly.

From March 23rd, it has been increased and kept at the constant value of 500 Watts until August 11th, where the accidental and definitive stop of heating circuit occured.

During the 90 first days, recorded temperature profiles, put on the same diagram, show discontinuity of experimental gradients, which can be due to existing initial voids at mandrell contact. Such a phenomenon was already observed on thermal bench. Furthermore, only the upper crown profile is different from the three other ones (Figure 2a).

The thermal conductivity, deduced from the curves, is about 2.5 W/m.K, value to be compared with the ones measured before thermal phase.

On the contrary, from about 100 days, a significant and continuous change is observed for gradients, up to an equilibrium (Figure 2b). They do not show any more discontinuities nearby the mandrell, and their slopes are noticeably lowered.

Corresponding thermal conductivity reaches 4 W/m.K, and is confirmed through effective measurement performed three weeks after thermal phase stop, with two sensors in normal operating condition.

To conclude, the following points can be drawn :

- Evolution of temperature profiles shows an improvement of contact between the heating mandrell and the engineered barrier candidate, and an important increase of its thermal conductivity as well.

- These effects may have for main origin the convergence of host ground, assorted of a possible rehydration of the material, and for consequence the breaking of connection between mandrell resistor and its power supply cable.

These elements justify the continuation of measurements in cooling down phase for some more time, and also the mock-up recovery in order to closely investigate it.

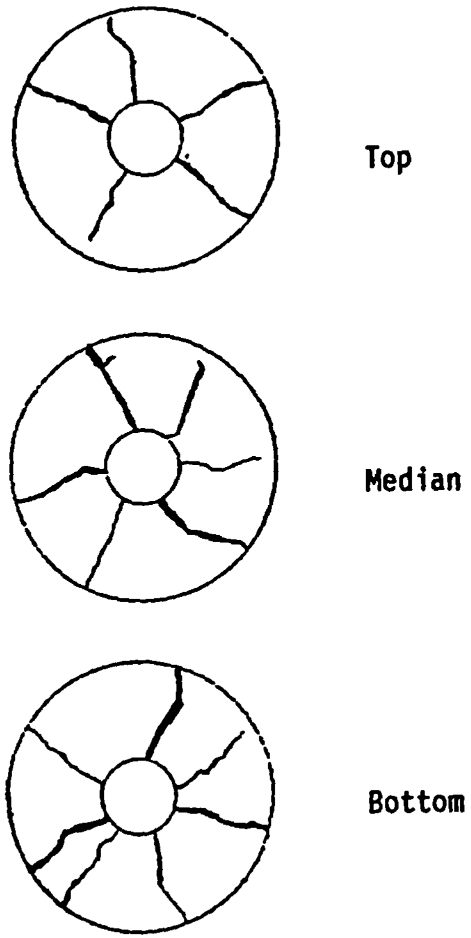


Fig. 1 a) Fissuration status of clay crowns observed after disassembling of thermal bench.

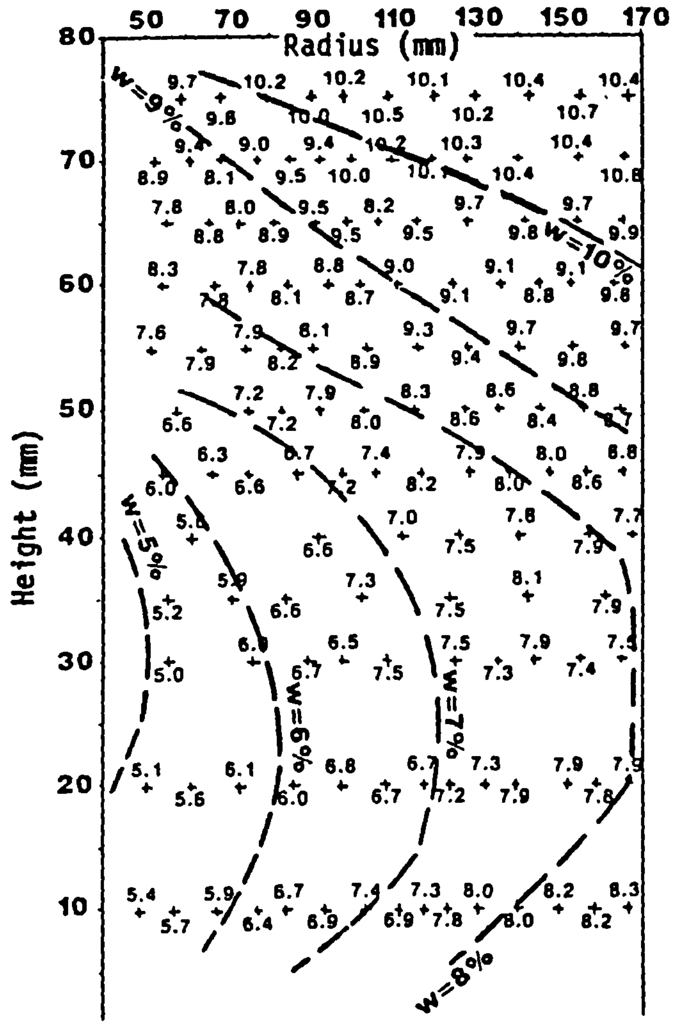
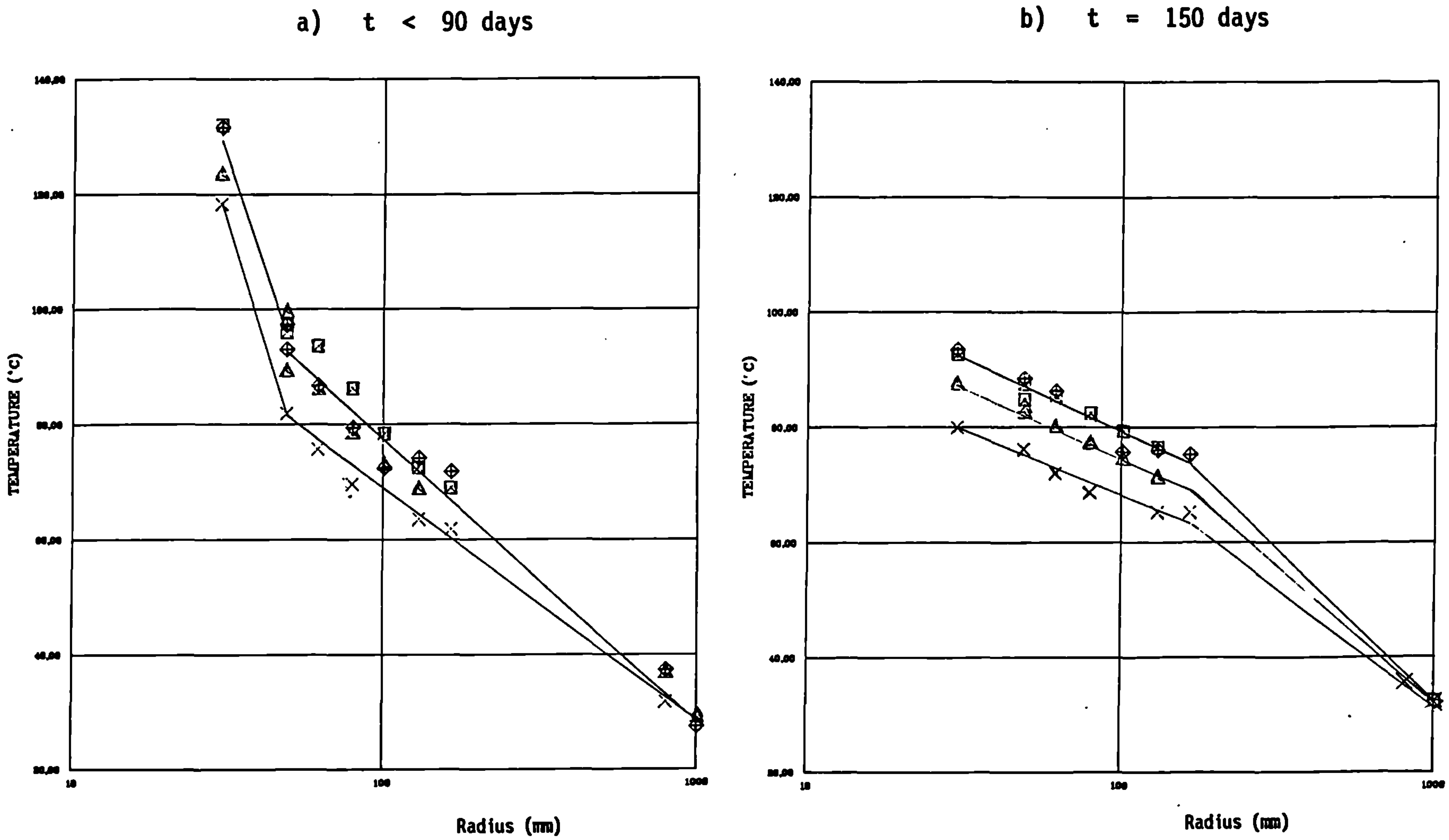


Fig. 1 b) Isovalue curves of water contents inside thermal bench, deduced from pinpoint measurements obtained on samples taken after disassembling.

Fig. 2 BACCHUS EXPERIMENT. Temperature profiles



DEVELOPMENT OF EFFECTIVE CONCEPTS FOR ATTENUATING THE NEAR-FIELD EFFECTS OF HLW IN ARGILLACEOUS ROCKS

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/00145-B
Duration of contract : from July 1987 through June 1990
Period covered : January 1989 - December 1989
Project Leaders : A.A. Bonne
Reported by : L. Noynaert

A. OBJECTIVE AND SCOPE

The aim of this study is to develop backfilling concepts with capabilities to attenuate near-field effects of mined repositories in argillaceous host rocks.

This study examines the characteristics and the performances of clay host rock and potentially favourable engineered barriers and assesses sytem components which contribute to their durability.

B. WORK PROGRAMME

1. Definition and quantification of near-field effects around HLW-packages in a clay environment for a limited set of selective representative emplacement configurations in mined repositories (in-gallery and in-floor concept).
2. Setting of disturbance allowances for the various components in the near-field (glass matrix, container material, concrete and in situ surrounding clay).
3. On the basis of the results of 1 and 2, definition of the attenuation factor to be achieved by engineered barriers with a particular attention to the thermal effects the radiation dose and the oxidation effects.
4. Definition of complementary requirements of there interface materials regarding their chemical compatibility their mechanical stability, their permeability, water saturation level and their emplacement feasibility.
5. Selection of appropriate materials on the basis of heat transfer and shielding properties, radiation resistance, hydraulic properties, chemical buffering, and radionuclide retention capabilities, mechanical characteristics, longevity, available methods of application and costs.
6. Design and dimensioning of a concept which meets the above set of requirements and establishment of appropriate emplacement techniques and procedures for the selected emplacement configuration.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Earlier work was devoted to the definition and quantification of near-field effects around HLW packages in clay environment and the setting of disturbances allowances for engineered barriers.

The study of the behaviour of the Boom clay under repository conditions was undertaken in 1989 focusing on the hydraulic field as well during digging works as after emplacement of HLW packages.

Progress and results

a. Hydraulic field disturbances around digging/drilling works

- observations made during the digging of the Test Drift, indicate that the radius of instantaneous influence can reach two times the excavated radius and more than 10 times at long term. The pressure drop in function of distances from the axis of the test drift is shown on figure 1 ;
- a program, called SOURCE, was developed to allow the computation in function of time and place of pore pressure changes around digging and drilling works. This program also allows to calculate the flow rate percolating through a filter and pressure drops at the surrounding filters. A rough validation test has been performed in the Boom clay. The best fit is actually obtained by using a permeability of 3.5 E-12 m/s and a consolidation coefficient of 15 m^2/y instead of 1 m^2/y which is the value adopted up to now (Fig. 2).

b. A program called TEMPPRES allows the following computations :

- temperature evolution around heat sources having the same center of gravity but with different dimensions and thermal power densities ;
- resulting pore pressures and stresses for plane stress and plane strain configurations.

Figure 3 shows the temperature and pore water pressure evolution in the near-field of a long pile of HLW after 50 years time.

List of publications

/1/ NOYNAERT, L., VOLCKAERT, G., Approaches for the backfilling and sealing of a radwaste repository in the Boom clay, NEA & CEC workshop on sealing of radioactive waste repositories, Braunschweig, May 22-26, 1989.

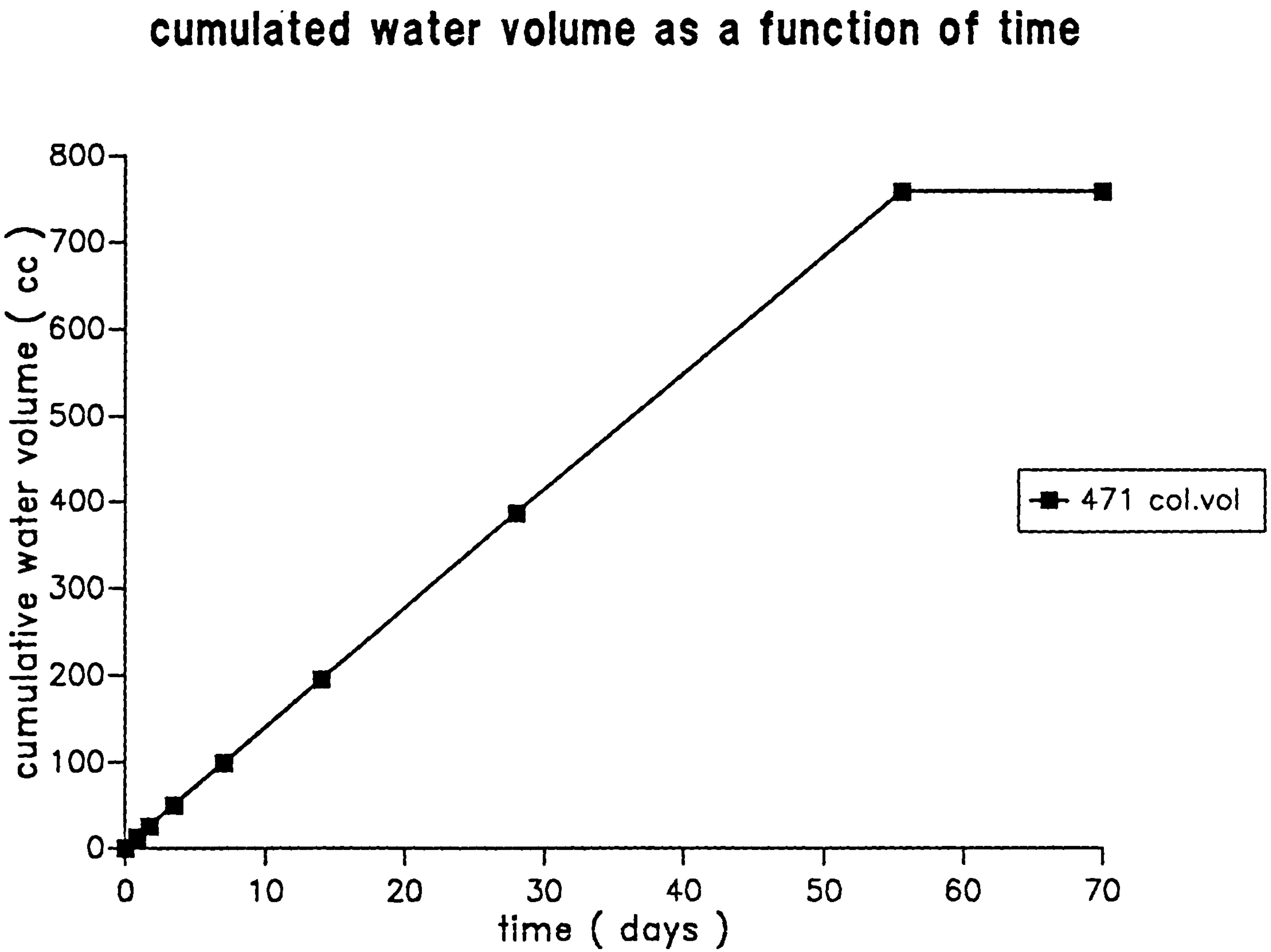


Figure 1.a. Collected water volume as a function of time computed by the program SOURCE

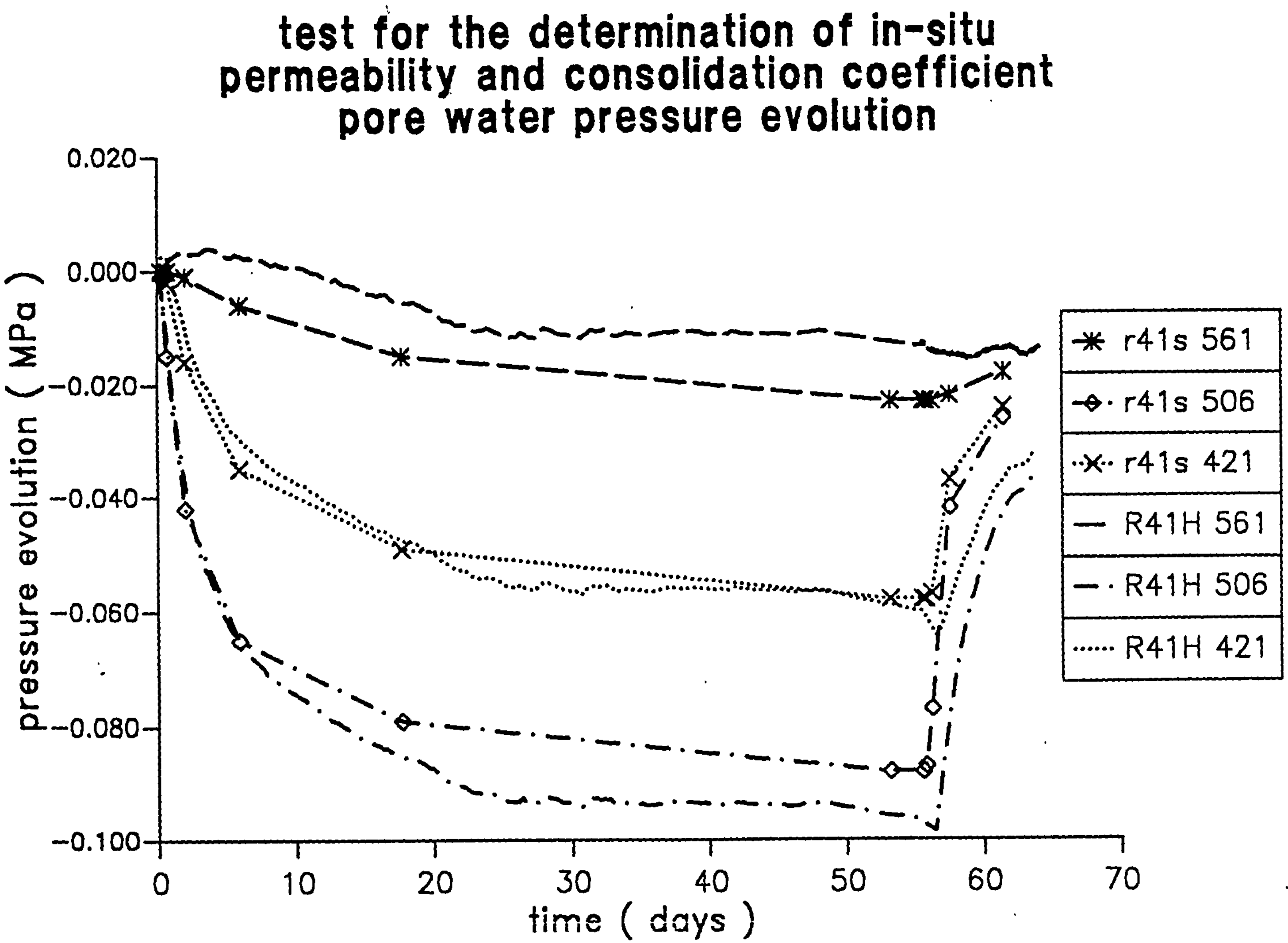


Fig. 1.b. Pore water pressure evolution at .35 m (screen 506), .50 m (screen 421) and .90 m (screen 561) from the screen 471.
Comparison between measured and computed values using the program SOURCE

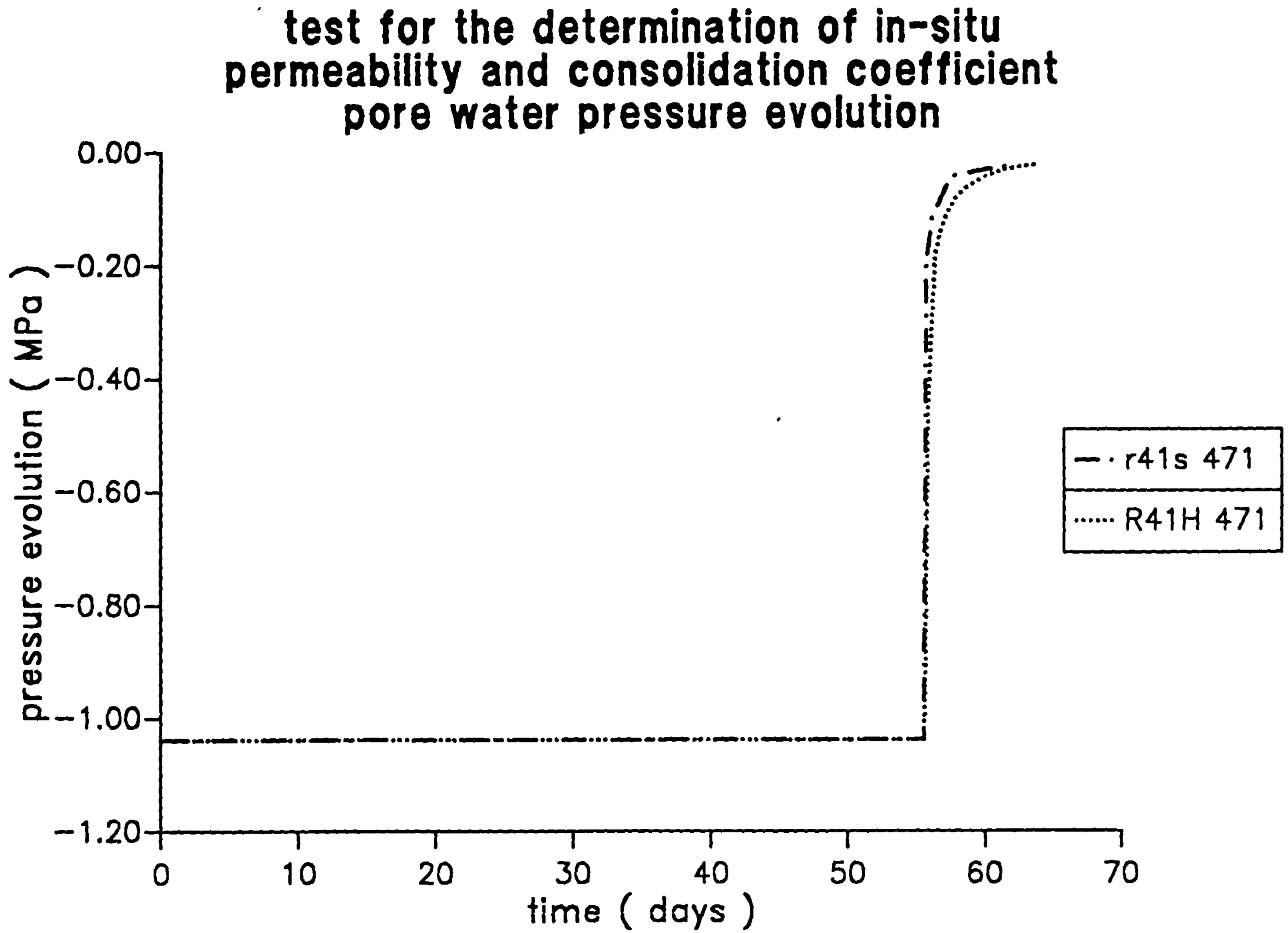


Fig. 1.c. Pore water pressure evolution at the screen 471 where flow rate was measured during 55.6 days
Comparison between measured and computed values using the program SOURCE

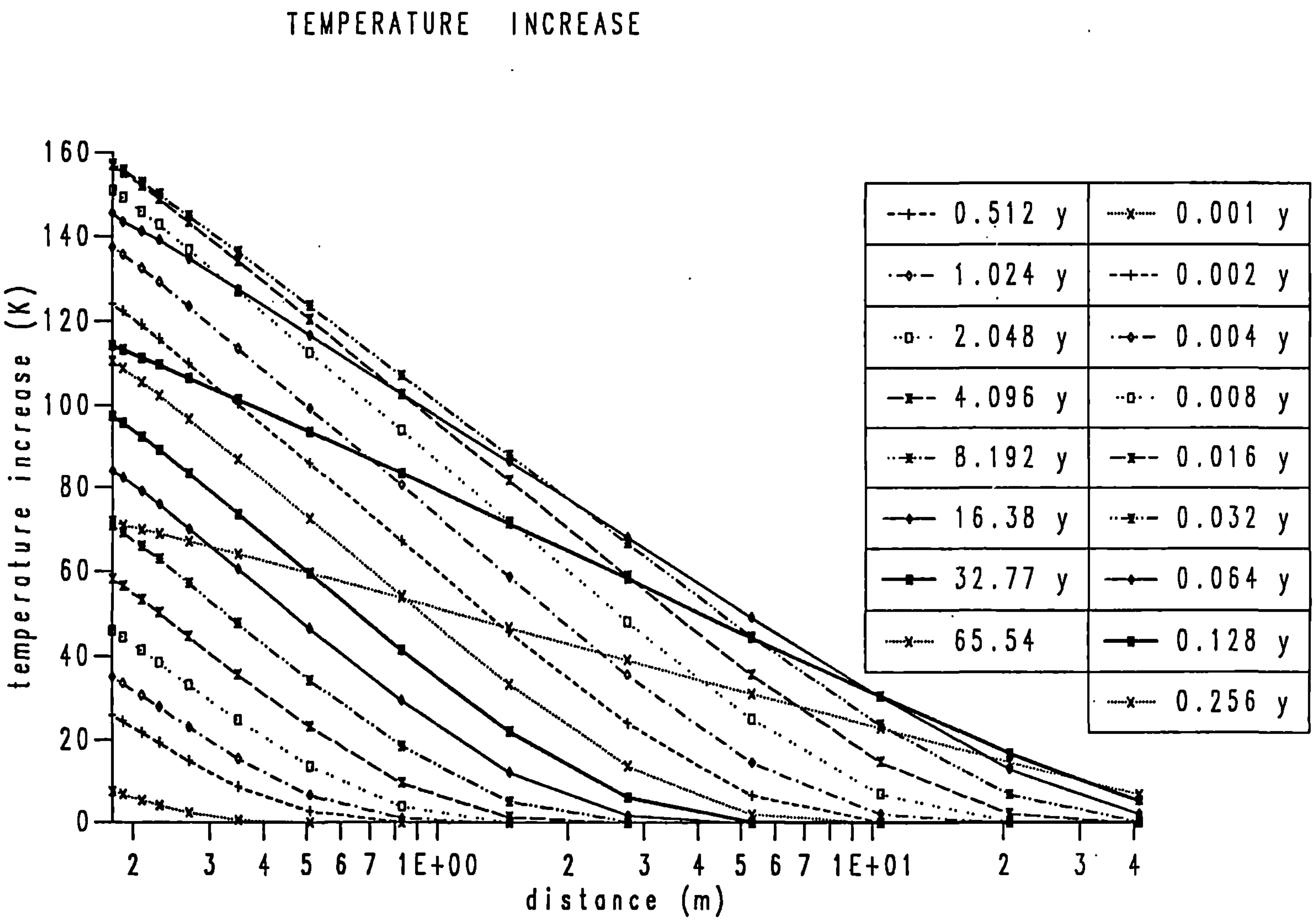


Figure 2. Temperature evolution around HLW stack computed by TEMPPRES program

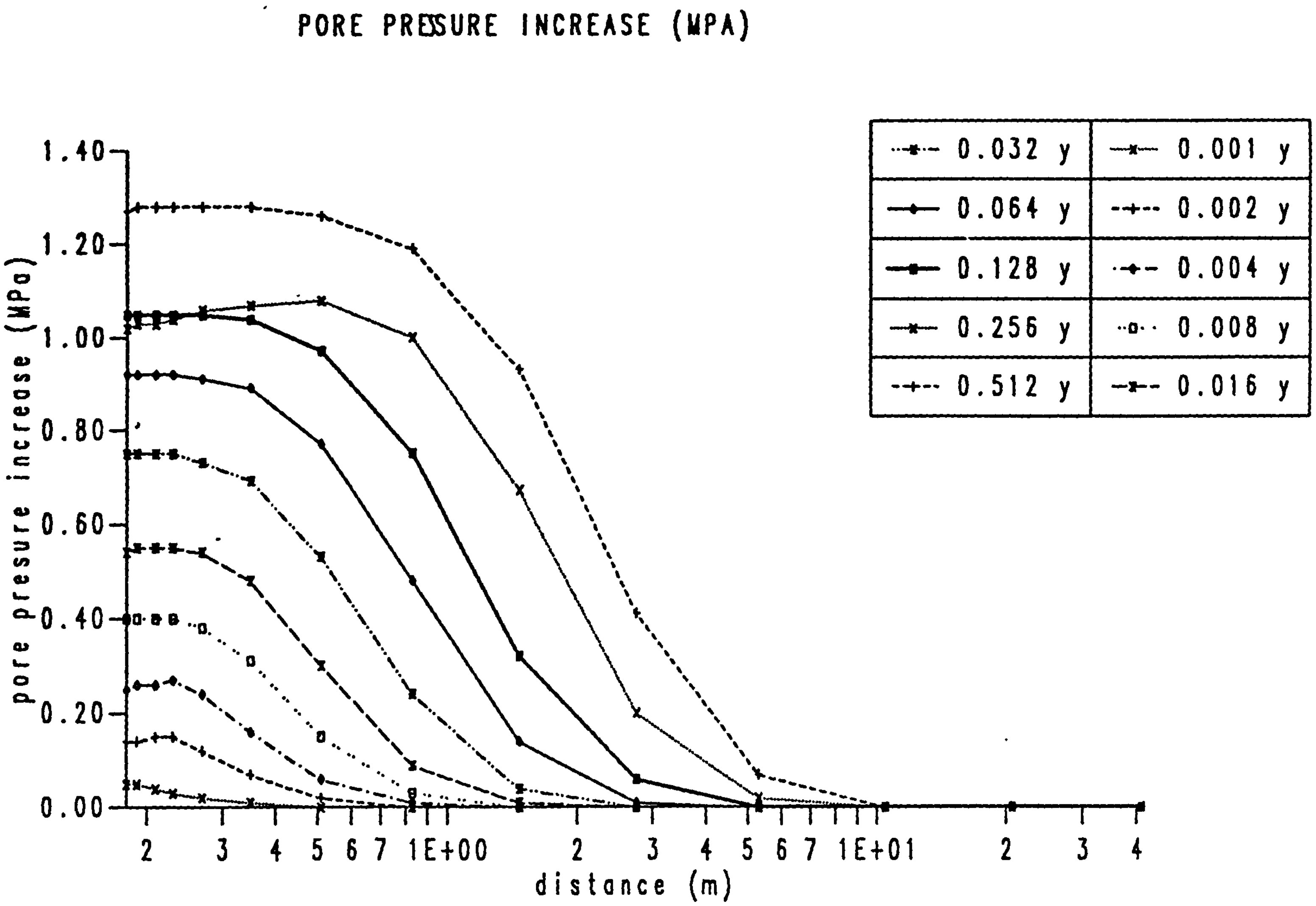


Figure 3.a. Pore pressure evolution around HLW stack computed by the TEMPPRES program

PORE PRESSURE INCREASE (MPA)

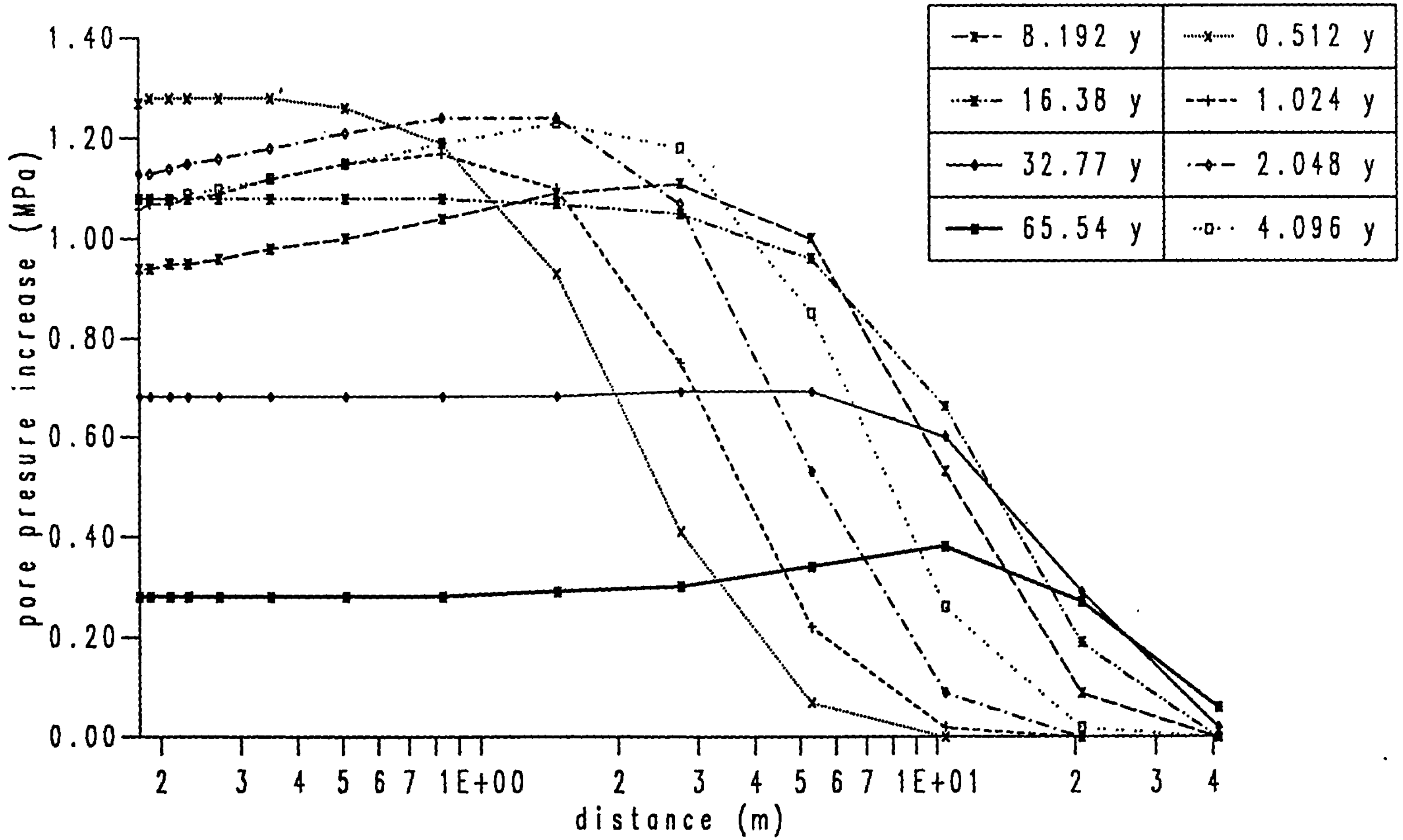


Figure 3.b. Pore pressure evolution around HLW stack comuted by the TEMPPRES program

TITLE: Studies of Historic Concrete
Contractor: Taylor Woodrow Construction Ltd
Contract No : FIIW/0158
Duration of Contract: January 1988 to June 1989
Period Covered: January 1988 to December 1988
Project Leader: Mr. T.P. Lees

A. OBJECTIVES AND SCOPE

Durability of concrete materials, either as matrix for low and intermediate level waste, or as backfilling/sealing materials for repositories, is a major concern in the preparation of both designs and safety assessments.
The objective of this study is to examine ancient concretes, and modern ones with proven durability; to identify the parameters which contribute to their durability. The work is an extension to that previously undertaken ('An Historical Examination of Concrete' (EUR 10 937) reported March 1986) in which a number of factors which appeared to be significant in relation to durability were identified. The results will be used to provide data to enable life predictions on modern concretes to be made.

B. WORK PROGRAMME

1. Sample retrieval

The following sources of samples will be investigated:

1.1 Ancient Materials
Preference will be given to obtaining samples from concretes containing natural pozzoloanic materials, where the sample is partially buried and from zones of the concrete where carbonation may be incomplete.

1.2 Medieval Materials (up to 1824)
Preference will be given to materials where a hydraulic binder is likely to have been used.

1.3 Modern Portland Cement Based Concretes (1824 - present)

Preference will be given to securing samples from large masses of underground concrete located in a moist environment and with a likely pozzolanic content.

2. Examination & Testing

The procedure set out below will be followed where appropriate, depending on the size and nature of the sample and the outcome of previous tests:

2.1 Visual examination
2.2 Assessment of depth of carbonation
2.3 Optical microscopy of thin sections taken at positions representing fully carbonated material, the carbonation front and uncarbonated material.
2.4 Scanning electron microscopy of polished sections and fracture surfaces in the same zones.
2.5 Individual phases identified above analysed by electron probe microanalysis.
2.6 Chemical and XRD analysis of portions representative of the different zones of the sample.

2.7 Pore structure analysis of similar portions.
2.8 Analysis of pore fluid expressed from the portions used in 2.7
2.9 Other tests may also be made to resolve issues arising from the results of the above procedure.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

All analytical studies have been completed, and a final report will be issued in the near future. Of the thirty-two samples of concrete obtained, four were chosen for extensive study and another ten for limited study. It was not possible to examine all samples obtained due to the confines of both budget and time.

PROGRESS AND RESULTS

1.0 Twenty seven ancient samples, five medieval samples and one modern sample were obtained.

2.1 Visual examination.
All samples have been visually assessed. In general materials were poorly compacted. Aggregates of two Roman samples from London contained fragments of clay tile, possibly included for their pozzalanic effect.

2.2 Assessment of Carbonation.
All ancient and medieval samples were fully carbonated, with the exception of several pockets of unreacted calcium hydroxide found in a sample from Hadrian's Wall.

2.3- Optical and Scanning Electron Microscopy.
2.4- Optical and S.E. microscopy of three Roman samples from
2.5 Vicus, near Rome, London and Hadrian's Wall, UK show evidence for containing, or having contained, calcium silicate hydrate gels similar to those found in modern concretes. A more modern, 18th century, sample from London was shown to contain calcium silicate hydrate gel very similar in appearance and composition to that found in modern concrete.

2.6 Chemical Analysis.
Twelve samples were analysed for calcium and silica contents and five of these were further analysed for sodium, potassium, barium, strontium, iron and aluminium. The results are currently under discussion. XRD analysis was not thought necessary.

2.7- Pore Analysis.
2.8 Samples were too porous to obtain meaningful data from this technique.

2.9 Other Analyses.
pH of the cementitious matrices was assessed. This was carried out because modern reinforced concrete has to maintain an alkaline environment (pH = 11-12) as slight increases in acidity can be deletrious to the steel reinforcement. Results indicate that with the exception of the relatively modern sample, alkalinity levels were less than desired for modern reinforced concretes. These pH levels are consistent with extensive carbonation. Results are given in Table I.

Table I Assessment of pH of the Cementitious Matrix

Site	Sample	Approx Date	pH
Thomas More Street, London.	Warehouse Foundation	Earlier than 1824	10.4
Roman Baths, London.	Concrete Floor	100-300	8.6
Roman Baths, London.	Wall Mortar	100-300	7.8
Hadrian's Wall.	Mortar Infill - surface	c.120	8.9
Hadrian's Wall	Mortar Infill - centre	c.120	9.2
Carnuntum Ampitheatre, Austria	Wall Mortar	100-300	8.6
Tor Paterno Villa, near Rome.	Wall Mortar	100-300	9.0
Vicus, near Rome.	Foundation Concrete	c.200	8.9
Vicus, near Rome.	Waterproof bath lining	c.200	9.2
Vicus, near Rome.	Concrete core of wall.	900-1100	8.7
Vicus, near Rome.	Foundation Concrete	900-1100	9.2
Anguillara, near Rome	Concrete core of vaulting	140-160	8.8
Anguillara, near Rome	Concrete wall Foundations	140-160	8.5
Anguillara, near Rome	Concrete Wall	140-160	8.9

The Development and Application of Mathematical Modelling Approaches to Interactive Effects for Concrete Backfill in Hard Rock and Argillaceous Hosts

Contractor: Taylor Woodrow Construction Limited (UK)
Contract No: FI.1W/0159
Working Period: February 1988 - 31st March 1990
Project Leader: P. Dawson

A. OBJECTIVES AND SCOPE

The research study relates to the possible use of concrete as a backfilling or sealing material in a radioactive waste repository. The main aim of the work is to develop a model for the creep and shrinkage behaviour of concrete applicable to both early age response to thermal loads and long term effects arising from host material in-situ stresses. Such a creep model would complement the ultimate load model for concrete developed, in part, within a previous study (Report EUR 10383 EN/II).

The intention is to devise a finite element based concrete model capable of predicting with reasonable confidence a wide range of load, temperature and time dependent effects.

The research has made use of the Adina finite element package as a workbench in which to build suitable models for concrete behaviours.

The research is co-funded by the UK Department of the Environment and Taylor Woodrow Construction.

B. WORK PROGRAMME

1. Literature Review - ongoing as appropriate.

2. Review of Adina Features and test runs - completed.

3. Development of creep model - substantially developed but to be tested.

4. Modelling of host ground (argillaceous and hard rock) - not yet commenced.

5. Draft report - to be commenced February 1990.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

A literature review of the subject matter covering the early age and creep behaviour of concrete identified a key theory first promulgated by Illston /1/ in the 1970's and supported in large measure by recent published texts /2/. The total strain in a loaded concrete specimen, is assumed to comprise additive components covering elastic strain, thermal strain, delayed elastic strain, viscous flow strain and thermal transient strain. The latter three components together constitute concrete creep strain which (with the exception of delayed elastic strain) are irrecoverable on unloading.

The research has taken into account the above creep strain components proposed by Illston but has also included age at loading and temperature change effects, both of which complicate creep in concrete.

It is believed that the model being developed will represent a significant advance in the present capability of analysing concrete performance and will be particularly beneficial by virtue of its incorporation within a commercially available finite element package.

Progress and Results

Commencement of the contract was delayed until April 1989 because agreement to a funding contribution was not initially forthcoming from the UK Department of the Environment.

A major product of the study is to be a material model for creep in concrete within the Adina finite element code. An early activity was to assess the characteristics of the creep models existing in the commercially available package. It became evident that the current models were only suited to metals and were inappropriate to concrete except for specimens under constant or monotonically increasing load. There was no provision for creep recovery under reduced load which is an observable occurrence with concrete.

Detailed literature review resulted in identification of a creep theory proposed by Illston /1/, work commonly referenced by recent documents /2/,/4/. Illston identified several creep strain components:-

(i) A viscous or creep flow which is a function of time and temperature and has no limiting value. This creep strain component is non-recoverable on reduction of load.

(ii) Visco-elastic or delayed elastic flow which is a function of age, time under load and temperature. This component is recovered gradually over time when the load is removed or partially if the load is only reduced. The term elastic indicates the recoverable nature of this part of total strain.

(iii) The third creep component is transient thermal creep. This again is non-recoverable. Evidence suggests that creep accelerates sharply with an increase in temperature provided the new temperature is in excess of that previously attained.

These, together with age and temperature effects on the elastic modulus, have been introduced into the Adina code and are to be tested against reported laboratory experiments /5/,/6/.

Suitable creep data to define the necessary material properties are sparse. The CEB report /7/does contain graphs and expressions for some of the necessary creep terms, although not in the precise form required for the Illston method. The relationships have been converted to suit, however, and the CEB document therefore provides a potential method for deriving material data for general concretes as opposed to only that specific mix used by Illston.

Future Work

The study is at the $^3/_4$ stage of completion and there remains the task of applying the creep model to backfill and sealing test scenarios for concrete. The complexity of the creep of concrete, and of the model therefore, has meant that a greater proportion of study time has been committed to the concrete model than originally anticipated. As a result, the derivation of a detailed model in Adina for Argillaceous and hard rock hosts will necessarily be curtailed.

Nevertheless, studies of concrete in a simplified repository environment will be carried out. These will by typified by a 5m diameter tunnel or shaft at about 1km depth. The tunnel will be assumed backfilled by concrete consistent with high strength. An appropriate heat output per unit volume of concrete from the hydration process will be assumed. Mechanical boundary conditions to the backfill will correspond to a very stiff host medium (an extreme granite) and to a free boundary (a very soft clay). These boundary conditions represent the extremes. It is hoped to analyse an intermediate case of a tunnel in soft clay with a support liner which is then backfilled with concrete. Analyses will include sample arrangements of canisters of HGW radioactive materials.

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5. Ross A.D., Creep of Concrete Under Variable Stress, ACI March 1958

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RESEARCH ON SWELLING CLAYS AND BITUMEN AS SEALING MATERIALS FOR UNDERGROUND REPOSITORIES FOR RADIOACTIVE WASTE

CONTRACTOR	:	Bullen and Partners Consulting Engineers 185 London Road Croydon CR9 1PT England
CONTRACT	:	FI 1W - 0161 - UK
DURATION OF CONTRACT	:	From February 1988 to February 1990
PERIOD COVERED	:	February 1989 to January 1990
PROJECT LEADER	:	Dr JA Allison

A OBJECTIVES AND SCOPE

Work previously carried out within the CEC's research and development programme indicates that swelling clay together with bitumen could be used to form a highly effective waste containment barrier for use in deep underground radioactive waste repositories.

This project seeks to identify relevant material properties and sealing mechanisms. Its objectives are :

- o To assess the potential behaviour of combinations of swelling clays and bitumen, and the potential effectiveness of synergistic combinations of these materials for sealing underground repositories against groundwater ingress, radionuclide release and gas release.

- o To assess the potential level of confidence in the long-term behaviour of such seals in the perspective of the quality assurance procedures that could be associated with the emplacement of backfill and seal materials.

B WORK PROGRAMME

The work programme consists of the following activities :

1 A review of available information on properties and behaviour of swelling clays (including bentonite and magnesium oxide) and information on bitumens as used in engineering structures.

2 Examination of the relevant properties of the materials and of combinations of the materials to establish whether seals are formed, and the nature of the seals.

3 Assessment of test results in the context of material properties and free swelling space that could be achieved in practice with current quality control systems in potential repositories.

C PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

The literature review was completed earlier in the year, but is being updated to incorporate more recent information.

Four sets of laboratory test rigs have been in operation for most of the year. Testing is substantially complete, and the final composite sealing test is in progress.

Confined swelling tests on compacted bentonite were completed early in the year, and bentonite intrusion testing is also complete. Difficulties experienced in casting well-formed bitumen specimens were overcome using facilities at Birmingham University. Creep testing of bitumen specimens containing imperfections has recently been completed.

Final analysis of the results and report writing is in progress.

PROGRESS AND RESULTS

1 Testing of Component Materials

Bentonite - Fully confined swelling tests have generated swelling pressures up to 40 MPa. Several relationships have been examined. These were carried out in the specially designed apparatus illustrated in Figure 1. Control testing has indicated the following :

- Calcium Bentonite is found to give much lower swelling pressures than the Sodium Bentonite.

- The swelling pressure is found to be related to the final (equilibrium) dry density as shown in Figure 2.

- The irrigation route (top, base or both) and the back pressure applied, have no effect on the swelling pressure, although irrigation from both ends and application of pressure reduce the time taken to reach equilibrium.

- Allowing limited free-swell of the bentonite reduces the final dry density and so reduces the equilibrium swelling pressure.

Tests have also been carried out to examine the extrusion properties of compacted bentonite. The bentonite was allowed to extrude into a hole within an adjacent perspect cylinder. A maximum extrusion of 41mm was obtained and the effect of variations in hole area and bentonite density have been examined.

Creep tests on bitumen samples with 'imperfections' in the form of triangular or rectangular slots, have been carried out. The apparatus used incorporates a Bellofram jack for application of constant load to laterally confined specimens. Closure was identified by passing water through the slot and measuring the flow rate. Variables examined were pressure applied, length of specimen, type of slot, and time for closure.

Gas permeability tests on bitumen have been carried out at Imperial College, London, on 38mm diameter samples. The results show no measurable permeability in the majority of cases, but inconclusive results were obtained on some grades. Time restraints have prevented further testing.

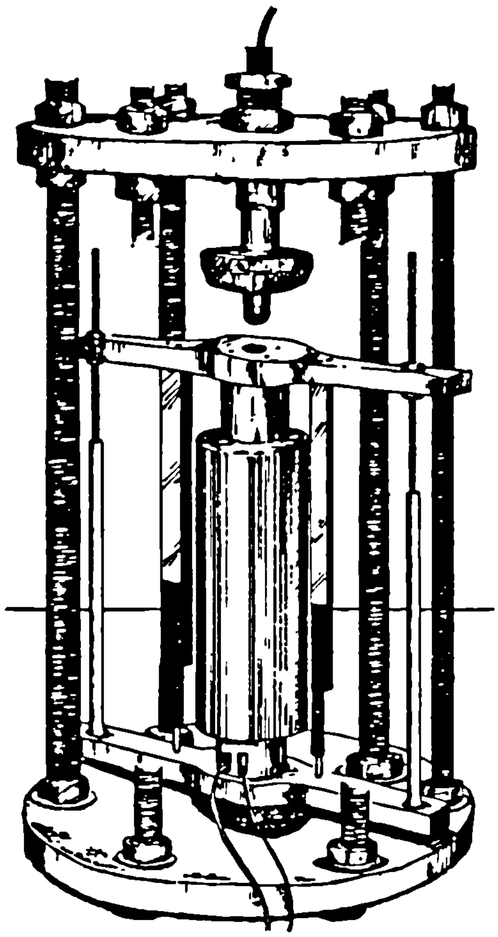
2 Composite Sealing Tests

Seven composite bentonite/bitumen sealing tests have been completed. These tests took up to 70 days for completion and this has led to a smaller number of tests than originally envisaged.

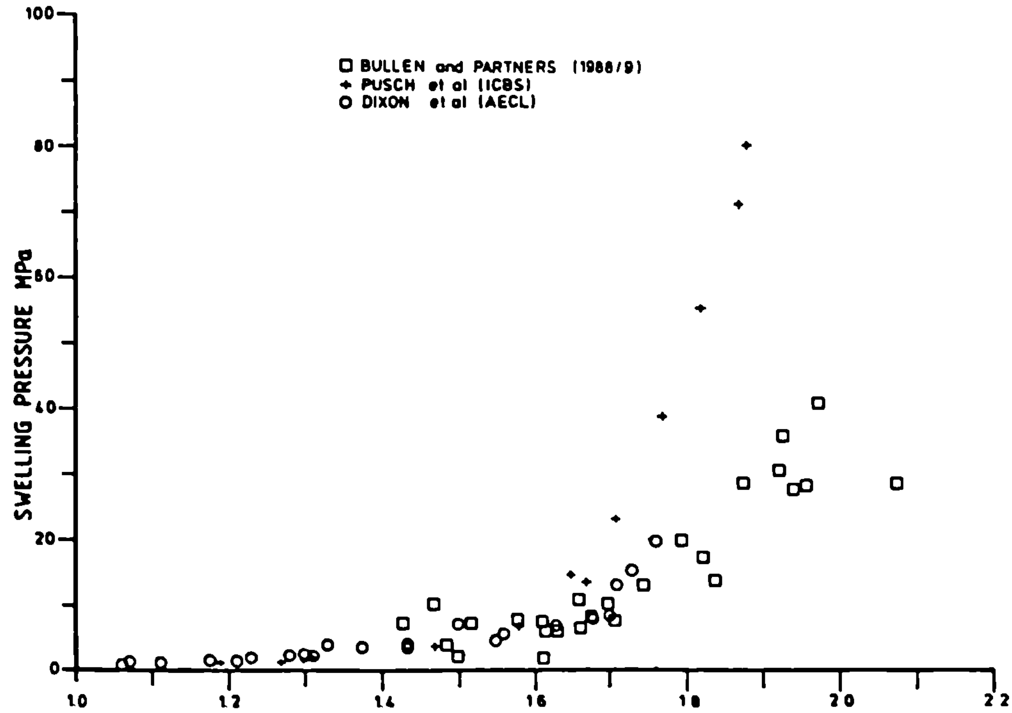
The test results are thought to provide an adequate basis for the laboratory-scale validation of the composite sealing concept.

Publications

Allison JA, Mawditt JM, Marsden JF, Wilson J
Development of an Engineered Backfilling and Sealing Concept Incorporating Swelling Clay and Bitumen. Proc NEA/CEC Workshop on Sealing of Radioactive Waste Repositories, OECD, Paris 1989.



APPARATUS FOR SWELLING AND COMPOSITE SEALING TESTS **FIG.1.**



RELATIONSHIP BETWEEN SWELLING PRESSURE AND EQUILIBRIUM DRY DENSITY **FIG.2.**

QUALITY ASSURANCE ASPECTS OF WASTE EMPLACEMENT AND BACKFILLING IN ILW AND LLW REPOSITORIES

CONTRACTOR	Bullen and Partners Consulting Engineers 185 London Road Croydon CR9 1PT England
CONTRACT	FI 1W - 0162 - UK
DURATION OF CONTRACT	From February 1988 to February 1990
PERIOD COVERED	February 1989 to January 1990
PROJECT LEADER	Dr JA Allison

A OBJECTIVES AND SCOPE

The objectives of the study are :

- To critically examine existing conceptual design proposals for the deep underground disposal of ILW and LLW in water-bearing (non-saliferous) host rocks, with particular reference to the quality control/quality assurance aspects of waste emplacement and backfilling.
- To examine the extent to which existing proposals enable effective monitoring and remedial action to be achieved.
- To identify the parameters which are most effective in describing backfill material properties and waste unit characteristics, both before and after placement.
- To identify appropriate measurement techniques and the means of application of such measurements in the development of a coherent quality control system.
- To examine the ways in which repository design details, methods of waste unit/backfill placement and the sequence of operations involved may be adapted to ensure that an effective quality assurance system can be established.

B WORK PROGRAMME

1 **Phase 1** comprises a brief review of the following aspects of the ILW and LLW disposal concepts developed in member states of the European Community :

- The types of waste units envisaged for deep underground disposal and the quality control/quality assurance systems currently adopted or envisaged for the pre-disposal stages.

- The waste emplacement systems currently proposed and the extent to which quality control/quality assurance procedures have been specified.

- The range of backfill materials under consideration, and the properties which relate most effectively to their intended functions.

2 **Phase 2** concentrates upon the generic deep-level repository design concepts for ILW and LLW disposal (including plutonium-contaminated wastes) which are incorporated within the current UK waste disposal strategy.

Consideration will be given to a range of backfill materials, reflecting the scope of current community research and development.

For these reference conditions, more detailed evaluation of quality control/quality assurance measures will be carried out, itemising the procedures and measurements required at each stage of development.

Where appropriate, the need for repository design modifications is to be considered as a means of improving the exercise of quality control in the waste emplacement/ backfilling processes.

The following are among the factors to be considered :

- The extent to which backfill materials can be prepared in solid 'fill block' form or as pre-batched 'fluid form' fills at off-site production centres, or at an adjacent surface facility.

- The scope for palletising waste units in appropriate spatial arrays, with partial interstitial filling, prior to emplacement and final sealing.

- The scope for exercising control over the geometry of waste unit arrays such that required full volumes are accurately known and directly comparable with volumes actually emplaced.

- Means of separate monitoring for groups or 'cells' of emplaced waste units/backfill, such that the origin of defects may be identified, and recovery or appropriate remedial action achieved.

- The extent to which the emplacement processes for different categories of waste (incorporated in a single repository) may require different approaches in exercising a consistent level of control.

3 The study will include recommendations concerning the development of quality control/quality assurance procedures for waste emplacement, backfilling and monitoring to complement and extend those which are being (or have been) developed in relation to the pre-disposal stages. Supplementary recommendations concerning the correlation of readily measured quality control parameters and specified performance properties will be provided.

C PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

Phase 1 - A literature study has been undertaken covering the aspects outlined in Phase 1. The following points are noted :

- o Several different types and sizes of waste unit have been identified and the quality control/quality assurance aspects of their production ascertained according to country of origin.
- o More detailed information on international proposals for waste emplacement systems has been obtained and enquiries in this line are now essentially complete.
- o Backfill materials, which are taken to include overpacks, buffering and repository linings in addition to the mass infill materials, are widely reported. Mass infill materials are found to be site specific and vary according to host rock type, groundwater regime and depth of burial. Phase 1 has now been essentially finished. Work has commenced on Phase 2.

Phase 2 - Work is in hand to clarify the factors most relevant to the implementation of effective quality control during waste emplacement and backfilling. Where appropriate, repository design measures which improve the scope for exercising quality control will be identified.

PROGRESS AND RESULTS

Waste Emplacement Systems

The waste emplacement systems proposed internationally are linked to site specific or host rock specific conceptual repository designs. As a result, the proposals of each country have been examined individually.
Quality control and quality assurance considerations relevant to waste emplacement and backfilling include :

- o ground stabilising measures
- o preparation of engineered backfill materials
- o preparation of disposal chambers prior to waste emplacement
- o preparation of waste units prior to disposal
- o placement of waste units
- o sealing of disposal chambers
- o sealing of access-ways and shafts
- o monitoring performance of completed construction
- o contingency plans for retrieval of waste units.

Reports defining specific quality control/quality assurance measures associated with the above aspects are generally lacking in detail.

Backfilling Materials

Repository backfill materials identified by various countries vary widely in composition, and include :

- o reconstituted spoil
- o cementitious-based material
- o clay-based material
- o compressed bentonite (pelletised or in block-form)
- o bitumen
- o zeolites
- o magnesium oxide

The prime purposes of the backfill are to :

- o inhibit the development of preferential groundwater flow paths
- o buffer the pH/Eh regime of the repository near-field environment
- o provide physico-chemical retention of radionuclides
- o control mechanical interaction between the geological host and the enclosed waste
- o control the dissolution of any gases generated within the vaults
- o provide physico-chemical stability to the projected geochemical environment of the repository.
- o provide a heat conduction medium in the vicinity of certain categories of waste.

The choice of backfilling material depends on a variety of factors, including :

- o geological host formation
- o hydrogeological features including groundwater perturbations during the construction and closure of the repository
- o layout of the access-ways, shafts and vaults of the repository
- o emplacement arrangements of the waste within the repository
- o radionuclide inventory and its time-related impact on the geochemical environment
- o heat generation characteristics of the wastes
- o swelling and consolidation characteristics.

Hence, backfill materials must be designed to cover a variety of purposes leading to the development of 'mix designs' pertinent to their function within a repository. 'Mix designs' are site specific and many countries (eg France, Belgium, Sweden, Switzerland, US and Canada) are now employing the use of underground research laboratories within potential host rock formations. One of their purposes is to obtain information on the desired characteristics of the backfill material and to evaluate placement techniques: for example, injection with or without pressure, air entrainment, gravity filling, rolling, vibration, etc.

The civil and mining engineering industries have wide experience in placing materials to achieve specific performance characteristics. However, further development work is required in order to formulate quality control systems capable of meeting the more stringent requirements of repository backfilling and sealing.

A quality assurance programme in a repository backfilling operation must include :

- o controls on the production of the backfill components, eg chemical composition, particle shape and size
- o environmental controls on the storage of the individual components prior to mixing
- o controls on mixing operations prior to emplacement
- o controls to be exercised during the emplacement operation
- o monitoring after completion of the emplacement operation.

Effective quality control procedures are likely to be based on index measurements which correlate with relevant engineering performance characteristics, e.g. particle size distribution, density, moisture content, porosity, void ratio, viscosity, etc. It is thought that simple mass-balance relationships will also provide a particularly useful means of regulation.

EMPLACEMENT FEASABILITY OF OPTIMIZED AIR PLACED MORTARS

Contractor : CEA, Fontenay-aux-Roses, FRANCE
Contract n° : FI 1W/0166
Duration of contract : October 1987 - December 1989
Period covered : January 1989 - December 1989
Project leaders : A. BERNARD, R. ATABEK

A. OBJECTIVES AND SCOPE

Air placed techniques - Gunite and Shotcrete - are commonly used in civil engineering for wall reinforcement with cement based materials. Gunite is a trade name to designate a mixture of PORTLAND cement and sand thoroughly mixed dry, passed through a cement gun and conveyed by air through a flexible tube, hydrated at a nozzle at the end of such flexible tube and deposited by air pressure. In the case of shotcrete, a proportioned combination of PORTLAND cement, aggregates and water is mixed by mechanical methods and pumped in a plastic state to the nozzle where air is added to expel the material.

These techniques are likely to be used for engineered barrier emplacement due to the facts that :

- spraying machines are commercially available and easily automatized for nuclear applications,
- their delivery ($\cong$ 10 m^3/h) is compatible with the french needs of gallery filling up.

The research programme, developed within the framework of this contract, is devoted to test air placed mortar ability to fill up the voids between the waste packages and the host rock. Materials and techniques will be optimized taking into account air placed mortar properties such as density, permeability, water transfer, radionuclide retention, etc...

B. WORK PROGRAMME

2.1 Literature survey: choice between the two processes (dry or wet); recommendations for the selection of the most appropriate equipment; definition of the test specifications.

2.2 Feasability tests: choice between different types of materials, taking into account cement types, additives (clays, silica fume), plasticizers, aggregate granulometry distribution.

2.3 Full scale study of the selected material: spraying cycle definition, rebound influence on air placed material homogeneity.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The literature survey allows the selection of the dry process technique and of two air placed machine manufacturers : ALIVA S.A and MEYCO. Feasability tests have been performed with different types of materials: mortars, cement-salt mixtures, as well as swelling clay and clay-sand mixtures. A full scale experiment is planned in the near future using an optimized mortar made of slag and fly ash cement (French reference: CLC 45).

Progress and results

2.1 Literature survey

The air placed techniques differ from one another in the stage at which water is introduced ; the mixture could be conveyed in dry or wet state. Taking into account the emplacement constraints (distance : 300 m, flow : 10 m^3/h) and the different types of materials (hydraulic binders as well as swelling clays), the dry process is selected as the most appropriate technique, considering the characteristics given in Table I. The main disavantage of this process is the production of dust, which may be prevented by material moistening before mixing. The inventory of the air placed machine manufacturers, performed by SOLETANCHE, heads to the choice of ALIVA S.A and MEYCO equipments.

2.2 Feasability tests

2.2.1 In the case of cement-based materials, six different types of mortar are studied, made respectively of Portland cement (OPC), slag and fly ash cement (CLC) and blast furnace slag cement (CLK), with and without silica fume. For each test, roughly 500 l of dry materials are needed, with the standard following composition:

Sand (0/4 mm)	≅ 1000 kg
Cement	≅ 300 kg
Silica fume	≅ 0 or 15 kg

MEYCO equipment with a rotor machine of 12 cells, a 50 mm in diameter and 20 m in length pipe and a compressor of 25 m^3/mn were used for these first feasability tests. The different mixtures were deposit into 150 l boxes (A.F.T.E.S type) specially designed to allow the core-sampling of four cylinders, 11 cm in diameter and 22 cm high. The nozzle flow was estimated between 1 to 2 $m^3/hour$.

The main properties of the air-placed mortars are evaluated. The mechanical strength measurements often reveal the presence of composition heterogeneity. Despite of these defects, the material nitrogen permeability appears to be low ($\simeq 1.3 \times 10^{-19}$ m^2) and its total porosity - using mercury intrusion technique - (figure 1) is roughly the same as the one of standard mortars (water/cement = 0.5, same curing conditions). There is no big difference in properties between the studied mortars. Salt aggregates lead to decrease the mechanical strength and to increase the total porosity of the air placed material.

2.2.2 In the case of pure swelling clays, the feasability tests are performed using 150 to 300 kg of material; the study is carried out on the french reference clay 4a, as a function of clay granulometry distribution and initial water content. A clay-sand mixture of 20 % sand is selected for preliminary experiments.

The ability of air placed technique to put in place pure clay is clearly demonstrated: a dry density of 1.50 kg/m^3 is reached in the case of coarse material, prepared at an initial water content in the range of 9 to 12 % (in weight). The final water content of the air placed material is relatively high, about 30 % (in weight).

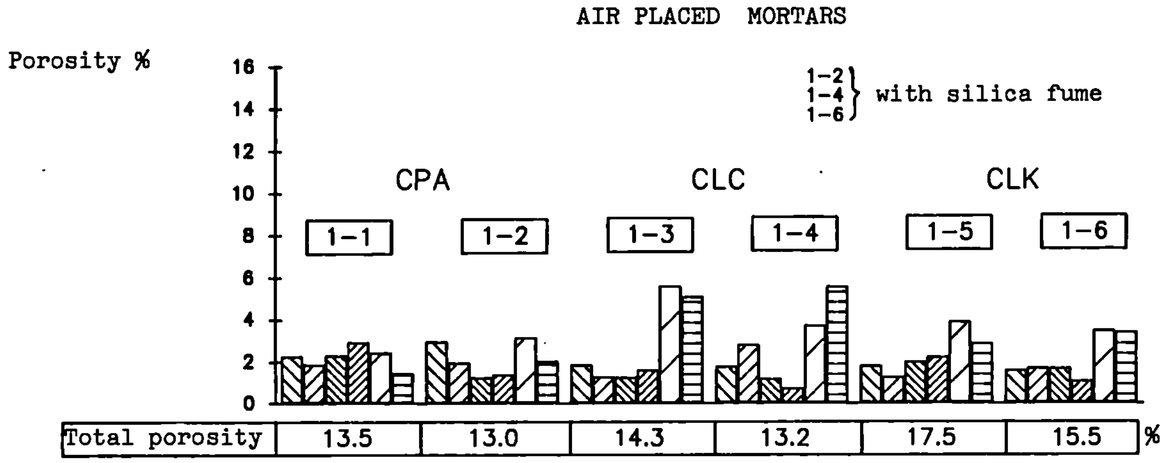
The two feasability tests, carried out on clay-sand mixture, leads to a plugging up of the nozzle, due to an unappropriate granulometry distribution of the sand.

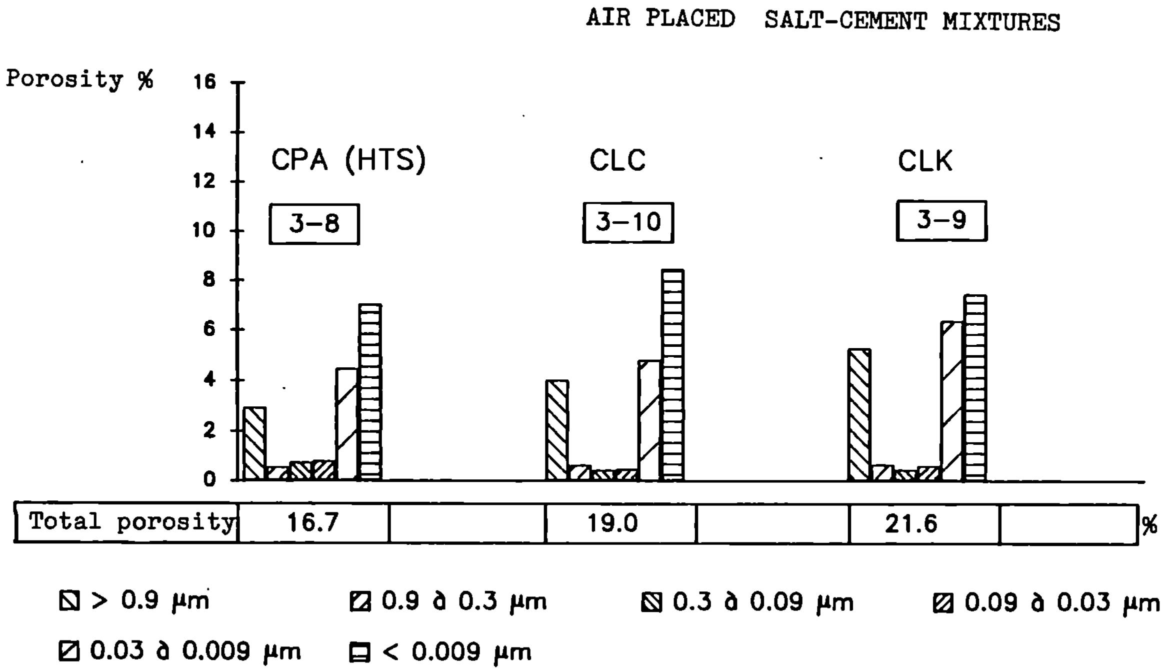
TABLE I : CHARACTERISTICS OF THE AIR PLACED TECHNIQUES

	Dry process	Wet process Diluted flow	Wet process Dense flow
Transport distance	***	o	*
Delivery	***	o	***
Compaction	***	**	**
Dust	o (*)	**	**
Maintenance Cleaning	***	o	o

o Pass ** Fairly good
* Average *** Good

FIGURE 1 : Air placed cement based materials: porosity and pore size distribution measurements (mercury intrusion technique) curing conditions: 90 days, sealed bag





UNDERGROUND LABORATORY VALIDATION OF VITRIFIED WASTE DISPOSAL IN A GRANITIC MEDIUM

Contractor : CEA, Fontenay-aux-Roses, FRANCE
Contract n° : FI 1W/0207
Duration of contract : August 1988 - March 1990
Period covered : January 1989 - February 1990
Project leader : M. JORDA

A. OBJECTIVES AND SCOPE

The objective of this research is the underground validation of the plugging back of a vitrified waste disposal pit in a granitic medium. The scenario selected, which is similar to that adopted in other studies carried out by the European Community, involves the stacking of twenty vitrified packages in a pit 30 m deep and 1 m in diameter.

The engineered barrier between the waste and the granite is composed of clay material, in the form of high density, compacted elements. Gaps are required to enable the lowering of the packages and of the barrier into the pit, but these gaps have the effect of reducing the density of the material introduced into the pit.

The uncertainty existing with regard to the definition of the minimum size of the residual gaps required for satisfactory installation can only be removed by performing an in-situ test. A demonstration pit will therefore be made in the mining region of Fanay-Silord, using state-of-the-art drilling technology, and then the pit will be plugged with the engineered barrier and the simulated glass containers.

B. WORK PROGRAMME

2.1 - Sinking of two demonstration pits, 1 m in diameter and 30 m deep, using the raise boring technique (one of the pits will be used as a reserve)

2.2 - Detailed characterization of the two pits bored, with verticality check and checks for surface condition, circularity faults and diameter variations

2.3 - Plugging back of the characterized pit using baskets filled with blocks of compacted clay, followed by testing of the installation of the glass containers.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of advancement

The barrier installation studies which form the basis of this research require the boring of a demonstration pit of 30 m depth minimum. Work authorisations have been granted by the local authorities of the mining region of Fanay and boring work proper started in April 1989. Detailed characterization of the two pits were performed, leading to the determination of the maximal diameter for the basket passing through.

Improvements in manufacturing techniques for the uniaxial compacting of bricks of the type which make up the barrier should enable an increase in the pit filling rate. Twenty eight baskets containing the engineered barrier were fabricated and lowered in the pit, without any difficulty, in February 1990. The density of the barrier after emplacement is under evaluation.

Progress and results

2.1 Boring of the demonstration pit

The tests are carried out at the mining center of Fanay, near Razes in the Haute Vienne Department (Silord Site). The site is separate from the mine, with accesses via galleries a) to the upper level and b) to the lower level 40 m below the upper level.

The upper level where the CEA work will take place, is enlarged and arranged so as to enable:

- the boring of two holes of a diameter of 1 m. The holes are spaced 4 m apart, center line to center line,
- about ten visitors to be received in order to monitor the installation tests under acceptable conditions,
- storage of about 20 dummy containers without hindering the free passage of personnel, the containers being 0.43 m in diameter and 1 m high approx,
- installation, over the holes, of a 3 tonne winch for the baskets and the containers,
- performance of the installation tests.

Primary aeration of the work gallery is provided by a 1 m diameter shaft leading to the surface. The distance between the surface (entrance to the shaft) and the test gallery (upper level) is 250 m.

The boring of the two pits is performed in two steps, using the Raise Boring technique (ECOFORE machine):

- drilling of a pilot vertical hole (23 cm in diameter) between the upper gallery and the lower gallery,
- boring of the pit it-self, starting from the lower level with cutting removal from the lower gallery.

The boring were carried out at Fanay-Silord in May 1989 with a drilling rate of 0.8 m/hour.

2.2 Detailed characterization of the two pits

The detailed characterization of the two pits allows the knowledge of the following geometrical parameters and observations:

- the two pits are almost circular, with a mean diameter of 99.3 cm and 98.3 cm respectively for pit 1A and pit 2B,

- their vertical deviations are not very important and of the order of magnitude of 0.4°,

- the rugosity, attributed to the cutting wheels of the boring machine, is regular and homogeneous all along the granite walls,

- pit 2B is less fissured than pit 1A.

The previous results lead to the selection of pit 1A for the faisability test it-self. A maximal diameter of 96.03 cm is calculated for the basket passing through (basket height = 1.335 m) taking into account the local deviations measured as a function of the pit depth. The lowering of basket gauges in the pit allows to precisely define the dimensions of the mould used for clay block compaction. An external diameter of 95.0 cm is required for a good emplacement of the barrier in the pit.

2.3 Back plugging of the pit - fabrication of the compacted clay blocks

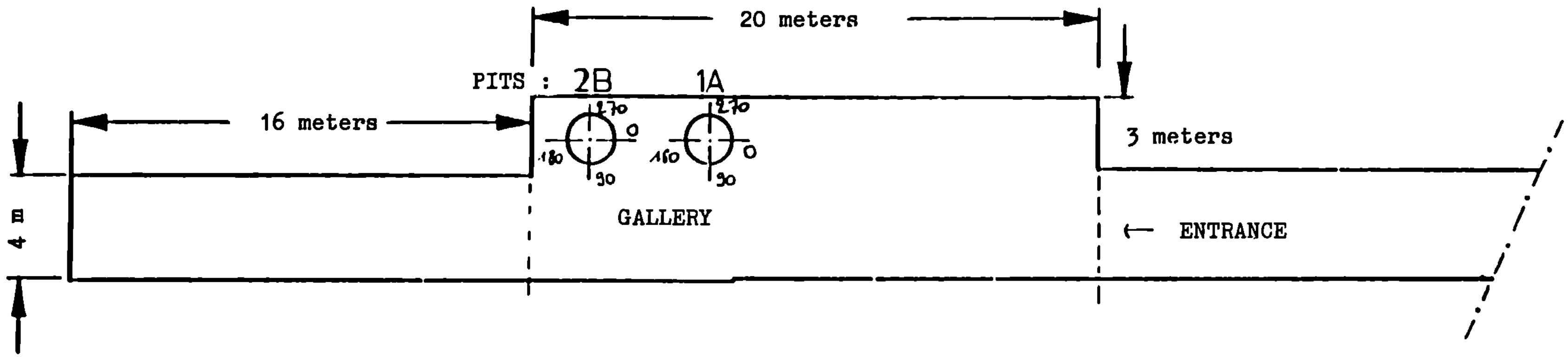
The compacted blocks are made from powdered material (of optimised particle size) and form the barriers which are installed using baskets. The technique used is compacting by uniaxial pressing. An assembly of six bricks forms a crown piece which does not require any further finishing work. Design of the mould used and of the compacting techniques have been improved, with:

1) Modification of the rate of descent of the upper piston and de-aeration of the powder
2) Height check of the pressed parts
3) Improvement of the mould release techniques.

The fabrication of the 28 baskets containing roughly 39 tons of clay material as well as their installation in pit 1A were carried out, in February 1990, without any difficulty. The exact dry density of the barrier reached after emplacement is under evaluation.

FIGURE 1   LOCATION OF THE PITS AND AXES NOTATION

LONGITUDINAL SECTION



FIELD TEST FOR THE DEMONSTRATION OF THE EMPLACEMENT FEASIBILITY OF CLAY BUFFER MATERIALS AS ENGINEERED BARRIERS IN CRYSTALLINE FORMATIONS

Contractor: ENRESA, Madrid, Spain
Contract No.: FI1W/0231
Duration of contract: December 1988-November 1990
Period covered: January 1989-December 1989
Project Leader: Julio Astudillo

A. OBJECTIVES AND SCOPE

the main purpose of the present study is to demonstrate the adequacy of the spanish bentonite materials, and the feasibility of emplacement of clay buffer as an engineered barrier in accordance with the present spanish conceptual design for deep geological disposal in crystalline rocks. This demonstration will be made in co-operation with CEA at the Fanay-Augères uranium mine in France.

Some natural spanish smectitic clays have unique characteristics for the purpose under discussion. Smectitic clays with high aluminium content (montmorillonitic type bentonite) are abundant in Cabo de Gata, Almería. Smectitic clays with high magnesium content (saponitic type bentonite) are abundant in the Madrid Basin. Reserves of these deposits are of the order of several million of cubic meters.

A present spanish disposal concept calls for the placement of the nuclear waste canisters in 7.5 m deep vertical boreholes of 1.5 m diameter drilled in galleries excavated in a granitic formation. The annular space between the canister and the borehole wall is to be filled with an adequate buffer material for which candidate materials are spanish natural clays.

In this project, six ore clay deposits will be studied, and two will be selected and tested as candidate.

B. WORK PROGRAMME

1. Study of the properties of the candidate material: Study of six bentonite deposits and selection of two clay types. Detailed characterization (geochemical, mechanical, and thermomechanical) and long term stability analysis. Study of clay-sand mixture to be used in the experiment.
2. Development of the industrial fabrication process for the buffer material: design of a brickcast. Design of the packing technique. Fabrication of approximately 100 bricks.
3. Field validation of the buffer concept: To develop and characterize the mechanics of the buffer materials emplacement, and to analyze the results. Tests will take place at the Fanay-Augères uranium mine. A new chamber, of 20m long by 7m wide and 4,5 high in which the test boreholes will be drilled, will be excavated.
   The construction and characterization of the disposal borehole (excavation, techniques, plugging technique and characterization of the roughness, slabbing, overbreaks on the disposal boreholes).
4. Analysis of results: Data evaluation, determination of possible techniques factor monitoring and selection and implementation of these techniques.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

During the last 1989, three bentonitic deposits in the Madrid Basin (Cerro del Aguila, Santa Bárbara and Yuncos) and three in the Cabo de Gata area (Serrata de Níjar, Los Trancos and Los Escullos) have been studied. On the basis of the data obtained, the saponites of Cerro del Aguila deposit (Toledo) and the montmorillonites of Serrata de Níjar (Almería) were selected as candidate clay.

Similar results were obtained by CEA, on the analysis of 12 samples (two per each ore deposit).

A detailed characterization (geomechanical and physico-chemical) has been concluded for the two clays selected.

In parallel, preliminary stability studies have been performed to define the analytical procedures for the final longevity studies.

A chamber for the spanish experimentation has been excavated in Fanay Silord and a study of the drilling alternatives to the rise-boring system is going on. The brick cast has already been designed.

Progress and results

1. Study of candidate material properties

1.1. Mineralogical and geochemical characterization

a) Madrid Basin:

Mineralogy: The study of the Cerro del Aguila bentonites shows a very high phyllosilicate content (93-100%). In the less than 2 m fraction the smectite content is 58-100% (x=82.5), sepiolite appears in 0-37% (x=8.2) and illite is present in 0-16% (x=9.7). The Biscaye index varies between 0.76 and 0.86, with a mean value of 0.81. There is a significant increase in the sepiolite content (reaching 72%) in the pink layers associated with the typical green bentonites of this deposits whereas illite is scarcy. The smetitie is trioctahedral (d(000)= 1.53A) and saponitic.

Phyllosilicat content for the Santa Bárbara bentonite (pink bentonite) ranges between 90-100%, with quartz and calcite traces in some samples. A wide XR diffraction band is observed between 9.6-20A in the less than 2 m fraction. the reflection corresponds to a trioctahedral smectite (d(000 = 1.52A) which belongs to an irregular kerolite-stevensite interstratification which has recently been described.

The Yuncos bentonites (brown bentonites) show a 90% phyllosilicate content and up to 10% content to detrital materials (quartz and feldspars. Smectite content in the less than 2 m fraction reaches 76%, with secondary quantities of illite (15%) and sepiolite (9%). The smectite is mainly trioctahedral (d(000) = 1.53A) although the presence of a little band at 1.50A suggests the existence os some dioctahedral ones. The Biscaye index is 0.77.

Chemical analysis: The Cerro del Aguila bentonites are characterized by a remarkable content of SiO_2 (48.5-52.0%) and MgO (13.3-25.5%), which are the most important components of the saponitic bentonites. Significant Al_2O_3, Fe_2O_3, TiO_2 and K_2O contents indicate an important contribution of detrital origin materials. Low carbonate, chloride and organic carbon content is characteristic of these bentonites.

The Santa Bárbara bentonites are characterized by a high MgO content (24.5-28.5%) and a lower incidence of detrital origin elements than in the green bentonites, which a higher content in a carbon and free silica.

The Yuncos bentonites show characteristics similar to those of the green bentonites, but with higher K O, CaO and Al O content. Low percentages of carbonate (0.10%), chlorides (0.01%), sulphates (0.05%) and organic carbon (0.04%) have been observed.

Physico-chemical properties: The CEC values in the Cerro del Aguila bentonites vary from 73.5 to 99 meq/100g with a mean value of 85.7meq/100g

It is important to mention the high values of the specific surface (316-385 m /gr) and liquid limit (129-237) measured in the green bentonites. The analytical data are presented in Table I to III.

Conclusions

Based upon the experimental results it can be concluded that for buffer candidate materials the Cerro del Aguila bentonites have the best characteristics of the three deposits selected in the Madrid Basin. Also, field data available indicates that the potential reserves and an easy to mine configuration of the deposits meet foreseen requirements.

b) Cabo de Gata bentonites (Almería)

. Mineralogy:

The smectite content is very high (>80%). Quartz and plagioclase are primary minerals, inherited from pyroclastic materials, whereas cristobalite and calcite are secondary minerals formed during and after the hydrothermal alteration process. In the fine fractions, cristobalite only appears as an accessory component; the content of this disordered mineral is high, specially in the Escullos samples.

From the X-Ray patterns, the crystalline size of smectites along the c-axis was deduced through the Scherre equation. In samples treated with sodium carbonate, these sized decrease some units, indicating the existance of small silica bonds between crystals. The Biscaye's index is in all cases higher han 0.8, value typical of hydrothermal smectites.

. Chemical composition:

The data obtained are in agreement with the mineralogical results and coincide with previous data from the same deposits. This indicates a great homogeneity in the chemical composition within deposits, since some of the previous sampling was carried out more than 25 years ago.

To deduce the structural formulae of these smectites, the amorphous silica content and the cation exchange capacity were determined. The silica content is low, except in sample E-6. CEC values are very high and in good agreement with the smectite content. The order of abundance of exchangeable cations is: Mg and Ca>Na>>K.

the structure was obtained for the smectites. It can be observed that the Los Trancos samples are beidellites, since their tetrahedral charge is higher than the octahedral one. In other cases, the smectites are montmorillonites.

. Physico-chemical properties:
Hygroscopicity and specific surface were analyzed.

The values obtained must be considered as normal ones if the smectite content of samples is taken into account. The values inferred for specific surface are in agreement with the theoretical value of 800 m /g accepted for pure smectite. It can be concluded that the water uptake by these bentonites is not seriously affected by the presence of amorphous silica.

. Conclusions:
The Serrata de Níjar Bentonite have the best characteristics for the buffer candidate material on the Almería area.

1.2. Mechanical and hydraulic properties

For the two selected clays, in parallel with the goechemical and mineralogical characterization, the mechanical properties were obtained. The parameters measured were: Natural humidity, molturation granulometry, specific weight and Atteberg limits, specific surface, uniaxial stress, swelling pressure, and hydraulic conductivity on function of dry density.

In Table III results are presented. On the basis of these data the Serrata smectite is selected as the main candidate clay for the buffer barrier. The Cerro del Aguila bentonite will be used as a second potential material.

1.3. Stability analysis

Preliminary studies for the Almería bentonite have been concluded. In these experiments, different ClK solutions and temperature range have been selected.

The selection of the K solution concentrations was done so as to be close to the ionic strenght of rivers, waters infiltrated through several rocks and sea water.

After reaction, pH values decrease with K molality, whereas K fixation increases. On the other hand, the silica content in the equilibrium solution is practically constant (Table VIII). The high K fixation at high electrolyte content indicates probably incipient neoformation of some illitic layers. This is corroborate by XRD data.

Although the diffraction patterns will be studied more rigorously, the Shultz's method offers very useful results. The ratio peak intensities between the (001) basal spacings of samples solvated with ethyleneglycol and those heated at 300ºC is a good measure of the percentage of illite layers. This ratio is near to 4.7 for pure smectite and goes down to zero when the number of illite layers increases. The decrease in this ratio can actually be observed in Table VIII.

. The results of the stability test in k-rich media, carried out at 175ºC during 21 days, point out the incipient formation of illite layers. Further studies are advised on this respect.

For the Madrid Basin bentonites the preliminary results are ongoing at this moment.

Activities in relation with tasks 2, 3 and 4 are going on in accordance with the chronogram.

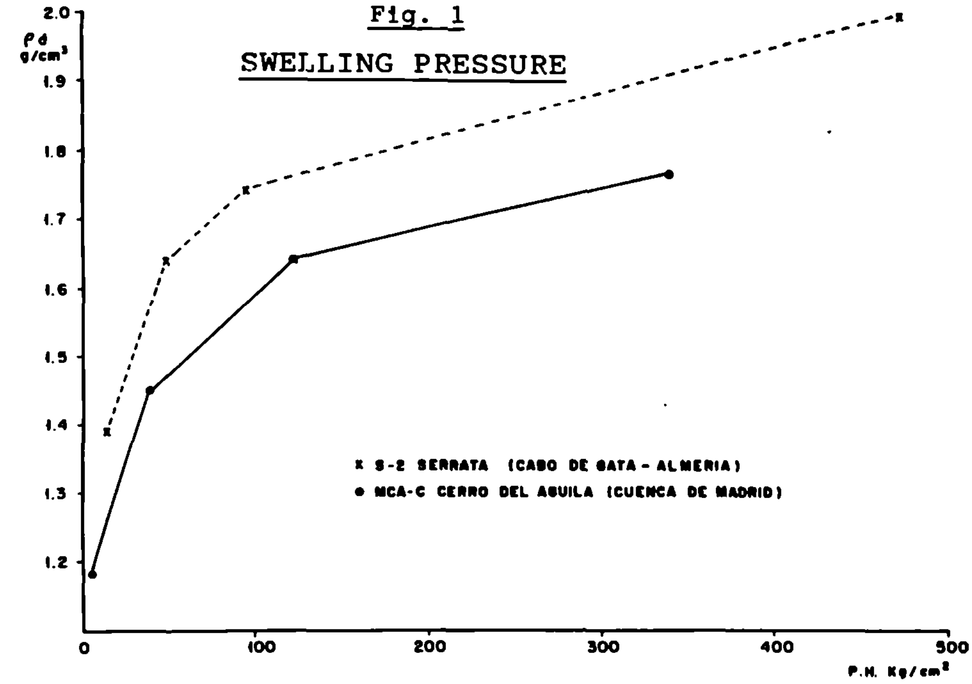


Fig. 1

SWELLING PRESSURE

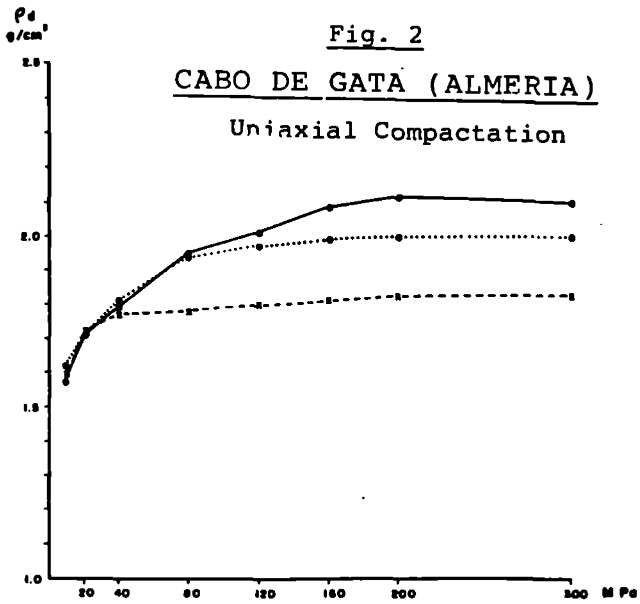


Fig. 2

CABO DE GATA (ALMERIA)

Uniaxial Compactation

o ≃ 9.5 % Humidity
o ≃ 15.0 % Humidity
x ≃ 20.0 % Humidity

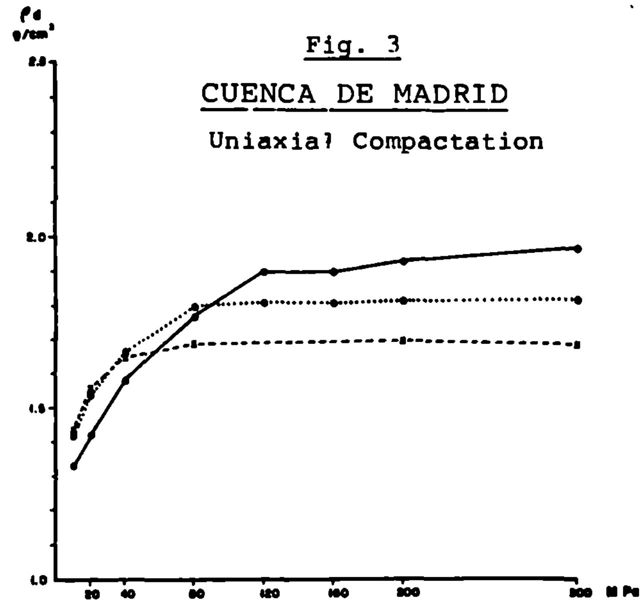


Fig. 3

CUENCA DE MADRID

Uniaxial Compactation

o ≃ 6.5 % Humidity
o ≃ 15.0 % Humidity
x ≃ 20.0 % Humidity

TABLE I: Chemical and Mineralogical composition of selected clay (Bulk samples)

PARAMETERS	A	B
Sand (%)	6,06	1,0
Lime (%)	4,67	12,7
Clay (%)	88,76	86,3
Smectite (%)	91,6	97
Quartz (%)	1,35	5
Plugioclase (%)	2,70	5
Crystobalite	2,20	-
Calcite	1,50	-
Biotite	,65	-
SiO_2 (%)	59,94	48,74
Al_2O_3 (%)	18,76	12,05
Fe_2O_3 (%)	3,60	4,98
CaO (%)	2,03	0,76
MgO (%)	5,68	13,20
Na_2O (%)	2,24	0,76
K_2O (%)	0,74	2,21
CO_2 (%)	0,72	0,1
H_2O^+ (%)	6,6	-
SiO_2 (amorphous)	1,92	0,135
Fe_2O_3 (amorphous)	0,11	0,02
Al_2O_3 (amorphous)	0,09	-
EXCHANGE CAPACITY		
Na (meq/100g)	30.	3,5
K	2,5	2,5
Ca	30,9	35,0
Mg	33,7	55,0
CEC	91,7	96

TABLE II: Structural Formulae

	A	B
Si(IV)	7,78	7,26
Al(IV)	,22	,74
Al(VI)	2,79	1,47
Fe(VI)	,33	,49
Mg(VI)	1,03	2,87

A: Serrata de Nijar bentonite (Almeria)
B: Cerro del Aguila Bentonite (Toledo)

TABLE III: Mechanical Properties

	A	B
Natural Humidity(%) (110ºC) 24 hours	8,69	22,68
Specific Weight (g/cm^3)	2,47	2,98
Liquid Limit (%h)	212,5	188,23
Plastic Limit (%h)	56,57	69,99
Plasticity index	155,93	118,24
Specific surface(m^2/g)	515,69	336,4
Uniaxial Stress(1051kg/cm^2) Humidity 10%) g/cm3	2,26	2,19
Swelling Pressure Kg/cm^2	W% g/cm SP 6,9 1,74 95	W% g/cm SP 6,8 1,69 121

4.3. RADIONUCLIDE MIGRATION IN THE GEOSPHERE (MIRAGE)

4.3.A. Actinide and fission product geochemistry in natural acquifer systems

CHARACTERISATION OF THE BOOM CLAY AND ITS MULTI-LAYERED HYDROGEOLOGICAL ENVIRONMENT WITH A VIEW TO RADIONUCLIDE MIGRATION

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0055
Duration of contract :
Period covered : January 1989 through June 1990
Project Leader : A.A. Bonne
Reported by : P.N. Henrion and M. Monsecour

A. OBJECTIVES AND SCOPE

The objective of the research is to determine conditions and parameters of importance for the prediction of radionuclide migration in the near-field and far-field of a nuclear waste repository in an argillaceous host formation. The assessment of the barrier performance is done by modelling and supposes that reliable in situ data are available.

The underground experimental HADES laboratory in the Boom clay at Mol allows to sample the clay and the pore water according to various procedures and to characterize them (pH and its buffering ; idem for the Eh ; oxidation, role of accessory minerals ; cation exchange capacity ; properties and molecular size distribution of the organic content of the clay, its capacity to form complexes under in situ conditions with Tc and actinides).

An active collaboration with the University of Leuven (K.U.L., Prof. A. Cremers and Dr. A. Maes) has been maintained on these matters.

Experiments performed from the gallery to determine in situ migration parameters are running in parallel with laboratory sorption and migration tests on reconsolidated clay plugs, with which they are compared. Though the transport mechanism is essentially diffusion particular features of the clay are revealed under conditions of surimposed convexion.

B. WORK PROGRAMME

1. Study of the organic matter fraction in the Boom clay with emphasis on Eu complexes.
   1.1. Characterization of the organic matter fraction. Mol.W^t distribution, density of complexing groups, mobility of the small molecules in compacted clay.
   1.2. Eu-humic acid complexes under in situ conditions.
   1.3. Radiolysis effects. Effects of gamma-radiolysis on molecular and complexing properties (using Eu as a reference element).
2. In situ short-term migration experiments in the Hades underground laboratory, using the important hydraulic gradient towards the gallery to cause water seepage through labelled clay plugs.
3. The characterization of a multi-layered aquiferous system.
   3.1. On the basis of hydrochemical data
   3.2. The on-going physico-chemical characterization of the Boom clay under in situ condition as a support for the above mentioned research.
   3.3. Emplacement of screens and filters in the formation around the gallery, to obtain various samples of pore water, the pressure distribution of this water, to be used as injection or collection ports for migration tests, and so on.

C. PROGRESS OF WORK AND RESULTS

State of advancement

The programme has been continued with little modification of course during the contract period, though with variations in pace and accentuation. 1989 saw the termination of laboratory diffusion tests on non-retarded species and the start of new series of long duration tests to confirm the excellent retardation of Np and Eu ($D \sim 10^{-10}$ cm^2sec^{-1}). A substantial contribution to the functional group characterization of the humic acids has been obtained (K.U.L.). The values determined for the complexation constants of Eu with normally immobile Boom clay humic acid molecules fully support the initial conclusions about migration reached by diffusion tests. The presence of reactive ferrous ions in the clay has been established and is believed to control the sensitivity to air oxidation and the ability of clay to react with molecular hydrogen.

As far as can be concluded from limited studies, credible irradiation doses to the Boom clay do not seem to entail detrimental consequences to the barrier function of the clay.

Convective in situ (from the gallery) migration experiments are in progress with labelled clay cores (with Cs and Am).

Progress and results

1. Characterization of the organic matter

A sophisticated acid-base titration method has been used to determine the proton capacity of soluble HA. The functional groups can be subdivided in three main families characterized by a pKa value. Humic materials from various origins are clearly distinguished. The interaction constant with Eu has been measured at several pH values. Under in situ conditions an identical log K ($\sim$ 13) is found for Am also. This result is most probably applicable to PuIII and the flux of the trivalent radionuclides in the formation will be controlled by the relative amount of small humic molecules, their size distribution and their diffusivities in the compacted clay.

A method combining acid-base titration, cobaltihexamine cation capacity measurements and chromatography on a hydroxy-apatite column was successful in characterizing gamma-irradiated Boom clay humic molecules (J. De Brabandere, K.U.L.). The main consequences of irradiation are an increase in the number of carboxylic functions and a decrease in the proportion of small molecules. The sorption of Eu and Tc tends to increase.

2. In situ migration experiments

Migration experiments on clay cores labelled with Eu and Sr have been completed while one experiment with ^{134}Cs has been running for more than two years without Cs breakthrough. To avoid the expensive overcoring required by this method a retrievable experiment has been designed and successfully tested. Similar experiments in a combined heat and radiation field are in preparation.

3. The characterization of a multi-layered aquiferous system

Steps have been taken to use a reconnaissance boring from the surface, conducted by the CEA, to core sands and clays in the transition zone between the Boom clay and the upper aquifer, and to proceed to a systematic physico-chemical characterization of about 30 m cores. These data would then be correlated to resistance diagraphies and other measurements in the borehole.

The presence of $FeCO_3$ in Boom clay has been demonstrated, and the distribution of Ca between $CaCO_3$ and ion exchange sites determined by a method involving selective extraction of solid phases, chemical analyses and the distribution in the system of Fe and Ca tracers. A slight exchange selectivity of Mg^{++} with respect to Ca^{++} is shown. "Available" ferrous iron in the clay (either sorbed, complexed by humic molecules or as $FeCO_3$) amounts to $0.7 - 0.11 \times 10^{-4}$ eq/gramme clay.

List of publications

/1/ Henrion, P., Put, M. and Van Gompel, M., The influence of the diffusion of non-sorbed species in Boom clay. 2nd International Conference on Chemistry and Migration Behavior of Actinides and Fission products in ghe Geosphere, Monterey, California, November 06-10, 1989

The Role of Organics in the Migration of Radionuclides in the Geosphere

Contractor: Risø National Laboratory, DK
Contract N°: FI1W/0066
Working Period: july 1986 - december 1989
Project Leader: Lars Carlsen

A. Objectives and Scope:

A review on the possible role of organics species in the ground water on the migration behaviour of radionuclides in the geosphere is required. Considerable amounts of data are available. They are, however, rather scattered throughout the literature.

An experimental study as well as theoretical considerations on the influence of organic complexing agents on the sorption, and hence migration behaviour of radionuclides are of fundamental interest in attempts to evaluate the possible transport of released radioactive waste with ground water.

Characterization of naturally occurring organics, e.g. humic and fulvic acids is of general interest, due to the omnipresence of these polymeric species in the terrestrial environment and to their known complexing abilities towards metal ions.

B. Work Programme:

B.1. Review of available literature on the influence of organics compounds including experimental and theoretical data obtained at Risø.

B.2. Batch-type experiments to elucidate the influence of organics on radionuclide migration.

B.3. Column-type experiments to elucidate the influence of organics on radionuclide migration.

B.4. Theoretical study to elucidate the effect of complex formation on radionuclide migration.

B.5. Characterization of humic acids partly within the frame of the joint european programme on humic acid characterization.

C. Progress of Work and Obtained Results

Summary.

The review paper on the role of organics on the migration behaviour of radionuclides in the terrestrial environment has been published by the Commision /1/.

The theoretical study on the influence of complexation on radionuclide migration has been completed and the results has been published separately in *Waste Management* /2/. The results are furthermore incorporated in the above mentioned review paper.

The laboratory has during 1989 participated in the joint european intercomparison and characterization study of humic acid complexation with europium. Further experimental effort has been devoted to the characterization of humic acids by determination of exchangable hydrogen with H/D exchange.

Progress and Results

1. Review (B.1.)

The review report summarizes factors of importance concerning the role of organics on the migration of radionuclides in the terrestrial environment and is now available /1/.

Following some introductory remarks (*chapter 1*), *chapter 2* describes the occurrence of organic compounds in the geosphere, taking both naturally occurring as well as artificially introduced compounds into account. The behaviour of organic compounds in the terrestrial environment is summarized (*chapter 3*) with special emphasis on sorption and persistence in the environment. Both chemical and microbiological degradation reactions are discussed. *Chapter 4* describes the fundamentals of complex formation in relation to migration. Stability constants for the interactions between relevant metal ions and low molecular weight ligands as well as with humic- and fulvic acids are summarized. *Chapter 5* is devoted to an evaluation, based on theoretical considerations, of the influence of organics on the migration behaviour of radionuclides in the terrestrial environment. Additionally, the behaviour of two special elements, *i.e.* technetium and iodine, is discussed. An overall summary is given in *chapter 6*.

The report contains a comprehensive list of references.

2. Theoretical work (B.4.)

The results of the theoretical investigations has been in detail been reported in the paper *The influence of complexation on radionuclide migration. A theoretical study* /2/.

The paper describes a theoretical evaluation of the influence of complexation on metal ion, *e.g.* radionuclide migration in environments containing an excess of complexing agents, *i.e.* the equilibrium between the free and the complexed metal ions can be regarded to follow pseudo first-order kinetics. It is shown that as long as the rate of interconversion between the free and complexed metal ions is rapid relative to the residence time in the system studied, the two species will migrate with the same speed, controlled by an "effective retention factor". It is clearly demonstrated that approaching zero complexation the effective retention approaches that of the more retarded species (the free metal ion), whereas the effective retention approaches that of the less retarded species (the complex) for increased complexation. The implication for the distribution of radionuclides in the terrestrial environment is discussed.

The described calculations unambiguously demonstrate the influence of complexation on the migration of metal ion in the terrestrial environment. Increasing the apparent stability constant of the complex, either as a results of high true stability of the complex or by increased ligand concentrations, significantly decrease the effective retention of the equilibrium system. Hence, it is concluded that the naturally occurring organic ligands, i.e. humic- and fulvic acids, which form strong to very strong complexes with metal ions, apparently must be expected to exhibit a pronounced influence on the migration behaviour of radionuclides, leading to an increased migration speed. Similarly the possible presence of artificially introduced potent ligands, as e.g. EDTA, forming stable complexes with most polyvalent metal ions, certainly should be avoided, due to the dominating role such compounds, even in very minor concentrations, will play in controlling the migration of the metal ions.

3. Humic Acid Characterization (B.5.)

The work in this area has been concentrated on the study of europium - humic acid complexation. The complexation of europium with different humic acids has been studied based on the conventional ion-exchange technique. The originally proposed dialysis-approach appeared less advantageous due to the fact the the different humic acids contained low molecular weight fractions, which could pass through the dialysis membrane.

Humic acids from Fanay Augeres has been studied in addition to further studies on commercially available humic acids (Aldrich) and Gorleben Humic acids.

The work on humic acid characterization based on isotope-exchange has been completed. It apperas that the determined content of exchangable protons was somewhat higher than the previously determined total proton capacity (determined by the $Ba(OH)_2$ method) in the cases of Aldrich-HA(H^+) and Gorleben Humic acid (GoHy 573-I), whereas significantly higher values were found in the case of Aldrich-HA(Na^+). This however, is not surprising since it should be expected to find values higher than the so-called total proton capacity. Hence, the H/D exchange technique will also give rise to exchange of protons located in e.g. amino- and mercapto groups. This taken into account the technique does not seem to constitute as an important tool in the estamation of humic acid complexing abilities, but only as an characterization of the humic acids with respect to functional group content.

It is, however, important to note that significant experimental difficulties appears due to the hydroscopic nature of D_2O/OD^- solutions. Thus, it seems necessary to carry out all operations in dry glove-boxes in order to obtain reasonable reproducible results.

<u>List of References</u>

/1/ Carlsen, L. EUR-12024 EN, Commision of the European Communities, Nuclear Science and Technology, Luxembourg, 1989, 76 pp

/2/ Carlsen, L., Nielsen, O.J., and Bo, P. Waste Mang. 9, 165-169 (1989)

ACTINOIDE MIGRATION PHENOMINA IN GROUNDWATER: COLLOID GENERATION AND COMPLEXATION WITH NATURAL ORGANICS

Contractor: Institut für Radiochemie, Technische Universität München
Contract No.: FI 1W/0067
Duration of contract: Sept. 1986 - Dec. 1989
Period covered: 1. Jan. 1989 - 31. Dec. 1989
Project leader: J.I. Kim

A OBJECTIVE AND SCOPE

Important geochemical processes that govern the migration of actinides in deep geological aquifer systems are: hydrolysis reaction, redox reaction, complexation with inorganics as well as organics and colloid generation. The colloid generation and complexation with natural organics, e.g. humic substances, appear to be the important geochemical phenomena with regard to actinide migration in a variety of aquifer systems. The contract research deals, therefore, with the colloid generation of representative actinides in different groundwaters and the complexation of Am and Pu with natural organics, particularly humic substances. The results are expected to give an insight into the migration mechanisms of actinides in the geosphere and hence to improve the safety assessment for the nuclear waste disposal in a given geosphere.

B WORK PROGRAMME

B 1. Actinide colloid generation in groundwater

- Characterization of colloids
- Generation mechanisms of actinide pseudocolloids
- Quantification of colloid generation in a migration medium for actinides

B 2. Actinide complexation with natural organics

- Characterization and complexation study
- Humic substances as organometallic colloids
- Mobility of complex species and colloids in aquifer systems
- Quantification of actinide mobility

B 3. Interlaboratory comparison exercise on complexation with natural ligands (COCO-group: TUM, CEN/SCK, KUL, CEA-FAR, Risø, BGS, JRC-Ispra and other new members)

- Intercomparison of characterization methods
- Separation and production of natural humic acids present in the reference sites
- Intercomparison of stability constants

C PROGRESS OF WORK AND OBTAINED RESULTS

Statement of advancement

The characterization of site specific humic and fulvic acids from the member countries (MIRAGE; COCO group) has been further proceeded in order to evaluate their complexation behaviour with actinide ions. The humate complexation of Am(III) and Cm(III) has been investigated for the reference and site-specific humic acids by different spectroscopic methods. These results are being applied for the geochemical modelling to assess the effect of humic acid on the solubility of Am(III) in Gorleben aquifer systems.

As for the colloid generation in groundwater, the interlaboratory cooperation with UKAEA has been undertaken for the characterization and the evaluation of the migration behaviour of colloids in five different Gorleben aquifer systems. Much effort is given to the elucidation of the behaviour of chemically actinide homologous elements (M(III) and M(IV)) and natural radionuclides (Th und U isotopes) between groundwater and colloids.

Progress and results

1. Characterization of humic and fulvic acids

A number of site-specific humic and fulvic acids from different geological origins of the COCO member countries have been characterized. A part of results for the whole characterization processes is given the progress reports [1,2] and the results of interlaboratory comparison exercise are summarized in the review paper of the MIRAGE plenary meeting [3]. The important part of the characterization results is the proton exchange capacity of each given humic acid, which is a primary parameter that goes directly into the evaluation of humate complexation constants for metal ions [4,5]. Such exchange capacities determined by a direct pH titration under inert gas atmosphere for different site-specific aquatic humic and fulvic acids are given below:

Reference humic acid	Aldrich-HA(H^+)	5.43 ± 0.16 meq/g
Gorleben humic acid	Gohy-573-HA(H^+)I	5.38 ± 0.20 meq/g
Gorleben humic acid	Gohy-573-HA(H^+)II	5.00 ± 0.05 meq/g
Gorleben fulvic acid	Gohy-573-FA(H^+)	5.70 ± 0.09 meq/g
Boom-Clay humic acid	Boom-Clay-HA(H^+)	4.22 ± 0.02 meq/g
Fanay-Augeres humic acid	Fanay-Augeres-HA(H^+)	1.85 ± 0.05 meq/g
Fanay-Augeres humic acid	Fanay-Augeres-FA(H^+)	6.93 ± 0.06 meq/g

2. Humate complexation of Am(III)

The Am(III) humate complexation has been studied by UV-spectroscopy for different humic acids [4,5], the reference humic acid (Aldrich-HA(H^+)), Gorleben groundwater humic acid (Gohy-573-HA(H^+)I) and Bradford Lake (USA) humic acid (Bradford-HA(H^+)). A careful analysis of the complexation process indicates that, under the condition in which the equivalent concentration of humic acid exceeds that of Am^{3+} (ratio >1), a tridentate complexation of Am(III) is dominant. Taking into account the Am(III) loading capacity of humic acid which varies with pH and ionic strength, the complexation constant of Am(III) with humic acid can be evaluated. The average values from multiple determinations for different humic acids are:

Aldrich-HA(H^+)	$\log \beta = 6.39 \pm 0.14$	(L/mol)
Gohy-573-HA(H^+)	$\log \beta = 6.44 \pm 0.15$	(L/mol)
Bradford-HA(H^+)	$\log \beta = 6.28 \pm 0.15$	(L/mol)

The values represent a tridentate complexation and are independent of pH and ionic strength.

3. Humate complexation of Cm(III)

Using a particular 5f transition state of Cm^{3+} which has a relatively high fluorescence yield, the humate complexation of this ion is investigated by time-resolved laser fluorescence spectroscopy (TRLFS) for Gohy-573-HA(H^+) [6]. The concentration of Cm^{3+} investigated is in the range of 10^{-8} mol L^{-1} in 0.1M $NaClO_4$ at pH=6.0. As is the case with the Am humate complexation the Cm^{3+} ion undergoes a tridentate complexation with humic acid. The experimental analytical results [6] are summarized in Table I.

Table I: The TRLFS determination of the humate complexation constant of Cm(III)

[Cm]tot (mol/L)	[HA(III)]tot (mol/L)	(CmHA(III)) / $[Cm^{3+}]$f	$[Cm^{3+}]$f (mol/L)	[CmHA(III)] (mol/L)	[HA(III)]f (mol/L)	log β [L/mol]
7.30×10^{-8}	6.65×10^{-8}	0.139	6.41×10^{-8}	0.89×10^{-8}	5.76×10^{-8}	6.38
5.52×10^{-8}	1.78×10^{-7}	0.449	3.81×10^{-8}	1.71×10^{-8}	1.61×10^{-7}	6.45
2.24×10^{-8}	3.38×10^{-7}	0.898	1.18×10^{-8}	1.06×10^{-8}	3.27×10^{-7}	6.44
6.11×10^{-8}	6.65×10^{-7}	2.130	1.95×10^{-8}	4.16×10^{-8}	6.23×10^{-7}	6.53

The average complexation constant is found to be:

$$\text{Gohy-573-HA}(H^+) \quad \log \beta = 6.45 \pm 0.06 \quad (L\ mol^{-1})$$

which is in excellent agreement with the value determined for Am(III) (see above).

4. Colloid generation

As a joint investigation with UKAEA [7], the groundwater colloid study has been carried out for 5 different Gorleben aquifer systems. The colloids separated by ultrafiltration with different pore sizes are characterized for their organic and inorganic constituents. The colloid size and population distribution in the groundwaters are determined by photoacoustic counting of laser light scattering (PALS) which has been developed recently in our laboratory [8]. The size and population distribution of colloids in one of the investigated groundwaters (Gohy-2227) are summarized in Table II. The results show that a major part of colloid population is found in the size range of 1 ~ 15 nm.

Table II: Determination of colloid population in Gohy-2227 by photoacoustic detection of light scattering (PALS): size fractionation carried out by ultrafiltrations

Fraction range (nm)	Average size used (nm)	Photoacoustic signal (μV/mJ)	Colloid population (particle/L)
1000-400	700	4.38 ± 0.08	$(3.9 \pm 0.1) \times 10^{9}$
400-100	250	6.54 ± 1.23	$(3.9 \pm 0.7) \pm 10^{11}$
100-80	90	2.75 ± 0.20	$(1.4 \pm 0.1) \times 10^{13}$
80-10	45	2.63 ± 0.26	$(5.5 \pm 0.6) \times 10^{13}$
15-2	8.5	3.21 ± 0.32	$(1.9 \pm 0.2) \times 10^{17}$
10-1	5.5	1.53 ± 0.15	$(3.4 \pm 0.3) \times 10^{17}$
Background signal		0.70 ± 0.10	

Table III shows the concentrations of inorganic elements in one of the investigated groundwater (Gohy-2227) and colloids fractionated by ultrafiltration at different pore sizes. The major part of trivalent and tetravalent elements, e.g. REE, Fe, Zr, Hf and Th, are found to be bound (80 ± 6 %) on colloids larger than 2 nm diameter, whereas only 10.6 ± 1.0 % of divalent elements are found as colloid-bound. Nearly 90 % of Th and U found in the groundwater is bound on colloids larger than 1.5 nm. It is conceivable from the present results that the elements with higher oxidation states ($Z \geq 3+$) may be transported by groundwater colloids.

Table III: Trace element concentrations in colloids collected by ultrafiltration of the prefiltered (1000 nm) groundwater Gohy-2227 at different pore sizes and in groundwater Gohy-2227 filtered at 0.4 μm pore size

Element	Concentration (mol L^{-1})*				
	Groundwater (filtrate at 400 nm)	Filter cake (colloids collected on ultrafilters)			
		400 nm	100 nm	15 nm	ca 2 nm
Ba (x 10^{-7})	3.38 ± 0.69	0.01	0.05	0.15	0.32
Ca (x 10^{-4})	5.12 ± 0.40	0.00	0.03	0.12	0.58
Ce (x 10^{-8})	4.41 ± 0.24	0.02	1.02	2.93	3.51
Co (x 10^{-9})	9.91 ± 0.78	0.13	1.18	2.40	5.86
Eu (x 10^{-9})	1.62 ± 0.02	0.00	0.18	0.61	1.18
Fe (x 10^{-6})	4.58 ± 0.52	0.66	2.73	3.00	3.44
Hf (x 10^{-9})	5.23 ± 0.06	0.01	0.30	1.00	4.11
La (x 10^{-8})	1.79 ± 0.01	0.00	0.38	1.27	1.33
Lu (x 10^{-9})	1.82 ± 0.07	0.00	0.11	0.36	1.28
Nd (x 10^{-8})	1.87 ± 0.12	0.01	0.46	1.31	1.57
Sb (x 10^{-10})	3.93 ± 0.77	0.10	0.54	0.93	2.23
Sc (x 10^{-8})	2.96 ± 0.24	0.01	0.22	0.76	2.36
Se (x 10^{-9})	2.20 ± 0.20	0.01	0.22	0.66	1.96
Sr (x 10^{-6})	2.34 ± 0.33	0.00	0.03	0.09	0.26
Tb (x 10^{-9})	1.33 ± 0.13	0.00	0.17	0.47	1.14
Th (x 10^{-9})	7.23 ± 0.73	0.03	2.02	5.52	6.68
U (x 10^{-9})	3.62 ± 0.10	0.06	0.52	1.39	2.57
Yb (x 10^{-9})	6.07 ± 0.23	0.01	0.40	1.35	4.74
Zn (x 10^{-6})	1.03 ± 0.29	0.03	0.14	0.24	0.28
Zr (x 10^{-7})	9.34 ± 1.50	0.01	0.41	1.34	6.97

* Elemental concentrations of the colloids are normalized to the solution volume of 1 L.

References:

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STUDY OF THE INTERACTIONS BETWEEN ORGANIC MATTER AND TRANSURANIUM ELEMENTS

Contractor: CEA-IRDI/DERDCA/DRDD/SESD/SCPCS

Fontenay-aux-Roses France

Contract n°: FI1W/0068

Duration of contract: from 1985 to 1989

Period covered: 1989

Project Leader: Mme Moulin, Mr Billon

A. OBJECTIVES AND SCOPE

The main objective of this programme is to study the importance of natural organic ligands (humic substances) as complexing agents of radioelements on the migration/retention processes of radioelements through the geosphere. Considering the humic substances as colloidal entities (due to their size and their interfacial properties), this programme is also devoted to the emphasis of the role of the natural colloids present in natural aquifers. Hence the objectives of this research programme are:

- to define the characteristics and properties of the natural colloids including more specifically humic substances,
- to determine the formation constants of the complexes formed between humic substances (humic and fulvic acids) and radioelements.

B. WORK PROGRAMME

B.1. Isolation and characterization of humic substances

B.1.1. Concentration of humic materials from natural groundwaters

B.1.2. Characterization of humic susbtances by physical and chemical methods

B.2. Interactions between humic substances and actinides

B.2.1. Development of the chromatographic method selected for the complexation studies

B.2.2. Determination of interaction constants by this method. Comparison with other methods, in particular spectroscopy (application and development)

B.3. Studies on natural colloids

B.3.1. Isolation of colloids from natural waters by ultrafiltration: sampling, water analysis, development of the technique.

B.3.2. Characterization of the colloids: size, population, composition. Use of scanning electron microscopy.

B.4. Intercomparison exercise (CoCo group)

C. PROGRESS WORK AND RESULTS OBTAINED

State of advancement

The characterization of natural humic substances isolated from a granitic water (Fanay-Augères/ Massif Central ; borehole located at 260 meters of depth) has been completed. Chemical and physical methods have been used to determine specific properties of humic and fulvic acids (elemental composition, mineral content, size, proton capacity, functional group content).

The binding properties of these specific humic substances towards americium (III) have been studied through chromatographic and spectrophotometric techniques and have been compared with humic substances of different origins. Formation constants have also been determined in order to compare the techniques and the origin of humic materials.

The interlaboratory exercise on the isolation and characterization of natural colloids from Grimsel water is completed ; a CEC report has been written /1/. The interlaboratory exercise on the characterization of humic substances from different specific sites is finished, a report is under progress.

Progress and results

Fanay-Augères water with 2 mg/l of Total Organic Carbon has been selected as an aquifer representative of a granitic geological formation. Humic substances isolated from this water (and distributed to the laboratories involved in the CoCo intercomparison exercise) represent 41% of the T.O.C.. Fulvic acids constitute the major part of the humic materials although humic acids represent a non-negligeable fraction which is somewhat different from what it is mentionned in the literature /2/.

Fanay-Augères humic substances have been characterized by different physico-chemical techniques. These characteristics (elemental and mineral composition, size, proton capacity, datation) are reported in Table 1. From these data, two features have to be underlined :

. the relatively high content of **uranium** present in humic acids which may be correlated to the environment of these materials (a uranium mine), and the affinity of uranium for humic acids,

. the fulvic acids **datation** (625 years) which seems to indicate that the residence time of the organic materials in Fanay-Augères water is relatively short. This could be correlated to the tritium datation (16 tritium units) of the water. Moreover Fanay-Augères water analysis (composition, datation, application of geochemical codes) seems to show that the water circulation in Fanay-Augères massif (fractured medium) is relatively complicated.

The interaction between a trivalent actinide, **americium (III)** and humic substances from different origins, and more particularly from Fanay-Augères has been investigated by two different techniques :

spectrophotometry and size-exclusion chromatography. Binding constants have been calculated assuming the formation of 1:1 complexes and complexing capacities (the amount of complexing sites present in the ligands) have also been determined experimentally (by spectrophotometry). Values are given in Table 2 expressing the binding constants in different unit systems. From these data it appears that:

. Fanay-Augères humic substances present relatively low complexing capacities compared with other humic/fulvic compounds,

. binding constants for the system Am(III)-Fanay-Augères are of the same order of magnitude than other data obtained for different humic materials,

. values obtained by chromatography and spectrophotometry are in fair agreement although higher data are observed with chromatography.

List of Publications

T.Dellis and V. Moulin
"Isolation and characterization of natural colloids, particularly humic substances, present in groundwater"
Proceedings of WRI-6, Ed D.L. Miles, Balkema, Rotterdam, pp 197-201

V. Moulin, M. Caceci and M. Theyssier
"Complexation behaviour of humic substances from granitic groundwater towards Am(III)"
Proceedings of the International Symposium of Aquatic Humic Substances in the Environment, August 1989, Linköping, Suède

References

/1/ Degueldre C., Longworth G., Moulin V. and Vilks P., PSI-Bericht Nr 39 (1989)

/2/ Thurman E.M., Organic Geochemistry of Natural Waters, M. Nijhoff and W. Junk, Dordrecht (1985)

	HUMIC ACIDS	FULVIC ACIDS
Elemental analysis		
% C	46.2	49.2
% H	6.3	4.8
% O	30.6	44.7
% N	8	1.4
% ash	<1	3.5
Mineral composition (ppm)		
Al	340	20
Fe	670	40
Ca	5080	2200
U	7760	30
Mg	2200	110
Acidity (meq/g)	3.4	5.7
Datation*	-	625 y
Molecular weight (Daltons)**	5000	5000
Size (nm)***	>1.5 and <25	<1.5

* determined by Svedberg Laboratory (via Professor B. Allard/ Sweden)

** determined by size-exclusion chromatography on Shodex column with a Tris buffer (0.1 M, pH 7)

*** determined by ultrafiltration in Tris buffer at pH 7

Table 1: Some characteristics of Fanay-Augères humic substances

	SPECTROPHOTOMETRY				CHROMATOGRAPHY	
	EC (meq/g)	logβ (1/eq)	logβ (1/g)	logβ (1/eq H^+)	logβ (1/g)	logβ (1/eq H^+)
HA Fanay	0.30	7.0∓0.2	3.5	6.0	4.6∓0.3	7.1
FA Fanay	0.45	6.5∓0.2	3.2	5.4	4.2∓0.3	6.4
HA Aldrich	0.96	7.0∓0.2	4.0	6.2	4.8±0.3	7.0
FA groundwater	0.88	6.4∓0.2	3.1	-		
FA surfacic water	1.22	6.0∓0.2	3.1	-		
HA surfacic water	1.20	7.0∓0.2	4.1	-		

Table 2: Binding constants of Am(III) with humic substances obtained by chromatography (pH 5 I = 0.1 M NaClO4) and spectrophotometry (pH 4.65 I = 0.1 M NaClO4)

DIFFUSION, SORPTION AND STABILITY OF RADIONUCLIDE - ORGANIC COMPLEXES IN CLAYS AND CLAY-ORGANIC COMPLEXES

Contractor: IMPERIAL COLLEGE, LONDON (UK)
Contract No: FI1W/0147

Duration of contract: January 1988 - June 1990
Period covered: January 1989 - December 1989
Project leader: L.V.C. Rees

Progress report not yet available.

EFFECTS OF NATURAL ORGANIC SUBSTANCES ON THE GEOCHEMISTRY OF A RADIOACTIVE WASTE REPOSITORY

Contractor:	UKAEA, Harwell Laboratory, UK
Contract Number:	FI1W/0156
Duration of Contract:	January 1988 - March 1990
Period Covered:	January 1989 - December 1989
Project Leader:	F T Ewart

A OBJECTIVES AND SCOPE

A significant proportion of the dissolved organic carbon in natural groundwaters consists of humic and fulvic acids. There is evidence that these may form complexes with some radionuclides which may result in increased aqueous concentration either due to increased solubility or decreased sorption on to surfaces. Previous work in this laboratory has involved the completion of a literature review /1/, an investigation of possible methods of characterisation and some preliminary studies of the effect of humic acid on the sorption of americium on cementitious materials. This programme is for a continuation of that research in those areas relevant to a radioactive waste repository.

There are four main objectives in the programme. Limited humic characterisation forming part of the "CoCo" intercomparison exercise will be made on groundwater samples obtained from geological structures which are typical of reference repository sites. The effects of humic acids on the solubility of Pu, Am and Np in waters representative of a cementitious repository will be investigated and the influence on sorption studied for these elements. A study will be made of the feasibility of including the effects of natural organic compounds in geochemical modelling codes.

B WORK PROGRAMME

B.1 Characterisation of humic substances
B.2 Effects of humic acids on Pu, Am and Np solubilities
B.3 Effects on humic acids upon the sorption of Pu, Am and Np on cement grouts
B.4 Modelling studies

C PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In the period covered by this report the size distribution of humic species present in a 0.1M sodium hydroxide solution extract of Boom Clay has been studied using a combination of gel permeation chromatography and ultracentrifugation. The extracted material consisted of two major fractions with molecular weights determined by ultracentrifugation of approximately 5,000 and 20,000. Studies of the solubility of americium in solutions of humic acid extracted from Boom clay in the presence of 9:1 Blast Furnace Slag/Ordinary Portland Cement have found that the size distribution of americium is dependent upon the inventory of the radioelement. It is therefore not possible to determine a single solubility value. Sorption studies of americium and neptunium on 9:1 BFS/OPC have also been made. These have shown that humic acid causes significant decreases in Rd values only over timescales of a few days.

The current state of the programme is as follows:

- B1 Work completed
- B2 Work ceased due to difficulties in determining single solubility values
- B3 Progressing normally
- B4 Progressing normally

PROGRESS AND RESULTS

Characterisation of Humic Substances (B1)

The size distribution of humic species present in a 0.1M sodium hydroxide solution extract of Boom Clay has been studied using a combination of gel permeation chromatography (GPC) and ultracentrifugation to reduce the polydispersity of the sample. The humic material was extracted with sodium hydroxide solution (clay:solution = 1:10 w/v) under nitrogen and the extract filtered through a 0.45μm filter prior to concentration above a 1,000 Molecular Weight Cut-Off (MWCO) ultrafilter. This concentrate, which contained approximately 2,500$\mu g.ml^{-1}$ Total Organic Carbon (TOC), was fractionated on a Sephorose CL-6B GPC column using 0.1M sodium hydroxide solution as the eluant. Two broad peaks were observed with molecular weights of $\approx$ 5,000 and $\approx$ 50,000 estimated from protein calibrations. The molecular weights of selected individual fractions were then determined by sedimentation equilibrium ultracentrifugation /2/.

Table I gives the molecular weights determined for the fractions and also for the acid insoluble and acid soluble fractions of the leachate. Because the calculations are sensitive to the value used for the partial molar volume of the solute two typical values were used. There are discrepancies between the weights estimated by GPC and those determined by ultracentrifugation and these increase as the weight increases. The resolution of the GPC column was lower at high molecular weights and calibration with proteins under highly basic conditions was only possible up to a maximum molecular weight of approximately 50,000. Both factors may contribute to the differences in molecular weights found by the two techniques. The molecular weights found by ultracentrifugation for the unfractionated acid soluble and insoluble samples were 3,000-4,000 and 8,000-10,000 respectively. These values were lower than expected from a comparison with individual fractions and arise from the polydispersity of the two samples. This highlights the importance of reducing sample polydispersity before making molecular weight determinations.

Effects of Humic Acid on Amercium Solubility (B2)

The effect of the inventory of americium upon its solubility in a Boom clay/BFS:OPC extract has been examined. Americium was added to give initial concentrations of 10,120 and 1000 $Bq.ml^{-1}$ ($\approx 3 \times 10^{-10}M$ to $\approx 3 \times 10^{-8}M$) and samples taken over a 20 day period by filtration through 0.45μm filters. Although there was a steady decrease in solution activity for each of the two lowest inventories over this period the solution which initially contained 120 $Bq.ml^{-1}$ of americium always maintained a higher concentration of the radioelement. However, when the initial concentration of americium was increased from 120 $Bq.ml^{-1}$ this resulted in a decrease in the concentration found in the 0.45μm filtrate after one day from $\approx$ 60 $Bq.ml^{-1}$ to $\approx$ 2 $Bq.ml^{-1}$. Unfiltered, agitated samples did not show a similar decrease. Analysis of the 1000 $Bq.ml^{-1}$ solution after 13 days found that the majority of the activity was associated with species between 0.45μm and 2μm in size. The concentrations found are given in Table II. These experiments were repeated with an initial americium concentration of $\approx$ 124,000 $Bq.ml^{-1}$ ($- 4 \times 10^{-6}M$). These were equilibrated for one day before filtration through a range of filters and the concentrations found are also given in Table II. Nearly all the americium is associated with species which are larger than 0.45μm. The concentration found in the filtrate which passed through a 0.45μm filter was $\approx$ 60 $Bq.ml^{-1}$, equivalent to a concentration of $2 \times 10^{-9}M$. This compares with a solubility of $3 \times 10^{-11}M$ for americium at pH12 determined after filtration through a 25,000 MWCO filter. If all the americium which passed through a 0.45μm filter was assumed to be in solution then this would correspond to a solubility enhancement of approximately 100 times.

These results in conjunction with these previously obtained, suggest that enhancement of americium solubility by humic substances is possible but that in the near-field of a cementitious repository the effect will be limited by the tendency of both calcium and the radioelement to cause aggregation of humic species.

Sorption Studies (B3)

The sorption of neptunium on 9:1 BFS/OPC in the presence of humic acid has been studied. The short-lived isotope neptunium-235 was used as a tracer to enable the experiments to be carried out at concentrations an order of magnitude below the solubility limit of neptunium (IV) at high pH. The neptunium was reduced to Np(IV) electrochemically and added to cement water extracts of Boom Clay containing approximately 10$\mu g.ml^{-1}$ TOC. After one hour the solutions were sampled and the cement added. The concentrations of neptunium in 0.45μm and 30,000 MWCO filtrates were monitored. Table III gives the Rd values calculated from the measured concentrations. The table also includes data calculated from parallel experiments in which humic acid was absent. In both sets of experiments the Rd values increase with time and values calculated from the concentrations in the 0.45μm filtrates are less than those from the corresponding 30,000 MWCO filtrates. Sorption is initially low in the presence of humic acid but then increases to approach the level observed in cement-equilibrated water. After 71 days the Rd value calculated from a 0.45μm filtrate in the presence of humic acid is approximately 3×10^4 $ml.g^{-1}$ compared to 1×10^4 - 6×10^4 $ml.g^{-1}$ in the absence of humic acid.

Modelling Studies (B4)

It had been hoped to use data obtained in the experimental programme to develop a model of humic acid complexation in the near-field of a cementitious repository. The data has, however, proved inadequate for this purpose and modelling is in progress using data available in the literature.

REFERENCES

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/2/ GARDINER, M.P., Harwell Laboratory Report AERE M3730 (1989)

TABLE I

MOLECULAR WEIGHTS OF BOOM CLAY EXTRACT FRACTIONS

		M_{wt} by Ultracentrifugation	
Fraction	M_{wt} by GPC	V~ = 0.71	V~ = 0.75
7	600,000 - 420,000	56,000	68,000
10	200,000 - 140,000	60,000	73,000
13	64,000 - 45,000	16,000	21,000
16	22,000 - 15,000	20,000	23,000
18	10,000 - 7,000	17,000	23,000
21	3,500 - 2,400	4,500	5,500
24	1,500 - 800		
Acid Soluble	-	3,000	4,000
Acid insoluble	-	8,000	10,000

NOTES:

M_{wt} = Molecular weight

GPC = Gel Permeation Chromatography

V~ = Partial Molar volume

TABLE II

SIZE DISTRIBUTIONS OF AMERICIUM-CONTAINING SPECIES IN A 9:1 BFS/OPC WATER EXTRACT OF BOOM CLAY HUMIC ACID

Filtrate/μm	Initial [Am] = 1000 $Bq.ml^{-1}$		Initial [Am] = 124,000 $Bq.ml^{-1}$	
	[Am]/$Bq.ml^{-1}$	[Am]/M	[Am]/$Bq.ml^{-1}$	[Am]/M
0.22	-	-	6	2×10^{-10}
0.45	2	7×10^{-11}	63	2×10^{-9}
1	650	2×10^{-8}	-	-
2	910	3×10^{-8}	119,000	4×10^{-6}
5	920	3×10^{-8}	130,000	4×10^{-6}
10	930	3×10^{-8}	125,000	4×10^{-6}
Unfiltered	930	3×10^{-8}	130,000	4×10^{-6}

TABLE III

SORPTION OF NEPTUNIUM ONTO 9:1 BFS/OPC

	Rd/ml g^{-1}			
Time/Days	Humic Acid Leachate		Cement equilibrated water	
	0.45μm	30,000	0.45μm	30,000
1	670/110	320/250	780/2200	>63000
2	610/330	1300/800	17000/19000	>63000
5	6700/2600	14000/8000	63000/>63000	>63000
9	32000/-	-	>63000	-
71	30000/35000	-	12000/57000	-

DETERMINATION OF THE MOLECULAR WEIGHT OF THE GORLEBEN HUMIC ACID.

Contractor : Université de Nantes, France
Contract N° : FI1W-0197
Duration of contract : January 1988-December 1989
Period covered : January 1989-December 1989
Project leaders : Prof. J. PIERI,

A. OBJECTIVES AND SCOPE

Humic and fulvic acids are important transfer vectors of radionuclides in the geoophere, particularly in the porous sedimentary systems. Such systems are obliged ways between the geological formationof the nuclear waste storage sites (salt or clay options) and the biospher. In the case of the granitic formation, weathering materials of fractures or mined cavities have a similar behaviour. Indeed these acids, always present in the underground waters, have complexation properties which can induce high mobility species in the water-table.

These are the goals of the project :

- measurements of humic acids molecular weight by means of ultracentrifugation and liquid chromatography (intercalibration exercice).
- fractionation and isolation of humic and fulvic species.

B. WORK PROGRAMME

1- The tested sample was send by Professor J.I. Kim (Institut für Radiochemie der Universitat Munchen) with the reference GoHy-573-HA (H+) and come from the Radioactive Waste Deposit Site of Gorleben (DBR).
2- Estimation of the competition of radionuclides on definite sites of ligands (native or partially denatured).
3- Taking part in the intercalibration exercise on the humic and fulvic acids.

PROGRESS AND RESULTS

1- DETERMINATION OF THE MOLECULAR WEIGHT BY ULTRACENTRIFUGATION ON PREFORMED SUCROSE GRADIENT

1.1 - METHODOLOGY

The method used and described in the annual CEE report (J. PIERI, 1988, /1/) is modified because of the low rate of penetration of the substance in a 0-15% sucrose gradient after a 8 hours, 110, 000 g run. So, the procedure was modified.

The gradients(0-5% sucrose) are submitted to a high gravitation field (110,000 g, 40 hours, Beckman L5-75 B Ultracentrifuge, SW 41-Ti rotor). The gradients are then divided into 25 fractions (Fraction collector LKB Superrac 2211) and the optical density is measured for the following wavelengths : 254, 465 and 665 nm (humic Acid), 410 nm (C-Cytochrome), 280 nm (S. B. Trypsin Inhivitor, A-Chymotrypsinogen). The gradient shape is measured with an Abbe-type refractometer.

Substances	Svedberg's coef. (sec)	M. W. (g)
C-Cytochrome	1.17 x 10-13	12,500
S. B. Trypsin Inhibitor	1.80 x 10-13	14,300
A-Chymotrypsinogen	2.54 x 10-13	25,000

Table I - Characteristics of the standards (from SOBER /2/, LEHNINGER /3/, and BOEHRINGER documentation.

1.2 - RESULTS

The results of the standards centrifugation are illustrated by the figure 1. A good penetration is obtained for these three substances, with a growing molecular weight sorting in the usable part of the linear gradient. A fitting is used to determine the peaks position.
The results of two successful experiments are illustrated by the figures 2 and 3. The peak positions are near and very clearly defined (repectively 7.0 and 7.6).

The E4/E6 ratio (O.D. 465 nm/O.D. 665 nm) which varies according to a straight line with a negative slope, indicates a behaviour similar to those found for the Aldrich Humic Acid and the Boom Humic Acid (/1/).

The Svedberg's coeffecient is obtained by determination of an exponential regression fitting of the relation : S = f (Fraction number) with the standards data. The extrapolated S values for the Gorleben Humic Acid are respectively 0.986 and 1.036 for both experiments (see figure 4).

The molecular weights are determinated by a second relation M.W. = f(S). The best approximation is an exponential regression with the standards data. By this way, the approximate Gorleben Humic Acid molecular weights are respectively 11,500 and 11,800 (see figure 5).

The molecular weights of the Gorleben Humic Acid is inferior to those found for the Aldrich Humic Acid (27,000 - 107,000 range) and for the Boom Humic Acid (54,000) /1/, but the value is in harmony with those given by POSNER and CREETH (1972) in STEVENSON (1982) /4/ for the substances extracted from the red-brown earth.

The method, improved by extension of run-time, seems very reliable, specially with well purified humic substances.

2 - DETERMINATION OF THE MOLECULAR WEIGHTS BY GEL FILTRATION CHROMATOGRAPHY

2.1 - METHODOLOGY

The methodology describe for the Aldrich Humic Acid and the Boom Humic Acid is reemployed.

The calibration of the column has been made with the substances characterized in Table II.

Substances	M. W. (g)	abs.(nm)	Elution volume cm^3	Kav*
Dextran Blue	2,000,000 (Vo)	660	15.91	0
Aldolase	158,000	280	20.08	0.199
Albumin	67,000	280	21.22	0.253
Chymotrypsinogen	25,000	280	25.64	0.464
A-Ribonuclease	13,000	280	27.91	0.572
K-Hexaferrocyanate	329.3 (Vi)	420	32.20	0.776

Table II - Physical characteristics of Sephacryl S-300 column standards.

- $Kav = \frac{Ve - Vi}{Vt - Vo}$; Ve = elution volume ; Vo = exclusion volume

Vt = total volume = 36.9 cm^3 ; Vi = interstitial volume

A 1 ml volume of a 0.05 g/l Humic Acid solution is disposed and the eluate solvant is collected into 40 fractions (Fraction Collector LKB Superrac 2211) and the optical density is measured for the following wavelengths : 254, 465 and 665 nm.

2.2 - RESULTS

The results of two successful attempts are presented on the figures 6 and 7, with the calibration curves of Dextran Blue (Vo) and K-Hexacyanoferrat (Vi). The two peaks are very near (respectively 28.5 and 28.3 cm^3). Contrary to the results of the Boom Humic Acid (/1/), no tailing effect appears with the Gorleben acid, attesting the high level of purification and the absence of gel-interacting substances. This fact seems confirmed by the reversed-V shape of the E4/E6 ratio curve inside the Vo-Vi range. The maximum of the E4/E6 ratio (7.24 for the first attempt, 5.61 for the second one) indicate a low level of the fulvic species (SCHNITZER (1971) in STEVENSON (1982) /4/).

The molecular weights are interpolated in the straight section of the calibration curve : log(M.W.) = f(Kav), illustrated by the figure 8. The estimated values are respectively 12,300 and 13,100 for the two attempts. These values are in harmony with those obtained by ultracentrifugation (see 1.2).

3 - REFERENCES

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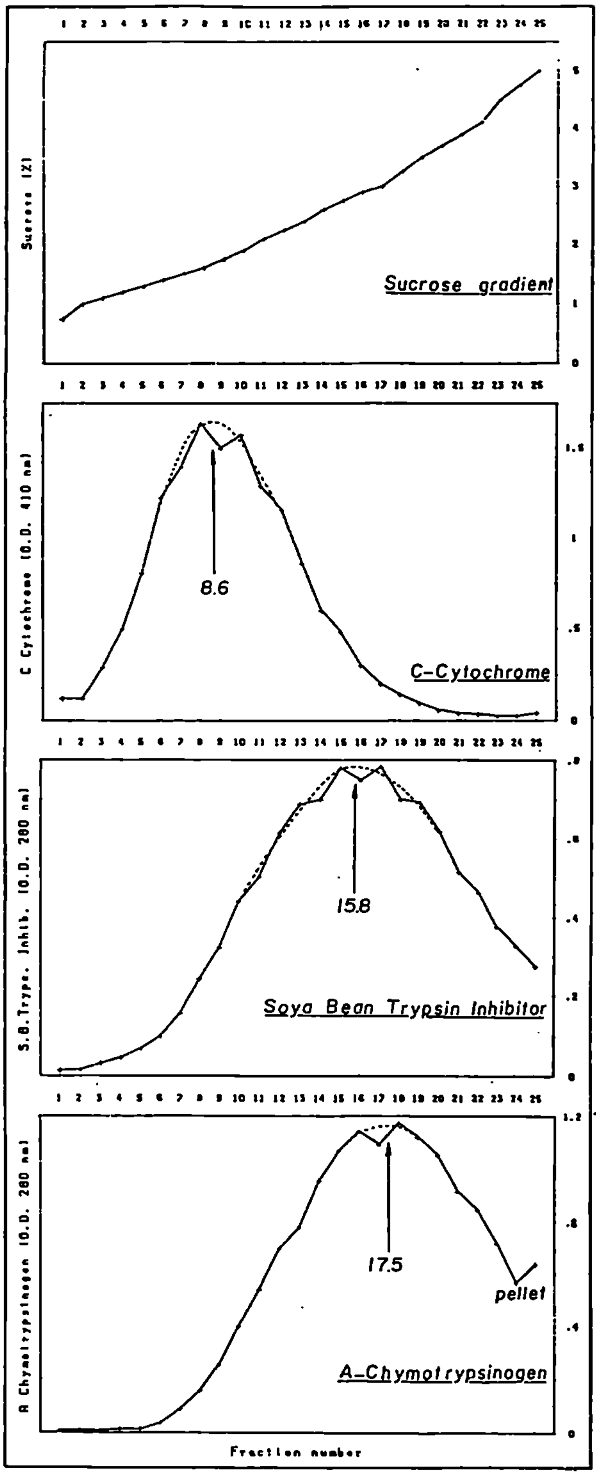

Fig.1 - Ultracentrifugation markers

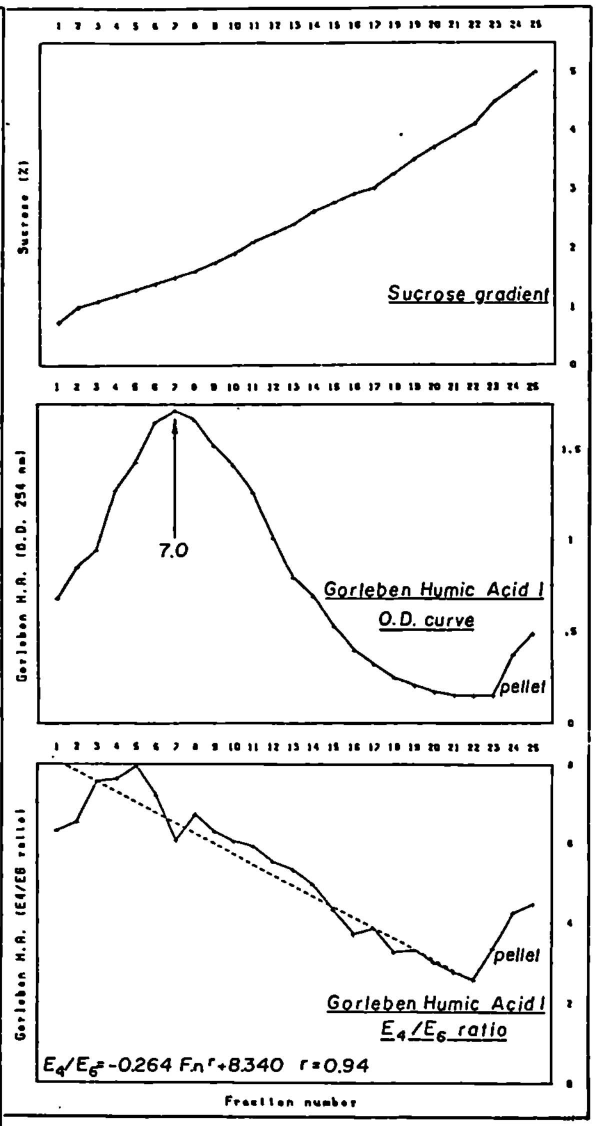

Fig. 2 - Gorleben Humic Acid ultracentrifugation results.

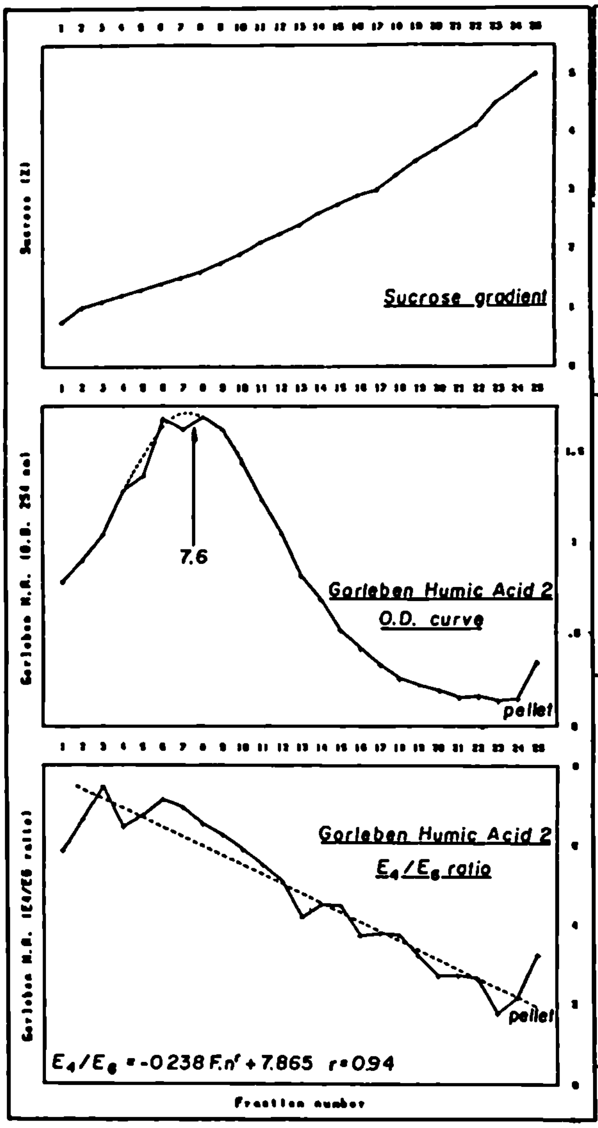

Fig.3 - Gorleben Humic Acid ultracentrifugation results

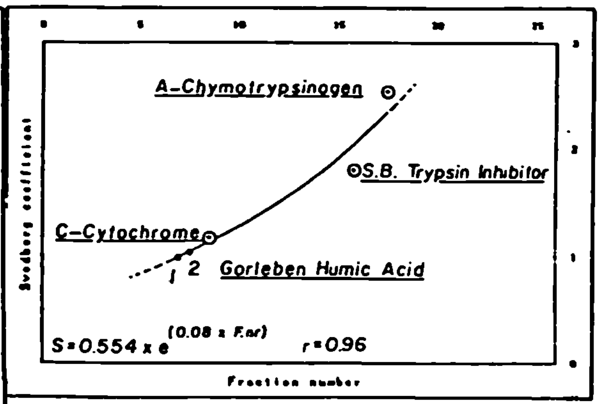

Fig. 4 - Determination of the Svedberg's coefficients

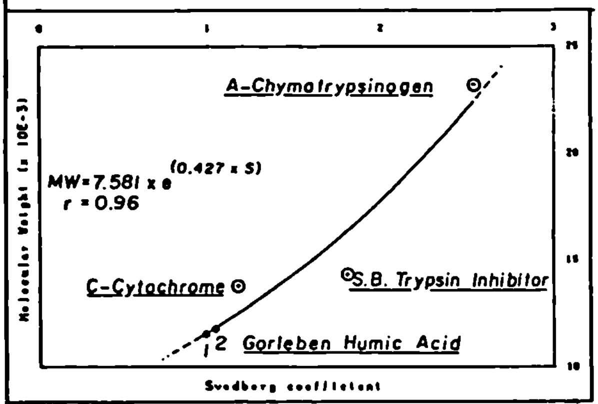

Fig. 5 - Determination of the Molecular Weights

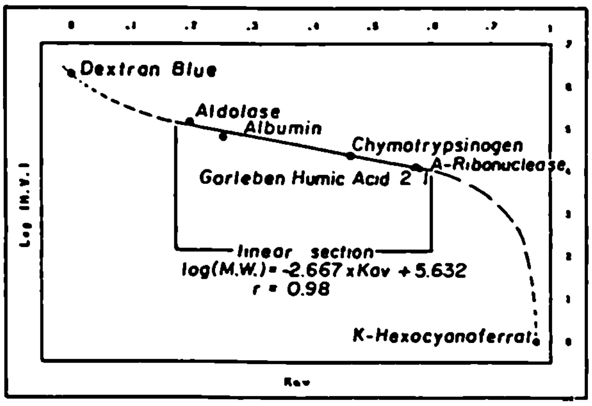

Fig 8 - Molecular Weight determination of the Gorleben H.A

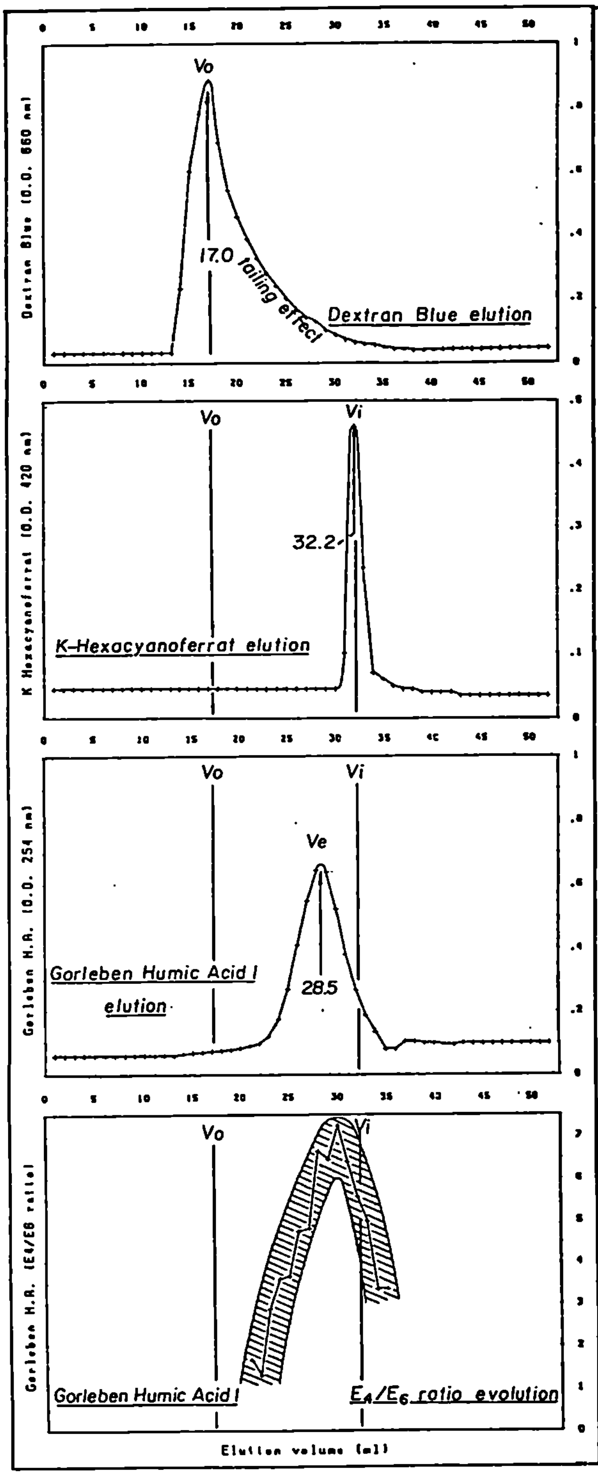

Fig. 6 - Gorleben Humic Acid I . Elution curves on Sephacryl

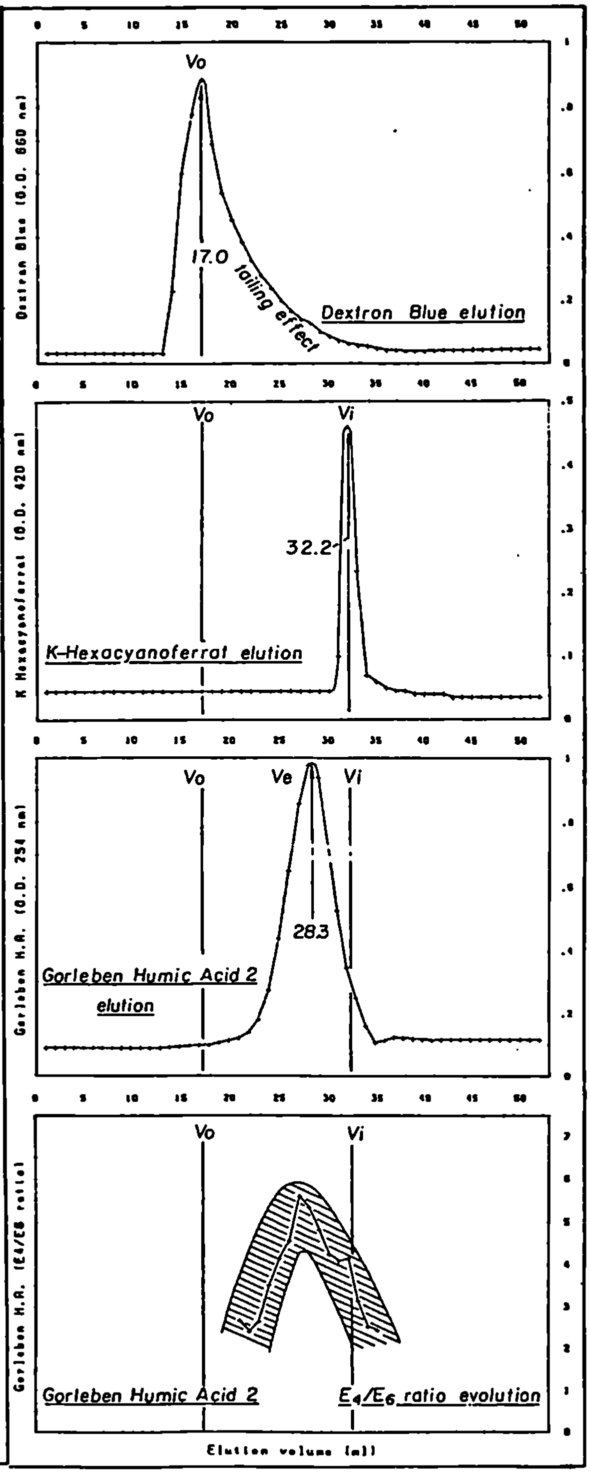

Fig.7 - Gorleben Humic Acid 2. Elution curves on Sephacryl S-

ASSESSMENT OF BASIC MIGRATION PHENOMENA OF ACTINIDES: COMPILATION, VERIFICATION, DETERMINATION OF THERMODYNAMIC CONSTANTS AND SPECIATION IN NATURAL AQUIFER SYSTEMS

Contractor: Institut für Radiochemie, Technische Universität München
Contract No.: FI 1W/0202
Duration of contract: Dec. 1988 - May 1990
Period covered: 1. Jan. 1989 - 31. Dec. 1989
Project leader: J.I. Kim

A OBJECTIVES AND SCOPE

The main objective of this research programme is to develope a validation possibility for the geochemical modelling of migration phenomena of actinides in deep aquifer systems. To realize this objective, two important basic works are envisaged in the programme. The first part of the research is to obtain reliable thermodynamic constants of hydrolysis reaction and carbonate complexation for actinides of different oxidation states, whose data are not well known or known with large uncertainties. This concerns Am, Pu, Np, Th and U ions. The species assumed for the evaluation of thermodynamic data will be verified with the help of spectroscopic or other methods. The second part of the programme aims at the speciation of actinides in various aquifer systems. The conditions would be those of the Gorleben aquifers as a MIRAGE reference site. The elements to be studied are Np, Pu and Am. The speciation will be carried out using laser-induced photoacoustic spectroscopy (LPAS) and time resolved laser fluorescence spectroscpy (TRLFS) with the help of ultrafiltration and ultracentrifugation.

B WORK PROGRAMME

B 1. Compilation, verification, determination of thermodynamic constants of hydrolysis reaction and carbonate complexation

- Critical assessment and compilation of thermodynamic data of An(III), An(IV), An(V) and An(VI)
- Experimental determination and verification of uncertain thermodynamic data
- Verification of species assumed for the evaluation of thermodynamic data

B 2. Speciation of actinides in natural aquifer systems from the MIRAGE reference site (e.g. Gorleben)

- Speciation under various geochemical conditions of different aquifers
- Comparison of experimental speciation with theoretical speciation based on thermodynamic data
- Development of a new spectroscopic method

C PROGRESS OF WORK AND OBTAINED RESULTS

Statement of advancement

Apart from a completion of the literature survey and compilation of thermodynamic data for hydrolysis reactions and carbonate complexation of Th, U, Np, Pu and Am, the verification experiments have been carried out for a number of thermodynamic data which are known with large uncertainity or conflicting among published values in the literature. The work hitherto carried out covers the determination of new thermodynamic data for the hydrolysis reactions of Np(V), Pu(IV), Pu(VI), Am(III) and Am(V), and the carbonate complexation of Np(V) and Am(III). Solubility and spectroscopic methods have been applied in parallel in order to validate the results of one method by another.

Speciation of soluble actinides in Gorleben groundwaters has been carried out by laser-induced photoacoustic spectroscopy (LPAS), time-resolved laser fluorescence spectroscopy (TRLFS) and photoacoustic detection of laser light scattering (PALS). The results are compared with those from a geochemical modelling in the same aquifer system. Such a comparison process is the first approach for the validation of the geochemical modelling.

Progress and results

1. Solubility products of Pu(IV) hydroxide and oxide

By the solubility experiment of well characterized amorphous $Pu(OH)_4$ and crystalline PuO_2 in 1M $HClO_4$ in support of spectroscopic speciation, the solubility products (K_{sp}) of the two solids are determined for the reactions:

$$Pu(OH)_4 \rightleftarrows Pu^{4+} + 4\ OH^- \qquad (1)$$

$$PuO_2 + 2\ H_2O \rightleftarrows Pu^{4+} + 4\ OH^- \qquad (2)$$

The results evaluated for the thermodynamic constants [1] are:

Reaction (1): $\log K_{sp} = -57.85 \pm 0.05$ (experiment)
Reaction (2): $\log K_{sp} = -60.20 \pm 0.17$ (experiment)
$\log K_{sp} = -63.8 \pm 1.0$ (calculated)

The free energy of formation for $Pu(OH)_4$(am) is calculated to be -1141 kJ mol^{-1} [1].

2. Carbonate complexation of Am(III)

Am(III) carbonate complexation has been investigated [2] in 0.1M and 0.3M $NaClO_4$ under Ar + 1 % CO_2 atmosphere in the pH range of 6.0 -

9.2. Under this condition the Am(III) precipitated is found to be $Am_2(CO_3)_3(s)$. The precipitate is characterized by X-ray diffraction, thermal analysis and IR-spectroscopy. Solubility and spectroscopic methods have been used in parallel to determine the solubility product (K_{sp}) and complexation constants (β_i). The results from both methods agree satisfactorily with one another. The average values determined by the two methods [2] are:

$$\log K_{sp}\ (Am_2(CO_3)_3) = -29.80 \pm 0.07$$
$$\log \beta_1\ (AmCO_3^+) = 6.22 \pm 0.09$$
$$\log \beta_2\ (Am(CO_3)_2^-) = 9.76 \pm 0.16$$

Under Ar + 1 % CO_2 atmosphere the contribution of Am(III) hydrolysis reactions is minimal, whereas this contribution becomes significant under normal atmosphere with a CO_2 partial pressure of $10^{-3.5}$. Fig 1 shows relative amounts of Am(III) species under two different CO_2 partial pressures, 10^{-2} and $10^{-3.5}$ [2,3]. The formation of mixed hydroxo-

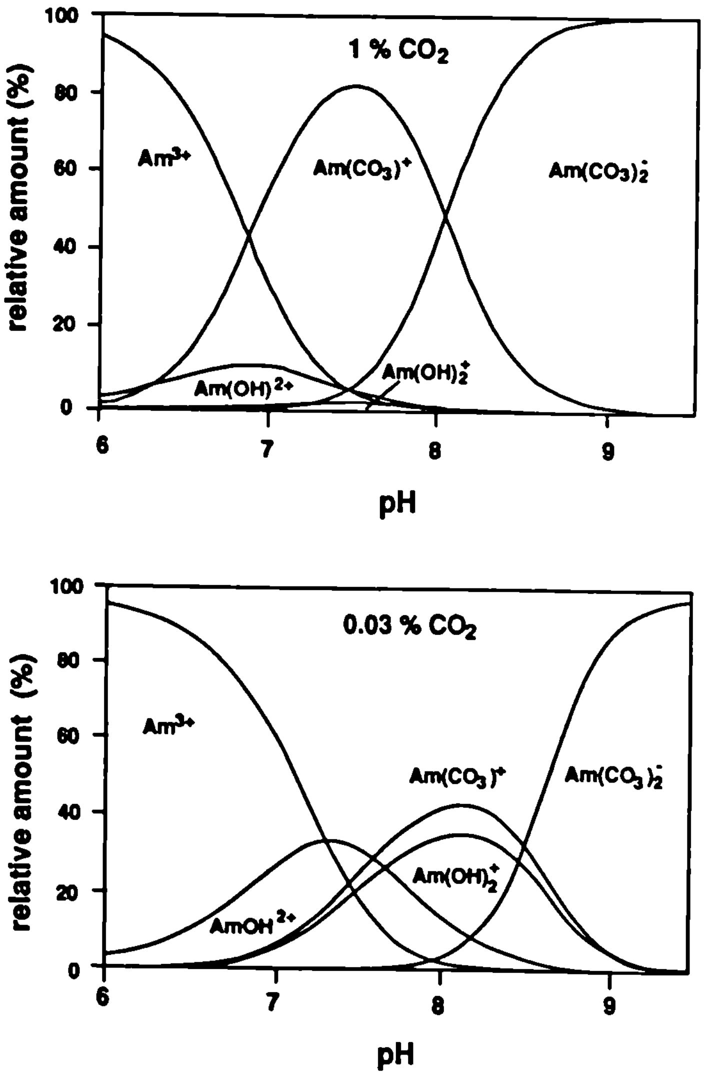

Fig. 1: Speciation of carbonate and hydrolysed ions of Am(III) in 0.1M $NaClO_4$ based on thermodynamic constants [2,3] for two different CO2 partial pressures.

carbonate species is possible under normal atmosphere with a CO_2 partial pressure of $10^{-3.5}$ (down part fig. 1).

3. Hydrolysis and carbonate complexation of Np(V)

Hydrolysis reactions of Np(V) have been investigated in 0.1M, 1.0M and 3M $NaClO_4$ in the pH range of 6-13 under Ar atmosphere [4]. The experimental results have corroborated the previous experiment which showed the presence of two hydrolysis species NpO_2OH and $NpO_2(OH)_2^-$ [4]. The new experiment at different ionic strength has resulted in a possibility of evaluating the effect of ionic strength on complexation reactions. The thermodynamic constants determined for the hydrolysis reactions of NpO_2^+ [5] are:

$$\log K_{sp}\ (NpO_2OH(c)) = -9.42 \pm 0.05$$
$$\log K_{sp}\ (NpO_2OH(am)) = -8.75 \pm 0.04$$
$$\log \beta_1\ (NpO_2OH) = 2.53 \pm 0.27$$
$$\log \beta_2\ (NpO_2(OH)_2^-) = 4.44 \pm 0.07$$

Carbonate complexation of Np(V) has been investigated in 0.1M $NaClO_4$ under Ar + 0.03 % CO_2 atmosphere in the pH range of 7-10 [6]. A combination of solubility and spectroscopic methods has been applied for the experiment. The precipitate is found to be $NaNpO_2CO_3(s)$. The solubility product and carbonate complexation constant obtained are:

$$\log K_{sp}\ (NaNpO_2CO_3) = -10.13 \pm 0.04$$
$$\log \beta_1\ (NpO_2CO_3^-) = 4.40 \pm 0.07$$
$$\log \beta_2\ (NpO_2(CO_3)_2^{3-}) = 6.40 \pm 0.18$$

4. Speciation of actinides in Gorleben groundwater

Speciation of actinides in selected Gorleben groundwaters has been carried out by different laser spectroscopies: laser-induced photoacoustic spectroscopy (LPAS), time-resolved laser fluorescence spectroscopy (TRLFS) and photoacoustic detection of laser light scattering (PALS). The first method was developed in the frame work of the MIRAGE project I and the latter two methods have been developed during the present research period [7]. A combination of the three modern spectroscopies enables a direct speciation of actinides in groundwater. The sensitivities obtained by LPAS is 7×10^{-9} mol L^{-1} for Am^{3+} and by TRLFS is 10^{-12} mol L^{-1} for Cm^{3+} and UO_2^{2+}. The PALS provides quantification of groundwater-colloids for the size range of 1 nm to 400 nm [8].

As a preliminary work for the validation of a geochemical modelling for the solubility of Am species in groundwater, one of the well characterized Gorleben groundwaters is chosen for the speciation work. In this groundwater (Gohy S113-1092) an excess of Am^{3+} is introduced, precipitated and maintained a solubility equilibrium for over six months. The spectroscopic speciation results of dissolved Am species are then compared with those from a geochemical modelling whch was carried out by the MINEQL programme using the Gorleben groundwater data and the thermodynamic constants for Am(III) determined at this laboratory [2, 3]. The preliminary results are compared with one another in Table I.

Table I: Comparison of the results of modelling (MINEQL, "open system" pH=7.00 Calcit-saturation) with the spectroscopic speciation by LPAS in Gohy S113-1092

Method	[Am]tot [mol/L]	[Am^{3+}] [mol/L]	[AmCO3] [mol/L]	[AmOH] [mol/L]	[AmOHCO3] [mol/L]	[AmCl] [mol/L]	[AmHA] [mol/L]
Modelling	1.1×10^{-6}	2.6×10^{-7}	3.1×10^{-7}	2.5×10^{-8}	1.8×10^{-9}	3.4×10^{-9}	-
LPAS	9.2×10^{-7}	3.0×10^{-7}	3.3×10^{-7}	$<9 \times 10^{-8}$	-	-	2.9×10^{-7}

AmHA : Am humate complex as a tridentate

References:

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COMPLEXATION OF RADIONUCLIDES WITH NATURALLY OCCURRING ORGANIC COMPOUNDS IN GROUNDWATER

Contractor: British Geological Survey, Keyworth, Nottingham.

Contract No: FI-1W-0203
Duration of Contract: from October 1988 to March 1991
Period covered: from June 1989 to January 1990
Project Leader: G.M.Williams

A. OBJECTIVES AND SCOPE

The broad objective of the programme is to study the ability of natural organic compounds present in groundwater to form mobile complexes with radionuclides. The work involves development of techniques to extract natural organic material from groundwater with minimal alteration, the separation of those organic fractions that complex with cations, and the development of geochemical speciation models to describe or predict metal binding. The modelling approach will be tested by applying it to the study of the mechanisms which enhance the migration of nickel from a disposal lagoon into which a wide range of organics and heavy metals have been discharged. Liaison with Loughborough University of Technology has been established to develop appropriate techniques to determine radionuclide speciation directly and to separate organic fractions.

B. WORK PROGRAMME.

The project is divided into the following research areas:-

B.1 Assessment and development of techniques to extract and separate natural organic material from groundwater.

B.2 The separation, characterisation and quantification of those organic fractions which complex with radionuclides.

B.3 Development of a model to describe metal binding capacity based on the characterisation of natural organic compounds.

B.4 Investigation of the mobility of nickel which has apparently migrated over 100 m in groundwater organically contaminated with industrial wastes, in an attempt to apply and validate the modelling approach.

C. Progress of work and results obtained.

C.1 State of advance.

Phase one of the work has been completed. Humic materials have been extracted from groundwater using methodologies developed during phase 1. The effect of different methodologies on the ability of extracted organics to complex radionuclides will be studied within phase 2 of the work which will be commenced when characterisation of the extracted material has been completed.

C.2 Progress and results.

Assessment and development of techniques to extract and separate natural organic material from groundwater (B1).

Extraction separation and purification of natural organic material from groundwater is required to allow further characterisation and complexation studies. Many techniques exist for the extraction of such material however no one technique is applicable to all organics and from a practical aspect it is necessary to concentrate on the separation of particular classes of compounds. Acidic species such as humic and fulvic acids have been strongly implicated to be the more potent species with respect to metal complexation and development of techniques for their extraction and purification has been concentrated on.

Organic compounds containing acidic functional groups have been extracted from 600 l of groundwater (from the glacial sand aquifer at the Drigg research site) using the weak anion exchanger, DEAE cellulose. The DEAE cellulose containing extracted material was split into two portions (approximately 300g in each). Bound organic compounds were eluted from the first of these portions using the standard methodology (based upon 0.1 m NaOH) whilst similar compounds were eluted from the second portion using a 50:50 V:V mixture of 0.1M HCl and MeCN. Fractions containing organic matter were collected, purified and separated into Fulvic and Humic acids. The fulvic and humic acids produced from these processes are being characterised by standard methodologies prior to assessing their complexing ability relative to each other and the original water sample.

The separation, characterisation and quantification of those organic fractions which complex with radionuclides (B2).

The application of high performance size exclusion and reverse phase chromatography to the separation, characterisation and quantification of organic fractions in groundwaters has been further developed. The previously described problems encountered when using high performance size exclusion chromatography has been overcome and results obtained are now more reproducible, the application of this technique to quantify the degree of metal complexation has been investigated and shows promise. The use of ion pair reverse phase chromatography has been further improved by the purchase of a fluorescence detector which together with UV absorption data adds weight to the separated components being of derived from material of similar origin. The degree of resolution achievable using this methodology is much greater for fulvic acids than for humic acids which show a greater affinity for the stationary phase under similar chromatographic conditions.

Further development work will be carried out to investigate the use of size exclusion chromatography for determination of radionuclide speciation and the use of reverse phase chromatography to study the effect of extraction methodologies on naturally occurring humic material.

Development of a model to describe metal binding capacity based on the characterisation of natural organic compounds (B3).

An electrostatic interaction approach to binding between organic macromolecules and protons/cations has been incorporated successfully into the speciation code PHREEQE. Owing to the concept of speciation codes, the discrete ligand model had to be retained whereas the binding intensities are modified using an electrostatic (surface complexation) model. To allow for different conformations of natural organic material two alternative concepts have been implemented: a) it is assumed the organic molecules form rigid, impenetrable spheres, and b) the organic molecules can be represented as flat surfaces. The former concept will be more appropriate for molecules in the smaller size range, while the latter will be more representative for larger size molecules or organic surface coatings. A paper concerning the model (Falck 1989) has been given at the International Symposium on Humic Substances, Linköping, Sweden, in August and has been accepted after review for the symposium's proceedings. A more detailed paper, including listings of the modified subroutines of PHREEQE has been submitted to Computers & Geosciences (Falck subm.).

Investigation of the mobility of nickel which has apparently migrated over 100 m in groundwater organically contaminated with industrial wastes, in an attempt to apply and validate the modelling approach (B4).

Anaerobically sampled groundwater from the Villa farm has been shown to contain Ni, 60% of which is associated with a size fraction between 1,000 and 500 daltons (by ultrafiltration). Characterisation of Ni-organic complexes that may be present in this groundwater by reverse phase and size exclusion HPLC was complicated by the low concentration of Ni in the sampled water, to overcome this problem two approaches have been tried

(a) Further boreholes have been sampled to attempt to obtain groundwater containing Ni at concentrations observed in previous studies (>10ppm). This has been unsuccessful and none of the subsequently sampled groundwaters have Ni concentrations higher than 3 ppm. This may either be due to sorption of the aqueous Ni or to further dispersion of the plume.

(b) Additional Ni (10ppm) has been added to the anaerobically sampled groundwater used in the ultrafiltration studies These spiked samples have been analysed by reverse phase and size exclusion HPLC, correlating the UV absorbance of the eluant (measured at 254 nm) with its Ni content (measured using graphite furnace atomic absorption spectroscopy). Results from these experiments show that Ni is predominantly associated with polar organic species that exhibit UV absorbtion spectra similar to naturally occurring humic material.

D. REPORTS

FALCK, W.E. (1989): The Incorporation of Natural Organic Matter–Cation Interaction into the Speciation Code PHREEQE.- Presentation given at the Internat. Symp. on Humic Substances in Aquatic and Terrestrial Environ., Linköping, Sweden, August 21-23, 1989, paper accepted for proceedings.

FALCK, W.E. (subm.): Multisite Binding Equilibria and Speciation Codes: Incorporation of the Electrostatic Interaction Approach into PHREEQE.- Submitted to Computers & Geosciences.

Role of Colloids in the Transport of Radionuclides in Geological Formations

Contractors: UKAEA, Harwell, UK/CEA, Fontenay aux Roses, France.
Contract No: FI 1W/0204
Duration of Contract: from July 1988 to December 1989
Working Period: January 1989 - December 1989
Project Leaders: J D F Ramsay, A Billon

A. OBJECTIVES AND SCOPE

Recent research has emphasised the potential importance of colloids in the migration of radionuclides from a waste repository into the geosphere /1,2,3/. Thus, if radionuclides are associated with colloids their fate may be different from that of simple ionic species in solution. In particular, depending on the size and charge of colloid species retention effects may occur due to filtration, or in contrast there may be enhanced migration due to differences in flow mechanisms and a reduction in the interaction with mineral surfaces.

Our objective here is to improve the understanding of the role of colloids in radioactive waste disposal. This is being achieved firstly by detailed characterisation of natural colloids derived from groundwaters, typical of those in a granite geology proposed for waste disposal, and secondly by investigating the interaction of natural and synthetic colloids (as model systems) with granite surfaces which have been characterised petrographically. This should provide an insight into retention mechanisms and would lead to subsequent investigations of the transport and hydrodynamic behaviour of colloids in these and other geological systems. Here expertise and techniques developed at both Harwell and Fontenay are being applied in a collaborative investigation.

B. WORK PROGRAMME

B.1 Preparation and characterisation of synthetic colloids.
B.1.1 Preparation of silica, iron oxide, alumina and humic materials.
B.1.2 Characterisation by light scattering, electrophoresis and electron microscopy.
B.2 Characterisation of natural colloids occuring in groundwaters.
B.2.1 Characterisation of host rock.
B.2.2 Characterisation of groundwater.
B.2.3 Characterisation of natural colloids by light scattering techniques, ultrafiltration and electron microscopy.
B.2.4 Isolation of organic colloids.
B.3 Studies of radiocolloid retention processes.
B.3.1 Association (incorporation, sorption) of actinides and rare earth elements with natural/synthetic colloids.
B.3.2 Interactions of colloids with well-characterised mineral surfaces.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

The main emphasis of work in the year has involved a detailed study of the mechanisms of retention of colloids onto mineral surfaces using Rutherford Back Scattering (RBS) techniques. Such novel investigations are of fundamental importance for establishing the potential role of colloids in radionuclide migration and provide a basis for developing predictive transport models. We have used a combination of techniques in the characterisation of a range of model colloid systems (ceria, silica, iron oxide, montmorillonite clay, Th(IV), U(VI)) and also in the pretreatment and detailed examination of pure mineral surfaces. It has been demonstrated that the uptake of colloids onto a mineral surface is marked when the surface charges are opposite. This leads to the formation of a densely packed monolayer. When the surfaces are of the same sign the uptake is considerably lower or negligible. The latter situation may pertain when "pseudocolloids" such as silica associated with radionuclides are in contact with mineral surfaces such as mica and quartz. In contrast "real" colloids, which arise from metal ion hydrolysis and polymerisation and which in general have a positive charge in natural groundwaters, have a high affinity for such minerals.

The RBS technique, which is highly sensitive and quantitative, is being extended to investigate retention from extremely dilute solutions of rare earth and actinides both in the colloidal and ionic state. Future investigations of the kinetics of adsorption/desorption processes are also planned to provide data for colloid transport models.

Important advances are also reported in the study of sorption of the actinides Np and Am onto characterised silica colloids. Here the marked difference in behaviour reflects the greater tendency of Am^{3+} to hydrolyse in solution compared with NpO_2^+. Such differences in sorption onto colloids are also noted with Th^{4+} and UO_2^{2+} and are also evident in the retention of the solution species onto mineral surfaces.

PROGRESS AND RESULTS

1. Preparation and Characterisation of Synthetic Colloids

Model or synthetic colloids have been characterised by a variety of techniques including PCS (photon correlation spectroscopy), neutron scattering, ultracentrifugation, microelectrophoresis, and TEM (transmission electron microscopy). The colloids, together with their properties (particle size, point of zero charge, surface area), are as follows:

(i) ceria, CeO_2 (7nm, pH ~6),
(ii) silica, SiO_2 (80nm, pH <2, 50 m^2 g^{-1}),
(iii) iron oxide (haematite), Fe_2O_3 (~80nm, pH 8.2),
(iv) montmorillonite clay-fine fraction (19-35nm).

The synthetic colloids are spherical particles with a narrow size distribution. The montmorillonite was obtained as a stable dispersion after a lengthy fractionation and purification procedure involving dialysis and ultracentrifugation. These colloids were used in the studies of radiocolloid retention processes (Section 3).

3. Studies of Radiocolloid Retention Processes

3.1 Association of Actinides with Synthetic Colloids

Preliminary investigations have established appropriate experimental procedures which are necessary for measurements of actinide uptake onto colloids at low concentrations in solution. Such measurements are complicated by actinide removal onto container walls for example. The sorption of Am(III) and Np(V) from solution onto a model silica colloid (80nm particle size) has been investigated as a function of pH at a fixed ionic strength (0.1 mol dm^{-3} $NaClO_4$). Figure 1 shows how the presence of the colloidal silica affects the removal of Am and Np by ultracentrifugation at different values of pH. With Np removal remains low and is independent of pH, although removal appears to be higher in the presence of the silica colloid which can be ascribed to limited sorption. In contrast the removal of Am increases markedly above a pH of 5, both in the presence and absence of silica colloid. In the absence of silica such an effect can be ascribed to the hydrolysis and formation of polynuclear Am(III) species which are sedimented on high speed ultracentrifugation. The presence of colloidal silica enhances the removal of Am probably due to sorption or retention of small polynuclear species with the silica surface.

3.2 Interactions of Colloids with Well-Characterised Mineral Surfaces

The interaction of inorganic colloids with mineral surfaces typical of the constituents of granite has been investigated to establish the mechanisms of retention of colloids by such surfaces. Several pure, well-defined mineral phases (muscovite mica, untreated and prehydrated silica, and haematite) have been exposed to well-characterised synthetic model colloids (both real and "pseudo"). The prehydrated mineral silica was prepared by bombardment with inert gas ions and subsequent water attack to give a gel layer. In addition some samples of the mica and silica were modified by pretreating them with an iron oxide (haematite) sol before exposing them to a second colloid. Sorption and retention of the colloids was then studied using charged particle backscattering techniques (RBS, RNRA).

Tables 1 and 2 list the mineral phases and give details of the various colloids used and the treatment conditions. They also include estimates derived from the RBS data of levels of retention by the mineral surfaces of the appropriate colloid elements. The pseudocolloids used included ceria-coated silica, U(VI)-coated iron oxide, Th-coated silica and Th-coated montmorillonite. These were prepared by pretreating the relevant model colloid with a second colloid (CeO_2) or ionic solution of a heavy metal (UO_2^{2+}, Th^{4+}, as nitrates). Details of the model colloids and their characterisation are given in Section 1. Some mineral specimens were exposed directly to very dilute, ionic solutions of $UO_2(NO_3)_2$ or $Th(NO_3)_4$, under pH conditions where polymeric or colloidal species might arise as a result of hydrolysis.

The observed retention levels (Tables 1 and 2) are generally consistent with the formation of a monolayer of colloid on the mineral surface. The presence of a hydrated gel layer on the silica leads to increased retention of both the real colloid ceria and the pseudocolloid ceria-coated silica. This probably reflects the larger surface capacity that should be available for sorption. The sorption of ceria alone is roughly an order of magnitude greater than that of the silica/ceria pseudocolloid (Table 1). Such a difference reflects the importance of the surface charges of both the mineral phase and colloid species in controlling the level of colloid retention on the mineral. Ceria is positively charged while silica (mineral or colloid) is negatively charged. Similar effects are observed with the mica (Table 2) which is also negatively charged.

Th(IV) is sorbed more readily than U(VI) by the minerals silica and mica as might be expected from its larger ionic charge and the possibility of polynuclear species at pH ~ 3.7. The level of retention of the Th increases as the pH is increased in accord with the tendency for Th(IV) to hydrolyse and form polymeric or colloidal species. The RBS spectra in Figure 2 show the increasing retention of the Th by mica as the pH is increased. The tail towards lower energy in the Th peak at pH 7 indicates multilayer formation.

Retention of the pseudocolloid silica/Th by the mineral haematite was considerable (Table 2). At the lower pH (3.8) the colloid and haematite are oppositely charged (+ve and -ve respectively). At the higher pH (9.1) the haematite is slightly above its point of zero charge (~8.2) and, although the consequent electrostatic repulsion is not adequate to prevent some retention, the level is lower than at pH 3.8.

List of Publications

/1/ RAMSAY, J.D.F., The role of colloids in the release of radionuclides from nuclear waste, AERE R 11823 (1985).

/2/ RAMSAY, J.D.F., The role of colloids in the release of radionuclides from nuclear waste, Radiochimica Acta 44/45(1), 165 (1988).
/3/ RAMSAY, J.D.F., AVERY, R.G. and RUSSELL, P.J., Physical characteristics and sorption behaviour of colloids generated from cementitious systems, Radiochimica Acta 44/45(1), 119 (1988).
/4/ BILLON, A., CACECI, M., DELLA MEA, G., DRAN, J-C., MOULIN, V., PETIT, J.C., RAMSAY, J.D.F., RUSSELL, P.J. and THEYSSIER, M., The role of colloids in the transport of radionuclides in geological formations - Progress report for the period 1st January to 30th June 1989, AERE R13691 (1989).
/5/ BILLON, A., DELLA MEA, G., DRAN, J-C., MOULIN, V., PETIT, J-C., RAMSAY, J.D.F., RUSSELL, P.J. and THEYSSIER, M., Proc. of Conference "Migration '89" Monterey, CA.1989 (to be published in Radiochimica Acta).

Table 1
Quantitative assessment from RBS data of the extent of sorption of various colloid elements by mineral SILICA

Colloid Treatment	Element Measured	Observed Retention Level (atom cm^{-2})	
		(a) Untreated Silica	(b) Prehydrated Silica
CeO_2,100μg ml^{-1};pH~6	Ce	1 x 10^{16}	1.6 x 10^{16}
SiO_2,500μg ml^{-1}/CeO_2,60μg ml^{-1};pH~6.1	Ce	2 x 10^{14}	1 x 10^{15}
U(VI),10^{-4} mol dm^{-3};pH~4.7	U	2.1 x 10^{14}	9 x 10^{13}
(i)Fe_2O_3;(ii)U(VI),10^{-4} mol dm^{-3};pH~4.7	U	5.4 x 10^{14}	2.7 x 10^{14}
Fe_2O_3,50μg ml^{-1}/U(VI),10^{-4}mol dm^{-3};pH~5	U	~3 x 10^{14}	~3 x 10^{14}
Th,2x10^{-4} mol dm^{-3};pH 3.7	Th	1 x 10^{15}	3.7 x 10^{14}
Th,2x10^{-4} mol dm^{-3};pH 5.1	Th	7.1 x 10^{15}	3.2 x 10^{15}

Table 2
Quantitative assessment from RBS data of the extent of sorption of various colloid elements by mineral phases MICA (muscovite) and HAEMATITE

Mineral Phase	Colloid Treatment	Element Measured	Retention Level (atom cm^{-2})
Mica	CeO_2,100μg ml^{-1};pH~6	Ce	7.5 x 10^{15}
Mica	SiO_2,500μg ml^{-1}/CeO_2,60μg ml^{-1};pH~6.1	Ce	1.8 x 10^{14}
Mica	U(VI),10^{-4}mol dm^{-3};pH~4.7	U	1 x 10^{14}
Mica	(i)Fe_2O_3;(ii)U(VI),10^{-4}mol dm^{-3};pH 4.6	U	2.2 x 10^{14}
Mica	Fe_2O_3,50μg ml^{-1}/U(VI),10^{-4}mol dm^{-3};pH~5	U	5 x 10^{14}
Mica	Th,2x10^{-4}mol dm^{-3};pH 3.7	Th	1 x 10^{15}
Mica	Th,2x10^{-4}mol dm^{-3};pH 5.1	Th	3.6 x 10^{15}
Mica	Th,2x10^{-4}mol dm^{-3};pH 7	Th	7.9 x 10^{15}
Mica	Mont.,1000μg ml^{-1}/Th,10^{-5}mol dm^{-3};pH 3.7	Th	not detected
Haem.	SiO_2,500μg ml^{-1}/Th,10^{-4}mol dm^{-3};pH 3.8	Th	3.6 x 10^{15}
Haem.	SiO_2,500μg ml^{-1}/Th,10^{-4}mol dm^{-3};pH 9.1	Th	1.8 x 10^{15}

Haem. = haematite; Mont. = montmorillonite.

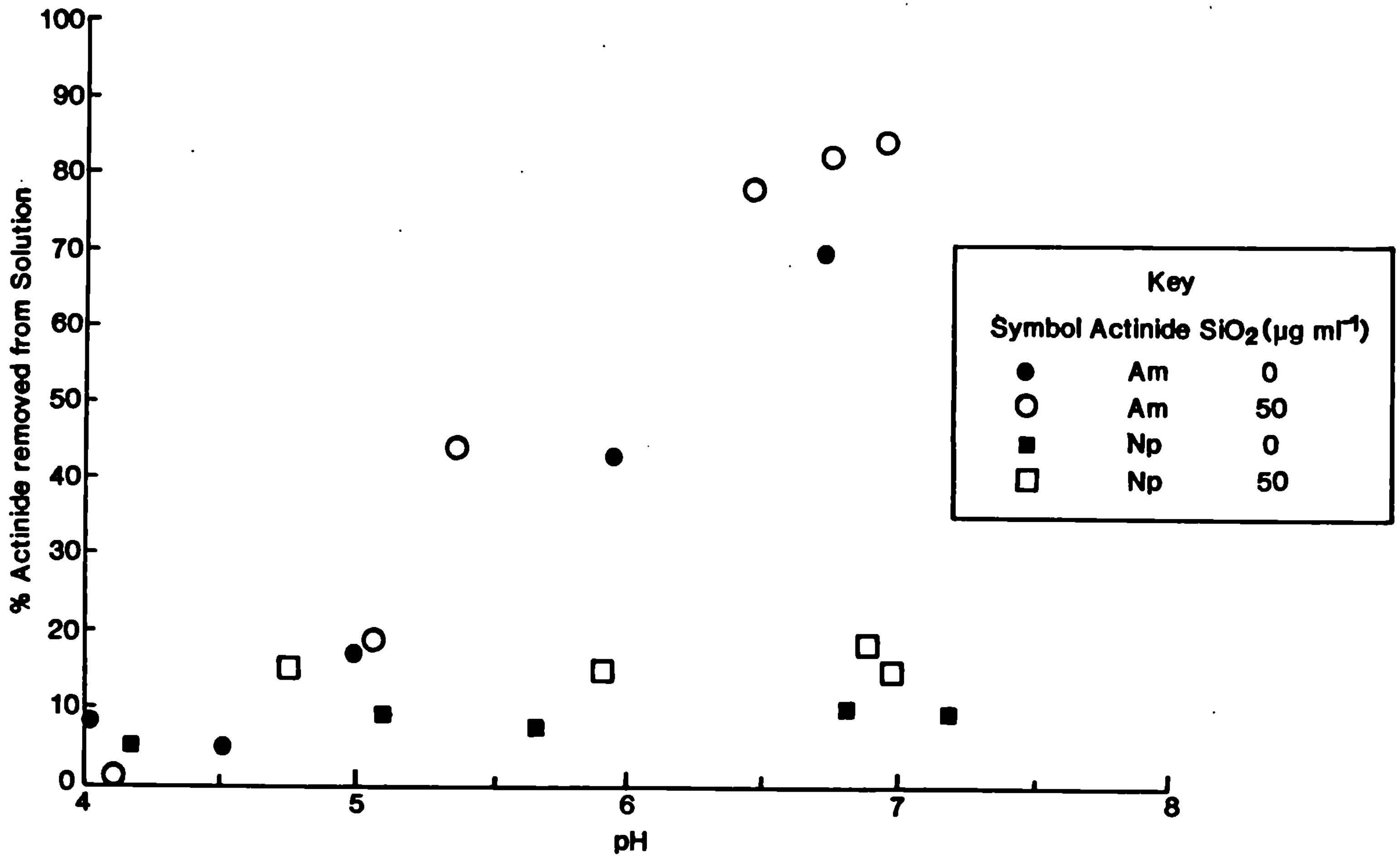

FIG.1. COMPARISON OF THE EFFECT OF pH AND THE PRESENCE OF SILICA COLLOID ON THE REMOVAL OF Am (10^{-8} mol dm^{-3}) AND Np(7×10^{-6} mol dm^{-3}) FROM SOLUTION AT FIXED IONIC STRENGTH

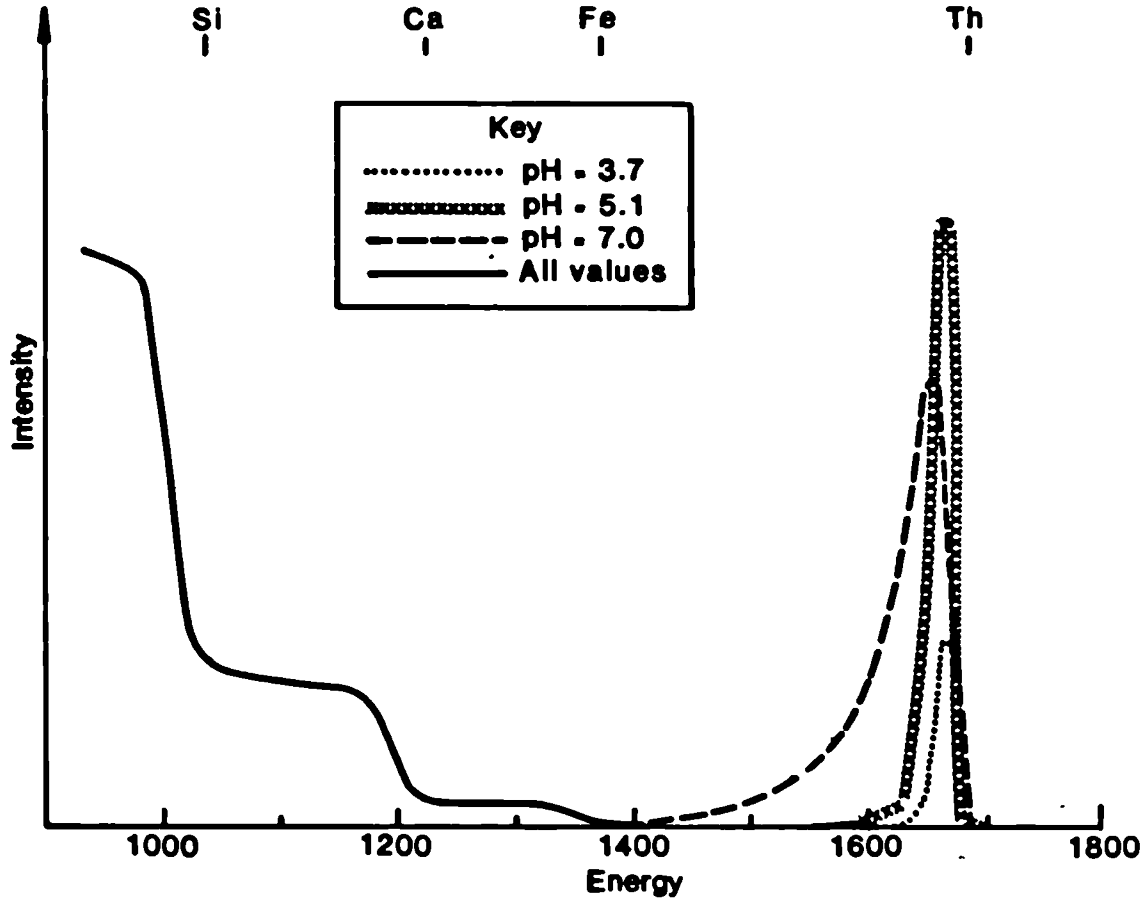

FIG.2. SIMPLIFIED RBS SPECTRA SHOWING THE SURFACE RETENTION OF Th BY MICA (MUSCOVITE) AS A FUNCTION OF pH

ASSESSMENT OF EXPERIMENTAL RESEARCH TECHNIQUES FOR THE INVESTIGATION OF RADIONUCLIDE MIGRATION IN AQUIFERS

Contractor: GSF, Neuherberg, Federal Republic of Germany
Contract No.: FI1W/0210
Duration of contract: from August 1988 to July 1990
Period covered: January 1989 - December 1989
Project leaders: P. Fritz, C. Wolfrum

A. OBJECTIVES AND SCOPE

The transport of radionuclides released from a repository to a surrounding aquifer is the key problem in the safety analysis of waste disposal in geologic media, it requires detailed knowledge of aqueous chemistry, mineralogy and specific mechanisms of solid/solution interaction. The primary objective of this project is to advance the understanding of the transport behaviour of complexes and colloids in aquifers. The study has been subdivided into two components: a) a comparison of laboratory techniques in migration studies used with the radionuclides 99-Tc, 152-Eu, 238-Pu and b) investigations into the relation between redox condition and formation of complexes with natural organic macromocules as well as generation of colloids. Based on experimental data a compilation and comparison of literature sorption data from natural systems will be undertaken.
In the present stage of the project, experiments focus on the influence of colloids and humic substances on the results of batch, diffusion and column tests. Of special interest was to study the behaviour of complexed radionuclides under the dynamic transport conditions.

B. WORK PROGRAMME

1. For comparison of the different experimental methods a series of laboratory tests were perfomed, parallel to the compilation of literature sorption data.

2.1. The experimental work was continued with samples from the Gorleben site. Experiments with Drigg trench sand and the corresponding natural water are still in progress.

2.2. Experimental procedures were the same as before. Behaviour of radionuclides both in pure groundwater and in contact with the corresponding sediment was investigated.

2.3. Sorption behaviour of 238-Pu depending on the contact time solid/solution and the equilibrium values for 85-Sr, 95-Zr/Nb and 152-Eu were determined in diffusion and batch tests.

2.3.2 The stability behaviour of 99-Tc and 238-Pu was studied in various Gorleben groundwater samples.

2.3.3 The influence of HA concentration on the Rs/Rd equilibrium values for 152-Eu and 85-Sr under aerobic conditions was investigated.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The study will take the dual approach of a) a critical comparison of laboratory techniques used in investigations of radionuclide migration in aquifers supported b) by own experiments.The work was continued with sediments and groundwaters from the Gorleben site which have already been well characterized with the aim to interpret and compare sorption results. The experiments focus on the radioisotopes of Tc, Eu, Pu,(Sr),(Zr),(Nb) and investigate the essential influences on the transport behaviour with regard to redox conditions, formation of complexes with natural organic molecules as well as formation and/or presence of colloids.

Examplary for the Eu^{3+} interaction with organic matter purified Gorleben humic acid was added in different concentrations to several sediment-groundwater-systems. In addition sorption experiments dealing with the effects of an artificial organic ligand (EDTA) on the Rs values of Eu were performed. Sorption coefficients for Sr, Nb, Zr and Eu determined in diffusion and batch experiments are compared.

Column tests were performed to get a qualitative description of the transport behaviour of Eu compared with that of Na, Cl, Ca and Sr. It was found that no measurable amounts of the complexed or colloidal or Eu pass the column. This will be discussed by considering various competetive processes, which have influence on the mobility of Eu^{3+} under conditions encountered in natural systems.

Program item 1 will be adjusted in the end of the report.

Progress and results

2.1, 2.2 SEDIMENTS, GROUNDWATERS AND EXPERIMENTAL PROCEDURES

The Gorleben sample description with analytical and physicochemical data are given in /1/-/3/. Experiments with Drigg trench sand and the corresponding natural water are still in progress. All experimental procedures such as sample storage and equilibration, filtration of groundwaters, addition of radionuclide solutions, realization of batch and diffusion tests with handling in glove boxes for anaerobic conditions and so on have been retained as described in earlier reports.

2.3 SORPTION EXPERIMENTS WITH REGARD TO KINETIC, EQUILIBRIUM AND THE EFFECT OF REDOX CONDITIONS

Sorption behaviour was investigated for different sediment-groundwater-systems from the Gorleben site in batch tests so as to obtain Rs values of 238-Pu under anaerobic conditions with regard to the contact time between solid and solution. In a time range of 20 weeks the sorption coefficients for the Gohy samples 8/1281, 9/1281, 9/1341 and 17/641 were monitored and found to be constant (Table I). The Rs values of 238-Pu agree with the data obtained by KIM /3/.

The dependance of the sorption/desorption equilibrium values of 152-Eu on the carrier $Eu(NO_3)_3$ was studied in the concentration range between 10^{-8} and 10^{-2} mol/l. For the sediment-groundwater-systems 4/2131, 5/1281 and 9/1341 the Rs/Rd values decrease with increasing Eu carrier concentration. This result can be interpreted in case of ion exchange as a competetive effect. In the case of precipitation sorption coefficients should increase with concentration.

2.3.2 SORPTION EXPERIMENTS WITH REGARD TO THE EFFECT OF COLLOIDS

To test formation and behaviour of colloids under anaerobic conditions, 99-Tc and 238-Pu were added to the Gohy groundwater samples 641, 1281 and 1341 as the compounds

- NH_4TcO_4 in a concentration of $1 \cdot 10^{-6}$ mol/l and
- $PuO_2(NO_3)_2$ in a concentration of $1 \cdot 10^{-10}$ mol/l.

In fixed time intervals and during about three months small sample volumes were filtered through 450 and 220 nm cellulose acetate filters. During this period the ß-activities of 99-Tc in the filtrates were constant within the time range, in contrast to the initial solutions. Filtration of the Gohy groundwaters through 450 and 220 nm pore size does not alter the Rs/Rd values for 99-Tc within the the error limits.
Accordingly, no colloidal influences were observed in the investigated sediment-groundwater systems. The stability behaviour of 238-Pu differs in the various Gohy groundwater samples. In the water 1341 no formation of Pu-colloids is recognizable, the effect in water 641 is small (c/co= 0.9 at t= 100 d), whereas in water 1281 a time dependant generation of Pu-colloids has been detected. Figure 1 shows the filtration curve (450 nm), whereby the ratio of c/c_0 (with c: radionuclide concentration after filtration, c_0: radionuclide concentration added) is plotted as function of time.
Column tests were performed to get a qualitative description of the transport behaviour of 152-Eu compared with that of 35-Cl, 22-Na, 45-Ca and 85-Sr. When 250 cm^3 of groundwater volume were pumped through the column) with the relatively high flow rate of 1 cm^3/h (to force the transport of particles) Eu was fixed strongly within the column (Figure 2). As shown in filtration experiments up to 80 % of the raionuclides added (Zr, Nb, Eu) should be present in colloidal forms with a particle size smaller than 450 nm. Taking into account that filtration of sediments will be in the same range, it was expected that larger portions of the colloidal or complexed radionuclides pass the column without significant retardation. This effect was not observed. On the contrary, within three weeks only about 1 % of 152-Eu added could be detected in the effluent of the column. The bulk of this radionuclide is sorbed onto the sediments. Various competetive processes may have considerable influence on the mobility of Eu under conditions encountered in natural systems:

- complexation by inorganic anions (e.g. carbonate, phosphate, chloride, sulfate, fluorite),
- complexation by organic compounds (e.g. amino, humic, fulvic acids),
- precipitation (at high concentrations),
- sorption on surfaces (including suspending particulates),
- hydrolysis.

Each of the possible Eu complexes will have a different mobility depending both on size and sorption characteristics.

2.3.3 SORPTION EXPERIMENTS WITH REGARD TO THE EFFECT OF NATURAL AND MAN MADE ORGANICS

In natural aquifer systems it is assumed that the migration of metal ions of higher valence states will be influenced by complexation with organics, especially humic and fulvic acids. They are thought to be responsible for a faster transport. As an example for the Eu^{3+} interaction with organic matter the purified Gorleben humic acid Gohy 573-HA (H^+) (characterization in /4/) was added in concentrations of 1, 10 and 100 mg/l to the samples 4/2131, 2/1281, 9/1281 and 9/ 1341. For comparison, a cation with a simple chemistry (Sr^{2+}) was investigated in the same batch tests. The 152-Eu-Rs/Rd values for the samples 4/2131 and 9/1341 are almost independant from the HA content. In the first case (Gohy 2131) the reason could be found in a high Fe^{2+}/Fe^{3+} content (11.3 mg/l from /2/) and hence the presence of a cation competetive to Eu with regard to the formation of HA complexes. In the second one (Gohy

1341) the high salinity (about 21 mS/cm from /2/) might have been responsible. The influence of HA on 85-Sr-Rs/ Rd values is negligible.
As Eu forms also strong complexes with man made organic ligands the influence of di-Na-EDTA in a concentration range between 10^{-8} and 10^{-3} mol/l was studied in two examples (4/1281, 9/ 1281). Under the given experimental conditions, the sorption/desorption coefficients decrease considerably.

1. COMPARISON OF DIFFERENT EXPERIMENTAL METHODS

For the Gohy systems diffusion experiments with 85-Sr, 95-Zr, 95-Nb and 152-Eu were performed under aerobic conditions. Selected were a) Sr^{2+} because of its simple chemistry, b) Zr^{4+} whose valence state does not change and which is thus an interesting element to study colloid formation and behaviour and c) Eu^{3+} as an analogue to Am^{3+}. Diffusion coefficients were calculated from breakthrough curves (Ficks first law - considering the steady state) and on the basis of Ficks second law (describing the non-steady state). Examples are graphically presented in Figures 3 and 4. In Table 2 Rs values determined in diffusion and batch experiments are compared. The data agree quite well.
However, comparison of the static (batch) with the dynamic (column) technique is much more problematic because of the various boundary conditions which have to be taken into account. These are in batch tests the dependance of sorption values on the volume/mass ratio as well as the nuclide concentration (resulting in different sorption/desorption reactions). Thus, the results of batch experiments with complexed radionuclides show that there is also an interaction of ligands with natural cations /5/. Therefore, in future work not only the concentration dependance of nuclide sorption but also the behaviour of complexing agents must be considered if a comparison between batch and column results is undertaken. To interpret the behaviour of colloid forming nuclides, the correlation between the solubility of nuclides and colloid generation has to be addressed. Experiments have shown that colloid concentration is lowered drastically if groundwater is in contact with sediment and if nuclide concentration dedecreased. In addition in the case of Zr and Eu it was found that no colloids have passed "ideally" even small columns. These experimental results seem to be contradictory and demonstrate the difficulties.

List of publications

LANG, H. and WOLFRUM, C.: Nuclide Sorption on Heterogeneous Natural Surfaces. Water-Rock Interaction WRI-6 (proceedings of the 6th international symposium, Malvern, U.K.) ed. D. L. Miles, A. A. Balkema, Rotterdam 1989, 417
WOLFRUM, C. and LANG, H.: Considerations on Sorption Behaviour of Colloids in Natural Systems. Water-Rock Interaction WRI-6 (proceedings of the 6th international symposium, Malvern, U.K.) ed. D. L. Miles, A. A. Balkema, Rotterdam 1989, 781
LANG, H. and WOLFRUM, C.: Mechanismen der verzögerten Ausbreitung radioaktiver Stoffe unter wassergesättigten Bedingungen. Kolloquium für physikalische und chemische Arbeitsmethoden in der Hydrologie, Ludwig-Maximilians-Universität München, 5.12.1989

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/3/ GRIMM, W.-D., Sedimentpetrographische Untersuchungen an 12 Sedimentproben aus dem Raum Gorleben, unpublished report, München (1988)
/4/ KIM, J.I., BUCKAU, G., Characterization of Reference and Site Specific Humic Acids, RCM-report 01588, München 1988
/5/ FALCK, W.E., A Review of Modelling the Interaction between Natural Organic Matter and Metal Cations, BGS Technical Report WE/88/49, Keyworth (1988)

TABLE I: SORPTION COEFFICIENTS Rs FOR 238-Pu AFTER 6, 9 AND 20 WEEKS CONTACT TIME IN GORLEBEN SAMPLES UNDER ANAEROBIC CONDITIONS.

Gohy sediment	Gohy water	V/m (cm^3/g)	pH	Eh (mV)	Pu R_s (cm^3/g) 6	9	20 weeks
8	1281	3.0	7.8	+201	154	150	149
9	1281	3.0	7.9	+188	182	155	163
9	1341	3.0	7.5	+233	258	191	199
17	641	4.0	7.5	+117	87	72	80

TABLE II: COMPARISON OF SORPTION COEFFICIENTS Rs FOR 85-Sr, 95-Zr, 95-Nb AND 152-Eu DETERMINED FROM DIFFUSION AND BATCH EXPERIMENTS UNDER AMBIENT CONDITIONS.

Gohy samples	R_s (cm^3/g) (diffusion) Sr	Zr	Nb	Eu	R_s (cm^3/g) (batch) Sr	Zr	Nb	Eu
6	(16)	57	73	(1110)	2.7	42	78	700
7	1.1	-	24	20	0.9	14	21	-
8(2)	5.3	-	21	-	5.5	190	390	220
9(2)	4.5	317	-	-	3.4	37	310	180
10	3.8	95	53	-	3.0	59	51	-
11	1.3	44	63	-	0.4	30	67	-

- not evaluable
() poorly evaluable

FIGURE 1: FILTRATION CURVE - c/c_0 OF ^{238}Pu IN GOHY GROUNDWATER 1281 AS FUNCTION OF TIME t WITH

c_0: RADIONUCLIDE CONCENTRATION ADDED

c : RADIONUCLIDE CONCENTRATION AFTER FILTRATION BY 450 nm

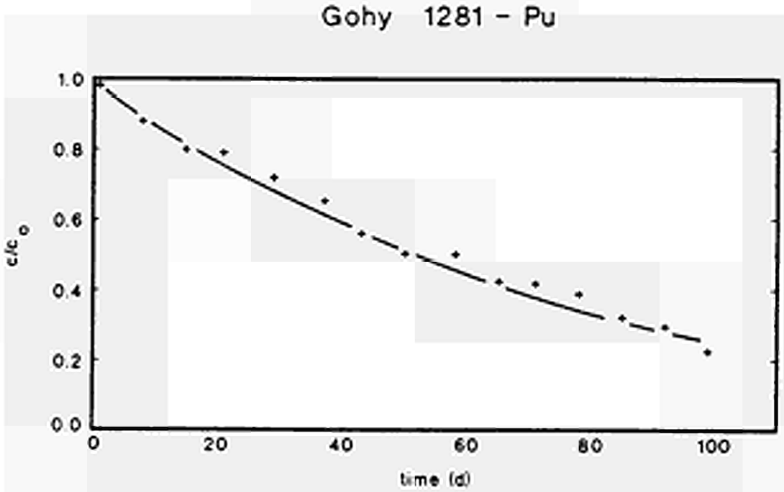


FIGURE 2: DISTRIBUTION OF 152-Eu IN THE GOHY SAMPLE 5/1281 AT THE END OF THE FLOW THROUGH COLUMN EXPERIMENT.

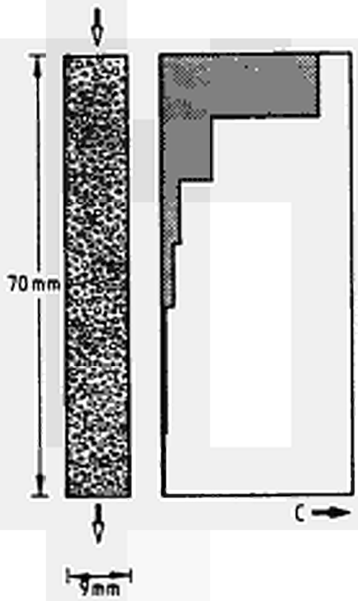


FIGURE 3: Sr^{2+}-DIFFUSION THROUGH THE GROUNDWATER-EQUILIBRATED SEDIMENT GOHY 8/1281.
EVALUATION ACCORDING TO FICKS FIRST LAW: INCREASE OF THE Sr^{2+}-CONCENTRATION c_i IN THE INITIALLY INACTIVE RESERVOIR AS A FUNCTION OF TIME t.

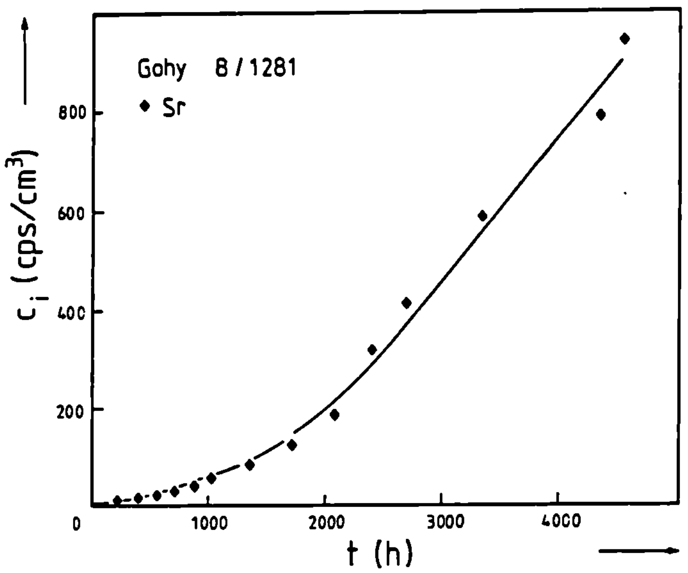
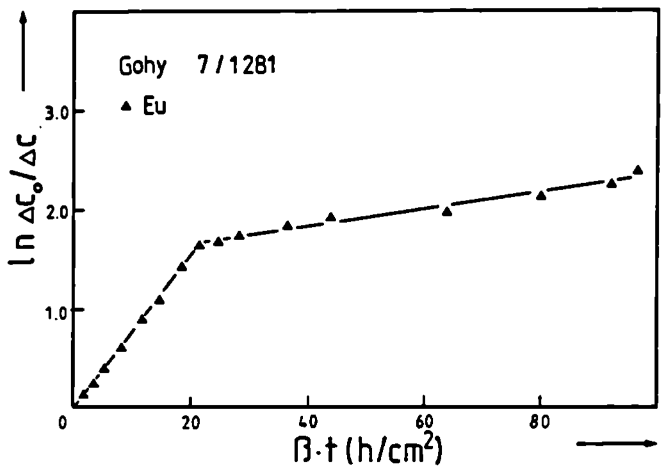


FIGURE 4: Eu^{3+}-DIFFUSION THROUGH THE GROUNDWATER-EQUILIBRATED SEDIMENT GOHY 7/1281.
EVALUATION ACCORDING TO FICKS SECOND LAW: $\ln \Delta c_o/\Delta c$ WITH Δc_o THE INITIAL AND Δc THE TIMEDEPENDENT CONCENTRATION DIFFERENCE OF Eu^{3+} BETWEEN THE TWO RESERVOIRS AS A FUNCTION OF $\beta \cdot t$.



CHARACTERIZATION OF ACTINIDES, COMPLEXES AND COLLOID MIGRATION IN GRANITE (EL BERROCAL)

Contractor: ENRESA, MADRID, Spain
Contract No.: FI1W230
Duration of Contract: December 1988-November 1990
Period covered: January 1989-December 1989
Project Leader: Julio Astudillo

A. OBJECTIVES AND SCOPE

The objective of the study is to characterize the migration processes in a fissured granitic formation by means of in situ and comparative laboratory experiments.

The studies will be conducted at El Berrocal uranium mine, located in a granitic batholith of the "Sistema Central" of Spain.

After an extensive characterization of the El Berrocal batholith, the natural migration processes of uranium from the intergranitic uraniferous veins and its desintegration products will be studied by sampling and analysing the natural colloids and the groundwaters. The objectives of this phase are to clarify the migration processes which have naturally occurred, as well as the system explanatory model, so that the sensibility of all different parameters involved in the radionuclides transport can be elucidated.

Granite and fissure material samples will be used in laboratory columns, under oxic and anoxic conditions, to investigate the migration of uranium in fissures and its diffusion through the rock matrix. The results will be compared with those of the in situ observations.

In parallel with the natural migration studies in the Berrocal, stability complexes and colloids analysis will be developed with natural uranium-humic acid complexes obtained from uraniferous lignites.

B. WORK PROGRAMME

1. Characterization of the Berrocal granitic Batholith: Full geological characterization; sampling from boreholes, galleries and mineralized fissures; physical parameter and natural radionuclide analysis and mineralogical and geochemical characterization.
2. Characterization of the liquid and colloidal environment: Sampling and analysis for ionic and colloidal phase (filtration, dyalisis, size distribution and in situ measurements of the physico-chemical parameters).
3. Stability analysis of actinide humic acid colloids and complexes: Isolation and characterization of uranium humic acids of other spanish sites detailed characterization and stability analysis.
4. Laboratory migration experiments: Development of column migration apparatus, with preliminary experimentation, migration experiments in oxic and anoxic conditions in rock matrix and fissures in several physicochemical conditions.
5. Data analysis using conventional models for chemical equilibrium and transport, to compare laboratory and in situ migration results.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

During last 1989, the characterization of the Berrocal Batholith was concluded (task 1). The colloidal phases, inorganic and organic was sampled and the detailed characterization is now going on. Sampling and analysis of ionic phase has already been concluded (task 2).

The urano-organic (humic) complexes were obtained from spanish uraniferous lignites. These complexes have been characterized and the stability studies are also going on (task 3).

Several granite columns have been tested in laboratory, for the migration experiments. The Berrocal groundwater and urano-humic complexes have been used in the preliminary experiment. At present, the specific conditions for the experiments, and the characteristics of the granitic columns have been stablished (task 4).

In order to perform all these activities, 20 meters of the new gallery were excavated, and 12 boreholes were drilled, two of them next to the "Tarica gulley" (70 and 46 meters) and the rest, a total of 205 meters have been drilled inside the gallery (Fig. 1).

Geophysical and hydrogeochemical testing were performed. At the Berrocal working gallery all the equipment for the sampling, ultracentrifuges concentrator and resin concentrator and other instrumentation have been installed.

Progress and results

1. Characterization of the Berrocal site

Uranium behaviour and distribution: The mobility and spatial distribution of uranium was determined by selectively leaching, in six phases from easily interchangeable uranium to retained in refractory minerals. In figure 2 the results are presented.

The area closest to the vein presents the highest concentration of mobile uranium in the form of uranyl phosphates and oxi-hydroxides of iron. The total uranium in this area is lower due to the intense circulation of fluids. Meteoric waters reach these levels with pH values between 4 and 6, making it easy for the uranium leaching. In the intermediate zone of the gallery, up to 70% of the uranium is retained in goethytes and clay minerals. In the area, away from the ore vein, the uranium (75-90%) is associated with refractory and primary minerals.

Porosity and permeability: These parameters were determined in fresh and altered granite, with mercury porosimeter, permeameter and by empirical means. The porosity ranged from 0,47% in the fresh granite to 4,8% in altered granite (Table 1). The equivalent permeability obtained by geophysical methods is $6,7.10^{-10}$ m/s. A variation of permeability on core samples is not observed with depth. There is a network of interconnected channels along which the fluids move; these channels are, generally, limits between grains and microfissures in the grains themselves. Two types of voids have been observed: one kind of more than 1µm and the second type of less than 1µm of diameter, representing the greatest part of the total pore volume.

2. <u>Characterization of liquid and colloidal environment</u>

The groundwater samples were obtained from Berrocal boreholes, after the cleaning, two months later of the drill operation. The groundwater was pumped directly to the analytical equipment under nitrogen atmosphere for the in situ testing. Other samples were sent to the conventional laboratory. At the same time, physico-chemical profiles were obtained in situ. The results indicate that for the nº7 borehole, a clear ox-red boundary is observed (Fig. 3). The analyses of hydrogeochemical data, physico chemical profiles, geophysical and lithological logs will make it possible to select the most appropriate areas for sampling and in situ migration test.

The colloid fraction was concentrated in an Amicon CH2A through a hollow fibre tangential filter which cut-off was 30.000 M.W. Preliminary analytical data are presented in Table 2.

The results of HPSEC showed three peaks corresponding to a molecular weight of 900, 250 and 100 Daltons. The IR spectrum of dried sample of/ concentrated water indicates a similar pattern to those of humic substances in the region 4000-1400 cm^{-1}, due to H-bonded OH stretching (3429 cm^{-1}), aliphatic C-H (2925 and 2857cm^{-1}), aromatic C=C conjugated with C=O and/or COO (1625 cm^{-1}) and bending vibrations of aliphatic C-H groups (1427 cm^{-1}).

The purified humic acids from the Berrocal soils give molecular weights that vary from 9,000 to 26,000 Daltons. In both the CIEMAT and JRC (ISPRA) laboratories, the colloidal fraction of the lignites was extracted in columns, under both oxic and anoxic conditions respectively, using distilled water and synthetic water (Allard water).

3. <u>Analysis of actinides, humic acids, colloids and complexes</u>

The extraction and characterization of uranium-bearing humic complexes from lignites, which will be used for stability studies and for column migration tests, is almos finished.

Extraction of the humic substances was carried out using alkaline solvent (NaOH 0.1M) and later precipitating the humic fraction by lowering the pH to 1. Fulvic acids were concentrated with amberlite XAD-8 and purified with DOXEX-H+. The results indicate that the fraction of humic acids represents between 20-30% W/W, and that approximately 10-30% of the uranium present is complexed with them.

The fulvic acids represent between 30-30% W/W, complexing between 25% and 30% of the uranium. A loss of up to 60% of free uranium occurs during extraction of the humic substances.

The last results indicate the predominance of aromatic structures with high levels of oxygenated groups. The tritation curves indicate that the humic acids are monobasic.

4. <u>Laboratory migration experiments</u>

Column migration experiments were carried out in the JRC (ISPRA) and CIEMAT laboratories using 60 hand drilled cores performed in the gallery by CIEMAT.

At the moment the CIEMAT results are not available. For this reason the result of JRC (ISPRA) are presented, although these works are not included in the contract but are covered in the global project of ENRESA.

The breakthough curves, for the first column migration tests, with Tritiu, Chlorine-36, Tecnecium-95 and Neptunium-239 have been carried out. Migration was accomplished using water from "El Berrocal", in equilibrium with the granite, for the first case, and a leachate obtained under anoxic condition with double-distilled water from two ore samples of lignite (Matilde and Biosca) and a second leachate obtained with artificial water, rich in sodium and with a pH of 8.3, in this case from a sample of Matilde (artificial Matilde). Work has also been carried out with a humic extract from the same samples obtained by means of solubilization with NaO4 and precipitation with ClH.

From the results it may be deduced that for this leachate, 239-Np, has no leachating power, unlike 238-Pu, which shows a very high leachating capacity even for 15 nm colloidal fractions. The 241-Am presents an intermediate value as regards leachating capacity.

Studies of distribution coefficients were carried out using ground samples of "El Berrocal" granite, the results showing that when the humic extract was used with, for example, 238-Pu, there was a reduction of almost 60% in Kd, and that this reduction reached almost 80% when the concentration of the humic extract was two and a half times greater. The same test, performed on the "artificial Matilde" leachate, showed a 70% reduction in the first case and a reduction of more than 90% with concentrations 2.5 times higher. This was for similar organic material contents in the humic extract and leachate.

<u>Publications</u>

ASTUDILLO, J.A. (1989) Ninth EURATOM/AECL information exchange meeting. Pinawa, September, 19-21, 1989. "El Berrocal Project", laboratory and in situ determination of the migration processes of actinide complexes and colloids in a fissured granitic environment.

GUTIERREZ, M.G., BIDOGLIO, G., AVOGADRO, A., MINGARRO, E., D'ALESSANDRO, M. (1989) "Experimental investigation of radionuclide transport through cored granite sample". Second International Conference on Chemistry and Migration behaviour of actinides and pression products in the Geosphere. Monterey, California, November, 1989.

MINGARRO, E., RIVAS, P., GOMEZ, P., SAENZ-GRANERO, F., HERNANDEZ, A., TURRERO, M.J., PARDILLO, J. "Complexation of uranium by humic substances from uraniferous lignites of Spain". International Simposium on Humic Substances in the aquatic and terrestrial Environment". Liköping (Sweden). August, 21-22, 1989.

TABLE 1 : POROSITY AND PERMEABILITY

A. GALLERY SAMPLES

FRESH GRANITE

SAMPLE	POROSITY (%)	PERMEABILITY (m/s)
4	0.77	3.2E-11
5	0.80	1.1E-10
6	0.98	1.5E-10
7	0.72	1.0E-11
8	0.96	3.1E-11
9	0.88	5.2E-11
12	0.93	8.1E-11
1-D	0.68	1.4E-11
3-D	0.60	1.8E-10
4-D	0.78	3.0E-10
5-D	1.06	3.9E-10
7-D	0.99	3.2E-10

ALTERED GRANITE

SAMPLE	POROSITY (%)	PERMEABILITY (m/s)
10	2.06	5.1E-11
11	2.84	7.0E-11
13	3.48	1.1E-10
14	4.18	1.0E-10
15	2.41	4.3E-11
16	3.67	1.7E-10
11-I	3.22	1.4E-09
13-I	3.10	1.6E-09
14-I	3.29	1.5E-09
17-I	4.12	4.8E-09
18-I	4.80	2.9E-09

B. BOREHOLE SAMPLES

SAMPLE	POROSITY (%)	PERMEABILITY (m/s)
1(9.64m)*	1.55	1.2E-11
2(9.77m)**	0.60	1.9E-11
3(15.46m)**	0.47	3.0E-11
4(16.85m)**	0.60	2.1E-11
5(45.80m)*	1.31	4.0E-11
6(47.85m)*	1.34	1.3E-10
7(59.09m)*	1.64	4.3E-11
8(59.34m)*	1.48	1.0E-10

* Altered granite near to fractures
** Fresh granite

C. EXPERIMENTAL PERMEABILITY

SAMPLE	PERMEABILITY (m/s)
3-D	5.5E-11
5-D	2.9E-09
6-D	1.4E-10
7-D	3.9E-10
101	4.8E-10
102	1.4E-07
103	1.1E-09
104	1.3E-08
105	1.8E-10
106	5.3E-10
107	4.4E-09
108	4.9E-09
109	3.9E-10
110	2.7E-09
111	1.4E-10
112	9.2E-09
114	5.6E-10

TABLE 2 : CHEMICAL COMPOSITION

IONIC PHASE (GROUNDWATER) mg/l			COLOIDAL PHASE (CONCENTRATE)	URANIUM CONCENTRATION µg/l	
ELEMENT	Borehole 2	Borehole 3	Borehole 2	Borehole depth	u
Al	0,06	0,05	< 0,5	S-1 10 m	71
Ca	33	26	20	S-1 30 m	196
Mg	7,2	5,1	2,4	S-1 50 m	170
Mn	0,49	0,57	0,05		
Na	10	9,8	4,7	S-7 10 m	21
Zn	0,4	0,11	0,08	S-7 30 m	21
K	0,39	0,47	1,6	S-7 50 m	35
HCO_3^-	166,1	131,5	58,6		
Cl^-	3,2	4,0	4,0	S-AT-1 10 m	21
$SO_4^=$	1,5	3,1	16,8	S-AT-1 30 m	16
PO_4^{3-}	< 0,5	< 0,5	0,94	S-AT-1 60 m	22
NO_3^-	0,4	1,2	10,5		
F^-	1,8	1,4		S-AT-2 10 m	14
U	24 µg/l	10 µg/l		S-AT-2 40 m	17
pH	6,9	7,4	7,3		
			E4/E6:1,78	S-3 10 m	14

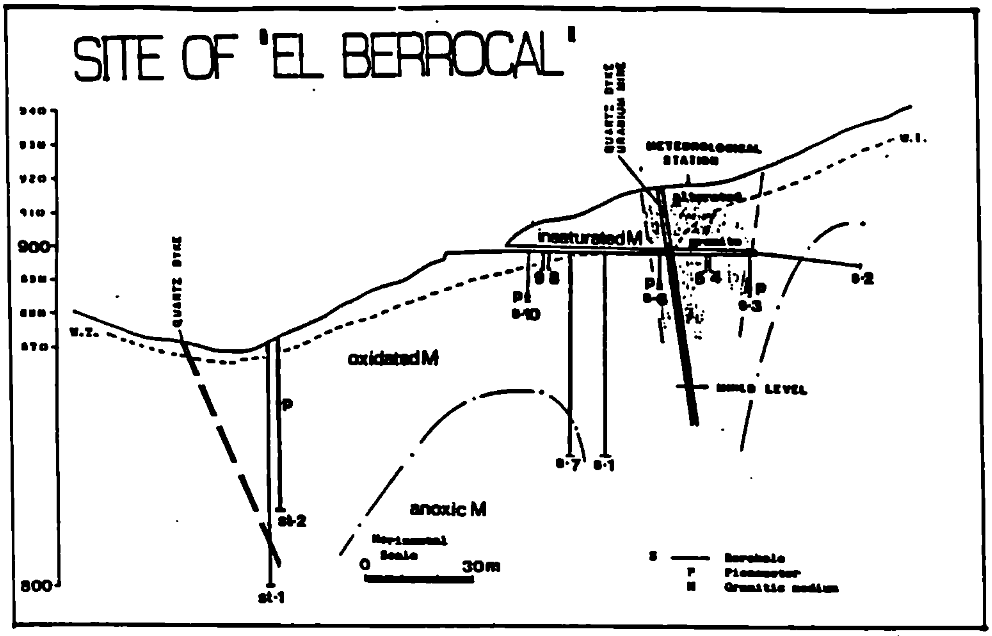


Fig. (1): Boreholes situation

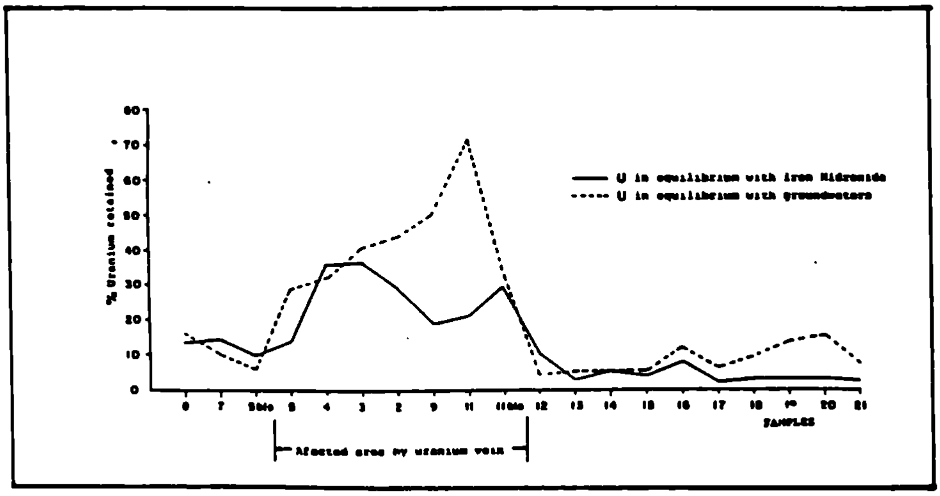


Fig. (2): Lixiviated uranium distribution

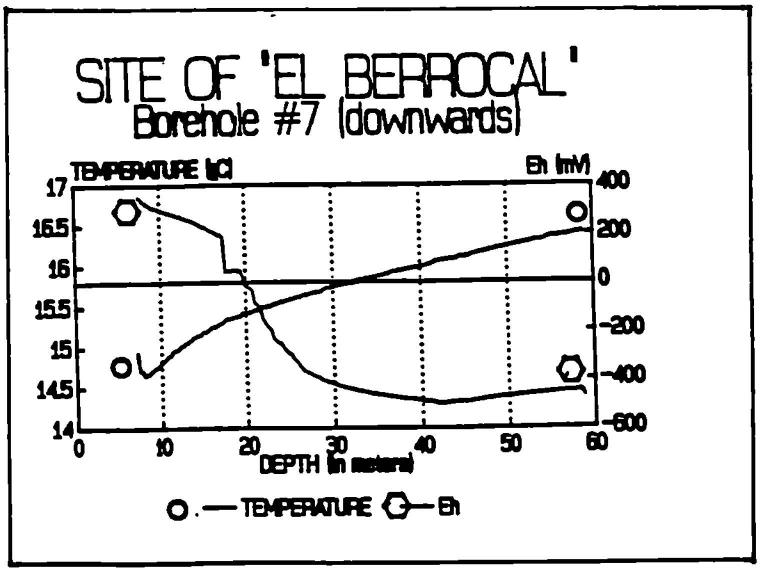


Fig. (3): Groundwater borehole testing.
Eh- Temperature distribution

IMPROVEMENT OF COLLOID SAMPLING TECHNIQUES IN GROUNDWATER AND ACTINIDE CHARACTERISATION OF THE GROUNDWATER SYSTEM AT GORLEBEN (FRG) AND EL BERROCAL (E)

Contractor: Harwell Laboratory, UKAEA, UK
Contract No.: FI1W/0234.
Duration of contract: October 1988-March 1990
Period covered: January 1989-December 1989
Project leader: M. Ivanovich

A. OBJECTIVES AND SCOPE

The apparent radionuclide solubilities in groundwater may be enhanced by their association with colloids, so that a realistic transport model for the migration of radionuclides at a radioactive waste repository requires a knowledge of the radionuclide loading of the colloids present as well as the extent of their transport through the geosphere. Ultrafiltration techniques have been developed to collect groundwater samples in the field, including the particulate, colloid and aqueous solution phases. The research described hereafter is concerned with uranium studies of natural colloids designed to quantify the effects of actinide/colloid association on the past and future migration of radionuclides at Gorleben (FRG) and El Berrocal (Spain). The aims are as follows:

To sample natural colloids from several boreholes at the above sites in collaboration with Technical University, Munich and ENRESA/CIEMAT staff.
To characterize particulate, colloid and solute phases in terms of their physical, chemical and actinide composition.
To use artificial colloids labelled by artificial radionuclides to study in greater detail the uptake characteristics of the ultrafiltering system used.

B. WORK PROGRAMME

1. Sampling and characterisation of Gorleben groundwater in terms of actinide inventory and physical, chemical characterisation of geocolloids, with intercomparison exercise (Harwell/TUM).

2. Study of colloid uptake on tangential flow ultrafilters using model colloids.

3. Characterisation of El Berrocal groundwater in terms of actinide inventory and physical/chemical characterisation of geocolloids.

4. Active participation in CoCo club both in terms of defining future actions and carrying out future benchmark exercises.

C. PROGRESS OF WORK OBTAINED AND RESULTS

State of advancement

Phase II samples, collected in June 1989 from five boreholes along two perpendicular flow paths, have been analysed. All the groundwaters are rich in organic material with DOC concentrations ranging from 30 to 110 mg/ℓ. One sample, GoHy 2227, was sampled in Phase I and was resampled in Phase II.

All samples show evidence for the association of U and Th isotopes with the groundwater organic colloids. The $^{234}U/^{238}U$ activity ratios for these samples all indicate a lack of chemical equilibrium between uranium in the colloids and in the solution phase. These results are consistent with those obtained from Phase I. Sample GoHy 2227 shows evidence of fluctuations in groundwater chemistry with time during Phase II sampling.

Samples of aquifer geomatrix, including brown coals, are currently being analysed for U, Th isotopic activities.

A study of colloid particle/element uptake on tangential flow ultrafilters is underway. Experiments are being carried out using unfiltered Gorleben groundwater samples GoHy 2227 and GoHy 1231 from Phase I. In addition, the uptake of model hematite and latex colloids loaded with known radionuclide concentrations is being studied.

Sampling of the El Berrocal mine site was completed in October 1989. A total of 12 boreholes were sampled for unfiltered groundwater. Boreholes S-1 and S-7 were sampled at 20 m and 45 m depth additionally for colloid concentrate and ultrafiltrate. All sample collection was conducted under anaerobic conditions (N_2 + 1% CO_2 atmosphere).

Progress and results

The results of analyses for groundwaters GoHy 2227 and GoHy 1231 in Phase I may be summarised as follows:

(1) Uranium and thorium are associated with humic colloids in GoHy 2227. In terms of U isotopes, the organic colloids in GoHy 2227 are not in chemical equilibrium with the solution.

(2) Uranium and thorium may be associated with fulvic acid colloids in GoHy 1231. These colloids are smaller than the humic colloids in GoHy 2227 and pass through the 10,000 molecular weight (MW) cut-off filter, and thus are present in the ultrafiltrate.

(3) Metals with a 3^+ and 4^+ valency are strongly bound to humic colloids whereas 2^+ valency metals are less well bound.

(4) A linear relationship exists between DOC concentration and colloid population in GoHy 2227.

(5) The colloid population in GoHy 2227 is 5.3 x 10^{17} particles/litre. The largest proportion of colloids occurs in the 1 to 15 nm particle size range. These have the largest surface area for potential

complexation with metal ions.

(6) The colloid population (1 nm to 1 μm range) in GoHy 1231 is approximately 10^{12} particles/litre. This is too low to determine element loading on colloids within the sensitivity of the analytical techniques. This sample is being further investigated.

The results for Phase II Gorleben samples may be summarised as follows:

(1) The organically-rich groundwaters again show the association of U and Th with the organic groundwater colloids. Elemental analyses currently available show that Fe, Mn, Ti and DOC are also associated with the organic colloid phase.

(2) The physico-chemical results for the groundwaters measured in the field are given in Table I. The results for U/Th isotopic partition between the colloid fraction, ultrafiltrate and ultrafilter wash are given in Table II.

(3) Sample GoHy 2226 has the highest conductivity but the lowest U/Th isotopic partition between the colloid fraction and ultrafiltrate. Sample GoHy 532 has the lowest conductivity and one of the highest U/Th isotopic partition values (Table II). However, there is no consistent relationship between conductivity and U/Th isotopic partition.

(4) Attempts to study isotopic activities in size-fractionated samples (10,000 to 100,000 MW, 100,000 MW to 0.1 μm and 0.1 to 1 μm) in sample GoHy 2227 are further complicated by fluctuations in groundwater chemistry with time during sample collection. The linear relationship between DOC and colloid population observed in Phase I and the fluctuations in DOC with time in Phase II imply that the colloid population in sample GoHy 2227 may also fluctuate during sample collection in Phase II. Sample colloid populations will be determined directly by LPAS to confirm this effect.

(5) The variation in U/Th isotopic activities in unfiltered groundwater samples from borehole GoHy 2227 collected at approximately 3 hour intervals is illustrated in Table III. Phase I results are also presented in Table III for comparison. It appears that the changes in activity are due to the humic colloid population. Thus, direct intercomparison between the results for Phase I and Phase II may not be possible.

<u>List of Publications</u>

DEARLOVE, J.P.L., LONGWORTH, G., IVANOVICH, M., KIM, J.I., DELAKOWITZ, B. AND ZEH, P. 1989. A study of groundwater-colloids and their geochemical interactions with natural radionuclides in Gorleben aquifer systems. Presented at the 2nd International Conference on Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere to be held at Monterey, USA, Nov 6-10.

DEARLOVE, J.P.L., LONGWORTH, G., IVANOVICH, M. KIM, J.I., DELAKOWITZ, B. AND ZEH, P. 1989. Sampling and characterisation of groundwaters at the Gorleben site, FRG. Phase I: Results for groundwaters GoHy 2227 and GoHy 1231. Harwell Report AERE-R 13628.

TABLE I: PHYSICO-CHEMICAL RESULTS FOR THE 5 BOREHOLES SAMPLED IN PHASE II FROM THE GORLEBEN SITE

	GoHy 532	GoHy 2211	GoHy 1271	GoHy 2226
pH	9.0	8.3	8.4	7.4
Eh (mV)	+86	+39	+11	+34
Conductivity (μS/cm)	975	5000	3190	11700
DOC (mg/ℓ)	30	110	44	49
Temperature (°C)	12.8	13.4	14.7	15.8

	GoHy 2227/1	GoHy 2227/2	GoHy 2227/3
pH	7.7	7.7	7.9
Eh (mV)	+115	+91.9	+69.2
Conductivity (μS/cm)	4850	-	-
DOC (mg/ℓ)	80	110	82
Temperature (°C)	14.9	15.1	14.7

TABLE II: URANIUM AND THORIUM ISOTOPIC PARTITION FOR THE 5 PHASE II BOREHOLE SAMPLES FROM THE GORLEBEN AQUIFER

Sample	^{238}U	^{234}U	^{230}Th	^{232}Th	^{228}Th
	(% of total in fluid phase)				
<u>GoHy 532</u>					
Colloid fraction	77.9 ±6.1	70.7 ±5.2	98.9 ±7.7	97.5 ±8.0	89.1 ±6.9
Ultrafiltrate	21.6 ±1.6	28.9 ±1.4	0.70 ±0.04	2.3 ±0.3	10.7 ±0.7
Ultrafilter wash	0.46 ±0.04	0.40 ±0.03	0.36 ±0.03	0.28 ±0.02	0.27 ±0.02
<u>GoHy 2211</u>					
Colloid fraction	80.1 ±5.4	74.5 ±4.9	84.2 ±5.6	94.2 ±6.9	67.4 ±4.8
Ultrafiltrate	19.1 ±1.2	24.8 ±1.1	15.2 ±0.8	5.1 ±0.5	32.1 ±2.3
Ultrafilter wash	0.8 ±0.1	0.7 ±0.1	0.63 ±0.04	0.7 ±0.1	0.54 ±0.03
<u>GoHy 1271</u>					
Colloid fraction	67.1 ±5.8	61.8 ±5.0	53.9 ±4.6	69.6 ±5.6	34.6 ±2.4
Ultrafiltrate	22.7 ±1.9	30.7 ±1.6	27.3 ±1.5	8.4 ±0.7	56.6 ±3.0
Ultrafilter wash	10.2 ±0.8	7.4 ±0.5	18.01 ±1.1	22.0 ±1.3	8.7 ±0.4
<u>GoHy 2226</u>					
Colloid fraction	55.0 ±3.6	36.4 ±2.2	56.8 ±3.8	2.7 ±0.8	25.8 ±1.7
Ultrafiltrate	35.3 ±2.1	57.5 ±2.1	34.7 ±1.7	30.5 ±3.8	70.1 ±4.0
Ultrafilter wash	9.7 ±0.6	6.1 ±0.3	8.5 ±0.5	66.7 ±4.1	4.42 ±0.2

Sample	^{238}U	^{234}U	^{230}Th	^{232}Th	^{228}Th
	(% of total in fluid phase)				
GoHy 2227 10,000 MW					
Colloid fraction	60.9 ±4.3	45.9 ±3.0	39.4 ±2.8	72.6 ±6.7	18.6 ±1.3
Ultrafiltrate	35.6 ±2.7	51.7 ±2.3	55.3 ±3.3	18.5 ±3.4	78.7 ±5.5
Ultrafilter wash	3.5 ±0.2	2.4 ±0.1	5.3 ±0.3	8.9 ±0.7	2.7 ±0.1
GoHy 2227 100,000 MW					
Colloid fraction	44.1 ±3.1	25.1 ±1.7	38.0 ±2.7	80.6 ±6.5	15.8 ±1.2
Ultrafiltrate	55.3 ±3.5	74.3 ±3.5	61.3 ±1.5	18.6 ±1.8	83.8 ±5.9
Ultrafilter wash	0.7 ±0.1	0.6 ±0.1	0.7 ±0.1	0.9 ±0.1	0.38 ±0.03
GoHy 2227 0.1 micron					
Colloid fraction	53.2 ±4.3	34.9 ±2.7	46.2 ±3.8	83.2 ±7.2	25.0 ±1.8
Ultrafiltrate	46.6 ±3.1	64.8 ±3.0	53.7 ±3.0	16.7 ±1.7	74.9 ±4.7
Ultrafilter wash	0.2 ±0.1	0.2 ±0.1	0.052 ±0.003	0.09 ±0.01	0.030 ±0.001

TABLE III: VARIATIONS IN U AND TH ISOTOPIC ACTIVITIES AND ACTIVITY RATIOS FOR UNFILTERED GROUNDWATER FROM BOREHOLE GOHY 2227 WITH TIME (PHASE II) AND THEIR COMPARISON WITH THE RESULTS FOR GOHY 2227 (PHASE I)

Sampling time	U (nmol/ℓ)	Th (nmol/ℓ)	$^{234}U/^{238}U$	$^{230}Th/^{234}U$	$^{228}Th/^{232}Th$	$^{230}Th/^{232}Th$
			Activity ratios			
PHASE I - GoHy 2227						
	2.4 ± 0.1	6.0 ± 0.4	1.5 ± 0.1	0.8 ± 0.1	20 ± 1	1.6 ± 0.1
PHASE II - GoHy 2227						
14:00 (GoHy 2227/1)	0.60 ± 0.04	1.5 ± 0.1	1.8 ± 0.1	0.6 ± 0.1	17 ± 1	1.3 ± 0.1
17:40 GoHy 2227/2)	2.4 ± 0.2	4.9 ± 0.4	1.5 ± 0.1	0.9 ± 0.1	25 ± 2	1.9 ± 0.2
20:55 (GoHy 2227/3)	2.3 ± 0.1	6.0 ± 0.3	1.6 ± 0.1	0.84 ± 0.04	21 ± 1	1.6 ± 0.1

4.3.B. In situ migration experiments and development of measuring techniques

In Situ Determination of the Effects of Organics on the Mobility of Radionuclides in Controlled Conditions of Groundwater Flow

Contractor:	British Geological Survey, Keyworth, Nottingham
Contract No:	FI-1W-0064
Duration of Contract:	July 1986 - June 1989
Project Leader:	G.M.Williams

A. Objectives and Scope

The broad objective is to verify by means of *in situ* field tracer tests, predictions of the mobility of radionuclides in a shallow glacial sand aquifer, having taken into account the potential effects of organics (natural and introduced) on radionuclide speciation and mobility.

The tracer tests will be undertaken in a remote part of the low level radioactive waste site at Drigg. Prediction of their outcome is based upon detailed hydraulic characterisation of the field site, coupled with laboratory studies of radionuclide sorption and organic complexation. Liaison has been established with Loughborough University (LUT) for direct speciation measurements, the University of Wales College of Cardiff UWCC (formerly Institute of Science and Technology (UWIST) for radionuclide speciation modelling, and Delft Geomechanics for solute transport modelling.

B. Work Programme

The project is divided into a number of research areas as follows:-

(1) Aquifer characterisation and instrumentation - Involves the determination of aquifer hydraulic properties, its geochemistry, mineralogy and groundwater composition, particularly the nature and amounts of natural organics (humic and fulvic acids) and colloids. Development of instrumentation to monitor groundwater composition and radionuclide migration.

(2) Characterisation of complexes and colloids - interlaboratory comparison within the CEC, to characterise, and determine stability constants with selected radionuclides, for commercially available humic acid and natural organics from Drigg, Mol, Ispra etc. A parallel excercise is underway for colloids.

(3) Laboratory sorption studies - includes various sorption experiments to determine the effects of natural organics on radionuclide sorption, kinetic measurements, and direct speciation determinations of radionuclide complexation in groundwater after equilibration with the sediment.

(4) Modelling - Speciation models will be used to predict the speciation of radionuclides in the sorption experiments and help to determine the important mobile species in the field test. Deterministic flow modelling will aid in the design of the borehole array for the tracer tests and form a basis for reactive mass transport models.

(5) Field tracer experiments - initial tracer tests will compare various conservative tracers (^{131}I, Cl, and ^{3}H) and provide background data on the hydraulic characteristics of the aquifer. Subsequent tracer tests will involve reactive radionuclide species with the addition of organic solutes.

C. Progress of work and results obtained

State of advance

Field tracer tests have been undertaken using ^{131}I and ^{3}H, and a second test using $^{57}Co(EDTA)/\ ^{131}I$. Column experiments have been undertaken with a Co-Fulvic Acid complex derived from natural fulvic material extracted from groundwater at Drigg. Difficulties have been experienced in extracting sufficient natural organic material for the COCO club but a small amount has been isolated for BGS/LUT use.

Progress and results

(1) Aquifer characterisation and instrumentation (B1)

The ^{131}I and ^{3}H test was performed satisfactorily using an *in situ* γ detector attached to a winch controlled by a IBM-PC. Two additional winch systems were constructed for use in the $^{57}Co(EDTA)/\ ^{131}I$ experiment with colimated probes.

(2) Complexes and colloids (COCO excercise, B2)

This phase of the work (inter-laboratory comparison excercise) has been completed, (see reference /1/).

(3) Laboratory sorption studies (B3)

Cobalt migration has been studied in small-scale laboratory columns designed to provide a controlled intermediate test between laboratory batch and field tracer experiments. Tests using a Co-Fulvic Acid complex (Co-FA) were conducted and strong attentuation of the Co was observed. The exponential profile of activity in the column after the experiment was interpreted to show that the Co-FA complex was dissociating at a first order rate of 1.8×10^{-8} sec^{-1}. This precluded the use of the Co-FA complex in the field test since breakthrough would not be easily detectable. Consequently attention was turned to the Co-EDTA complex which was used in similar column experiments. Again the Co activity was attenuated but not to such an extent as with Co-FA. A complicated distribution of activity was observed along the column after passage of Co-EDTA suggesting that a number of Co species (possibly colloids) were present.

4. Modelling (B4)

Solute migration in the tracer test has been analysed using a one-dimensional analytical model. These showed longitudinal dispersion to be small with D_L ranging from 5×10^{-2} to 1×10^{-3} m. None of the breakthrough curves, even in the main flow path, showed peak heights comparable with those predicted by the one-dimensional model, suggesting that tracer 'loss' is occurring due to lateral dispersion. This shows that a one-dimensional model does not describe the observed solute migration behaviour adequately, and that account must be taken of dispersion transverse to the direction of flow. Simulation of tracer breakthrough in the pumping well based on a multi-layered aquifer gave the best fit to the observed response (Visser and Van Meurs, Delft Geotechnics, reference /2/). The variation in hydraulic conductivity of the layers rather than the dispersivity of individual layers exerted greatest control on the overall dispersion observed in the fully penetrating pumping well.

Attempts have been made to simulate the distribution of Co in the column experiments assuming the presence of a number of species/ colloids. A model has been constructed assuming the Co-EDTA to dissociate and form separate species each with a different retardation coefficient and dispersivity. Modelled results showed good agreement with the distribution of activity observed. However, additional work is required to confirm the mechanism for Co-EDTA dissociation.

5. Tracer tests (B5)

A field test was undertaken using ^{57}Co(EDTA) in conjunction with the conservative tracer ^{131}I. EDTA (8 x 10^{-5} mol dm^{-3}) and a prepared complex of ^{57}Co-EDTA were added to 20 litres of groundwater and allowed to equilibrate for 36 hours. The activity of the Co-EDTA was 1.85 x 10^{8} Bq (5 mCi) which gave a concentration in the release chamber of 1.36 x 10^{-10} mol. dm^{-3}. The Co-EDTA complex mirrored the ^{131}I breakthrough but with refernce to ^{131}I, only 60% of the cobalt complex was recovered (as observed in the laboratory columns). During the field test groundwater samples from the multi-level samplers were subjected to ultra-filtration to determine the association of cobalt activity with colloids. Up to 13% of the mobile cobalt appeared to be associated with the colloidal phase but the nature of these colloids has not been determined.

Reports

The following reports have been prepared during the contract:-

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Bloodworth A J and Inglethorpe S D J, 1988. Mineralogy of shallow borehole material from Drigg, Cumbria. British Geological Survey, Mineralogy and Petrology Report No WG/88/5C.

Falck W E 1988. Modelling the interaction between natural organic matter and metal cations: a review. British Geological Survey Fluid Processes Research Group Report No.WE/88/49, 62pp.

Falck W E, 1989. Natural organic - cation interaction and speciation models. Int Sym on Humic substances in the aquatic and terrestrial enviroment. Linkoping, Sweden. 21 -23 Aug 1989.

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Haigh D G, Higgo J J W, Williams G M, Ross C A M, Falck W E, Allen M A and Warwick, 1989. The influence of organics on the sorption of Np, Eu, U, I, Sr, and Cs by glacial sand in laboratory batch experiments. British Geological Survey Fluid Processes Research Group Report No.WE/89/16.

Haigh D G, Williams G M, Higgo J J W, 1989. The groundwater chemistry of the Drigg tracer test array. British Geological Survey Fluid Processes Research Group Report No.WE/88/41. (DRAFT)

Haigh D G, Williams G M, Hooker P J, Ross CAM 1988. The influence of organics on the sorption of cobalt by glacial sand in laboratory batch experiments. British Geological Survey Fluid Processes Research Group Report No.WE/88/20.

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Sen M A and Williams G M, 1989. Analysis of the migration of Cl, ^{131}I and ^{3}H tracers in a shallow sand aquifer. British Geological Survey Fluid Processes Research Group Report No.WE/89/9 (DRAFT).
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Warwick P, Shaw P, Williams G M and Hooker PJ, 1988. Preliminary studies of cobalt complexation in groundwater. British Geological Survey Fluid Processes Research Group Report No.WE/88/12. also in Radiochemica Acta, 44/45, 59 - 63.
Wealthall G P, Hallam J R and Williams G M, 1988. Design and installation of a borehole array for *in situ* radiotracer experiments. British Geological Survey Fluid Processes Research Group Report No.WE/88/17.
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West J M, Rowe E, Wealthall G P, Allen M, 1988. The geomicrobiology of the BGS in situ migration experiment. British Geological Survey Fluid Processes Research Group Report No.WE/89/8.
Williams G M, 1989. In situ determination of the effects of organics on the mobility of radionuclidess in controlled conditions of groundwater flow: Summary Report March 1989. British Geological Survey Fluid Processes Research Group Report No.WE/89/22.
Williams G M, Higgo J J W, Sen M A, Falck W E, Noy D J, Wealthall G P and Warwick P. The influence of organics in field migration experiments: Part 1 *In situ* tracer tests and preliminary modelling. Paper submitted to Radiochimica Acta (CEC conference Migration 89 Monterey California).
Williams G M, Roberts P D, Wealthall G P, Mooney P, 1989. A downhole ranging gamma spectrometer system for radio-tracer tests. British Geological Survey Fluid Processes Research Group Report No.WE/88/47 (DRAFT).

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/1/ Peachey D. and Williams G. M. 1987. Characterisation of humic material for inter-laboratory comparison. British Geological Survey, Fluid Processes Research Group Report No. FLPU 87-5 19pp..
/2/ Visser and Van Meurs, Delft Geotechnics 1989. Fluid Verification of Advanced Transport Models for Radionuclides in Heterogeneous. EEC Progress Report Contract No. FI1W/0083 - NL

Title : DEVELOPMENT AND APPLICATION OF A RETENTION PROPERTIES MEASUREMENT SYSTEM IN A GEOLOGICAL ENVIRONMENT USING RADIOACTIVE TRACERS IN THE DRILL-HOLE (self-contained probe FORALAB).

Contractor : CEA/IPSN CEN CADARACHE
F 13108 St. Paul-lez-Durance

Contract No. : F11W/065

Working period : November '86 - November '89

Project Leader : J. PORCHERON

A. OBJECTIVES AND SCOPE

Radionuclides from a subterraneous waste storage place have to force their way through, and interact with, several barriers prior to reaching the geological medium itself.

They are diluted by the suberraneous water which, by modifying their chemical structure, settles them into a final balance with the medium.

The purpose of this study is to determine the delay term of the radionuclides during their migration through the deep geological environment.

It became evident that it was preposterous to attempt in-laboratory duplication of the prevailing parametric conditions of the natural medium, whether physical, chemical or biological.

To avoid the uncertainties connected to laboratory experiments, the probe "FORALAB", whose performances had already been ascertained during the preceding contract (EUR 11141), was developed to permit studying the radionuclide sorption-desorption phenomena in a geological environment in a condition of equilibrium with undisturbed subterraneous waters.

The probe need not simulate the environment as it is plunged into it.

The contract scope is the "*in-situ*" qualification of the probe, using a dual tracing system, i.e. Tritium and Eu on the one hand, Pu and Np on the other hand.

The probe will then be operated in 3 geological sites, of some interest for the Community, i.e. AURIAT (granite), MOL (clay) and GORLEBEN (salt), in order to assess the containment properties of these environments.

The probe is essentially composed of a pump, a syringe, a 20 mm dia., 200 mm long test column and 40 sampling pots. Its double insulation is a safety against drill-hole pollution.

B. WORK PROGRAM

B1. Probe Qualification

The probe will be checked for performance in the hole drilled in the granitic site of AURIAT.

The column, filled with Fontainebleau sand, will be traced by means of Europium and Tritium.

The drill-hole water will be circulated in the column for one day before the tracer injection.

B2. Tests on the Reference Sites

The tests will be performed in the deep holes at AURIAT, MOL and GORLEBEN.

The columns will be filled with a mixture of Fontainebeau sand and 1 to 3 % clay from the site.

The radionuclides used will be am, Pu and Np.

Each column will be Tritium-calibrated before each individual test.

B3. Finally the results from each individual test will be mathematically processed to yield the delay terms and the absorption isotherms of each pollutant used.

C. WORK PROGRESS AND RESULTS OBTAINED

C1. Work Progress

The scientific program began in 1989 with experiments using europium at the Auriat site. Columns were place in the probe containing the following materials.

a) Sand from Fontainebleau (the reference material) with 5 % 10^{-4} g of europium ; 2,9 % of the tracer was recovered with a zero time-lag (the water and tracer flowed out simultaneously).

b) Unaltered ground granite from Auriat core samples, with but with a time-lag term (ratio between the water and tracer flow rates) of 7.3 (Figure 2).

c) Unaltered granite with 3 % alteration products sampled on the site, and with 6×10^{-2} g of europium ; 30 % of the tracer was recovered, apparently without any time-lag, although this cannot be confirmed because of a technical incident.

Laboratory Tests :

The field tests are being repeated in the lab using water from the borehole sample under pressure at a depth of 400 meters.

C2. Drawings

After another experiment in granite together with alteration products, the granite site field tests have now been completed.

List of Publications :

J. Porcheron and J.M. Vinson. "Borehole Field Measurements of Chemical Parameters of the Natural Environment and Pollutant Transfer Coefficients". Hydrogeology and Safety of Radioactive and Toxic Industrial Waste Deposits : Orleans (France) : June 7-10, 1988.

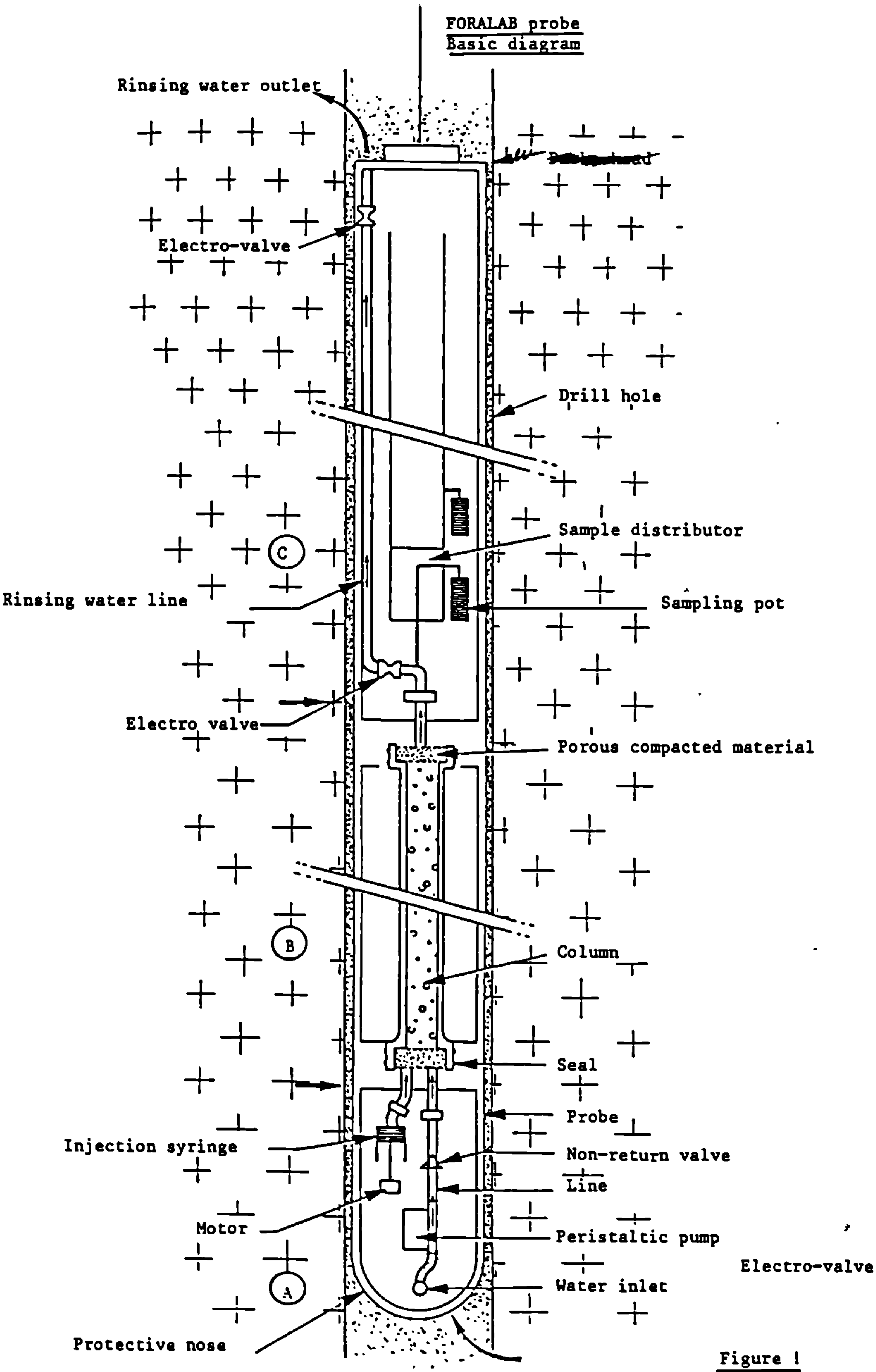
FORALAB probe
Basic diagram
Rinsing water outlet
Electro-valve
Drill hole
Sample distributor
Rinsing water line
Sampling pot
Electro valve
Porous compacted material
Column
Seal
Probe
Injection syringe
Non-return valve
Line
Motor
Peristaltic pump
Electro-valve
Water inlet
Protective nose
C
B
A

Figure 1

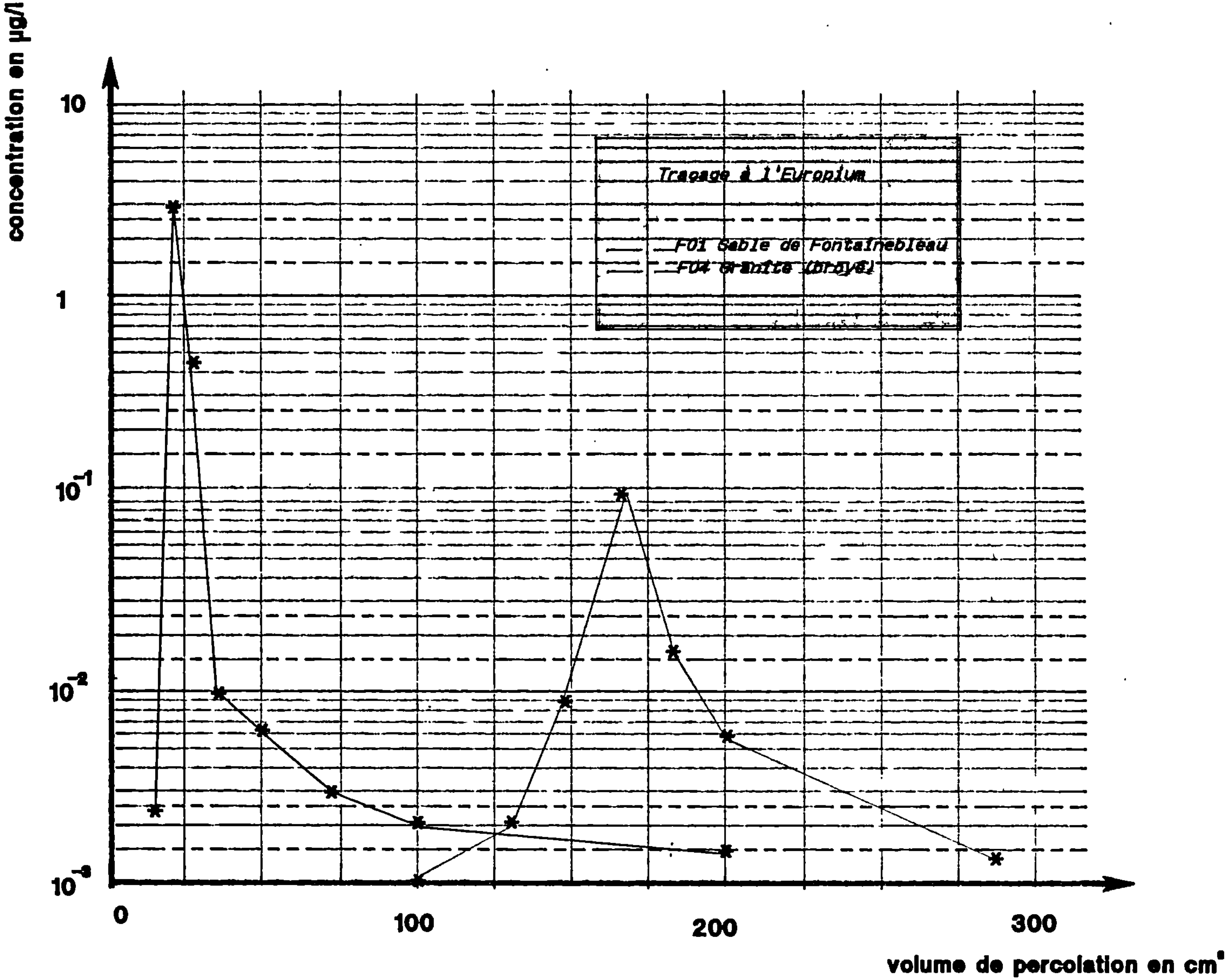

Fig. 2 : RETENTION COMPAREE DE L'EUROPIUM DANS LE SABLE DE FONTAINEBLEAU ET LE GRANITE (BROYE)

FIELD VERIFICATION OF ADVANCED TRANSPORT MODELS FOR RADIONUCLIDES IN HETEROGENEOUS SOILS

Contractor : Delft Geotechnics, Netherlands
Contract No. : FI1W/0083
Duration of contract : from march 1987 to december 1989
Period covered : februari 1989 to december 1989
Project leaders : Dr. Ir. M. Loxham, drs. W. Visser

A. <u>OBJECTIVES AND SCOPE</u>

The retardation of contaminants by adsorption on soil components such as clay and iron oxides is usually the most important mechanism that has to be taken into account when making the safety assessment of a toxic landfill. This holds for radioactive components as well as other chemicals. The mobilisation of the retardation capacity in any given soil profile is not only a function of the mass of adsorption sites but also of the distribution of the adsorption sites throughout the system. Because the adsorption sites are usually available in the less permeable zones of an aquifer it is important whether or not mass transfer can take place significantly from the advective flow towards the adsorption sites. In other words, the effective retardation capacity is a function of the structure or heterogeneity of the soil and of the length scales relevant to the problem. Earlier studies by the present researchers, supported in part by the CEC (phases I and II) have demonstrated the key importance of these phenomena both theoretically and at the bench scales.

The objective of this study is to verify the concepts developed in the earlier programs at a field scale using the data generated by the communities investigation conducted by the British Geological Survey (BGS) at Drigg in the United Kingdom. Whilst the study is directly related to the problems associated with the geosphere safety assessment technologies for the shallow burial of low level radioactive waste, the problem definitions, solution methodologies and problem solving experience are directly applicable to all toxic waste and contaminant migration studies.

B. <u>WORK PROGRAM</u>

I. Data collation and critical review.
II. Predictive modelling of the field experiment
II.1 Calculations at level 1 (Homogeneous Approximation)
II.2 Calculations at level 2 (Stratified Approximation)
II.3 Internal modelling
III Evaluation of the Modelling Exercise
III.1 Inter-model level comparison
III.2 Comparison with the results obtained from the Drigg experiments
III.3 Analysis of dispersion lengths in relation with groundwater velocities and soil heterogeneities

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In the last years progress report we reported results which were obtained with data available from expermiments and analysis carried out at that time. These data consisted of the results of some first permeability measurements and limitted data about the texture and mineralogy of the aquifer under consideration and the results of a chloride tracer test. At that time there was no recording of an average breakthrough in a fully screened observation well. As this is crucial in conducting the studie required under our contract the average breakthrough was measured in the water recovery well for the tracer tests with iodine 131 and tritium as well as for the tracer test with CoEDTA. Ultimately all data needed for completing the project became available at Delft Geotechnics. The database has been defined and reviewed. The average breakthrough at the recovery well has been modelled for the conservative tracers using several modelling techniques. The results are compared with the measurements in the field as well as at an intermodel level. In the next few months the breakthrough of a slightly retarding tracer (CoEDTA) will be modelled, although a first evaluation of the measurements did not show hardly any retardation at all. Further analysis will be made of dispersivities encountered in relation with groundwater velocities and soil heterogeneities. The final report is expected to be ready in spring 1990.

progress and results

I. DATA REVIEW

Available data can be grouped into:

- data on hydrogeologic setting and system dimensions.
- data on soil properties.
- data on flow conditions during tracer test
- data on tracer injection
- data on tracer breakthrough

During the period covered in this progress report new data became available at Delft Geotechnics, which further enabled us to redefine our modelling approach. Undisturbed soil cores taken during installation of the well screens /1/ showed that the effective thickness of the aquifer was 1.0 m instead of 1.5 m obtained with penetrometer focussed resistivity probings /2/. Mean porosity of the effective part of the aquifer is 35 % instead of 32 % for the whole aquifer. These differences contributed to the rather bad fit of our first modelling results compared with the field measurements.

In addition to the permeability data already available some new data were obtained by BGS through a pressure wave dissipation test. These data gave an accurate value of the ratio $k*D/s$ ($k*D = T =$ transmissivity, $s =$ elastic storativity) for the different horizontal layers directly in the line of the tracer array. From a pumping test at D219 a value for s could be obtained using Theis's method /3/, so the permeabilities could be calculated for each horizontal layer. All permeability data available are presented in table 1.

To verify whether the physical presence of the sampling apparatus in the tracer array would significantly alter flow conditions the groundwater flow was modelled in detail using an analytical element model /4/. The results show that the influence on measured breakthrough will only be small. The streamlines starting from the tracer injection well will arive about 0.5 - 1.0 hrs later at the water extraction well if the presence of the sampling apparatus is taken into account (≈2-4% of total traveltime, see Figure 1). The variation in arrival times is about 1 hour in both cases because of the radial component in flow. Due to the representation of the presence of the sampling apparatus, a few streamlines penetrate these less permeable elements and contribute to the breakthrough as a kind of tailing effect in the results with obstacles.

Because there is no fully screened observation well in the part of linear flow at Drigg, average breakthrough had to be measured in the water extraction well. A major disadvantage of this is the large dilution in the well because of radial inflow of water. Average breakthrough was measured for iodine^{131} and CoEDTA.

II. PREDICTIVE MODELLING OF THE FIELD EXPERIMENT

II.1 Homogeneous approximation

Average breakthrough of iodine^{131} at the extraction well was modelled using a 2-D horizontal numerical approach. The parameters used were: discharge Q(in)=Q(out)= 2.88 m^3/day, thickness (D) = 1 m, porosity (n) = 0.35, α_L = 0.25 m and α_T = 0.025 m. The results are compared with the measurements in Figure 2. The tail in the measurements is caused by the reinjection of contaminated water (closed system).

II.2 Stratified approximation

This approach involves 2-D vertical modelling of the aquifer (taking transverse dispersion into account), as well as multilayer 1-D modelling (convoluted, without transverse disperion). The permeabilities and thicknesses used are shown in Figure 3. Other parameters are: n = 35%, α_L= 0.25 m, α_T = 0.025 m, gradient = 0.24 m/m. The approach only works for linear flow so the effect of radial flow at the extraction well had to be derived from the dilution between the 2-D horizontal results and 1-D modelling with variable velocities (decreasing the area of flow along the streamline). The concentration levels in the extraction well are shown in Figure 4 for the calculations as well as for the measurements.

III. EVALUATION OF THE MODELLING EXERCISE

Figure 4 shows that the peak arrival time is best matched with the 2-D horizontal numerical approach. This is to be expected because in this case the breakthrough will be defined by the (accurately measured) discharge, independently from permeability, as long as the thickness used for the aquifer is correct. The shape of the measured breakthrough curve is best matched with the multi 1-D approach, indicating that in the 2-D vertical numerical approach the dispersion values used are to big, and that the permeabilities measured are lower then those derived from peak travel times.

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/1/ BLOODWORTH, A.J., INGLETHORPE, S.D.J., Minarology of shallow borehole material from Drigg, Cumbria, BGS Report No. WG/88/5C (1988)
/2/ WEALTHALL, G.P., HALLAM, J.R., WILLIAMS, G.M., Design and installation of a borehole array for radiotracer experiments at Drigg, BGS Report No. WE/88 (1988)
/3/ KRUSEMAN, G.P., RIDDER, N.A. de, Analysis and evaluation of pumping tests data, ILRI bullitin 11 (1979)
/4/ STRACK, O.D.L., Groundwater mechanics, Prentence Hall (1989)

TABEL I: RESULTS OF DIFFERENT PERMEABILITY MEASUREMENTS

LOCATION	K (M/SEC)	RANGE	METHOD
D202, D203, D204 (nearfield)	4.75E-6	2.70E-6 - 7.70E-6	slugtests, fully screened
D205, D207 (local field)	3.67E-5	2.70E-7 - 1.80E-5	fallinghead, several screen sections
D219	2.88E-5		steady state pumpingtest, Thiem´s method
D219	3.80E-5		unsteady state pumpingtest Theis´s method, s=1.6E-4
D226/12	1.83E-5		pressure wave dissipation
11	5.53E-5		s= 1.6E-4
10	2.04E-5		mean K=
9	2.37E-5		(Σk*d/D)=
8	4.94E-5		3.58E-5
7	3.12E-5		
6	4.11E-5		

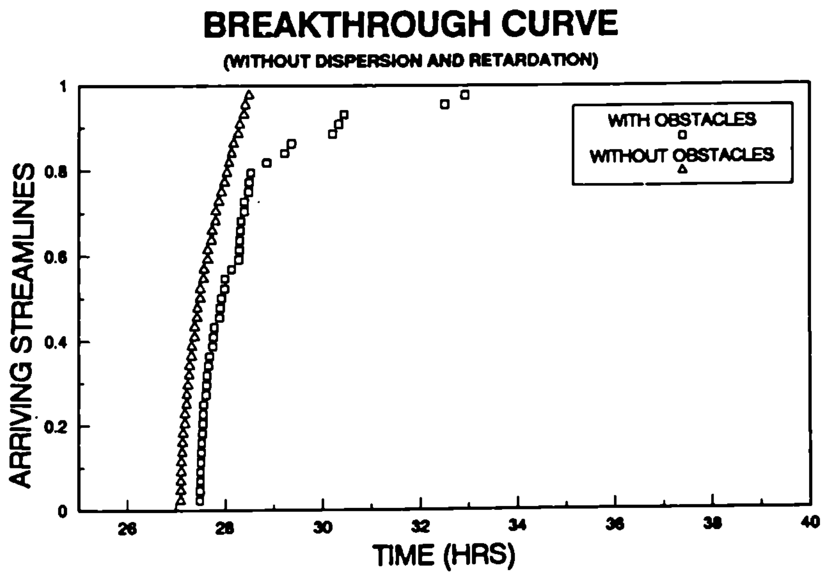


Figure 1

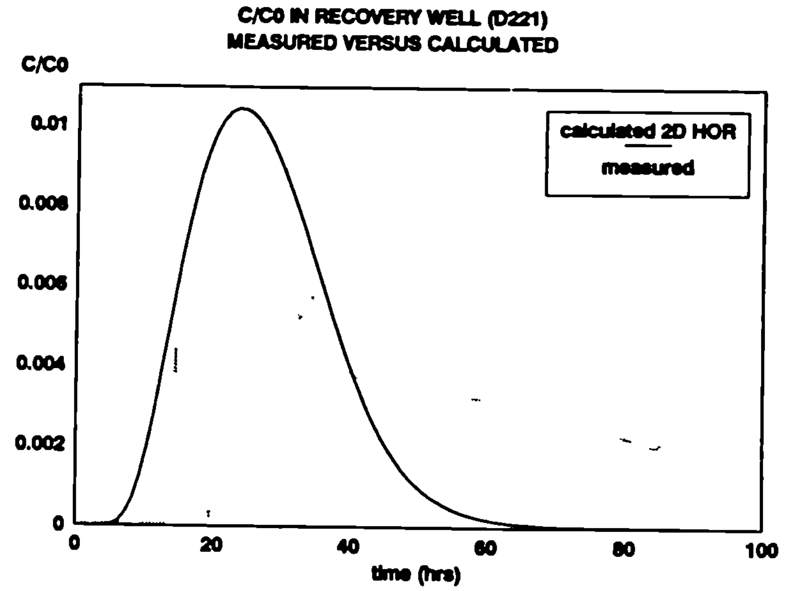


Figure 2

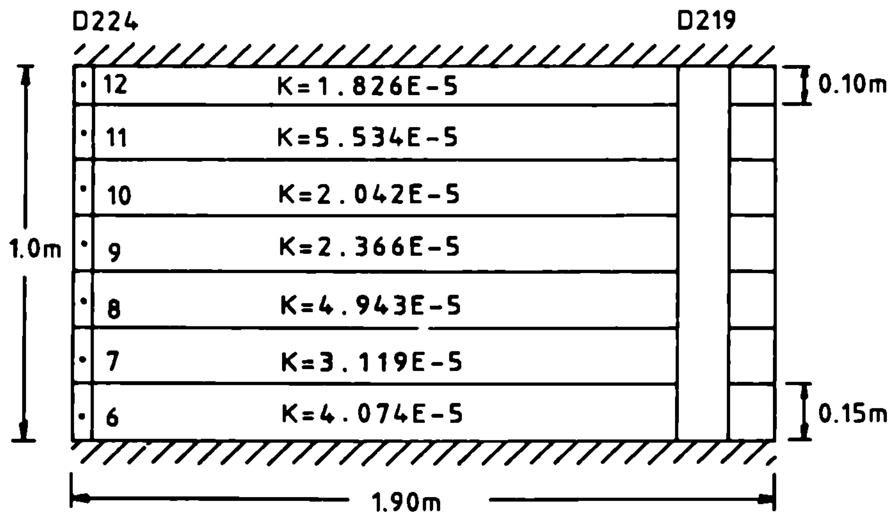


Figure 3

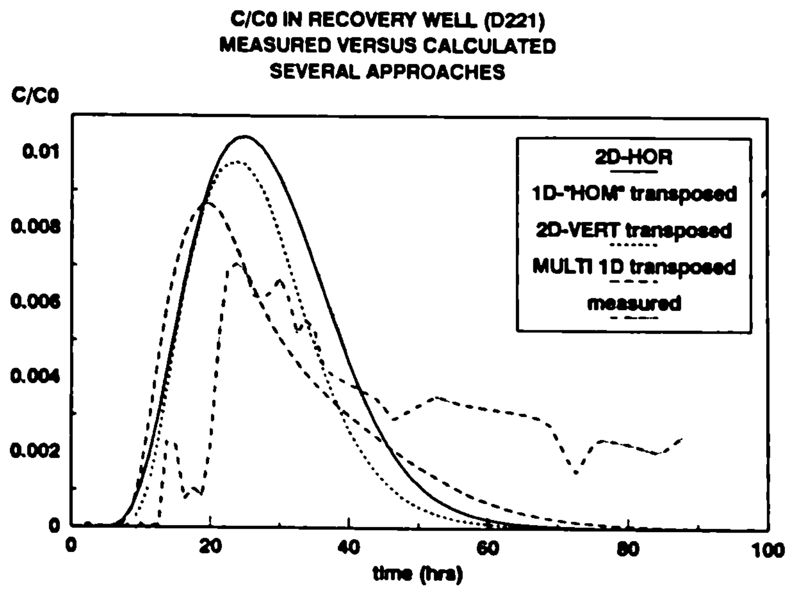


Figure 4

Title : *IN-SITU STUDY OF RADIONUCLIDE DIFFUSION IN CLAYS BY MEANS OF THE AUTOLAB PROBE*
Contractor : CEA/IPSN CEN CADARACHE
F 13108 St. Paul-lez-Durance
Contract No. : FI1W/0144
Working period : January 1988 - July 1990
Project Leader : J. PORCHERON

A. OBJECTIVES AND SCOPE

The storage of nuclear wastes in a deep geological formation calls for that a dependable knowledge of the site safety is gained. Significant data on the radionuclide diffusion throughout argillaceous materials should be acquired by *in-situ* observations and measurements in a drill-hole, provided that the conditions of the receptor environment are safeguarded.

B. WORK PROGRAM

B1. AUTOLAB is a probe designed for measuring the delay term of the radionuclide *in-situ* in the drill-hole clays.

B2. Specificity of the AUTOLAB probe

The probe is designed for diffusion measurements, which entails particular characteristics for grappling the slowliness of the phenomena.

The specification data required from the probe are as follows :

- operability in a drill-hole without time limitation (in practice, 6 months),
- complete autonomy, no supervision, no power feed,
- good exchange between the drill-hole water and the inside of the probe under static conditions (no pump operating),
- no pollution of the environment.

B3. Description of the probe (Fig. 1)

The probe is composed of a reaction chamber, of 5 l. capacity (65 mm dia., 1500 mm high), capable of containing 10 samples (approximately 10 mm dia., 50 mm high).

The tracer is contained in a glass bulb which will be ruptured by a weight (messenger) sliding along the carrier cable.

The reaction chamber may be put in communication with the drill-hole by windows (36 cm^2 surface) which may be actuated using the drill-hole pressure. It suffices for this to lower the probe by a few meters, which acts upon a hydraulic valve held closed by a spring set at a pressure corresponding to the drill-hole depth ; the valve, when opening, permits the action of the hole water on a piston opening or closing the windows.

B4. Experiment

The probe will be used for measuring the *in-situ* migration of radionuclides in clays.

a) **Equilibrium stage**

In a first stage the argillaceous material samples (in 10 mm dia., 50 mm high tubes) are left to come into an equilibrium with the drill-hole water. The water diffuses into the material ; a diffusive front of equilibrium is formed and will slowly progress for several months.

b) **Experimental stage**

When the foregoing stage is completed, the windows get closed, the tracer (double tritium-lanthanide or actinide marking) is released from the glass bulb using the messenger (this also puts an electric cell-driven agitator into action).

The tracers are diffused into the clay. A second diffusion front is formed, offset with respect to the first one and propagating in a zone already settled to a chemical balance with the drill-hole water.

B5. **Tests planned**

- Qualification of the probe on the AURIAT site.

Samples are selected with various sand contents and various natures of argillaceous materials.

The tracers will be Tritium and Europium.

The experiments will take place :

. in the granitic site of PARTENAY,
. in the saline site of GOERLEBEN,
. in the argillaceous site of MOL.

Use will be made of Tritium, of an actinide or, if unavailable, of a lanthanide.

The number and composition of the samples will be determined for each particular case, filling materials may be used, as necessary.

C. WORK PROGRESS AND RESULT OBTAINED

C1. Work Progress and Results Obtained

The probe was modified for use in a shallow (30 m) borehole at CCR/ISPRA : the probe is operated by borehole pressure and a hydrostatic pressure of 5-6 bars is necessary to allow for friction (fig. 2).

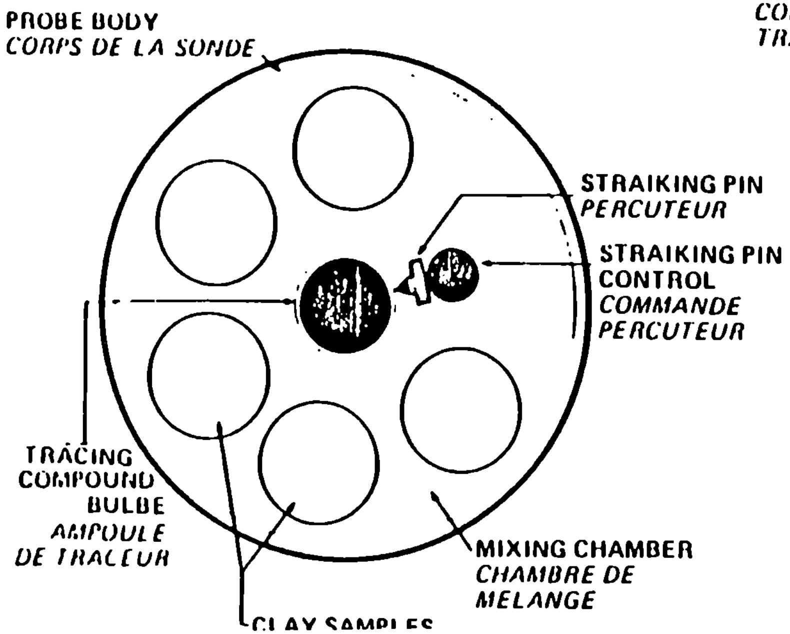
The system can be operated at a depth of 30 m under nitrogen pressure. The sample recovery system, based on slow depressurization of the reaction chamber, is now completed, although some problems have occurred due to sample outgassing (Fig. 3).

C2. Drawings

A radioactive tracer test will be conducted at CCR/ISPRA.

AUTOLAB PROBE
SONDE AUTOLAB

MESURMENT OF RADIOELEMENT MIGRATION
IN SITU IN DIFFUSING MODE
MESURE DE LA MIGRATION DES RADIOELEMENTS
IN SITU EN REGIME DIFFUSIF



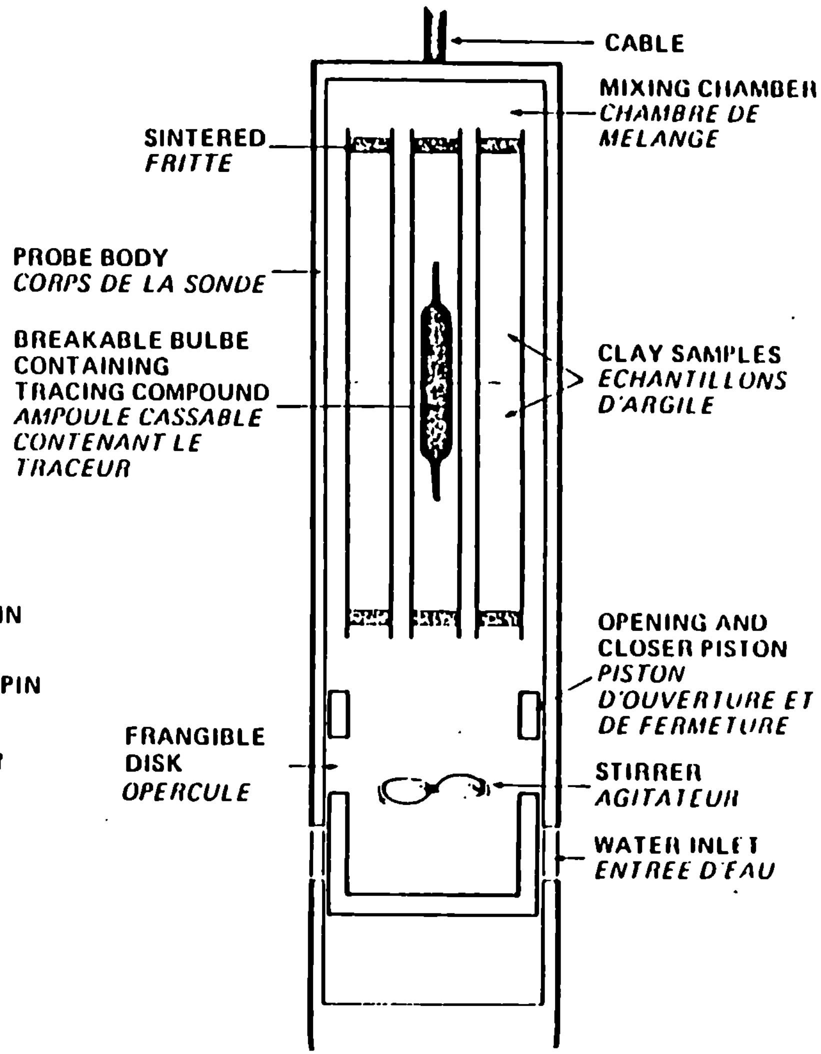


Figure 1

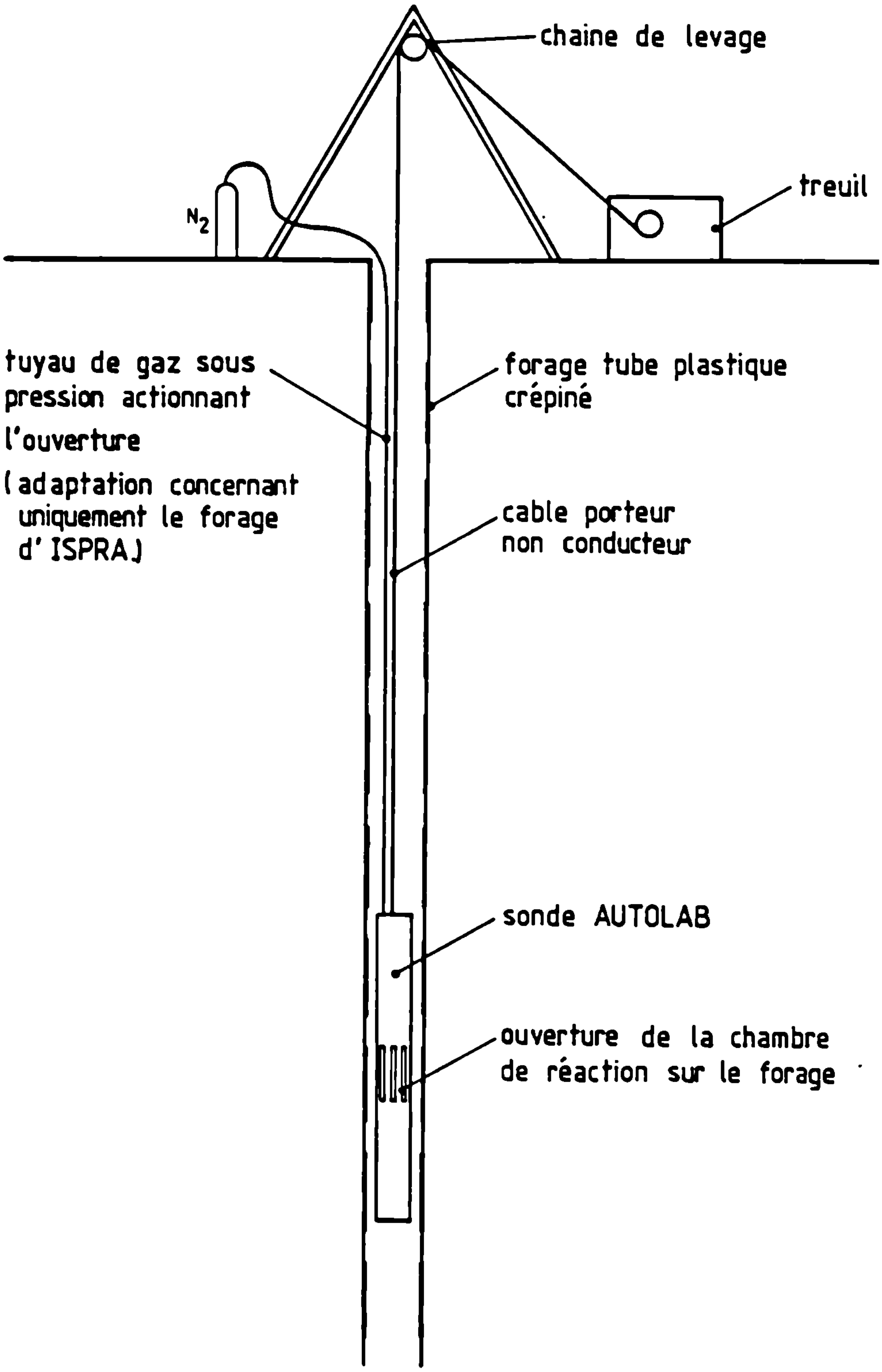
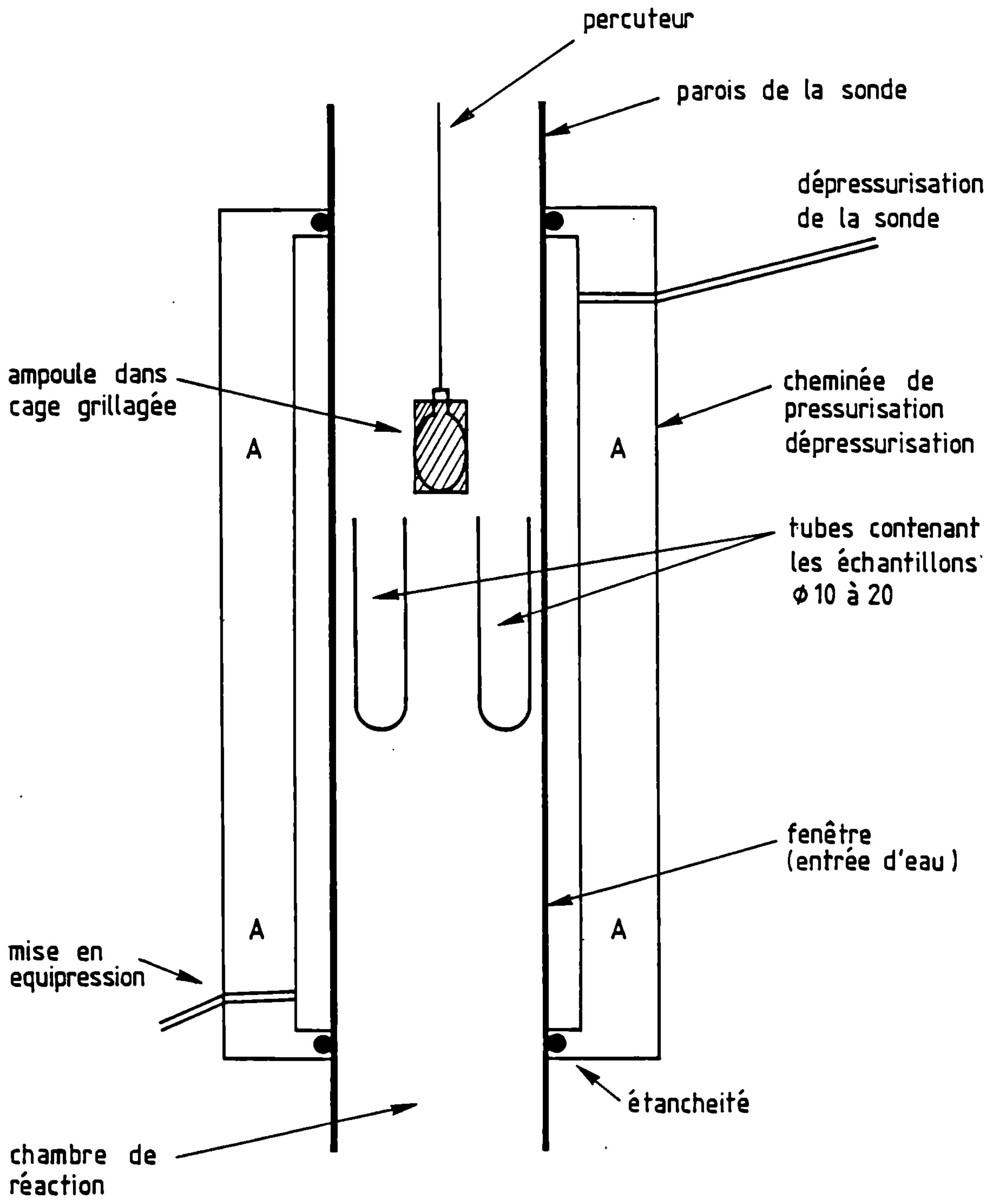


Figure 2 : Schémas de principe de l'adaptation de la sonde

Figure 3 : Système de dépressurisation lente de la sonde



Remarque : La partie A ne fait pas partie de la sonde mais sert à la dépressurisation de la sonde.

IN SITU DETERMINATION OF THE MACROPERMEABILITY OF A CLAY FORMATION IN VIEW OF ASSESSING LEAKAGE AND MASS TRANSFER IN A DEEP ARGILLACEOUS FORMATION

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0145
Duration of contract : from October 1986 through June 1990
Period covered : January 1989 - December 1989
Project Leader : A.A. Bonne
Reported by : J. Bronders

A. OBJECTIVES AND SCOPE

The hydraulic conductivity of the Boom clay situated at Mol (Belgium) will be determined by means of a macropermeability test. Knowledge of this parameter is an important factor in the study of the dispersion and travel of the radionuclides in the deep argillaceous formation.

A method of defining hydraulic characteristics of tight formations is found in the literature /4/ and transformed into a test which can be conducted in the clay formation in the underground gallery at Mol. The presented method is a modification of a convention slug test /1/, /2/ and /3/. The technique has been used in very tight formations, for example, permeabilities corresponding to values of 10^{-13} - 10^{-16} m/s have been reported.

At this stage of the research the theoretical and practical description of the test has been made. A global work scheme has been set-up for conducting the set-up and the measurements.

B. WORK PROGRAMME

1. Detailed design of the experimental set-up and development of the appropriate research model for the optimisation of experimental set-up.
2. Purchase of the instrumentation and equipment.
3. Calibration, testing and mounting of the instrumentation and equipment.
4. Performance and follow-up of the experiment.
5. Further adaptation/development of the research model for the final interpretation of the experiment.
6. Termination of the experiemnt and reinstatement of the initial conditions.
7. Final interpretation.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The test programme has been redefined during the year 1989. Till now there are no measurements available to calculate the parameters since the set-up of the test is not yet accomplished.

Progress and results

The principal experiment for determining the hydraulic characteristics exists of a test where the decay of a head change caused by pressurizing the volume of water stored in a shut-in well is recorded. Some variations of this test will also be conducted in the same well set-up. The hydraulic conductivity (K in m/s) and the storage coefficient (S dimensionless) can be determined by matching observed data with type curves or by calculation techniques as used for slug tests.

Set-up

In figure 1 the set-up of the experiment is given. A sintered inox filter of 15 m length with a diameter of 0.15 m is placed in a vertical borehole of 25 m depth. Around the filter two series of piezometers are installed, in this way it is possible to follow the pressure distribution around the filter during stabilisation and experiments

Figure 2 gives and overview of the tests which will be conducted.

a) During stabilisation of the borehole, discharge of water from the formation into the filter will be measured (Figure 2.a).
b) For the measurements where a pressure puls is used, packers are installed to create a shut-in part of the well. The pressure change in the pressurized part is observed and will yield the calculation of K and S (Figure 2.b and 2.c).
   Following measurements are planned :
   - after a sudden pressurization the pressure change is measured,
   - a second measurement is done by keeping the pressure in a shut-in part of the well constant by continuously pumping in water. If the discharge is known, parameters can be calculated ;
   - measuring the pressure stabilization after a sudden lowering of the pressure in a part of the well ;
   - pumping in a constant discharge and measure the pressure rise (Figure 2.d).

Theory

The determination method of the hydraulic parameters K and S is dependent on the type of measurement. Three methods are used :

- "Steady state" : K is calculated with following formula
  $K = Q/FPo$
  Q = discharge
  F = filter shape factor
  Po = pressure difference
- graphical methods : type curves are used to determine K and S for test under pressure
- pressure calculations in anology with models used for thermal calculations

If the presented method gives good results it is planned to conduct more tests of this type in different directions.

<u>List of publications</u>

/1/ Cooper, H.H., Bredehoeft, Jr., J.D. and Papadopulus, I.S., Response of a finite diameter well to an instantaneous charge of water, Water Resources Research, 3(1), pp 263-269 (1967).

/2/ Papadopulus, S.S., Bredehoeft, J.D. and Cooper, H.H. Jr., On the analyses of "slug test" data, Water REsources Research, 9(6), pp 1048-1089 (1973).

/3/ Dax, A., A note on the analysis of slug tests, Journal of hydrology, 91, pp 153-177 (1987).

/4/ Bredehoeft J.K., Papadopulus, S.S., A method for determining the hydraulic properties of tight formation, Water Resources Research, 16(1), pp 233-238 (1980).

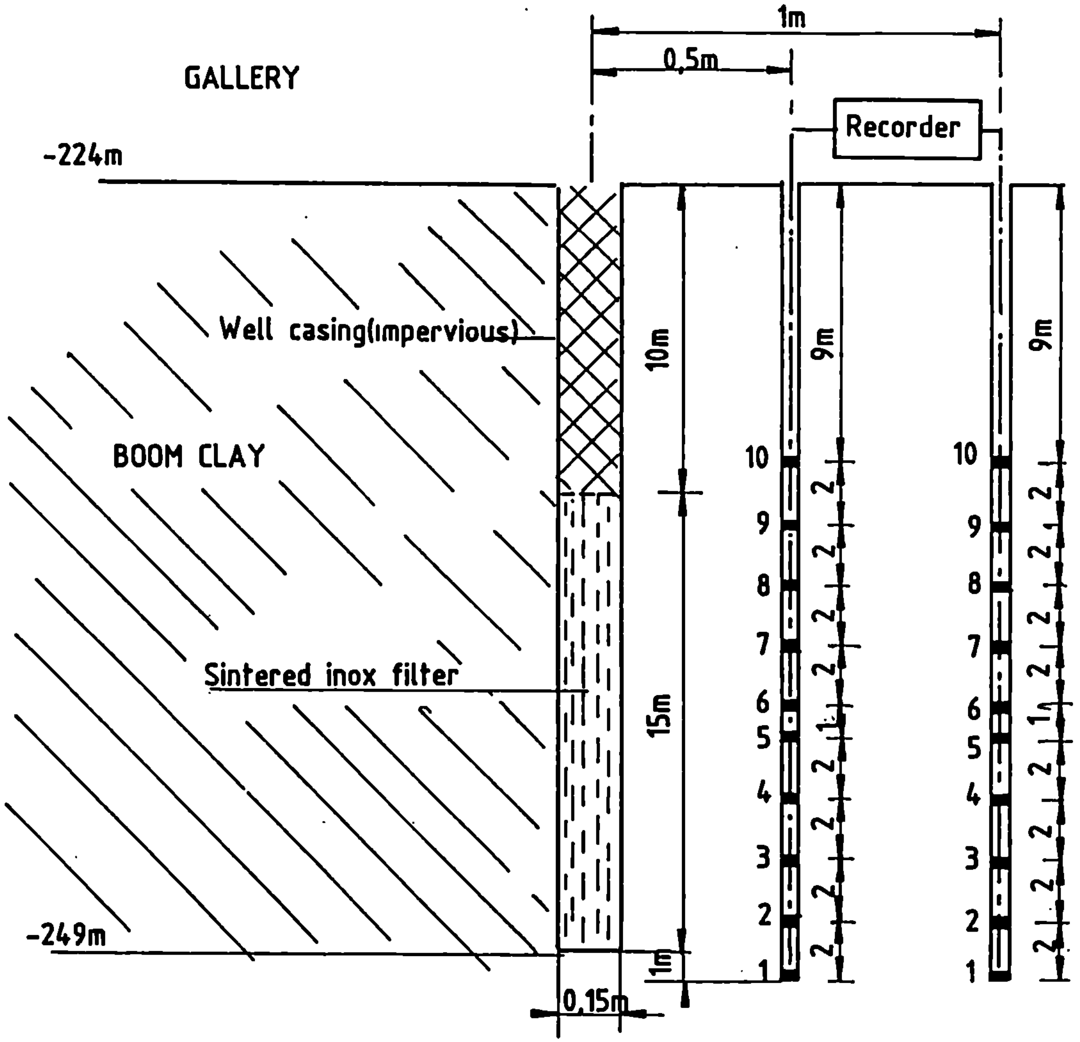


FIG. 1 SUGGESTED SET-UP OF THE TEST WELL

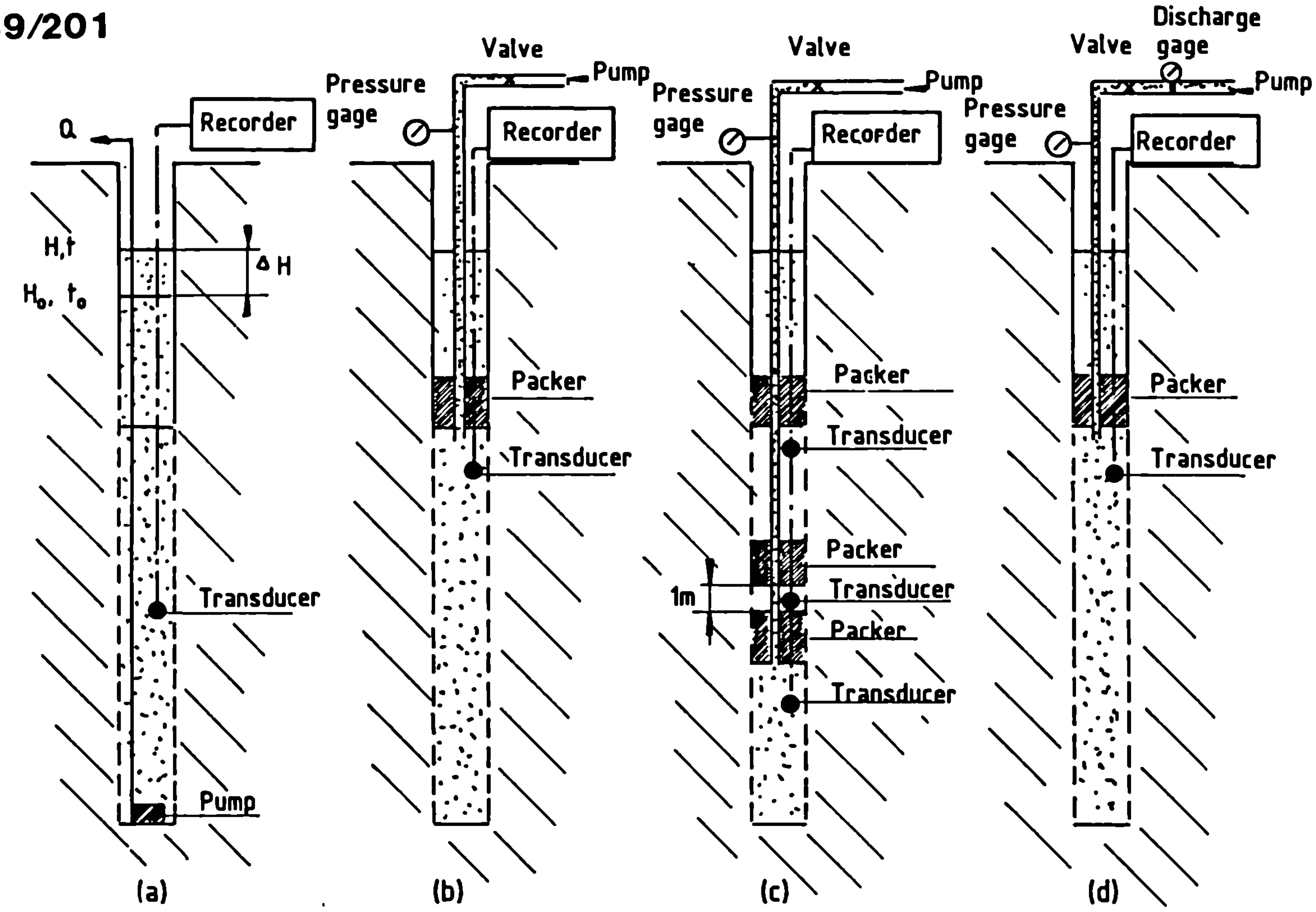


FIG. 2 SUGGESTED ARRANGEMENTS FOR THE DIFFERENT TESTS IN THE BOOM CLAY

LABORATORY AND FIELD TESTS FOR RADIONUCLIDE MIGRATION AND HIGH FLOW PATHS IN CLAY

Contractor : UKAEA, Harwell Laboratory, UK
Contract No: FI1W/0154
Duration of Contract : 1 January 1988 - 31 December 1989
Period Covered : January 1989 - December 1989
Project Leader : Mr P J Bourke

A OBJECTIVES AND SCOPE

Two field programmes are contained within this task. Their objectives are to investigate mass transfer in clay-rich geological materials.

The principal investigation is at Culham Laboratory, where it is proposed that water flow within the Kimmeridge Clay is measured. The objective is to determine if silt-rich or carbonate-rich horizons within the Kimmeridge Clay act to provide fast transport paths for water flow through this unit.

A subsidiary investigation is to be undertaken at SCK/CEN Mol, Belgium, where an in-situ measurement of solute transport by diffusion will be made.

B WORK PROGRAMME

Culham

- B1 Geological characterisation of Kimmeridge Clay unit, using cores recovered from boreholes.
- B2 Development of suitable techniques for measuring water flow in the Kimmerige Clay.
- B3 Measurement of hydraulic conductivity at different horizons in the Kimmeridge Clay, in order to investigate potential fast flow horizons.
- B4 Comparison of in-situ and laboratory measurements of hydraulic conductivity.

Mol

- B1 Design and manufacture of suitable apparatus to install in Port B, Underground Research Laboratory.
- B2 Design and manufacture of automated, computer controlled collimated radiation detection apparatus.
- B3 Installation of apparatus in Port B, Underground Research Laboratory.
- B4 Laboratory-scale trial tests to provide data for comparison with in-situ test.
- B5 In-situ test to measure diffusivity of Boom Clay using I-131 tracer.

C. PROGRESS OF WORK AND OBTAINED RESULTS

Two field investigations have been undertaken in this programme. Their objectives are to investigate mass transfer in clay-rich geological materials.

One investigation is at Culham Laboratory, Oxfordshire, England where it was proposed that water flow within the Kimmeridge Clay was measured. The objective is to determine if silt-rich or carbonate-rich horizons within the Kimmeridge Clay act to provide fast transport paths for water flow through this unit. The other investigation was undertaken in the Underground Research Laboratory at SCK/CEN Mol, Belgium where an in-situ measurement of solute transport by diffusion was proposed.

Drilling and field measurements to determine the hydraulic conductivity of a silty layer in the Kimmeridge clay at Culham have now been completed as planned.

These exploratory holes were drilled and cored on a 15m circle to depths of about 40m. These penetrated the Kimmeridge clay at depths of about 10m and the silty layer of about 0.5m thickness was identified in all three cores at depths of 27.5, 27.8 and 28.0m.

A fourth hole was drilled from the centre of the circle to a depth of 0.5m above the expected depth of the silt. A thin walled, sharp edged tube was then driven down from this depth until perforations in its wall were level with the silt, and the clay and silt inside the tube were augured out. Water was pumped from the tube at a steady pressure and flowed into the silt and analysis of the results has shown that the hydraulic conductivity of the silt to be $\geq 10^{-8}$ m s^{-1}. Comparative tests in the clay showed its conductivity to be at least fifty times less.

No progress has been made on this in-situ diffusion experiment in the Underground Laboratory at Mol, during this year. Previous progress reports detail the problems that have arisen following the installation of the liner tube and experimental flange at Mol, in particular the state of clay in contact with the flange. The relaxation of the clay to fill the void space behind the experimental flange was accompanied by considerable water softening. This disturbance has rendered the experiment of little scientific use, as undisturbed clay cannot new be tested with the present equipment without further significant engineering modification.

4.3.C. Natural analogues

FIELD INVESTIGATION WITH REGARD TO THE IMPERMEABILITY OF CLAY FORMATIONS.

Helium-4 in soil gas in sedimentary basins: a tentative study of secondary permeability in clayey formations (Paglia Valley).

Contractor : ENEA, CRE Casaccia, Rome (Italy)
Contract No : FI1W/0063-I
Duration of contract : January 1987 -December 1989
Period covered : January 1989 - December 1989
Project leader : C. Polizzano

A. OBJECTIVES AND SCOPES

The main aim of this work is to asses surficial methods for detecting the secondary permeability in clay and shale in sedimentary basins by means of direct in situ observations and detection of some noble gases in soil-gas as tracers of fractures, faults etc.

The chosen gases are helium 4 and radon 222. Both these gases are used from several years for locating ore, oil and geothermal fluids as well as earthquake precursors.

In this report only helium will be discussed. The helium in the atmosphere is a mixture of helium of different origin: radiogenic helium mainly from the metamorphic basement, primordial helium from the mantle. The rate at which helium escapes from the crust is lower than the rate of its production so that the crust itself may represent an accumulation zone for this element. Helium reaches the surface using fractures and faults as preferential routes for escaping. The choice of this element for detecting fractures relays on:

- it is a chemically fully inert element;
- its atomic radius is very small, comparable with that one of hydrogen.

These characteristics make helium one of the most mobile elements and therefore it can behave as an excellent tracer of geological continuities as well as of the primary permeability of the rocks.

This report presents the results of the survey done in the zones of Val di Paglia and in Val di Chiani (central- western Italy), as a continuation of the preliminary researches carried out in the past year as well as the preceding one conducted in Val d'Era and at Vasto.

B. WORK PROGRAMME.

1 - Preliminary survey of helium in soilgas at a regional scale in sedimentary basins characterized by distensive tectonics and, for comparison, by compressive ones;
2 - In situ observations and information from operators about the hydrological meaning of fractures in clay.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

In the preceding reports, related to the studies conducted in Val d'Era (central western Italy), a structural deep characterized by distensive tectonics, and at Vasto, close to the Adriatic Sea, a basin in the adriatic foreland, interesting relationships between helium leakage and structural features were individuated. In both areas the helium leakage did not seem related with the lithology of the outcropping formations (clay and sand of Pliocene age) nor with their thickness. The highest helium positive anomalies in Vasto were found in a zone were the pliocenic clay formation reaches the maximum thickness (1400 m). As a complementary result it was demonstrated that, excluding particular situations depending on local factors, tectonic dislocations seemed not to represent preferential pathways to groundwater migration at depth.
More than 2000 soilgas samples were collected and analyzed in the precedently surveyed areas, including the preliminary survey in the Paglia Valley.
The Paglia Valley (figure I) coincides with a structural depression attributable to the post-orogenic faulting phase, which involved the region chiefly from Upper Miocene, causing first the formation of lacustrine basins during the Messinian and then the marine transgression in the lower Pliocene.
Two structural highs, due to distensive tectonics, border the Paglia Valley according to the appenninic, NW-SE, direction in the

northern part and to a N-S direction in the central and southern part.
Quaternary vulcanism, geothermal fields and thermal springs are distributed aside or underlay the pliocenic clay basin.

In the Chiani-Paglia basin more than 1700 soilgas samples were collected from 1987 to 1988. Other supplementary samples (400) were taken along profiles crossing the Paglia Valley during the summer of 1989. The Chiani is a tributary of Paglia river and its valley is located on the eastern side of Paglia itself.
Samples were collected according to a regular grid with an average density of about 1.5 sampling points per square kilometre.
The soilgas samples were collected by means of hollow probes inserted into the soil at a depth ranging from 40 to 60 cm.
The ^4He contents in soilgas were detected by means of a mass spectrometer.
The helium content (5240 ppb) in the atmosphere is taken as a reference standard.

Table 1 reports a comparative picture of the helium distribution from the different surveyed areas.

Figure 2 shows the frequency distribution of the obtained results for the single part and for the whole set of data.

Filtering the data, subtracting the regional trend to the regular grid of average data, it is possible to obtain a map (fig. 3) showing the residual anomalies. This map enhances the results; it can be seen in fact that:
- values higher than the background are present in both valleys;
- values lower than the background are present in the westernmost margin of the surveyed area , in correspondence of the high structure of the Cetona Mt. and, east of this, in a wide area elongated N-S. These lower values correspond to the outcrop of chaotic clay (argille scagliose) or to the outcrop of fractured and permeable formations that act as recharge areas for the ground water. It seems that both chaotic clay and permeable, by fracture, formations may form a barrier to the deep origin helium leakage. If it is so, when impermeable lithotypes are not present, it is possible to use the lower values (negative anomalies) because they indicate impermeable (to deep origin gas) formations that could potentially contain radioactive waste.

According to the Val d'Era and Vasto results, in both areas the helium leakage does not seem related with the lithology of the outcropping formations nor with their thickens. The highest helium positive anomalies were found in a zone where the pliocenic clay formations presumably reach the maximum thickness. In both areas the major anomalies are aligned according to the main fault and fracture systems directions.
From the comparison between these two areas and those already studied it seems possible the conclusion that the open fracture and fault systems have the main control on the ^{4}He distribution in soil gas. The highest ^{4}He anomalies were usually found in zones where deep ^{4}He rich reservoirs exist: natural gas and oil in Vasto region; geothermal fluids in Paglia Valley. The data, so far collected, strongly support the hypotheses of a possible use of the helium as faults tracer in clayey formations where other geochemical tracers may fail. These results are, also, in agreement with those obtained by other researchers that use soil gas geochemistry for detecting faults in clay formations.

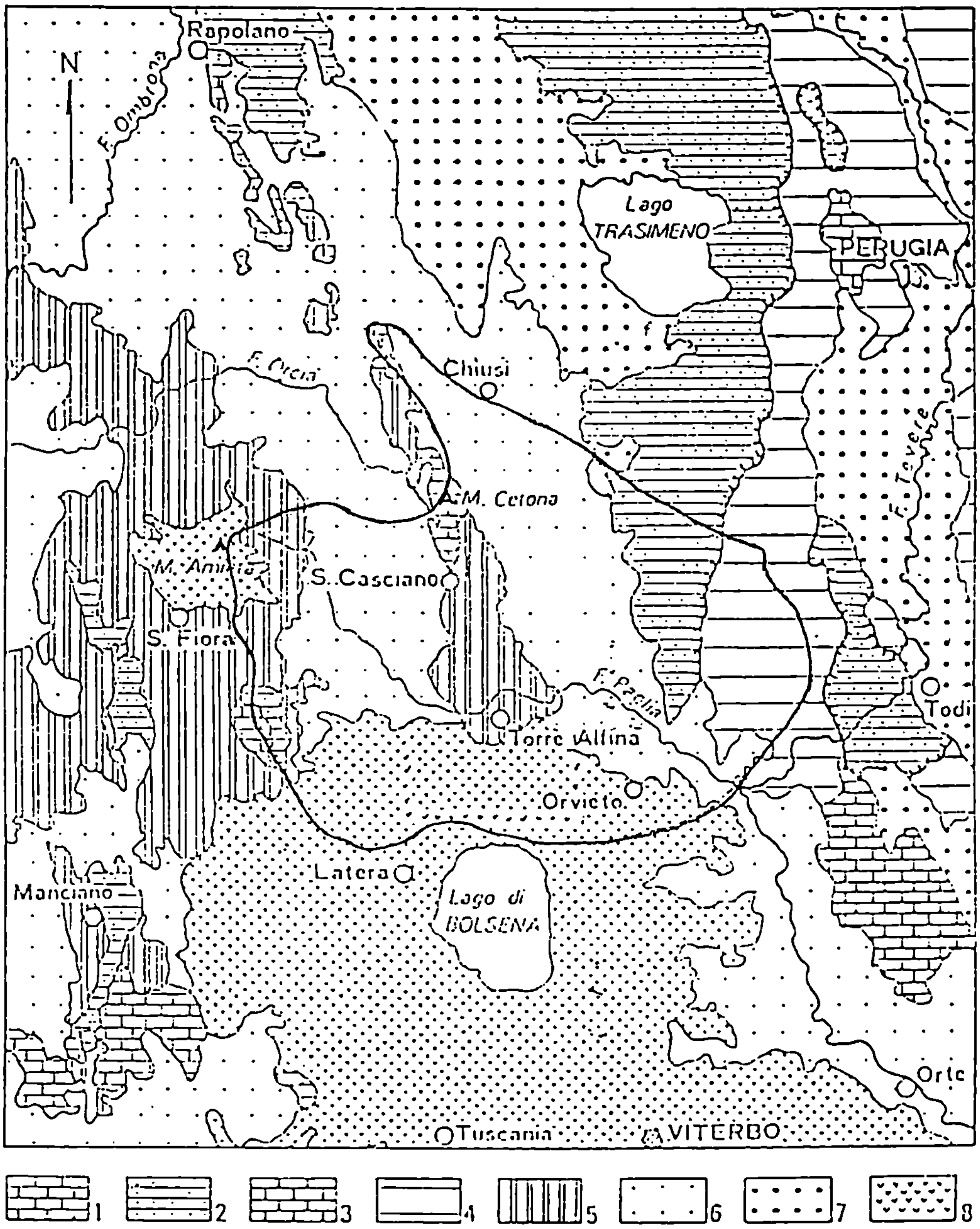


Figure n. 1 - Geological outline of the surveyed area. Legend. The complexes of Tuscan facies: Mesozoic carbonatic succession (1); Mesozoic-Tertiary succession (2). The complexes of Umbro-Marchigiana facies: Mesozoic carbonatic succession (3); Mesozoic-Tertiary terrigenous succession (4). Facies Ligure and internal Austroalpine complexes (5). Marine Miocenic deposits (6). Continental Plio-Quaternary deposits (7). Volcanic complex (8). The solid line delimits the surveyed area.

Area:	Mean:	Std.Dev:	Mode:	Min.Val.:	Max.Val:	Total samples:
Paglia	432	393	450	- 500	+ 3600	912
Chiani	270	186	300	- 460	+ 760	797
Pa.+Chi.	357	325	300	- 500	+ 3600	1709
-------	-------	-------	-------	-------	-------	------
V.d'Era	212	129	180	- 350	+ 670	919
Vasto	800	1114	----	-1300	+11030	824

Table n. 1 - In this table the obtained results are summarized. The data are expressed in ppb (v/v) as difference between the helium contents in the samples and the atmospheric helium. For comparison, the results of Val d'Era and Vasto surveys are also reported. As it is possible to see the results obtained in the Chiani Valley are similar to those found in Val d'Era whereas those observed in the Paglia basin are closer to the results of Vasto region.

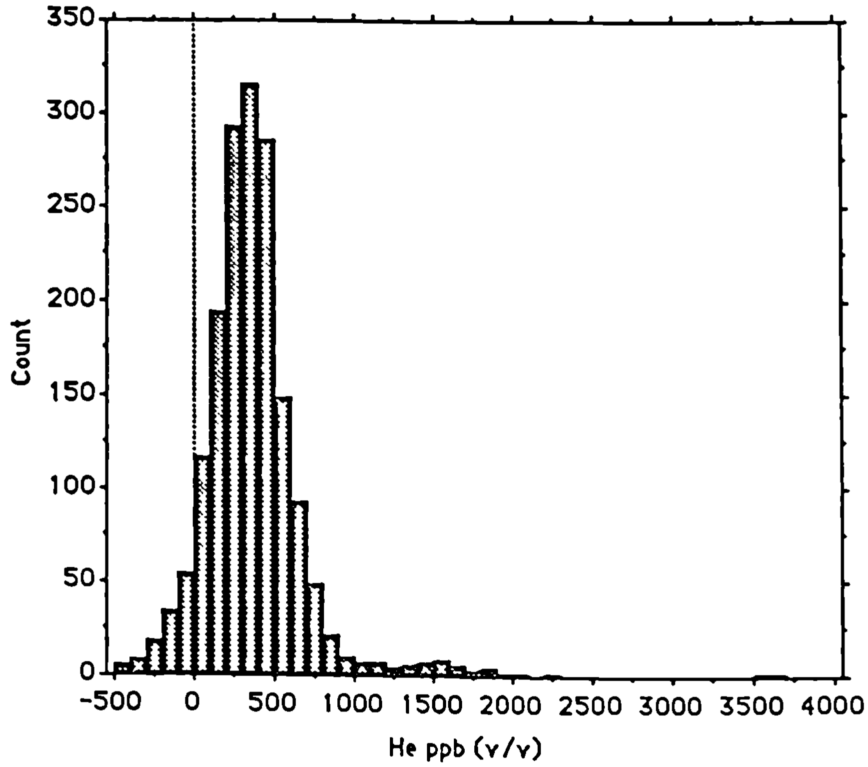


Fig. 2 - Frequency histogram showing the whole set of helium data collected in the Chiani -Paglia Basin.

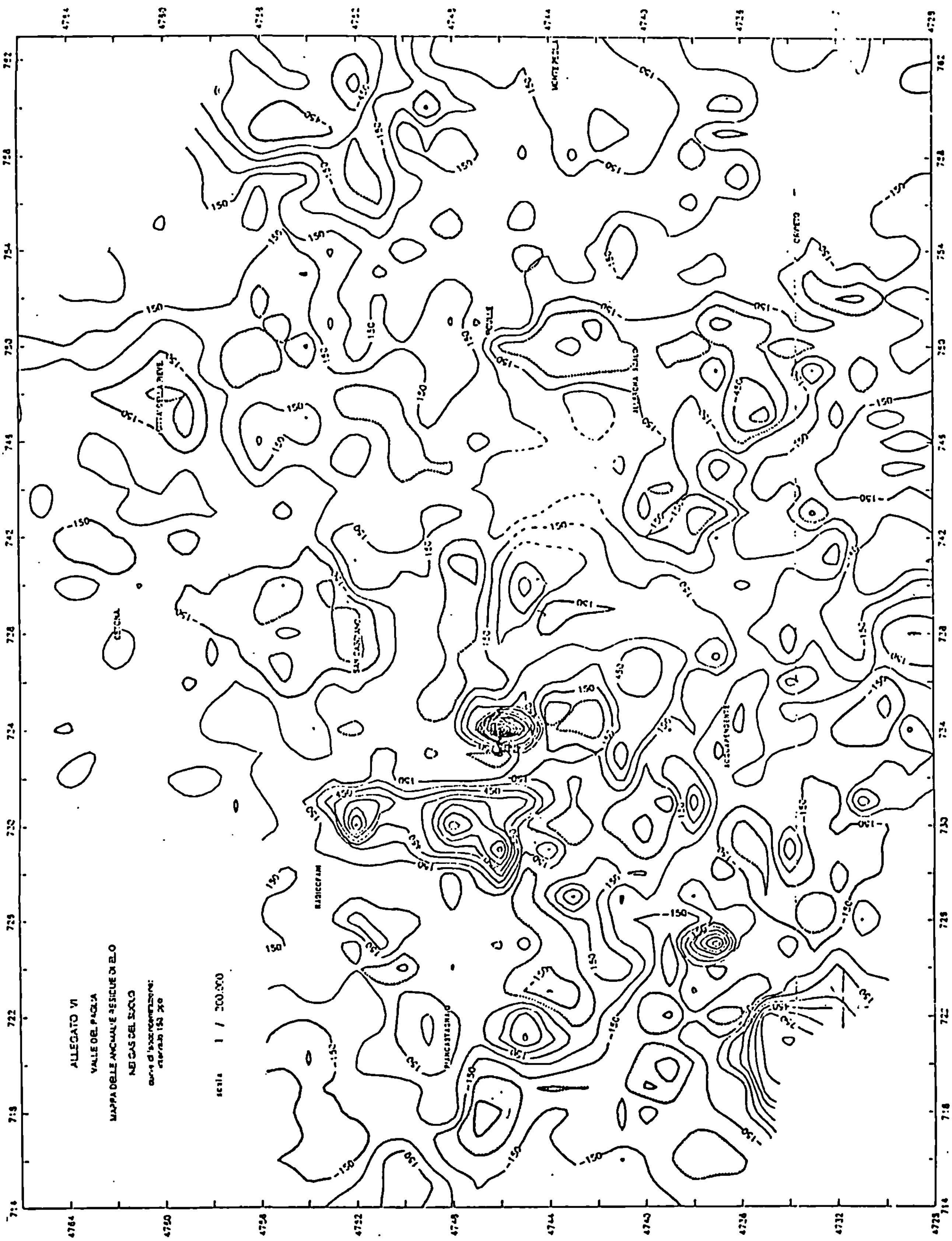


FIG. 3 - Helium in soilgas residual anomalies in the Chiani-Paglia Valleys. The contour line interval is 150 ppb. The coordinates are kilometrics.

MODELLING OF RADIONUCLIDE MIGRATION IN THE GEOSPHERE : NATURAL ANALOGUE STUDIES

Contractor : Centre d'Etude Nucléaires de Fontenay-aux-Roses, CEA/IPSN/DAS/SAED - FRANCE

Contract n° : FI1W/0070

Duration of contract : January 1987 - June 1989

Period covered : January 1989 - June 1989

Project leader : P. Escalier des Orres

A. OBJECTIVES AND SCOPE

The study of the migration processes of elements in geosphere through natural analogues is one of the best way to validate the calculation tools developped to predict radionuclides transfers on long time periods.

It has for objective to quantify these phenomena with slow kinetics, to show the thoroughness of the mechanisms taken into account in the models, or to permit their modification, if necessary.

The present study is based on the interpretation of the data obtained by the British Geological Survey on different "natural analogues" sites in the U.K.

B. WORK PROGRAMME

1. Modelisation of the elements transfers in the argillaceous sediments from Loch Lomond.

2. Pre-modelisation of natural analogues sites on the first sets of data gathered by the BGS.

3. Interpretation through models of the complete data sets : transfer in cristalline rocks (matrix diffusion), transfer in clay quaternary sediments, for example.

C. PROGRESS OF WORK AND OBTAINED RESULTS

1. State of advancement.

The 1989 objectives of the Needle's Eye natural analogue study were to bring the modelling work of the site to a successfull conclusion. This aim involved the three following requirements :

. To build a flow model of the site, according to the available hydrogeological data,

. To build a geochemical model, according to the chemical and geochemical data collected,

. To merge the two previous aspects into the building of a coupled model to reproduce the observed uranium concentration field.

2. Progress of work and obtained results.

In the hydrogeological model, and due to the very few hydrogeological and piezometric data, we tried to reproduce a qualitative behaviour of the aquifer in the silts. An upward flow is observed in the northern part of the area whereas the southern part seems to be well drained.

A regional model was build first, including the whole watershed of the silts aquifer where the flow was assumed to be a two dimensionnal (N-S) one. The geological units taken into account, north to south, were the Criffell Batholite (Clifton Craig), the Silurian Hornfels and the Carboniferous Limestones. The last two units are separated by a major hercynian fault which is finally overlaid by early quaternary silty sediments. The extension of the regional model was 2 kms, from which an area of silts of 80 m long was extracted for the purposes of the local hydrogeological modelling.

The local model was totally constrained by the computed head values from the regional model. The permeability field within the silts was adjusted in order to reproduce the observed flow configuration. This was made possible by assuming a semi-permeable fault and an anisotropic permeability of the silts.

Both modelling were done using the METIS flow model.

On the geochemical side of the work, the samples collected during the December 88 campaign were computed with the CHIMERE code, enlightning the role of uraninite in the control of the dissolved uranium. The work done in 1988, on the geochemically active minerals was thus resumed and it was concluded that the role of uraninite precipitation in the humics was a potential process to explain the lowering amounts of dissolved uranium within a few meters.

The uranium transport was first simulated with the METIS code, assuming reasonable uranium concentrations ($\approx$ 100ppb) to emerge from the hornfels zone (where the actively leached uranium minerals are situated). It was shown that the specificity of the groundwater flow in the silts was largely responsible for the dissolved uranium field resulting in a northern uranium-rich area and a southern poorly contaminated zone. The K_d approach was then used, based on laboratory results of experiments involving humic materials extracted from the site. It was shown that this approach cannot account for the uranium depletion within a few decimeters as seen in the two northern boreholes.

Finally, the STELE coupled model was used to test the uraninite precipitation, assuming an Eh depletion of 200m V in the humics, a value that was shown (with the CHIMERE code) sufficient enough for the whole dissolved uranium to disappear from the aqueous phase. The computed dissolved uranium stratification in the two boreholes north of the site are in good agreement with the field data, when assuming a precipitation kinetic of 1 day $^{-1}$.

The steady state is obtained after only one month, giving a total accumulated uranium in the sediments of 500g/ton in 5000 years (which is a probable duration of the observed accumulation process). This order of magnitude is close to the one found in commercially mined uranium. However, it was not possible to compare this value to measured ones.

3. Conclusions.

The obtained results are interesting for several reasons :

First of all, on a methodologic aspect, the collaboration and exchanges between the geologists, the experimentalists and the modellers has shown usefull to bring an important international sceintific project to a fruitful end. The complementary tasks that have been undertaken made it possible to characterize (even if some uncertainties remain) the main features of the coupled processes involving both the transport and the geochemical phenomena.

Secondly, the applicability of a coupled model to a large scale real case has been tested. This is an important point when considering the whole validation and devopment work on this tool in view of their final use in the framework of safety analysis of nuclear waste disposals.

Finally, the modelling work on the Needle's Eye project has shown that simple geochemical hypothesis in a roughly known hydrogeochemical context may account for the observed uranium concentration field. However, an important laboratory, field, modelling, if not conceptual work is needed in order to cover the remaining areas of uncertainty. The main important aspect to be carried out would be to determine whether the geochemical processes taken into account here are relevant or not to the problem in question. This will probably be included in a new CEC-programme to come.

4. LIST OF PUBLICATIONS.

JAMET Ph., LACHASSAGNE P., DOUBLET R.,
Modélisation hydrogéochimique du site de Needle's Eye, rapport d'avancement au 31.12.1988. Rapport CIG-LHM/RD/89/66

JAMET Ph., LACHASSAGNE P., DOUBLET R., LEDOUX E.,
Modélisation hydrogéochimique du site de Needle's Eye, rapport final, in print

JAMET Ph. LACHASSAGNE P., DOUBLET R., LEDOUX E.
Migration of the radionuclides in the geosphere, MOdelling of the Needle's Eye Natural Analogue, final report 43 p., rapport CIG/LHM/RD/89/81.

JAMET Ph., LACHASSAGE P., E. LEDOUX, P.J. HOOKER, P. ESCALIER DES ORRES,
Modélisation hydrogéochimique de l'analogue naturel de Needle's Eye (Ecosse). To be presented to GEOVAL-90, Stockholm, May 14/17, 1990.

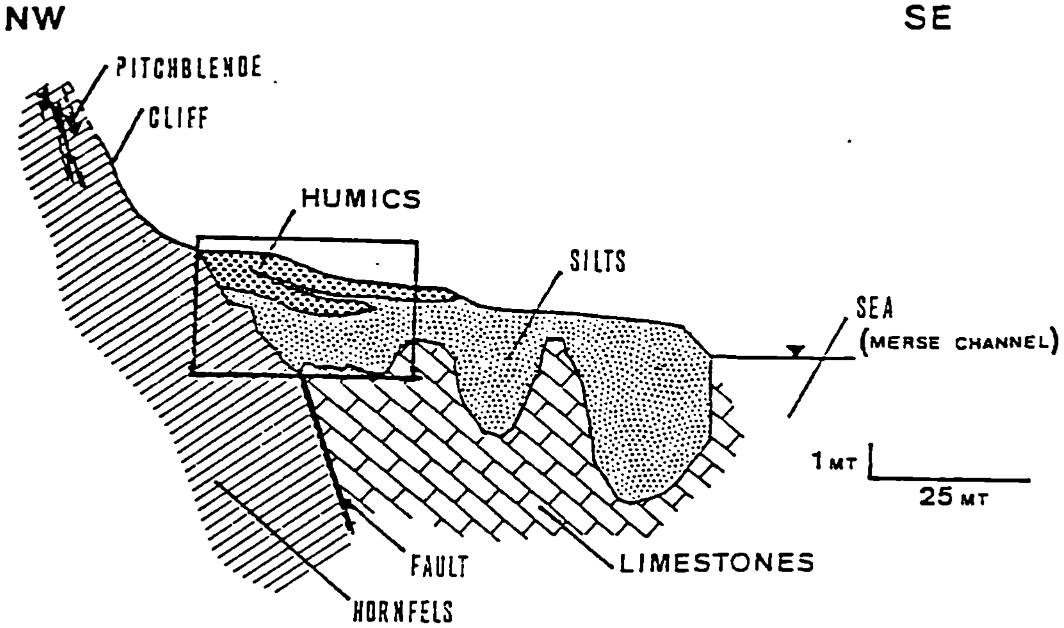


Fig. 1 : The Needle's Eye Site

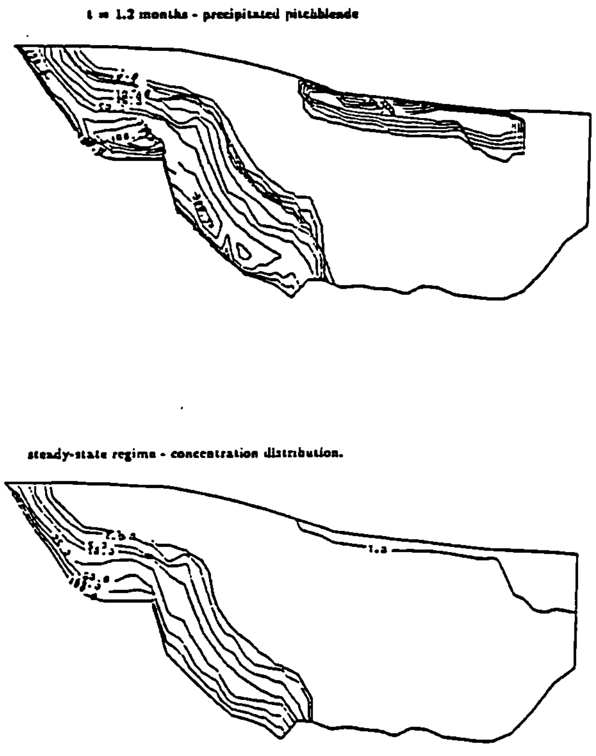


Fig. 2 : Pichblende precipitation calculated distribution of concentrations in metastable steady-State regime. Quantities of precipitated pitchblende values in ppb per unite volume of fluid phase.

STUDY OF MIGRATION PROCESSES IN CLAY FORMATIONS OCCURRING IN NATURE.

Contractor : ENEA, CRE Casaccia, Rome (Italy)
Contract No : FI1W/0071
Duration of contract : January 1987 - December 1989
Period covered : January 1989 - December 1989
Project leader : A. Brondi

A. OBJECTIVE AND SCOPE

The permeability state of clay may be affected by tectonic events. Fractures and faults may indeed give rise to a secondary permeability within argillaceous rocks. Ochraceous bands aside from fracture planes evidence circulation of meteoric water within fractures systems in clay. The penetration depth of these water in clay should reveal the thickness of clay affected by secondary permeability due to tectonics. The present work is a contribute aimed at ascertaining the real influence of tectonics on clay in particular formations in the Siena basin in central western Italy. The tectonics of the basin, a typical graben, is well defined.
Complementary researches in other situations, i.e., in Pasquasia mine, where clay are highly fractured, offer the opportunity of investigating a clay system isolated from surficial water.

B. WORK PROGRAMME

Three main lines have been envisaged:
1 - Selection of the most appropriate situations referred to the general tectonic frame;
2 - Studies on the variations of the geochemical system of clay because of the penetration of meteoric water;
3 - Investigation on the extent and causes of the secondary permeability of clay and their importance with regard to the geochemical stability of clay formation.

C. PROGRESS OF WORK AND OBTAINED RESULTS.

State of advancement.

Among the many possibility offered by the italian territory the Siena basin, located in central western Italy, has been considered as highly significative for the envisaged research.
The Siena basin is made by a deep tectonic distensive trench, parallel to the Appenninic chain, filled by a thick series of pliocenic clay and overlying sand. Because of the orogenic uplift still in course, the pliocenic series undergoes to accelerated erosion. Clay unloaded by the overlying masses, removed by erosional activity, expands in volume without a contemporaneous mass increase. This causes latent fractures to be opened and penetrated by surficial waters. A major effect of surficial water circulation is the oxidation of the fractures walls accompanied by a centimetric-decimetric alteration of the clay body. The clay oxidation along fractures may develop to a depth of some tens meters. These values may indicate the maximum penetration depth of surficial oxidizing waters within the clay.
It is general opinion that the tectonic elements affecting clay formations are very hardly recognizable in field. So the fracture systems are difficulty attributable to precise events. The data recently acquired in the Siena basin on the contrary prove that fracture systems in clay conform both to tectonics trends and to the landscape morphology. This correspondence may be also ascertained by means of an accurate use of satellite images.
The aim of this study is to reach an understanding of the causes and mechanisms underlying the formation and spatial distribution of the fractures systems in the clay of the neogenic basin of Siena, by evaluating the influence of tectonic and geomorphological factors, and biostratigraphic, chemical, mineralogical and physico-mechanical parameters.
In Tuscany, the orogenesis of the Apennine chain which began in the Oligocene, terminated in the upper Tortonian when the compressive was replaced by a tensional regime with the consequent relaxation of the upper crust end the creation of many tectonic depressions trending NW-SE and NNW-SSE (fig. 1).
The Siena basin is located in the intermediate stretch of a long depression system between the Monteriggioni "soglia" to the NNW and the Pienza "soglia" to the SSE. It corresponds to a true graben in which the maximum depth (> 100 m.) of pliocene sediments lies along an axis slightly to the east of the axis of symmetry. These sediments consist almost entirely of marine clays with a small percentage of sands and conglomerates variously interbedded. The main fault of the graben is on its eastern border and has a displacement which varies from 2000 to 600 m.

Twenty-nine measuring stations were selected in places in which the clay substrata is naturally or artificially exposed. The outcropping clays are crossed by nearly vertical systems of fractures of variable orientation, which do not show evidence of movement. These fractures continue to a depth of 10-15 m. The superficial levels normally have yellow-ochre coloured bands of oxidation, with a mean thickness of about 5 cm. With increasing depth the thickness affected by oxidation decreases gradually and finally disappears. In fresh artificial outcrops, the fractures are usually open and gradually narrow with increasing depth until they finally close. Close to the surface, the fractures extend horizontally for a distance of the order of decimetres to metres, and are straight or curved without any preferred orientation; dip is always 90° as shown in fig. 2a. This distribution and morphology is reminiscent of the vertical fracture systems of overconsolidated clays. At a depth of a few metres there is a gradual transition to a more regular distribution of the fractures. There is a system of variably spaced (20-30 cm. to 2-3 m.) parallel joints and a series of small fractures cutting the main system at about 90° (fig. 2b). The pattern of fractures shown in figure 2c occurs at even grater depth, usually more than 5 m. below the surface; minor fracture systems disappear, only the main system remaining. When observed on a 10 m. scale these outcrops are characterized by the constant parallel disposition of the fractures. Individual fractures vary in strike by about 20° and dip may vary from the mean near-vertical position by 10-15°. Thus the overall pattern is slightly arcuate, both in the vertical and horizontal planes, but substantially parallel.
Three zones were chosen in the area containing the most measuring stations and a detailed photogeological study of the lineations performed, using aerial photographs on a 1:13.000 scale. Lineations were considered to be alignements of elements, even of small dimensions, which generally signify precise morphological features. Elements which alone or in combination were used to identify a lineation were as follows:
- straight stretches of alluvial valleys;
- small straight steams generally corresponding to "anomalous" features of the hydrographic network implying structural control;
- rectilinear transitions between the foot of a slope and an alluvial plane;
- alignment of small saddles on ridges;
- sharp changes in shade in the photo, probably corresponding to changes in soil mixture;

- linear elements arranged in a lozenge in groups of "biancane" (clay domes rapidly eroding on the southern side); these are probably bands of intersecting fractures, sometimes slightly relatively displaced.
Considering the attitude of the main families of fractures in all the outcrops described (figure 3), it is evident that despite substantial dispersion, all the orientations cluster around the E-W direction. This suggest some link between the attitude of the fractures and the tectonic lineations.
If the fractures are compared to the pattern of the surrounding morphology, in most cases the attitude of the main system of fractures is parallel to the mean direction of the slope on which the fractures are formed. A relationship between fractures and morphology is also confirmed by the fact that:
- gradual changes in strike of the slope correspond to similar changes in the attitude of the fractures;
- this phenomenon is also found in areas in which the photointerpreted lineations show a well-defined parallel pattern;
- in areas of complicated morphology with many eroded valleys and slopes of variable direction, the relationship between fractures and morphology is uncertain.
In summary the following points emerging from the work performed are evidenced:
a) the systems of fractures studied in clays of the Siena basin have the characteristics of constant near-verticality, absence of movement and they extend down to a limited depth;
b) the complex geometry observed close to the surface is probably due to surficial physical phenomena (such as creep and changes in volume). These phenomena are superimposed on the dominant event which generated the main fracture system. At deeper levels where the fractures are nearly parallel, only the main system is recognizable;
c) although the situation appears very complex, it seems that a direct relation exists between fractures and tectonics, especially on a regional scale;
d) there seems to be a relationship between fractures and morphology. This relationship could be explained according to the results expected from the remark c); it can also account for a genetic mechanism of the salient features of the fractures based on tensile stresses from increased horizontal and vertical tensions due to erosion.

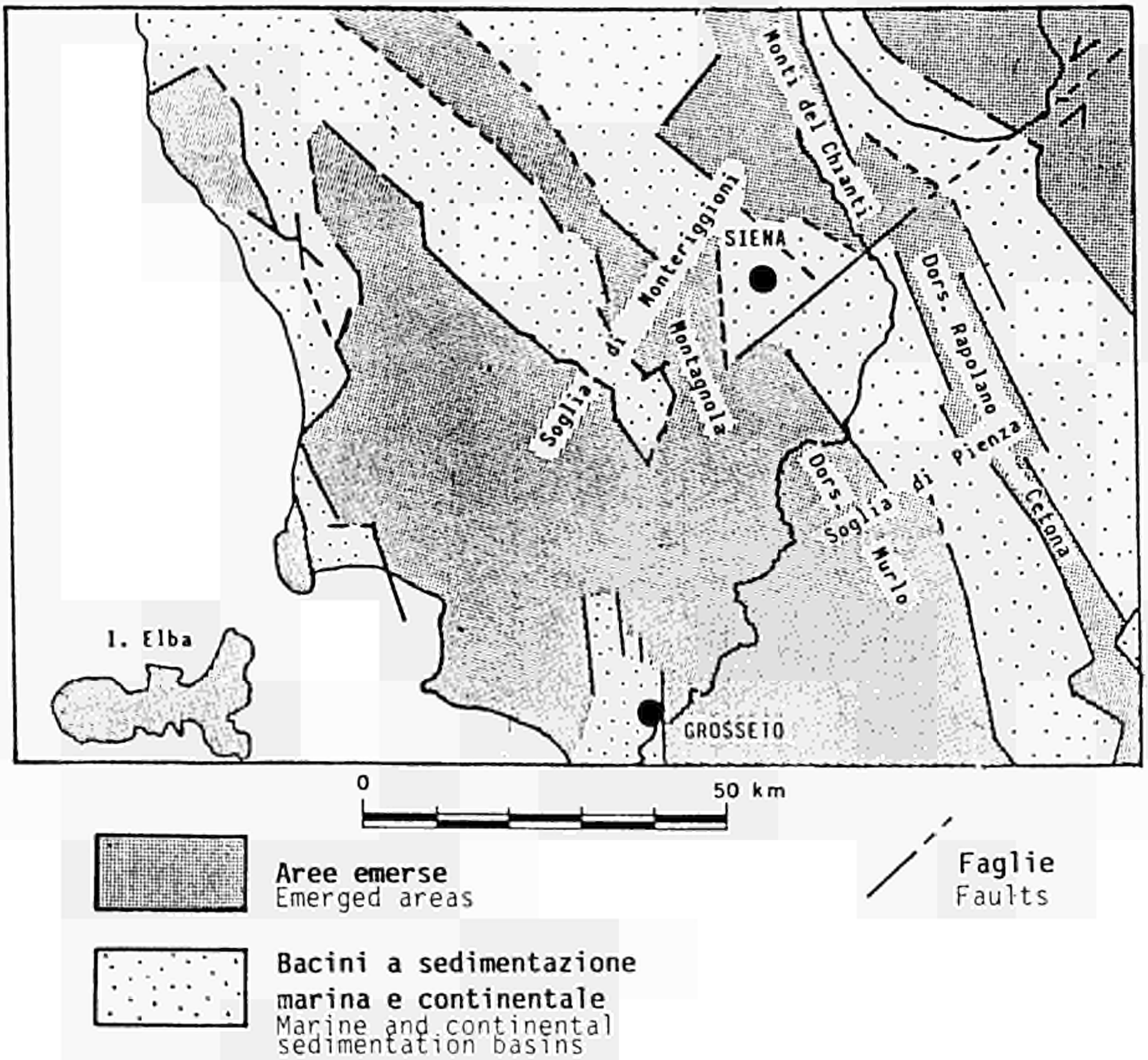


Fig. 1 - Distribution of the main neogenic sedimentary basins.

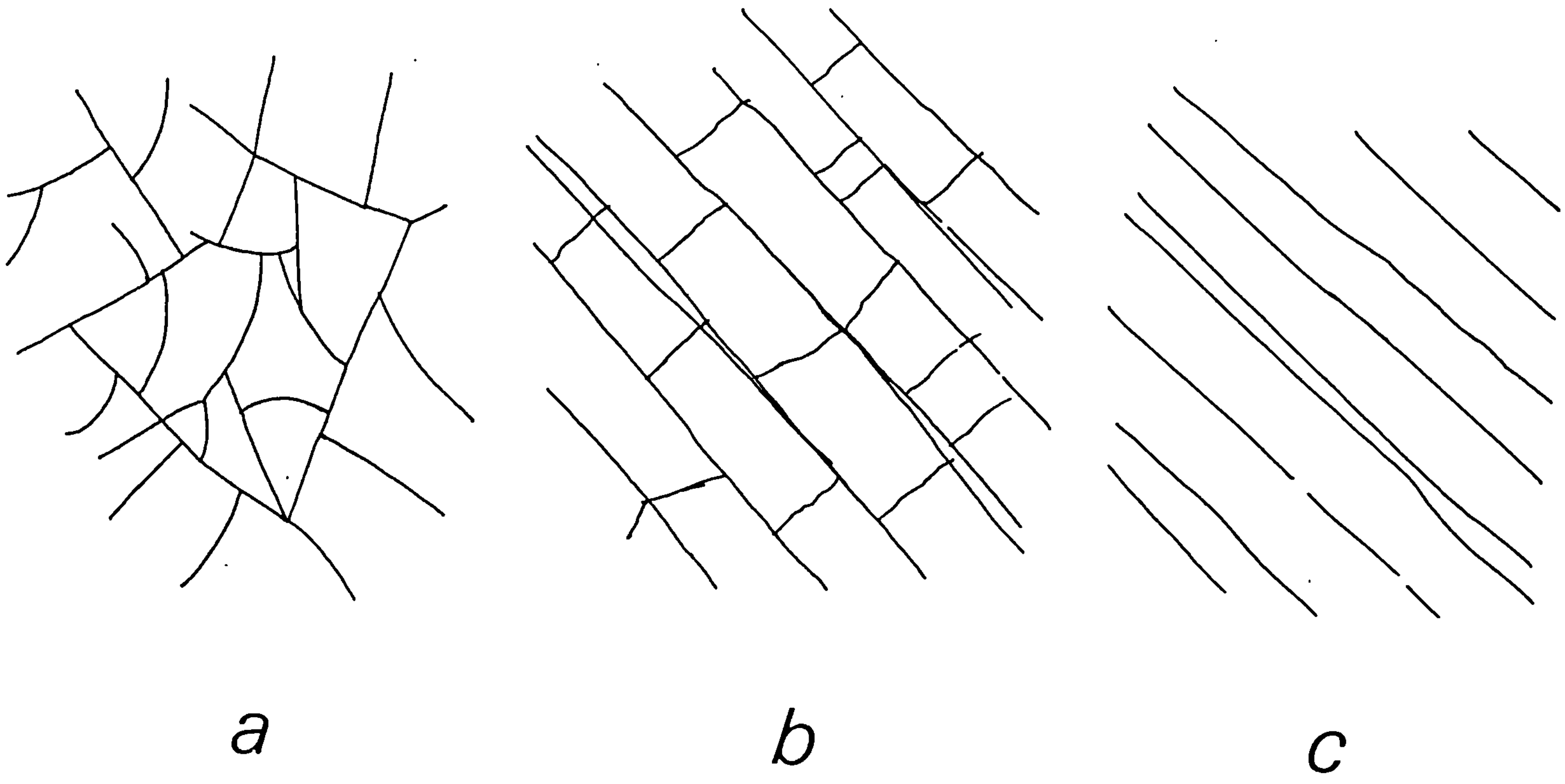


FIGURE 2

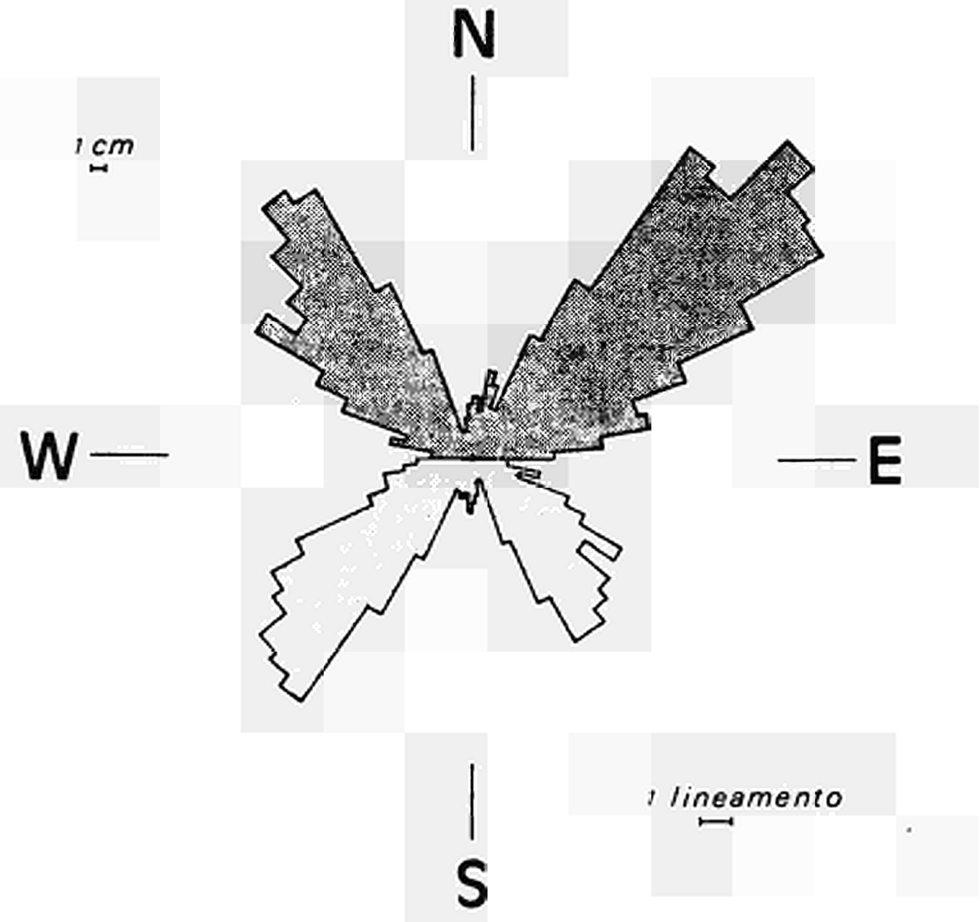


FIGURE 3

NATURAL ANALOGUE STUDIES OF RADIONUCLIDE MIGRATION

Contractor: British Geological Survey/NERC, Keyworth, Nottingham, UK
Contract No: FI1W/0073
Duration of contract: October 1986 to September 1989
Period covered: 1989
Project leader: PJ Hooker

A. OBJECTIVES AND SCOPE

It is important to be able to validate and support models of long-term predictions of radionuclide migration in the geosphere. The main aim of this research is to examine natural geochemical discontinuities and gradients as analogues of radionuclide transport in sediments. The mechanisms of processes of mobilisation, advection, diffusion and retardation for natural decay series elements and iodine and bromine will be addressed. This will entail some development of the techniques for measuring small concentrations and the speciations of these elements in both the solid and pore water phases. Analytical determinations by alpha spectrometry and neutron activation analysis will be carried out by SURRC, East Kilbride, and UKAEA Harwell under sub-contracts. Support in modelling will come from co-operation with the Ecole des Mines de Paris, Fontainebleau, and from a sub-contract with WS Atkins Engineering Sciences.

B. WORK PROGRAMME

B.1 Phase 1986-1987.

B.1.1 Site investigations.

B.1.1.1 Collection of fresh Loch Lomond sediments; analysis of I and Br depth profiles; preliminary modelling for effective diffusion coefficients.

B.1.1.2 Pilot investigation of I, Br, U and Th gradients across marl/clay boundaries in a well characterised sediment core from Lundin Castle, Fife, eastern Scotland.

B.1.2 A desk study of surface diffusion as a solute transport process for major cations through clays, with implications for trace radionuclide migration.

B.2 Phase 1988-1989.

B.2.1 Site investigations will be concentrated on measuring the speciation and mechanisms of distribution of I, Br, U and Th in sediments from Loch Lomond, Needle's Eye, near Dalbeattie in SW Scotland, and from Broubster in Caithness, N Scotland. (The investigation of Lundin Castle sediments was dropped in favour of the more fruitful work on the Needle's Eye and Broubster analogue sites.)

B.2.2 Modelling of the results and data from the field investigations for migration through sediments. (The application of the surface diffusion desk study has been dropped in favour of modelling the Needle's Eye and Broubster sites.)

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

SURRC investigations of the Loch Lomond sediments have led to the determination of halogen concentration-depth profiles in interstitial water samples. These have been understood in terms of conservative movement by diffusion. Work on the geochemistries and hydrogeologies of the Needle's Eye and Broubster sites has continued in order to understand the controls on the distributions of U and Th in the systems. Sediment samples from both sites were examined by XRF, alpha and gamma spectrometry, autoradiography, fission track registration and SEM methods to define U distributions. The results demonstrated the important role of organic matter in fixing U in the sediments. The groundwater analysis results and sediment characteristics were used by Ecole des Mines de Paris, Fontainebleau (EMP) for modelling the Needle's Eye site with the coupled transport code STELE, incorporating uraninite precipitation as the retardation process. The CHEMVAL database and the coupled transport code CHEMTARD were employed by WS Atkins Engineering Sciences to model the behaviour of U and Th at Broubster. Despite limited hydrochemical data, the uncertainties in the redox potentials, the limited understanding of the role of organic matter in fixing U and the complication of colloid transport, as demonstrated by UKAEA, much has been revealed by a thermodynamic equilibrium approach to modelling the groundwaters at Needle's Eye and Broubster [16]. It has indicated potential rock sources for the dissolved components, confirmed likely flow paths and provided ideas for coupled transport modelling. On the negative side, this approach cannot deal with disequilibrium and is inadequately prepared for describing the role of organics towards uranium movement in the sediments.

PROGRESS AND RESULTS

B.2.1 Field investigations

Loch Lomond

Loch Lomond sediments consist of a marine horizon, approximately one metre thick, underlain and covered by freshwater deposits that have accumulated a thickness of about 3m since the retreat of the sea around 5400 years ago. Previous studies of the sediments from the point of view of diffusion of Br, I, U and Ra from the organically associated enrichments in the marine layer into the overlying deposits led to further sampling and the development by SURRC of a new sensitive low-blank technique for analysing halogens in 0.1 ml of pore water [1]. The consequent concentration-depth profiles for chloride, bromide and iodide [2] are being interpreted and modelled in terms of diffusion and constant release rates from the marine horizon enrichments. Most of the halogen profiles can be explained in terms of diffusion without retardation.

Needle's Eye. A wide range of soil and water samples from the flood plain deposits of the Merse have been collected and analysed [3,4]. The U/Th series data from alpha spectrometry [5] have been interpreted to derive loss rate estimates of 5×10^{-5} to 10^{-6} per year for U from the cliff source area to the Merse sediments, depending on the time-scale assumed, and a halving-distance of 10 m for surface flow of U towards the tidal creek [6].

Integration of autoradiographic and fission track techniques has been used to locate and quantify U in samples of soils and silts proving that U is significantly retarded by organic matter [7]. Accumulation occurs at two distinct levels in the sediment profile, at around 50 cm depth and between 100-150 cm. Fission track registration indicates that at the 50 cm level uranium is located within the fine organic matter of the peaty matrix and is strongly enhanced around open root channels where concentrations reach several hundred ppm. This suggests fixation due to reduction. In the deeper accumulation, U is located almost exclusively within plant roots, and is particularly concentrated (often >1000 ppm) within specific cellular structures related to water transport. In addition, U-As-Cu and Bi bearing minerals found on root surfaces indicate that metal-fixing fungi may be important in arresting mobile uranium.

These data, combined with analyses from groundwaters, indicate two main inputs of dissolved uranium(VI) into this sediment system, namely surface flow of groundwater from the exposures of the mineralisation in the cliff, and upward flow from the bedrock below.

The fixation of uranium in the sediments is controlled by organic matter in the upper humic layers of the section, and by organic matter (roots) and associated iron oxyhydroxides in the deeper silts. This concentration of U in the silt is divorced from the sub ^{234}U daughters. In contrast, thorium is coherently associated with its daughters within detrital resistate minerals.

Broubster. The site is divided into two areas by a single-track road running north-south. To the west is the source area for uranium, a laminated limestone of the Caithness Flagstone Group belonging to the Orcadian lake sediments of the Middle Old Red Sandstone. Here, the area has been quarried on a small scale in the last century, and backfilled with quarry spoil. Approximately 100 m away downslope east of the road is a peat bog (<10,000 years old), underlain by boulder clay and the siltstone bedrock. This is the sink for uranium transported by groundwater flows along divergent paths from the source area. The peat contains up to 0.1 wt% of uranium . Field work [8] at the site had the objectives of: assessing the hydrogeology, collecting and describing samples of the 'source-term' uraniferous mineralisation, collecting groundwaters for chemical analysis, for U/Th speciation measurements with a portable chromatographic separation kit, and for U/Th series determinations at UKAEA, and collecting samples of the peat (sink-term) for metal analysis, C-14 dating and organic characterisation work [9].

A major source-term has been identified as a thinly laminated limestone approximately 40 cm thick at a depth of 90-130 cm. The lamellae consist of repeated units often less than 1mm thick, built up of a micrite (or algal carbonate mud) band and an organic silt band. The siltstone lamellae consist essentially of detrital quartz plus minor K-feldspar, plagioclase, muscovite, chlorite, biotite and mudstone pellets. The quartz and feldspars have developed euhedral overgrowths which cement the laminae. Authigenic pyrite is common as very fine octahedra or framboidal aggregates. Organic material is intermixed with this detrital component. Concentrated by biological and mineralogical processes the limestone is hence considerably enriched in uranium with up to 30 ppm U compared with the regional siltstone lithologies of 2-7 ppm U. The principal hosts for uranium in the limestone horizon are the carbonate bands, which comprise 80% of the whole rock with an evenly dispersed U concentration of 25 ppm, and the minor organic-silt layers with up to 60 ppm U. Hydrocarbon contained within the silt layers enclose crystalline phosphate/titanate minerals which have retained U. It is important to note too that a carbonaceous fossil fish fragment found within an organic silt layer had much U associated with it and had remained in place unaffected for 400 Ma [10]. Recent to present day weathering processes are effective in mobilising uranium. Oxidation of pyrite is of significance, causing acid leaching. Dissolution of calcite by percolating groundwaters also provide direct release of uranium. Little evidence exists from mineralogical work for Th mobilisation. However, rare earth element mobilisation is significant as evidenced by the REE patterns [8]. Uranium passes out of the quarried area complexed with carbonate, and is removed from solution by complexing principally with organic matter within the waterlogged peat deposits. Iron sesquioxides are also important, acting as scavengers of uranium.

B.2.2 Modelling the field investigations

Needle's Eye.

The controls governing uranium solubility were modelled by EMP on the basis of reduction-precipitation of U(VI) to U(IV) as uraninite. Uranium concentrations in the groundwaters and sediments contained in the section nearest the cliff were simulated using the coupled code STELE [11]. The mobile species are U(V1)-carbonate complexes which are stable at +400 mV, pH 7.8 and CO_2 pressure of $10^{-3.2}$ at. On contact with the organic-rich sediments with a lower Eh of +250 mV, U(V1) is reduced and uraninite can precipitate with a rate constant of 1 d^{-1}; this produces a steady state after a month. The calculated aqueous U concentrations agree well with the measured values. Also the computed accumulation of U in the sediments over 5000 years is 500 ppm, not much different from observed concentrations. The model has predicted the overall effects observed by measurement. It was not possible to reproduce the observed 2-D distributions of U with METIS and the Kd concept using laboratory-derived K_d values [12]; in this model the groundwater U concentration declines too slowly from the input concentration of $4x10^{-7}$ M, and gives a pessimistic prediction for U transport [11].

Broubster. Experimental measurements of groundwaters for U and Th using the portable chromatographic separation kit and ICPMS analysis indicate a soluble humate complex of U(VI) that is sorbed onto the peat. Modelling studies with PHREEQE and CHEMTARD [13,14], attempted to predict the speciation, retention and transport mechanisms for the actinide elements. Used in conjunction with the REE distribution data and complementary experimental measurements, a coherent interpretation of U, Th and REE behaviour at the site was possible. The applicability of equilibrium codes and thermodynamic data in the field situation at Broubster was demonstrated, but the inadequacies in modelling organic-metal interactions have again been highlighted. A number of conclusions can be drawn; thus, in the case of uranium:

- oxidation-reduction reactions do not necessarily control U fixation
- U levels in soils are not solubility constrained
- carbonates (and possibly phosphates) dominate the inorganic speciation of U
- organic complexation becomes dominant below pH 6
- U is present in 'true' solution and thus amenable to simulation by aqueous speciation models.

These results compared well with experiment and concur with the known distributions of uranium at the site. For thorium, the situation was less clear owing to probable co-precipitation of Th and Fe hydrous oxides and the likely presence of Th in colloidal form. U/Th series work [15], including ultra-filtration, has confirmed that colloids are moving Th through the system; also, whereas the bulk of the ^{238}U and ^{234}U activities in the peat are associated with the organic matter component , there is evidence for a repartition of the daughter ^{230}Th away from the organic phase into amorphous iron oxyhydroxides.

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MICROSTRUCTURE AND STATE OF RADIOACTIVE EQUILIBRIUM OF CRYSTALLINE ROCK IN RELATION TO THE RETARDATION OF RADIONUCLIDES BY DIFFUSION INTO THE ROCK MATRIX

Contractor: Univ. of Oviedo (E) and Univ. of Exeter (UK)
Contract N°: FI1W/0143-
Duration of contract: 01.10.87 - 30.09.90
Period covered: 01.01.89 - 31.12.89
Project leader: M. Montoto - E. Durrance

A. OBJECTIVES AND SCOPE

In most radionuclide transport models it is assumed that radionuclides migrating in solution along fractures will diffuse into water held in pores within the rock matrix adjacent to fractures, and will thus be retarded relative to the flow of water. The possibility that there exists adjacent to fractures a zone to which diffusion might be restricted, or that the pores adjacent to fractures might be clogged by the products of alteration and/or mineralisation, has not generally been included in radionuclide retardation models.

The objective of this study is to assess the effectiveness of diffusion as a mechanism for the retardation of radionuclides, by examining the microstructure of crystalline rock adjacent to and remote from fractures, and by determining the extent of disequilibrium in the uranium-238 decay series in rock adjacent to fractures. The effect on porosity and potential diffusion of the fractures themselves is also being assessed, with particular regard to stress-relief effects and to pore-clogging by the products of alteration, mineralisation and precipitation. It is hoped that the results of the study will help to resolve a major area of uncertainty in existing radionuclide retardation models.

B. WORK PROGRAMME

(1) Measurement of the porosity and permeability of intact rock adjacent to and remote from water-conducting fractures; determination of the permeability of fracture walls.
(2) Examination by optical, electron and acoustic microscopy of the microstructure of rock adjacent to fractures and the availability of pores for diffusion, using digital image processing techniques.
(3) Examination of the mineralogy of the rock and of fracture and microfracture linings and infillings.
(4) Investigation of the penetration of modern waters into the rock adjacent to fractures by measuring the state of radioactive disequilibrium in the uranium-238 decay series using alpha spectometry.
(5) Determination of the state of oxidation of iron in rock adjacent to fractures using Mossbauer spectroscopy.
(6) Assessment of the effect of in situ stress conditions on the potential for diffusion.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Work during the report period can be described under the following headings:
(1) Microstructural analysis: mineralogical and textural studies and quantification of microcrack density; other methods of sample preparation for microscopic studies has also been used.
(2) Determination of the physical properties of samples, both following international standards (ISRM, ASTM, etc) and using mercury porosimetry techniques.
(3) Determination of the ultrasonic parameters for a more precise petrophysical characterization of the rocks.
(4) Uranium series disequilibrium studies and geochemical analysis.

Items 2 and 3 have been included as another way of characterize the microstructure of the rock.

Samples studied during this period included El Berrocal granite, EB, (Spain), Stripa granite, ST, (Sweden), and preliminary results from Whiteshell granite (Canada).

Progress and results

1. Microstructural analysis.

The microstructural analysis was focussed on the quantification of some petrographic components:
* Measurement by digital image processing techniques of the form factor,FF, of grains in each of the sections of Stripa sample ST-5. There is no variation in the values of FF measured in the seriated thin sections, the mean value for the whole sample being 1.68.
* Quantification of crack density under reflected-light fluorescence microscopy in both Stripa and El Berrocal granites. Regarding ST-5, the linear crack density (LCD) from the section, located at 95 mm from the fracture and parallel to it) is 0.28 mm^{-1}, 12.5 % of the traces observed being of intragranular type, 61.7 % of transgranular type, and 25.8 % being intergranular. In the other sections almost no accessible cracks could be seen.

In core EB1B, Fig. 1, the thin section closest to the fracture shows the highest microcrack density. Perhaps in this slice weathering products are more abundant than in other slices, and they fill the microcraks avoiding the fluorescent resin to penetrate into the rock.

2. Physical properties

The measured properties included: dry density (ρ_d), grain density (ρ_s), porosity (both total and open one, n and n_o), degree of saturation (S_r), water content after 1 hour (Void index, I_v) after ISRM 1981; water content after 2 days (W_2), (ASTM, 1988); water absorption, water desorption, water content in saturation (W_s); water content in a long term (W_i), (CNR-ICR, 1981). They have been measured in each of the parallel slices cutted from each core, in order to assess whether or not there is any variation with distance from the fracture. Table I shows the mean values of each property for both El Berrocal and Stripa granites.

The relationship between the values of the physical properties and the distance to the rock fracture is not always consistent. So, regarding dry density (ρ_d), total porosity (n), degree of saturation (Sr) and water content after desorption, no relationship can be established between this property and the distance to the fracture, for both Stripa and El Berrocal granites.

On the other hand, void index, open porosity, water content after 5 days and water content in saturation, show a decrease in the value of the property as the distance to the fracture increases, although ST-7 vary in an irregular way regarding the void index and the open porosity. As an example the variation of the open porosity in the different slices from each core (scaled to the open porosity of the slice closest to the fracture in each core) is plotted in Fig. 2; the line joins the points corresponding to the mean values of the open porosity in the slices located at the same distance from the fracture.

Mercury porosimetry

Mercury porosimetry studies of El Berrocal granite samples have been carried out in the Laboratoire de Microanalyses Nucléaires, in the Faculté des Sciences et Techniques de Besançon (France). Final porosities (in the range 1.2 - 1.4 %) are similar to those measured by water saturation. Results from the slices cutted from core EB1F appear in Fig. 3; for each pressure (which is an indirect measure of pore size), the larger the distance to the fracture, the highest the porosity.

3. Ultrasonic parameters.

Ultrasonic parameters such as wave energy, amplitude, count number, velocity of longitudinal waves (V_p), etc, have been evaluated for a proper petrophysical characterization; more information on mineralogical composition, density, texture, porosity, microfractography,... is so intented.

An anisotropy in the Stripa granite, not previously refered in the literature, was so detected; Vp values along two orthogonal directions range from 3416 to 4038 $m.s^{-1}$ and from 4679 to 5000 $m.s^{-1}$.

In the El Berrocal granite the V_p values in dry ones specimens were 4967 to 5597 $m.s^{-1}$, and 6024-6452 $m.s^{-1}$ in the saturated ones. A less significant anisotropy has also been obtained along three orthogonal directions: 4642, 5441 and 5416 $m.s^{-1}$.

The variation of the Vp values (saturated specimens) in the different slices from each core (expressed as the percentage with respect to the Vp in the slice closest to the fracture in each core) is plotted in Fig. 4; the line joins the points corresponding to the mean values of the Vp in the slices located at the same distance from the fracture.

4. Uranium series disequilibrium studies and geochemical analysis.

Alpha spectrometry is being employed following separation of uranium and thorium by ion exchange techniques. Early experimental difficulties have been overcome and good separations are now being obtained.

The uranium series of alpha spectra from Stripa sample ST6 are plotted with selected geochemical data as a profile across the rock adjacent to the fracture in Figure 5. Although error values have not been yet determined, it appears that a U-234 excess has been identified close to the fracture surface and that the U-234/U-238 ratio approaches equilibrium values within about 20 mm of the fracture surface.

The geochemical analysis show no significant variation in total uranium or thorium values as the fracture is approached, with no identifiable uranium deposition on the fracture surface. The Fe_2O_3/FeO ratio increases at the fracture surface, but Fe_2O_3 values fall rapidly with increasing distance and continue to fall throughout the 100 mm profile. Partial uranium series data from El Berrocal granite appear in Table II.

Geochemical analysis of El Berrocal samples has revealed a strong uranium enrichment at the fracture surface (119 ppm uranium in slice EB1F.OX, 46 ppm U in slice EB1F.O), compared with a constant 7-8 ppm of thorium throughout the profile. The strong excess of both U-234 and U-238 over Th-230 shows that this uranium deposition is a recent phenomenon. The U-234/U-238 ratio for slice 1F.0 shows a U-234 excess but that for sample EB1F.0X is near equilibrium.

The uranium enrichment observed on the fracture surface in core EB1F does not penetrate into the rock beyond the first 5 mm slice and that enrichment in other metals (e.g. Mn and W) is also confined to this narrow zone. The pattern of variation in the Fe_2O_3/FeO ratio, with increased values around 1.0 very close to the fracture surface falling away to values of around 0.5 about 10 cm from the fracture, is very similar to that observed in Stripa sample ST6.

Disequilibrium/geochemical profiles are now being obtained for a number of cores from El Berrocal and from Whiteshell. The geochemical data for Whiteshell samples show no systematic variation in total uranium or thorium or in the degree of iron oxidation.

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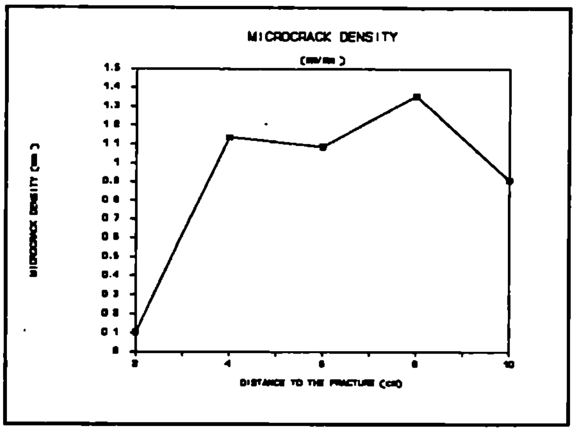

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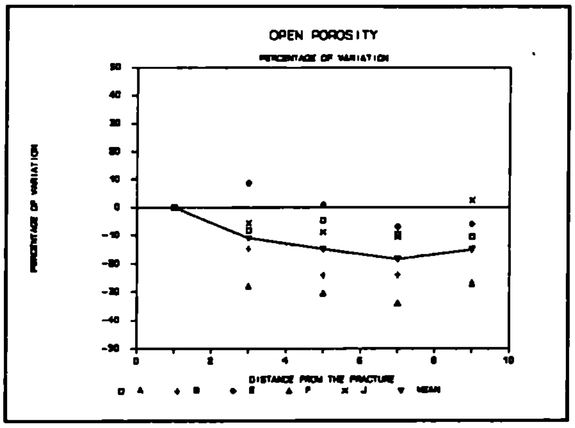

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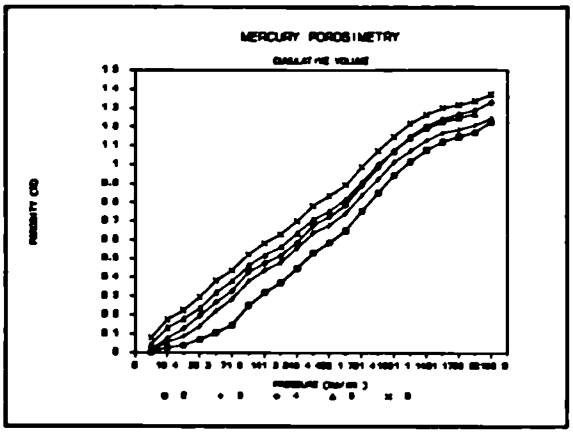
Fig. 1.- Density of microcracks in the series of thin sections of El Berrocal sample EB1B, plotted as a function of the distance to the fracture.
Fig. 2.- Variation of the open porosity with respect to the distance to the rock fracture (El Berrocal granite, samples EB1A, EB1B, EB1E, EB1F, EB1J, and their mean value M).



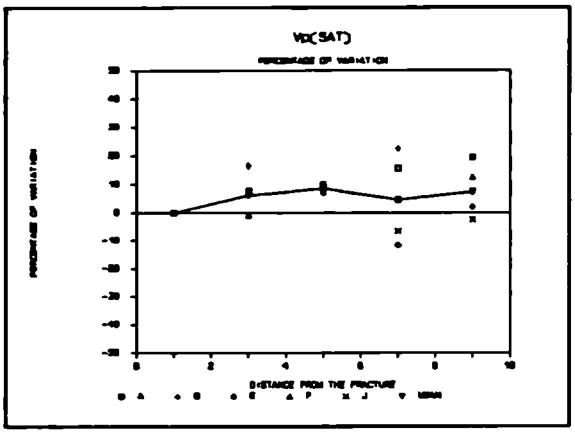


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Fig. 3.- Cumulative porosity plots of the serie of slices from El Berrocal sample EBF.
Fig. 4.- Variation of Vp (saturated conditions) with respect to the distance from the fracture (El Berrocal granite, samples EB1A, EB1B, EB1E, EB1F, EB1J and their mean value M).

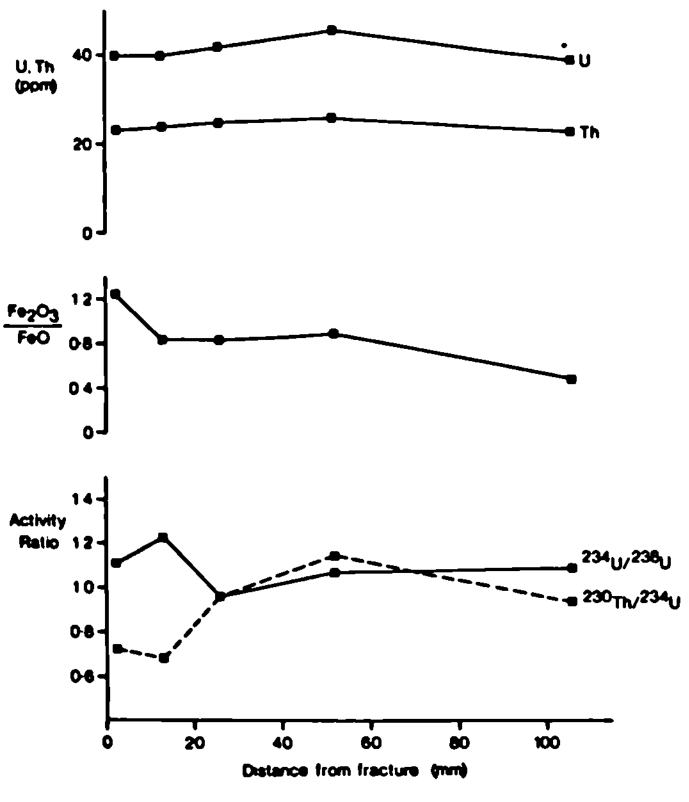

Fig. 5.- Variation in selected geochemical/radiochemical parameters with increasing distance from fracture surface, Stripa sample ST6.

TABLE I

	ST-6	ST-7	EB
Dry density, ρd (kg/m^3)	2,624 ± 2	2,624 ± 3	2,627 ± 4
Grain density, ρs (kg/m^3)	2,638 ± 3.20	2,638 ± 2.26	--
Accesible porosity, n_o (%)	0.39 ± 0.03	0.37 ± 0.03	1.20 ± 0.04
Total porosity, n (%)	0.52 ± 0.10	0.52 ± 0.11	1.33 ± 0.06
Degree of saturation, Sr (%)	76.74 ± 12.68	74.36 ± 15.34	91.18 ± 2.71
Saturation water content, Ws (%)	0.15 ± 0.02	0.14 ± 0.01	0.46 ± 0.02
Void index, Iv (%)	0.11 ± 0.02	0.09 ± 0.01	0.35 ± 0.01
Two days water content, W_2 (%)	0.13 ± 0.01	0.12 ± 0.01	0.37 ± 0.02
Five days water content, W_5 (%)	0.13 ± 0.01	0.12 ± 0.01	0.39 ± 0.02
Water content after desorption, Wd(%)	0.01 --	0.002 --	0.11 ± 0.02

Table I.- Physical properties of the intact rocks. Stripa samples ST-6 and ST-7, EB: mean values for the whole batch of El Berrocal samples. (95 % confidence interval)

TABLE II

Sample number	Distance from fracture (mm)	U-234/U-238	Th-230/U-234	Th-230/U-238
EB1F.OX	-5 - 0	1.03	0.34	0.35
EB1F.0	0 - 5	1.14	0.60	0.68
EB1F.2	22 - 37	1.20	0.65	0.78
EB1F.8	113 - 133	1.10	0.83	0.75

Table II.- Uranium series data for some slices of El Berrocal sample EB1F.

MIGRATION OF URANIUM DAUGHTER RADIONUCLIDES IN NATURAL SEDIMENTS

Contractor:	Natural Environment Research Council of the United Kingdom
Contract No.:	FI1W/0146.
Duration of Contract:	1 December 1987 to 31 May 1990
Period Covered:	1 January 1989 to 31 December 1989
Project Leader:	Dr. J. Thomson

A. OBJECTIVES AND SCOPE

This project utilises a characteristic uranium profile shape developed in deep-sea turbidite sediments to examine the behaviour of uranium and its daughter isotopes over time-scales desired for the isolation of radioactive waste. The geochemical background is that homogeneous organic-rich turbidites, emplaced in the deep-sea, experience oxidation from the top downwards by bottom water. Uranium redistributes in the turbidite in response to the oxidation front, but this redistribution ceases when the turbidite is isolated from bottom water on emplacement of the next turbidite. Two long cores (MD24 and MD10) are available from a 1985 NEA cruise with several such units up to 750 ky old preserved. Good stratigraphic, geochemical and geotechnical data are available, and pore water advection in the cores is believed to be negligible.

The goals of the project are:

Determination of uranium and its longer-lived daughter radionuclides in individual turbidite units with ages between 250 and 750 ky to compare the observed profiles with those predicted by radioactive ingrowth systematics.

Utilisation of the experimental data to estimate effective diffusion coefficients of the different elements for modelling purposes. Such data will be relevant to the *in-situ* geochemical conditions of the sediments over the long time scales indicated.

B. WORK PROGRAMME

2.1 Development and verification of a radiochemical analytical scheme for the analysis of uranium-238, uranium-234, thorium-232, thorium-230, protactinium-231, radium-226 and polonium-210.

2.2 Determination of activity versus depth profiles of the above radionuclides for different turbidites of different ages (250-750 ky) in core MD24.

2.3 Determination of a gross uranium profile for core MD10 to guide sampling and comparison of corresponding turbidite unit profiles with those of core MD24.

2.4 Chemical partitioning studies of selected samples (if necessary).

2.5 Model data obtained.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

Analysis of radionuclides (including ^{231}Pa) in samples from turbidite *s* in core MD24 is complete.

Three samples from turbidite *s* have been leached to investigate the feasibility of removing authigenic U from an older turbidite.

A mathematical model has been developed and has been applied successfully to the ^{226}Ra data from core MD24 to produce values for the effective diffusion coefficient of ^{226}Ra. Experimental work has confirmed model findings that only a proportion of ^{226}Ra is available for pore water diffusion.

PROGRESS AND RESULTS

2.2 Uranium, thorium, polonium-210, radium-226 and protactinium-231 analysis of core MD24

Radiochemical analysis of samples from turbidite *s* has been completed. Excellent chemical closure for the sequence $^{238}U \rightarrow {}^{234}U \rightarrow {}^{230}Th$ is demonstrated (mean $^{234}U/^{238}U = 0.997 \pm 0.004$, mean $^{230}Th/^{234}U = 0.976 \pm 0.006$). ^{226}Ra is deficient in turbidite *s* around the ^{230}Th maximum indicating that migration of ^{226}Ra has taken place. Investigation of five samples from the U maximum in *s* showed that ^{231}Pa was not present in high enough concentrations to give analyses of acceptable precision.

2.4 Chemical partitioning studies of selected samples

Three samples from turbidite *s* have been subjected to a mild acetic acid leach to determine whether it is possible to remove the authigenic U fraction from an older turbidite (Table 1). Broad agreement was found between predicted and measured authigenic U contents except for the sample from the U peak. The leach removes ^{234}U preferentially from the detrital fraction. The total analyses show that ^{234}U does not, in fact, migrate despite its potential mobility.

2.5 Model data obtained

The model developed approximates the parent nuclide profile as a series of strips. Each strip diffuses for the mean life of the daughter nuclide using the equation

$$C(z,t) = \frac{C_o}{2}\left\{ \mathrm{erf}\ [(h-z)/2\sqrt{(Dt)}] + \mathrm{erf}\ [(h+z)/2\sqrt{(Dt)}] \right\}$$

and substituting a suitable value for the effective diffusion coefficient D. The modelled daughter profile is built up by summing contributions made by each strip at certain depth intervals. The fit between the model and experimental data profiles is optimised by varying D and the proportion of the daughter available for migration. Figure 1 shows model and experimental profiles for ^{226}Ra through turbidites *s* and *t*, using $D = 6 \times 10^{-9} cm^2 s^{-1}$ and allowing 35% of total ^{226}Ra to migrate. Similar results were found for turbidite *w* (Fig. 2).

LIST OF PUBLICATIONS

COLLEY, S., THOMSON, J. & TOOLE, J., 1989. Uranium relocations and derivation of quasi-isochrons for a turbidite/pelagic sequence in the Northeast Atlantic. Geochim. Cosmochim. Acta 53, 1223-1234.

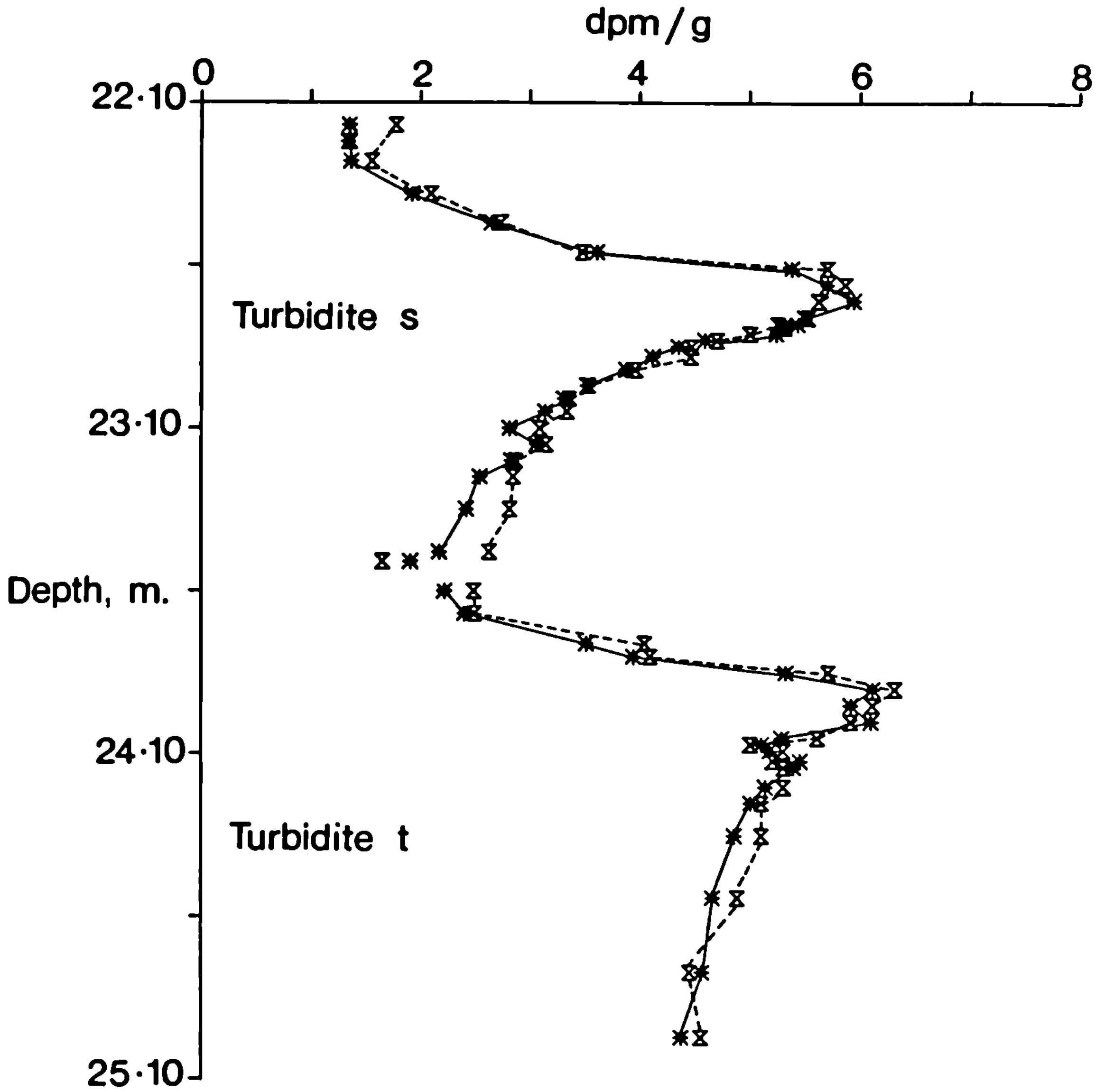

Figure 1 ^{226}Ra data (bow-ties) and model (stars) profiles for turbidites s and t, with 35% of ^{226}Ra free to move. The best fit gives an optimum value for D of $6 \times 10^{-9} cm^2 s^{-1}$.

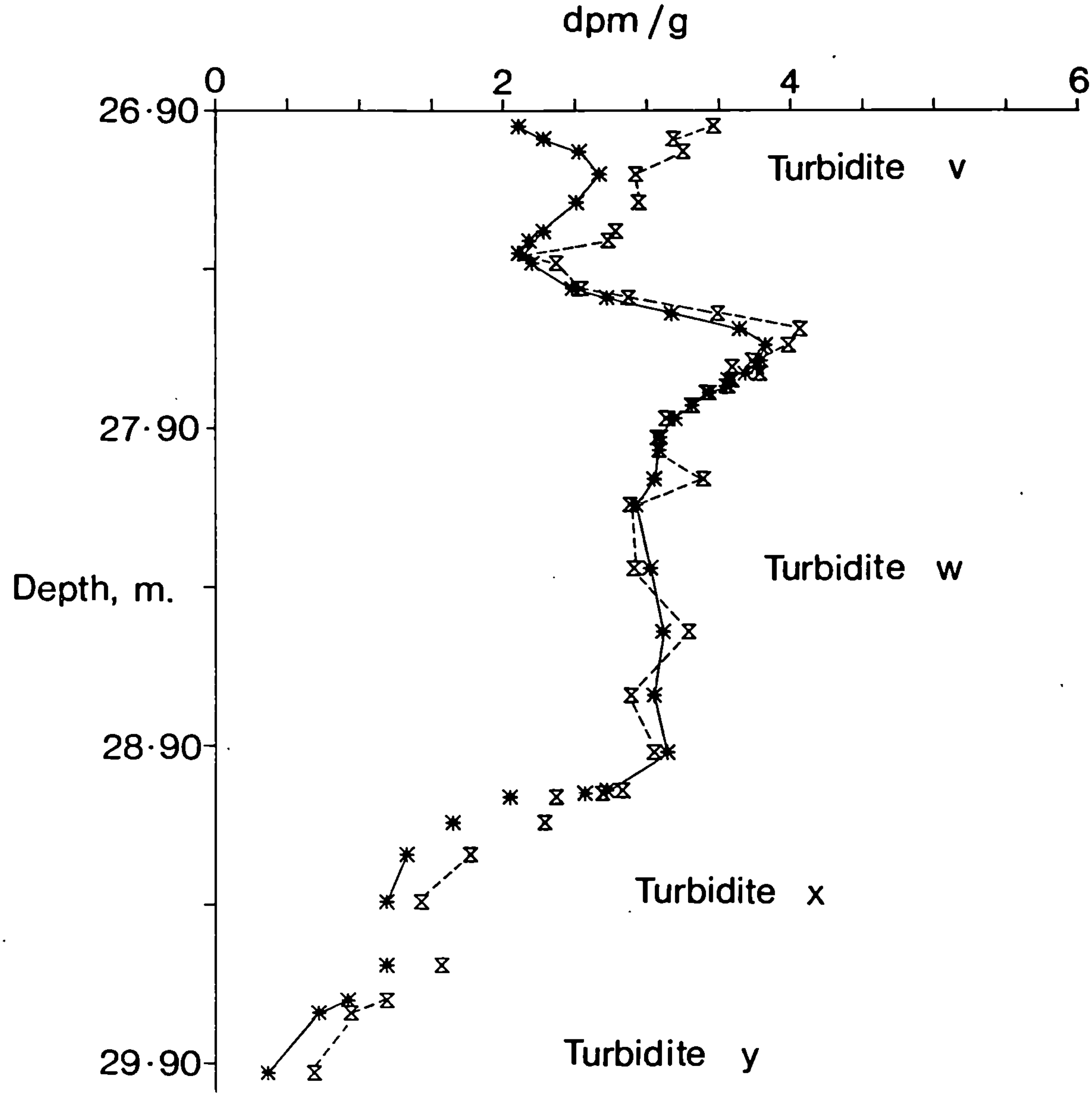

Figure 2 ^{226}Ra data (bow-ties) and model (stars) profiles for turbidite w and surrounding units, with 35% of ^{226}Ra free to move. The best fit gives an optimum value for D of $3 \times 10^{-9} cm^2 s^{-1}$.

TABLE 1 - Comparison of predicted and observed (HOAc/NaOAc leach at pH 5) U fractions in samples from turbidite g in core MD24.

			TOTAL			DETRITAL		AUTHIGENIC	
Sample No.	Depth cm	$CaCO_3$ (%)	U ppm	$^{234}U/^{238}U$ activity ratio		U ppm	$^{234}U/^{238}U$ activity ratio	U ppm	$^{234}U/^{238}U$ activity ratio
284	2228	45.1	1.62 ± 0.04	0.98 ± 0.03	Predicted*	1.46	1.0	0.16	1.02
			(1.51 ± 0.04	0.96 ± 0.03)	Observed	1.22 ± 0.04	0.76 ± 0.03	0.29 ± 0.01	1.83 ± 0.09
290	2271	51.3	8.80 ±0.2	1.02 ± 0.02	Predicted*	1.29	1.0	7.5	1.02
			(8.80 ± 0.1	1.02 ± 0.02)	Observed	2.58 ± 0.08	0.84 ± 0.03	6.2 ± 0.1	1.08 ± 0.02
312	2320	52.0	3.46 ± 0.07	0.98 ± 0.02	Predicted*	1.27	1.0	2.19	1.02
			(3.35 ± 0.06	0.98 ± 0.02)	Observed	1.46 ± 0.05	0.82 ± 0.03	1.89 ± 0.04	1.18 ± 0.03

* Predictions based on (i) detrital U = 2.7 x (1 - %$CaCO_3$/100, (ii) ^{234}U at secular equilibrium with ^{238}U in detrital component, (iii) authigenic U with maximum possible $^{234}U/^{238}U$ activity ratios of 1.02 (seawater value of 1.14 decayed for 650 kyr).

The total U contents and $^{234}U/^{238}U$ activity ratios quoted are from (i) a single analysis of whole sediments and (ii) in parenthesis the sum of the observed detrital and authigenic fractions.

STUDY OF THE MIGRATION OF U, TH AND REE IN AN INTRAGRANITIC URANIUM DEPOSIT

Contractor: CEA-IRDI/DRDD/SESD/SCPCS/LECALT- Fontenay-aux-Roses-France
Contract n°: FI1W/0149
Duration of contract: from December 1987 to December 1989
Period covered: January 1989 to December 1989
Project Leader: M.T.MENAGER

A. OBJECTIVES AND SCOPES

The intragranitic uranium deposit of the Jalerys (Granite de Grury-Morvan-France) is used as natural analogue of a high-level radioactive waste repository for studying the physico-chemical processes of transport for some trace elements of interest. The project is focussed on near-field migrations.

More specifically, the aims of this investigation are:

* To obtain information on the elemental mobilisations of U, Th and REE through the argillaceous and granitic barriers which surround the ore body considered as a highly concentrated source-term.

* To determine the geochemical mechanisms involves during the transport of the radionuclides and to evaluate, as far as possible, their migration rates.

* To confirm the analogy in the geochemical behaviour of transuranic elements and their natural counterparts.

* To characterize, the retention capacity of a repository, that would be realized in a similar geological formation.

* Finally, to model the interaction between the granite, the fluid and the ore body by means of the geochemical code EQ3/6.

This project is conducted in collaboration with COGEMA and CREGU.

B. WORK PROGRAMME

B.1. Sampling of the Grury granite intruded by uraniferous mineralizations (the Jalerys mine and the Jacquot open quarry)
B.1.1. Samples are taken at regular intervals along a direction roughly perpendicular to the vein, from the mineralized body to the fresh granite.

B.2. Mineralogical studies at the laboratory
B.2.1. Petrographical and mineralogical works on primary and secondary parageneses, specially on U-Th-REE-rich ones, with optical and electron microscopy (scanning and transmission coupled with X-rays analysis) as well as fission track micromapping.
B.2.2. Study of the geochemical properties of the granite and of their evolution during alteration (major and trace elements analysis by ICP and neutron activation analysis).
B.2.3. Determination of the physico-chemical properties of fluids responsible for the alteration by the study of fluid inclusions (microthermometry, Raman microprobe).
B.2.4. Evaluation of temperatures active during the functioning of the system by the study of the thermal annealing of fission tracks registered in appropriate minerals.

B.3. Isotopic studies at the laboratory

B.3.1. Study of radionuclide mobilizations using uranium and thorium-series disequilibria for dating the last opening of the system (alpha and gamma spectrometries, ion microprobe).

B.3.2. Dating of alteration minerals, whenever feasible.

B.3.3. Evaluation of the sorption efficiencies for U, Th and REE of granite-forming minerals for various degrees of alteration corresponding to different surface states (Rutherford backscattering, specific nuclear reaction analysis).

B.4. Modelling

B.4.1. Modelling of interactions between solids and solutions by means of the EQ3/6 geochemical code and comparison with data on the natural system.

B.4.2. Simulation with this code of the evolution with time of a high level radioactive waste intragranitic repository.

C. PROGRESS OF WORK AND RESULTS OBTAINED

STATE OF ADVANCEMENT

During this year, the elemental behaviour of major and minor components of the granite have been studied along the profile. Concurrently, the determination of physico-chemical properties of fluid inclusions associated with hydrothermal alteration steps have been conducted and compared with microfissuration of the rock. The measurement of radioactive disequilibria in U and Th-series was obtained by means of alpha-spectrometry on powder samples. The sorption efficiencies for trace elements of granite-forming minerals was evaluated with Rutherford backscattering spectrometry. Finally, the modelling of water/rock interactions is in progress.

PROGRESS AND RESULT

Geochemical behaviour during hydrothermal alteration

The alteration phenomena associated with the water/rock interactions lead to the complete dissolution of the granite near the U-mineralized vein. The initial rock is substituted by a smectitic selvage.

The elemental mobilization is very important near this U-vein and in the first 0.5 metre all major elements of the rock (except Si) are depleted. For trace elements, an enrichment of U, Cs, Sb and As is observed in the granite on both sides of the vein. It is probably correlated with the composition of hydrothermal solutions. All the other analyzed traces are depleted over one metre or more. The "fresh" granite is located at thirty metres of the vein.

Composition of hydrothermal solutions

The determination of physico-chemical properties of fluid inclusions trapped in the quartz of the Jalerys granite lead to distinguish three types of fluids in this area. Two types are composed of aqueo-carbonic solutions and are associated with the early history of granite. The third type is composed of very abondant aqueous inclusions. These inclusions are associated with hydrothermal events and form a network in the host rock. Near the U-vein, the trapping-temperature range of hydrothermal fluid (230-240°C) is in good agreement with the mineral parageneses of the second step of alteration. The trapping-temperature range of fluids decreases below 200°C over a few metres. On the other hand, a good agreement is observed between the structural directions of fissures and the fluid inclusion network.

Isotopic studies

The alpha-spectrometry data on U-series disequilibrium exhibit a large radioactive disequibrium in the first metre on both sides of the vein, namely a decrease of the $^{230}Th/^{234}Th$ ratio and an increase of $^{226}Ra/^{230}Th$ and $^{210}Pb/^{226}Ra$. Over one metre, the disequilibrium is lower and the secular equilibrium is achieved in the fresh granite. An excellent correlation is observed between the perturbed zone and the iron oxidation state. Except in the contact zone, no major perturbation has hence occurred in the granite for about 2.5 My.

Evaluation of the retention capability of the granite

Elemental migration is large in the contact zone with the U- vein, but the main species are trapped in the near field of the mineralized-brecchia. If the retention capability of the granite seems good, the smectitic selvage is a bad trap. For example, its uranium concentration is noticeably lower than that of its parent rock which reflects a preferential leaching during its formation. This argument is even stronger when one considers that this selvage has been permeated by U-rich solutions originating from the ore body.

Rutherford backscattering spectrometry studies have also been conducted with individual granite minerals. We have shown that the retention efficiency for U, Th and REE of mineral surfaces is particularly high when these elements are associated with colloïds (of charge inverse to that of the surface) and whenever a porous hydrated layer has developed on the solid as a result of hydrothermal alteration. Coprecipitation on mineral surfaces (e.g. calcite) also appears as a major retention mechanism.

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PARNEIX, J.C., MENAGER, M.T., TROTIGNON, L., PETIT, J.C., Hyrothermal alteration in the Auriat granite (Massif Central, France): analogy with a radwaste disposal. CEC Report n° EUR 11037 EN (1987) 13pp.

MENAGER, M.T., PETIT, J.C., MENET, C., Elemental remobilizations around the U-minearlized vein of the Jalerys (Morvan). Proceedings of the Internationnal Congress of Geochemistry and Cosmochemistry, Paris, August 29-September 2 1988, Chemical Geology pp136.

MENAGER, M.T., MENET, C., PETIT, J.C., COME, B., U, Th, REE mobilization during water-rock interactions in a U-mineralized granite. Water-Rock Interactions WR6, Proceedings, Miles (Ed), 475-478.

4.3.D. Development of calculation tools for the description of radionuclide migration

MODELLING OF MIGRATION PHENOMENA IN THE BOOM CLAY AND OF HEAT DISSIPATION FROM A HLW REPOSITORY IN THE MULTI-LAYERED HYDROGEOLOGICAL SYSTEMS SURROUNDING THE BOOM CLAY

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0055/B
Duration of contract : from October 1986 through June 1990
Period covered : January 1989 - December 1989
Project Leader : A.A. Bonne
Report by : M. Put

A. OBJECTIVE AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high-level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Several specific investigations still need to be further undertaken in order to characterise more accurately the argillaceous formation in view of assessing its appropriateness for hosting radioactive waste as well from engineering point of view as for long term safety and performance evaluations. In support of these also further modelling efforts are required in order to improve and confirm our prediction capability.

B. WORK PROGRAMME

B.4.1. Implementation and adaptation of the analytical code MICOF
B.4.2. Evaluation of the thermal impact due to HLW disposal into a stratified clay formation in a multi-layered aquifer system (Mol site) (completed and reported)

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The concept of the "diffusion accessible porosity" has been further investigated and applied.

The MICOF-code has been updated to include these concept. The code allow for 3-dimensional calculations in a homogeneous anisotropic porous media, and has been applied for the predictive calculations for a large scale (order of meters) 3-dimensional in situ migration experiment in the Boom clay.

Progress and results

Experimental evidence of the validity of the concept of "diffusion accessible porosity" have been obtained /1/, /2/. Figure 1 shows the diffusion conductivity constant $D_k = \eta RD$ as a function of the diffusion accessible porosity η for non-sorbed species (R=1). The Archie equation holds for different orders of magnitude, showing the influence of the diffusion accessible porosity on the diffusion constant. The value of the diffusion accessible porosity may be influenced by the consolidation pressure, the molecular mass and the electrical charge of the diffusing species, the ionic strength of the solution, etc.

The new conceptual model has been included in the MICOF-code. The code have been used for predictive calculations (by the start of the experiment) for an in situ 3-dimensional migration experiment on a scale of a few meters /3/. A quantity of 9.25 E+8 Bq HTO have been injected in the formation (filter CP1(5)) and the concentration is monitored at distances of 1, 2, 3 and 4 meter. Figure 2 gives the predictive calculations at a distance of 0.9 m together with the measured concentrations up to now.

For the interpretation of the results of the migration experiment with technetium consisting of a plug labelled with technetium in contact with an unlabelled, a model which takes into account different parameters for both plugs have been established /3/. The model fits nicely the experimental results (see Figure 3).

For the determination of the mobility of the humic acids (HA), in the interstitial clay water, percolation experiments have been set up. These percolation experiments have been modelled and preliminary results have been obtained /3/. Figure 4 shows the results.

List of publications

/1/ PUT, M.J., An improved mathematical model for the interpretation of the flow-through type diffusion test with influence of filterplates. "Migration 89", Monterey, USA

/2/ HENRION, P.N., PUT, M.J., VAN GOMPEL, M., The influence of compaction on the diffusion of non-sorbed species in Boom clay. "Migration 89", Monterey, USA

/3/ MONSECOUR, M., YOSHIDA, H., HENRION, P., PUT, M., DE REGGE, P., Migration studies in the Boom clay at Mol. "Migration 89", Monterey, USA

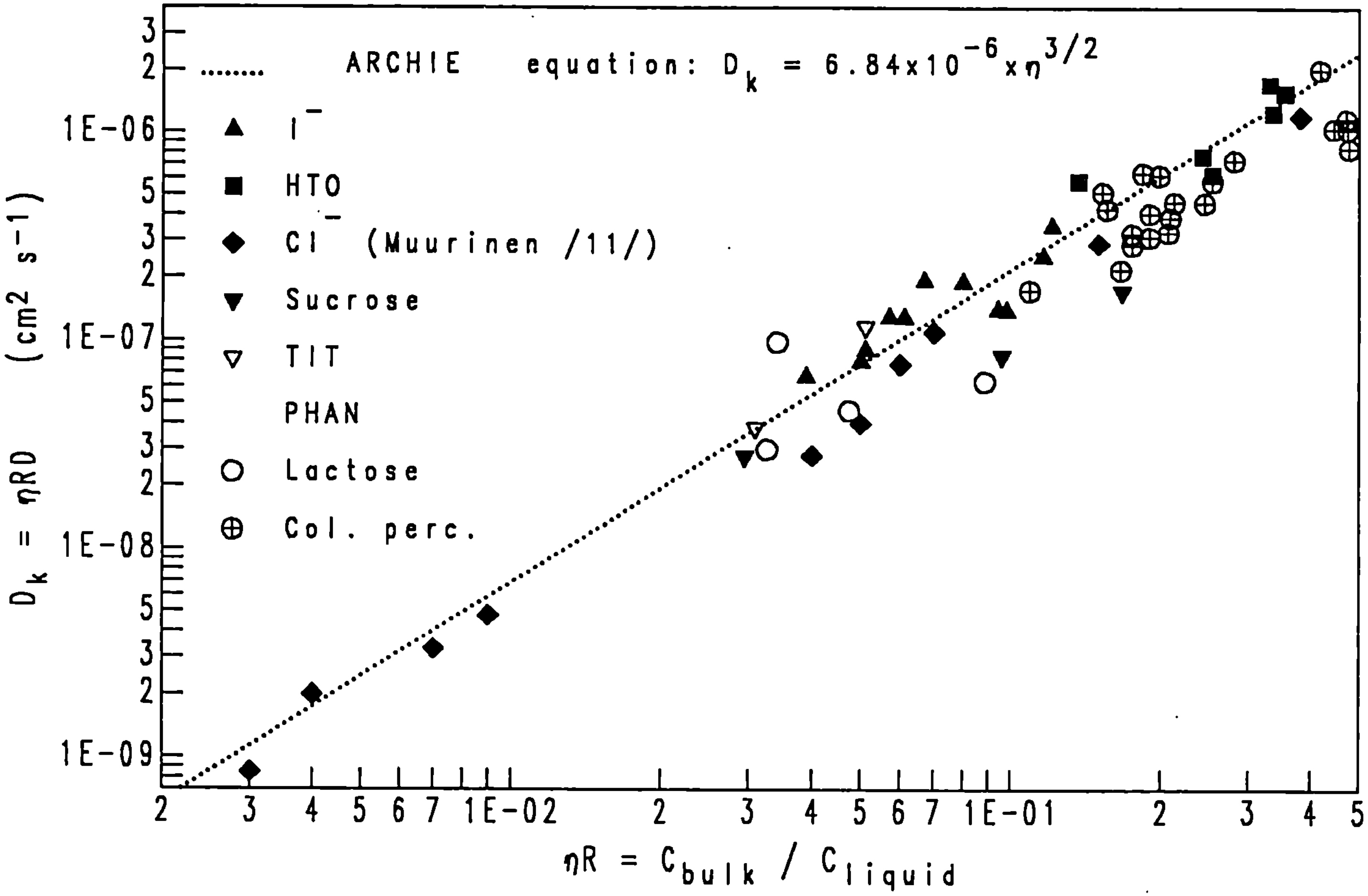

Figure 1: Diffusion conductivity constant D_k as a function of ηR values as determined by migration experiments (FT and percol.) for non-sorbed species.

FTCORRNU/mp/89

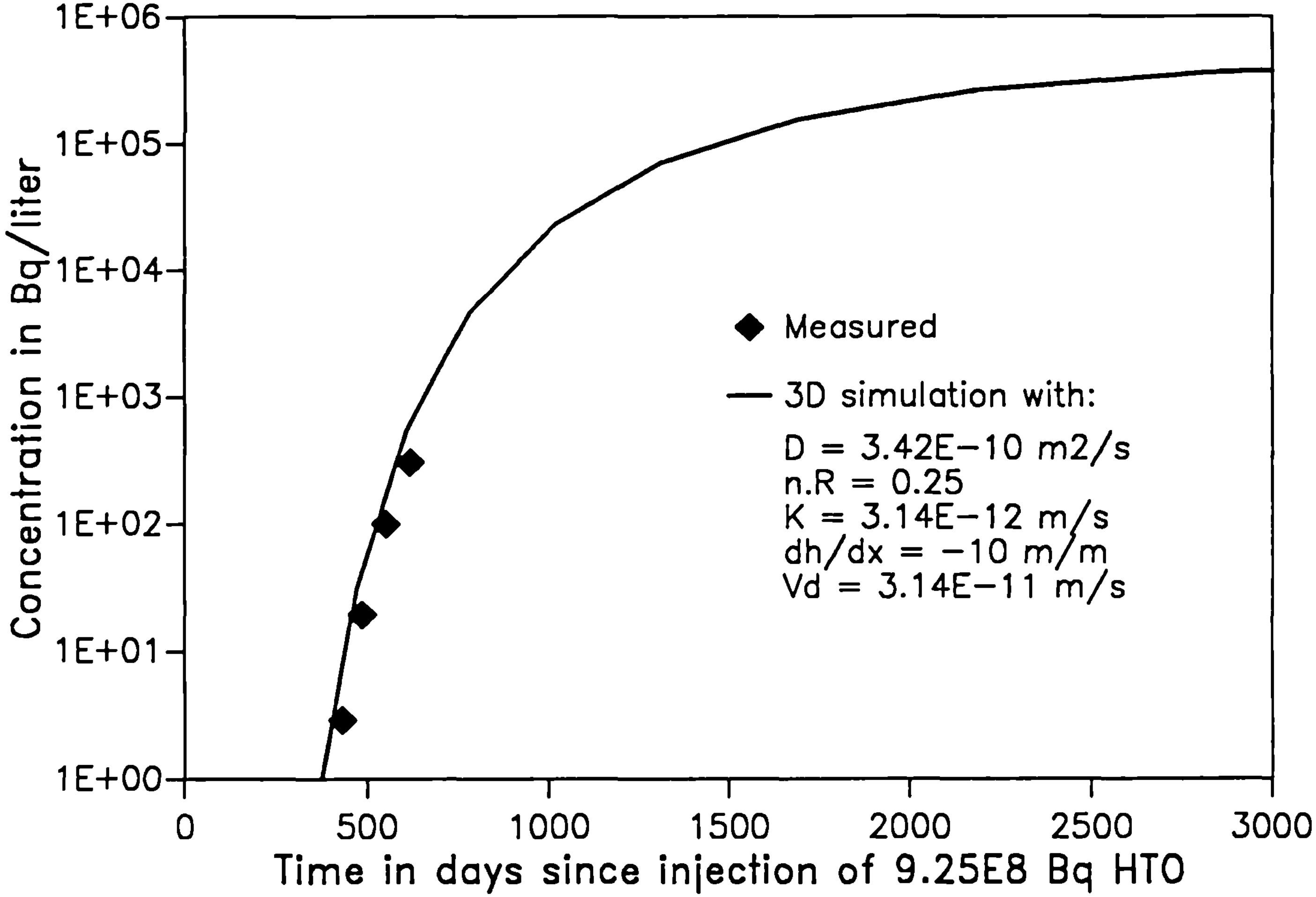

Figure 2. **Concentration in the liquid at a distance of 90 cm from the injection point.**

MIG89D4/mp/89

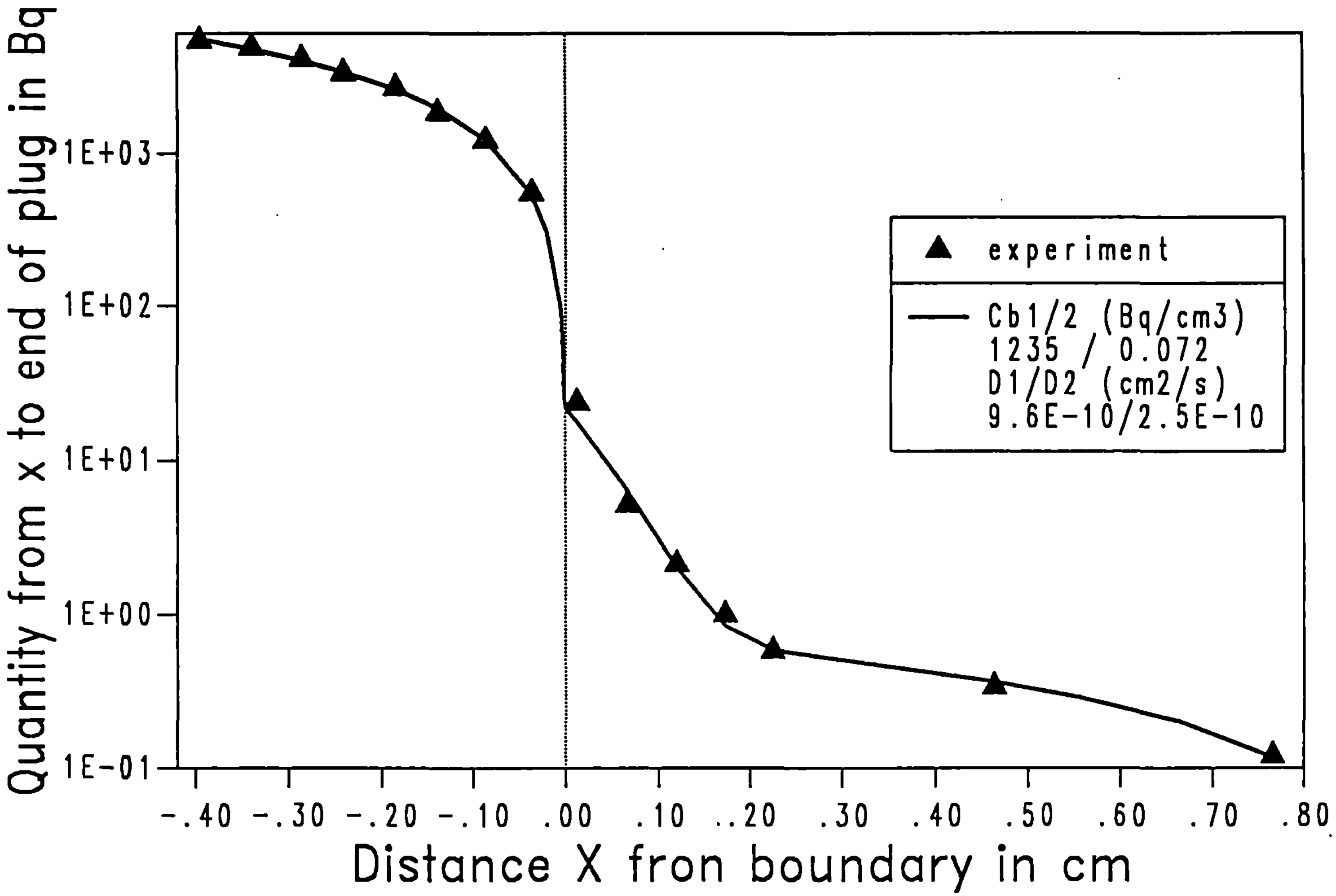

Figure 3. **Results migration experiment with Technetium. Labeled plug in contact with unlabeled.**

MIG89D5/mp/89

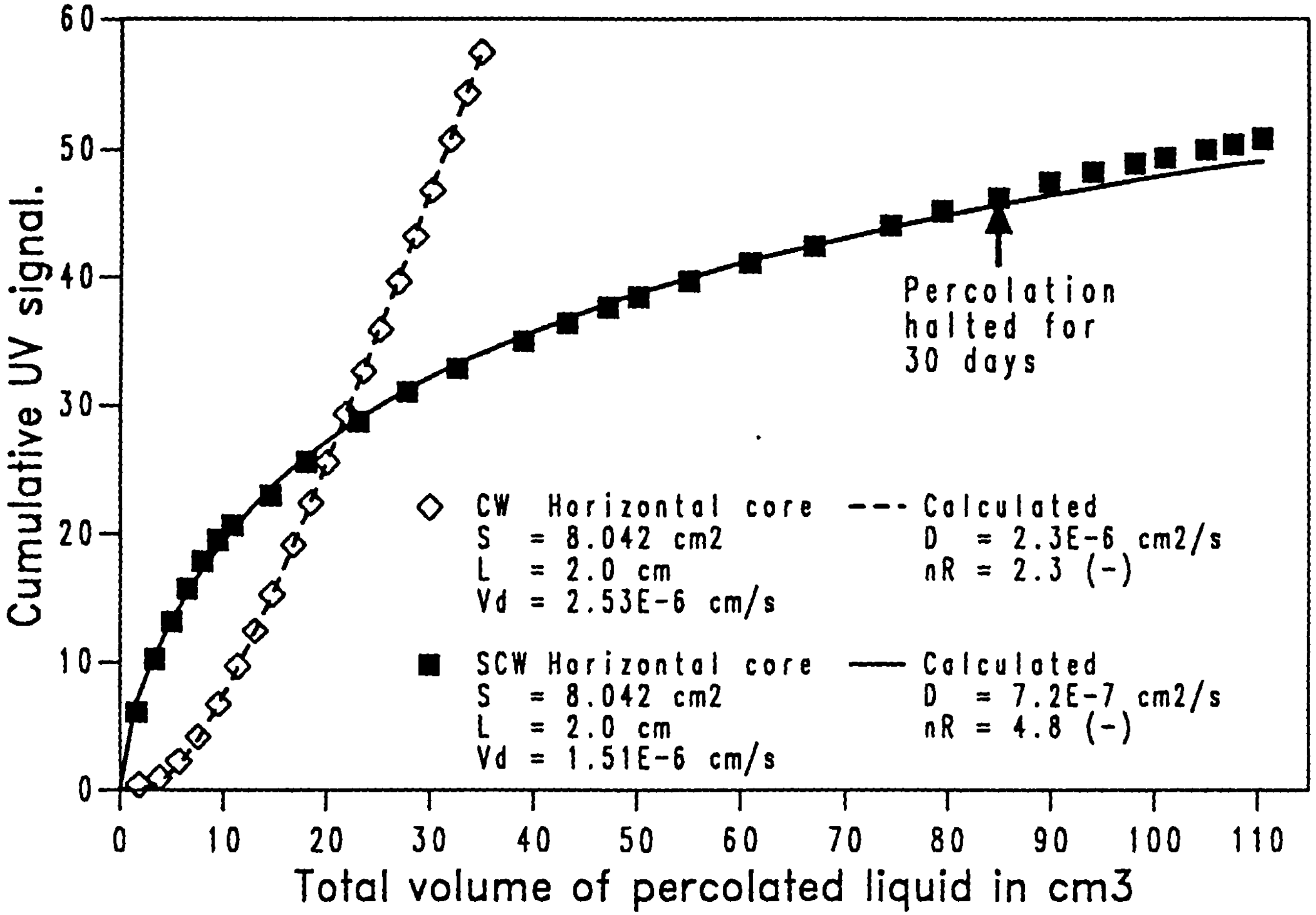

Figure 4. **Results of the percolation experiment for the measurement of the mobility of the Humic Acids present in the clay water. (preliminary test)**

MIG89D9/mp/89

INTERCOMPARISON OF PREDICTIVE COMPUTER PROGRAMS FOR RADIONUCLIDE MIGRATION IN THE GEOSPHERE

Contractor:	W.S. Atkins Engineering Sciences, Epsom, U.K.
Contract No:	FI1W/0077
Duration of Contract:	February 1987 - March 1990
Period Covered:	January 1989 - December 1989
Project Leaders:	T W Broyd, D Read

A. OBJECTIVES AND SCOPE

Prediction of the transport of radionuclides through the geosphere requires detailed knowledge of aqueous geochemistry, host-rock mineralogy and the specific mechanisms of solid-solution interaction. Such knowledge often needs to be obtained using computer models and associated databases, which must be shown to provide accurate results, i.e. are validated for use. International code verification and proving exercises have become accepted as an important means of assisting the validation process.

This contract will establish an international project called CHEMVAL, aimed at reviewing current progress and establishing research needs in the areas of chemical and chemical transport modelling. The objectives of CHEMVAL are as follows:

i) to produce the best possible overall thermodynamic database for use with aqueous speciation and coupled chemical transport codes, consistent with project resources and time-scales.

ii) to apply aqueous speciation computer models to a range of realistic waste disposal situations, and hence to establish and/or confirm areas of research requirement.

iii) to provide validation both for aqueous speciation models and coupled chemical transport codes.

B. WORK PROGRAMME

1. Comparison, sensitivity and extension of databases. The results of other parts of the work will be used in determining the most appropriate course of action.
2. The application of available aqueous speciation programs and databases to a range of hypothetical, though realistic, waste disposal situations.
3. The application of available programs coupling chemistry and waste transport to a range of test cases.
4. Validation of aqueous speciation and coupled codes by comparison, where possible, with a) experiments, b) field tests and c) natural analogues.

The exact scope and nature of work will be as agreed at plenary meetings of CHEMVAL participants, to be held throughout the duration of the project.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

CHEMVAL may be considered in terms of two related subject areas. The first deals with aspects of computer program verification/ validation and is designed to increase confidence in chemical speciation modelling. It involves the active participation of 14 research organisations within EC member states, Finland, Switzerland and Sweden. The remainder of project resources are devoted to reviewing, extending and improving the body of thermodynamic data available to participants, concentrating on those elements of greatest perceived relevance to radiological assessment. Within CHEMVAL, aspects of thermodynamic data are being considered primarily by the University of Manchester and the University of Wales. The organisation structure of the project is shown in Figure 1.

The modelling components of CHEMVAL comprise four main stages:-

STAGE 1 : application of aqueous speciation codes to representative cement water and groundwater compositions.

STAGE 2 : attempts at validation of aqueous speciation models by comparison with experimental field and laboratory data

STAGE 3 : verification of coupled chemical transport codes

STAGE 4 : attempts at validation of coupled models by comparison with experimental field and laboratory data.

At the end of 1989, Stage 1 has been concluded and reported, and Stages 2, 3 and 4 are substantially complete. Database activities continue throughout the project. A report on overall progress has recently been published /1/.

D. PROGRESS AND RESULTS

STAGE 1 - The objectives of CHEMVAL Stage 1 may be summarised as:-

- to assess the status of computer codes employed within Europe and to verify them on a variety of problems relevant to radioactive waste disposal.
- to highlight differences in thermodynamic data used at each organisation and the importance of such differences in determining radioelement speciation and solubility.
- to confirm areas where further work is required.

An agreed methodology was adopted for the verification study in order to distinguish variation in results caused by differences in thermodynamic data from those caused by other factors, such as coding/operator error or decisions made by the user at input. Participants were asked to perform calculations first with their own "in-house" database and then to repeat the simulations with a standard CHEMVAL database, issued by Atkins ES.

Test cases were based on actual field or laboratory analyses and were constructed to reflect the various disposal options envisaged by EC member countries. Radioelements included were those considered important during earlier radiological assessment programmes, namely americium, plutonium, uranium and technetium, plus possibly, caesium, strontium and cobalt in the case of rapid release.

Excellent agreement has been obtained not only for straightforward speciation calculations but also for conceptually difficult reaction path simulations (e.g. Fig 2). Other cases, including those concerned with redox reactions and organic complexation, displayed poorer agreement, however, and the lack of consistency especially in predicted trends requires further investigation.

A report on Stage 1 activities /2/ has been publised within the CEC's EUR series.

STAGE 2 - Stage 2 is concerned with the performance of models and databases for simulating real field or experimental datasets. Four sites are being studies:

Mol - representing a clay goundwater surrounding a repository.
Gorleben - simulating a saline water above a repository in salt.
Maxey Flats, Kentucky - an existing, near-surface repository.
Oman - natural, highly alkaline spring water providing an analogue to a cementitious near-field environment.

Test cases of varying complexity have been derived for each of the sites in question. The modelling work is proceding in two phases. In the first, participants were asked to provide "blind" simulations, that is in the absence of field results. The second phase entailed participants trying to match field results, and reporting on the methodologies employed. The results demonstrated that equilibrium models can produce a reliable estimate of experimental data when used within their known frame of reference, as exemplified by the predictions of Am and Np in synthetic Mol clay water shown in Figure 3.

STAGE 3 - Only a relatively small subset of participants have the capability to simulate coupled chemistry and transport. An initial phase of Stage 3 showed that candidate codes of a variety of complexity and solution algorithms can produce very similar results for uncoupled cases of aqueous speciation and solute migration. Test cases fully coupling the chemistry and transport included the reaction of cement with carbonated groundwater, the alteration of Na-bentonite by diffusion of Ca-rich groundwater and hypothetical instantaneous injection of NaOH into a siliceous aquifer.

STAGE 4 - It has proved difficult to identify appropriate test cases which were suitably and unambiguously defined. Two cases were finally chosen, the first involving Np migration through glauconite sands and the second concerning "flow through" column experiments in Fontainebleau sands.

Reports on Stages 2, 3 and 4 are currently in an advanced state of preparation.

Database Activities - Work on the production of a reviewed thermodynamic database will continue throughout the project. An extensive literature review of thermodynamic constants is being backed up by the use of model control cases and sensitivity studies, aimed at establishing the effect of proposed modifications to the database. Updated editions of the CHEMVAL database have been issued to coincide with each of the modelling Stages (Fig 4). The Stage 3 database contained 525 aqueous complexes and solubility products for 319 solids.

References

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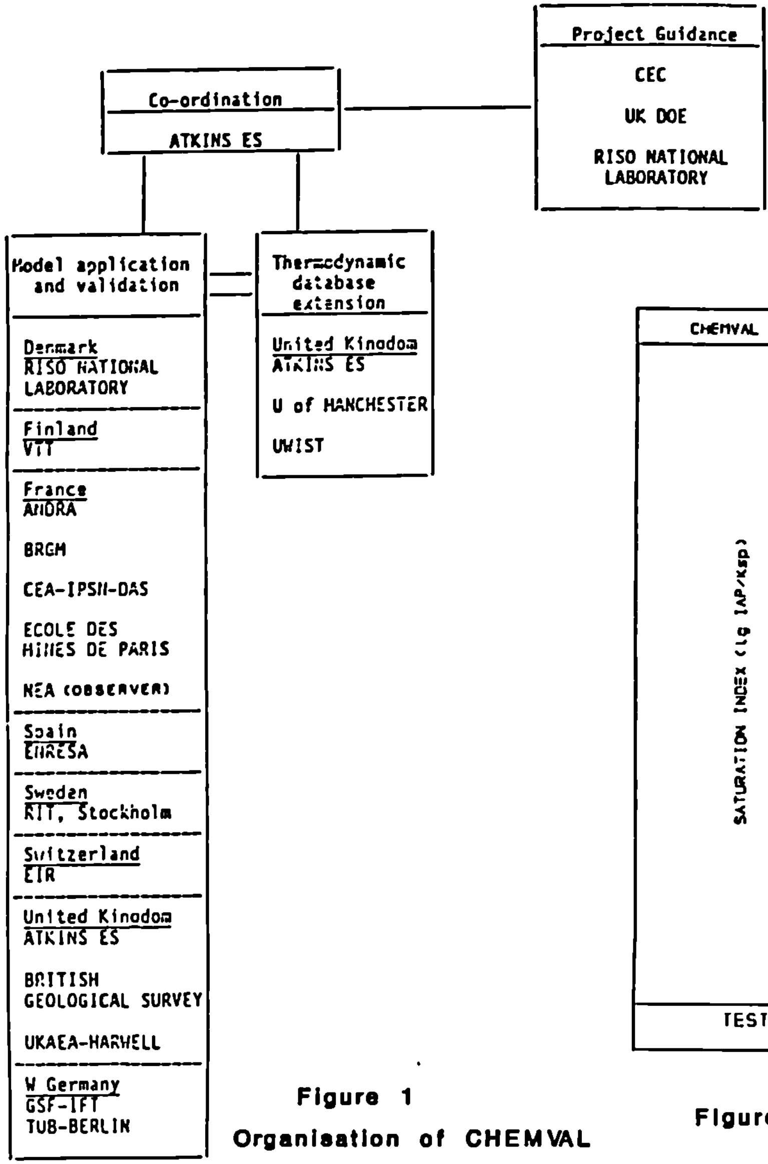

Figure 1
Organisation of CHEMVAL

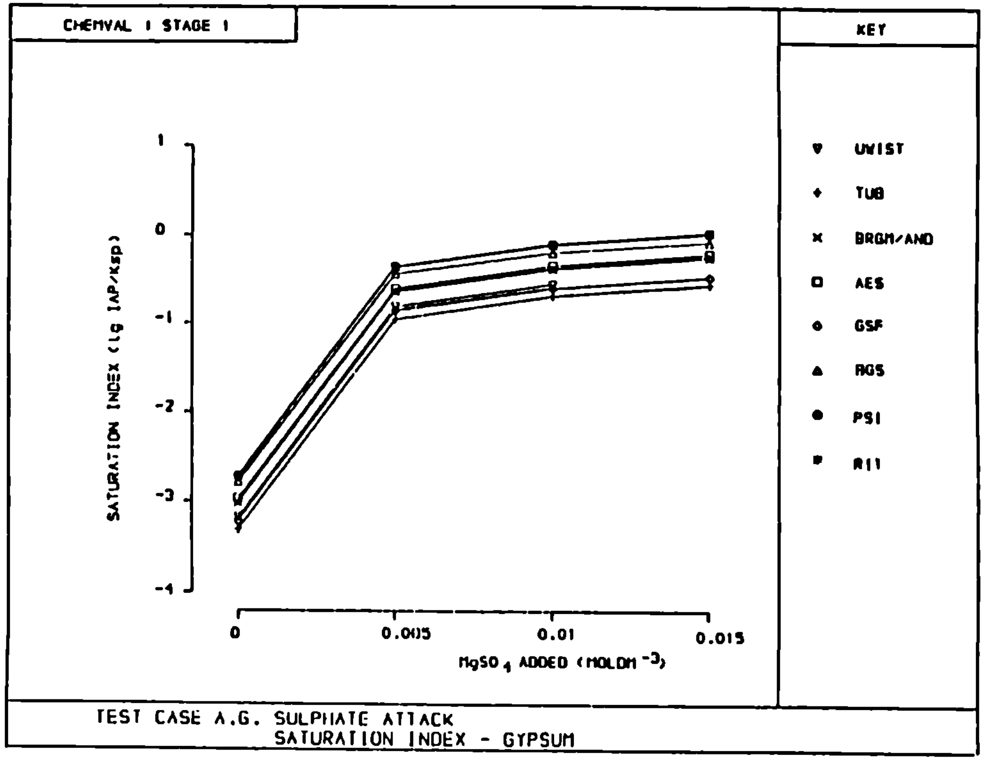

Figure 2 **Sulphate attack on cementitious water**

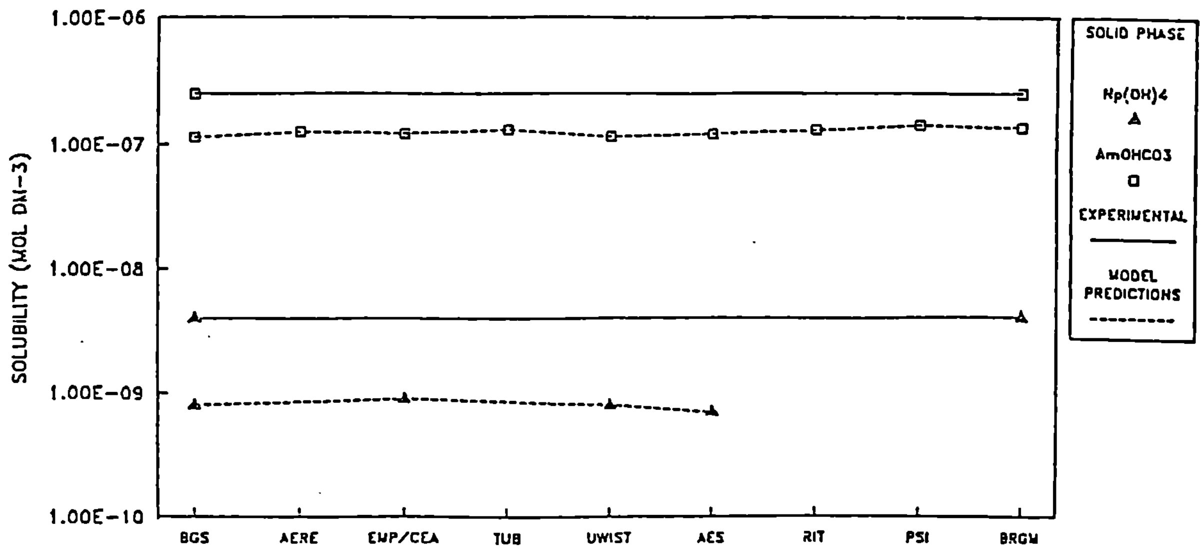

Figure 3 CHEMVAL Stage 2: Am and Np solubility for Mol

CHEMVAL - DATABASE

Stage	Release Date	Aqueous Species	Solid Species	Format
1	June 1987	376	259	PHREEQE MINEQL
2	May 1988	521	327	PHREEQE MINEQL
3	April 1989	525	319	PHREEQE MINEQL EQ 3/6
4	January 1990	≈675	≈390	dBase III+

Figure 4 CHEMVAL database releases

FAR-FIELD MODELLING OF RADIONUCLIDE MIGRATION

Contractor : Harwell Laboratory, United Kingdom
Contract No. : FI1W/0078
Duration of contract : January 1987 - December 1989
Period covered : January 1989 - December 1989
Project leader : J D Porter, K H Winters

A. OBJECTIVES AND SCOPE

Numerical simulation of groundwater flow and solute transport is an essential element of any assessment of the performance of a proposed repository for nuclear waste. The accuracy and realism of such simulations must be clearly demonstrable and also defensible against critical public scrutiny. Detailed simulations are expensive so any increased computational efficiency offers the opportunity for more extensive sensitivity analysis as part of a comprehensive repository assessment.

The project aims to improve the capability, efficiency and realism of the NAMMU code, which simulates groundwater flow and solute transport through a porous medium. Our detailed objectives are to discover and exploit superior methods for solving significantly non-linear problems; to introduce into the code a capability for modelling chemical reactions; to identify and test improved techniques for simulating the progress of sharp fronts in solute concentration; and to identify better-founded representations of solute dispersion. The last two topics will be covered by sub-contracts placed at Universities in the United Kingdom.

The NAPSAC code presently calculates flow through a three-dimensional network of fractures. Our aim is to decide upon and implement the most appropriate method for calculating solute transport through a three-dimensional network, so NAPSAC could be used to investigate radionuclide migration from a repository in hard fractured rock.

B. WORK PROGRAMME

1. Improved methods for solving significantly non-linear groundwater-flow problems will be implemented in NAMMU because those currently available, although robust, are expensive.
2. The NAMMU code will be enhanced to provide a means of modelling the impact of chemical reactions on solute transport in a porous medium.
3. Methods will be assessed and tested that offer prospects of simulating efficiently the advance of sharp fronts in solute concentration, without spurious dispersion.
4. Models of inhomogeneous materials will be explored to provide better understanding and descriptions of solute dispersion.
5. The NAPSAC fracture-network code will be enhanced by incorporating a judiciously chosen technique for calculating solute transport.

C. PROGRESS OF WORK AND RESULTS OBTAINED

State of Advancement

There has been substantial progress in all work areas in 1989 and the work on the first four tasks is now complete. In the "Progress and Results" section we will therefore summarise the overall conclusions from these four studies as well as describing this year's work.

During the current year:

The Quasi-Newton method due to Broyden has been applied to test cases which require the use of parameter stepping and a successful approach to using the Broyden method in such circumstances has been demonstrated.

The idea of coupling NAMMU and PHREEQE has been evaluated. It was decided that this was not a profitable approach at this time. The ternary ion exchange model incorporated into NAMMU has been further developed and tested.

Cranfield Institute of Technology have further tested Roe's Superbee method by application to convection-diffusion test cases. The advantages and disadvantages of this approach to the solution of sharp front problems have been identified.

Newcastle University have carried out Monte Carlo simulations of solute transport in a heterogeneous porous medium and have demonstrated that in the early stages of plume development the dispersion process is non-Fickian. An evaluation has also been made of schemes for the calculation of effective permeabilities.

A suitable method for the representation of transport in fracture networks, based upon a particle tracking approach, has been identified and the coding of this method into NAPSAC is underway.

Progress and Results

Improved Solution Methods for Non-Linear Problems (Task 1)

The Quasi-Newton method due to Broyden /1,2/ had been found to be robust and to give a useful saving in computation time. In the period of the present report the application of this method in conjunction with parameter stepping has been investigated. A discussion of parameter stepping can be found in /3/.

The test case used was again a saline convection cell, but with much smaller amounts of dispersion than the original example. Starting from a zero initial guess for all variables it was not possible to find a solution to the new problem directly using Newton-Raphson iteration on a coarse mesh. Parameter stepping was therefore used, giving the sequence of solutions shown in Figures 1.1-1.6. A similar sequence was obtained on a finer mesh. These Newton-Raphson solutions were then used as a benchmark against which the performance of the Broyden method could be judged. Two ways in which the Broyden method can be used in conjunction with parameter stepping are:

(i) At each parameter step, perform an initial Newton-Raphson iteration, followed by a number of Broyden iterations.

(ii) At the first parameter step perform one Newton-Raphson iteration followed by Broyden iterations. For all subsequent parameter steps use only Broyden iterations.

Both methods were implemented in NAMMU and the results obtained are summarised in Table 1. It can be seen that the first approach gives the greatest reduction in computing time on both meshes and this method is therefore recommended.

Overall the following conclusions can be drawn from this study:

1) The Broyden method gives a useful reduction in computing time and appears to be robust.

2) The relative saving obtained with the Broyden method increases with the problem size.

3) When the Broyden method is used in conjunction with parameter stepping, approach (i) above should be used.

4) The BFGS method was unsuccessful when applied to our test cases.

5) Line searches did not improve the performance of either the Broyden or BFGS methods.

Incorporating Chemical Reactions in the NAMMU code (Task 2)

A copy of the geochemical code PHREEQE was obtained /4/ and some sample timings made on the Harwell CRAY 2. These suggested that the CPU time requirements of a coupled PHREEQE/NAMMU code would be so great that such a code would not be a useful tool for assessment calculations. Work in the present year has therefore concentrated on the extension to two-dimensional cases of the ion exchange model described in the previous report and further testing of the model. The enhanced version of NAMMU has now been used successfully for two-dimensional calculations of transport with competitive ion exchange, including a case with heterogeneous chemical properties (eg different exchange capacities in different rocks). Other sample calculations include an investigation (loosely based on /5/) of the effect of calcium concentration on the migration of strontium (see Figure 2). Overall the results of this study are:

1) An enhancement of the empirical sorption models used in NAMMU.

2) The incorporation of a ternary ion exchange model in NAMMU, verification of this model and its use for some example calculations.

3) An assessment of the desirability of coupling NAMMU to an existing geochemical speciation code.

Developing Numerical Methods to Simulate the Advance of Sharp Fronts (Task 3)

As noted in a previous report this work was sub-contracted to Prof. Roe at the College of Aeronautics, Cranfield Institute of Technology. In the period of this report work has concentrated on the application of Roe's Superbee method /6/ to a number of test cases of two-dimensional convection-diffusion on a regular mesh. The last of these was based upon an INTRACOIN radionuclide transport test problem /7/ and included media with sharply contrasting hydraulic properties. The conclusions which can be drawn from the study are:

1) The Superbee method gives impressive results on some high Peclet number test problems.

2) The method is not so useful for cases with very different velocities in different parts of the solution domain (eg because of large permeability contrasts). This is because the timestep may be forced to be much shorter than the timescale of the process of interest. Impractically large numbers of timesteps would then be required to solve the problem.

3) The method could be useful for cases of contaminant transport in a single aquifer.

4) The method has only been tested on regular meshes.

A report on a theoretical study of the application of Superbee-type methods on unstructured meshes is awaited from Cranfield.

Methods for the Representation of Solute Dispersion (Task 4)

As noted in a previous report this work was sub-contracted to Prof. O'Connell at the Department of Civil Engineering, University of Newcastle upon Tyne. The dispersion of a solute in a heterogeneous porous medium was investigated using a Monte Carlo simulation technique. Realisations of the conductivity field were generated and then the flow through the system was calculated. Particles were tracked through the flow field and the dispersion of a particle cloud was analysed. Four sets of simulations were carried out, corresponding to two different flow rates and different statistics for the conductivity field. This study differs from previous work in this area in that plume development is considered in three dimensions and more emphasis is placed upon the analysis of the distributional properties of the generated concentration fields. The main conclusions from the study were:

1) The concentration distribution does not settle down to a multivariate normal distribution within the length and timescales of the numerical experiments.

2) The deviations from normality increase with the flow rate and the correlation length of the conductivity distribution.

A study was also carried out of methods of calculating effective hydraulic conductivity values for use in a multi-scale modelling exercise. It was found that the choice of integration volume and of weighting functions (even within the set of apparently suitable functions) can be very important. The results suggested that using an integration volume larger than the "application volume" on which the conductivity value was to be used could give improved results.

Transport in Fracture Networks (Task 5)

In order to account for the geometrical dispersion arising from the many different routes through a network of flow conducting fractures, it is appropriate to model the fractures directly within a stochastic fracture network model such as NAPSAC. We have assessed the alternative approaches to developing a transport algorithm within the data structure of NAPSAC.

The transport algorithm will be based on a particle tracking approach. We retain the concept of single fracture 'response functions' used in the flow model, and will calculate an appropriate number of pathlines between intersections on each fracture. These paths tell us which intersection locations are connected to which, and also the time a particle takes to travel between them. We must next assign appropriate probabilities that a particle arriving at an intersection will follow each particular path from that intersection. Once these have been set we can use the 'responses' on each fracture to efficiently track very many particles through the network. This approach has the advantage that it is relatively straightforward to substitute alternative conceptual models of single fractures: parallel-plate; variable aperture; or channelled fracture models may be incorporated within the modular code structure.

The outline data structure is now available and we are currently encoding the calculation of 'single fracture response' pathlines. This is the most difficult stage of the development since these calculations must be performed many times on each fracture: the algorithm must be very robust and efficient.

Subsequent examples will focus the work on areas where we can expect the code to be applied in the field. In particular, we will consider a test case involving a realistic number of random fractures, and we hope to develop a test case involving a typical borehole experiment. It is in the interpretation of such field experiments that we believe NAPSAC, and its transport option, will find its most important application.

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/6/ ROE, P.L., Characteristic based schemes for the Euler equations. Ann. Rev. Fluid Mech 18, pp 337 (1986).

/7/ INTRACOIN : International Nuclide Transport Code Intercomparison Study : Final Report Levels 2 and 3 Swedish Nuclear Power Inspectorate, Stockholm, Sweden (1986).

Table I : Comparison of Newton-Raphson and Broyden Methods for Parameter Stepping

Grid (Elements)	(v) Parameter Step Number (Value of D)	Number of iterations (ii) (NR)	(iii) (NB)	(iv) (BY)	Time in Solver (i) (NR)	(NB)	(BY)	Time (NB) / Time (NR)
5x5	Initial (10^{-6})	5	1+20	1+20	4.41	3.93	4.07	0.89
	1(8.0×10^{-7})	2	1+1	9				
	2(7.0×10^{-7})	2	1+1	5				
	3(5.6×10^{-7})	2	1+1	5	25.4	20.6	37.0	0.81
	4(3.6×10^{-7})	3	1+2	7				
	5(8.0×10^{-8})	4	1+4	57				
10x10	Initial (10^{-6})	4	1+15	1+15	38.7	19.9	19.6	0.51
	1(8.0×10^{-7})	2	1+1	7				
	2(7.0×10^{-7})	2	1+1	4				
	3(5.6×10^{-7})	2	1+1	7	157	81.2	93.7	0.52
	4(3.6×10^{-7})	3	1+2	7				
	5(8.0×10^{-8})	4	1+6	35				.

Notes:
- (i) Solver times in seconds
- (ii) NR means full Newton-Raphson method at each step
- (iii) NB means 1 Newton-Raphson iteration followed by Broyden iterations at each step. 1+N means 1 Newton-Raphson iteration followed by N Broyden iterations
- (iv) BY means 1 Newton-Raphson iteration followed by Broyden iterations are used to obtain the initial solution. Only Broyden iterations are used at subsequent parameter steps
- (v) Value in brackets is the Diffusion Coefficient at that parameter step

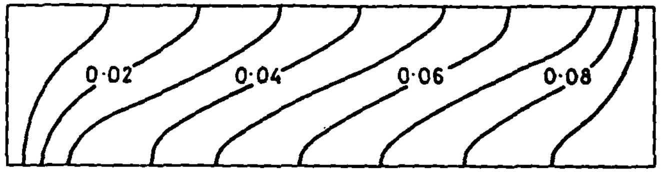

FIG. 1·1. INITIAL SOLUTION ($D = 10^{-6}$): CONCENTRATION.

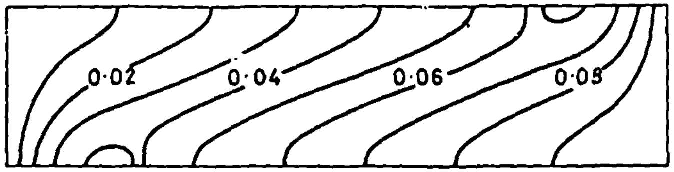

FIG. 1·2. FIRST PARAMETER STEP ($D = 8 \times 10^{-7}$): CONCENTRATION.

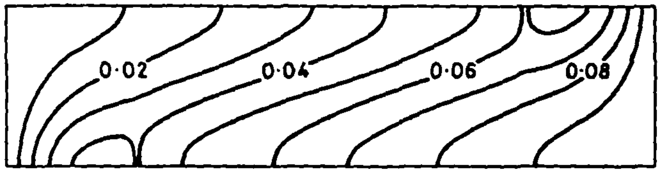

FIG. 1·3. SECOND PARAMETER STEP ($D = 7 \times 10^{-7}$): CONCENTRATION.

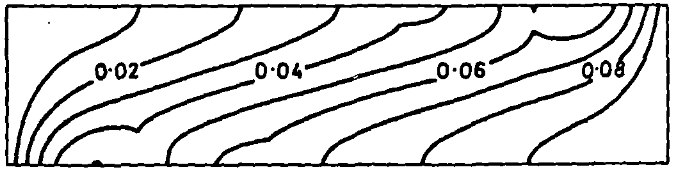

FIG. 1·4. THIRD PARAMETER STEP ($D = 5{\cdot}6 \times 10^{-7}$): CONCENTRATION.

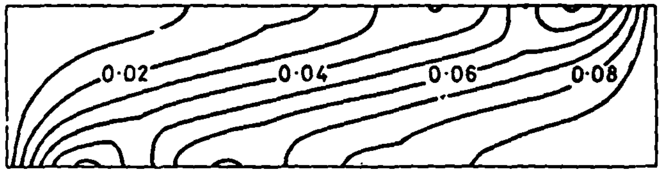

FIG. 1·5. FOURTH PARAMETER STEP ($D = 3{\cdot}6 \times 10^{-7}$): CONCENTRATION.

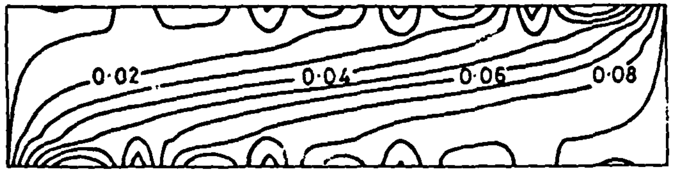

FIG. 1·6. FIFTH PARAMETER STEP ($D = 8 \times 10^{-8}$): CONCENTRATION.

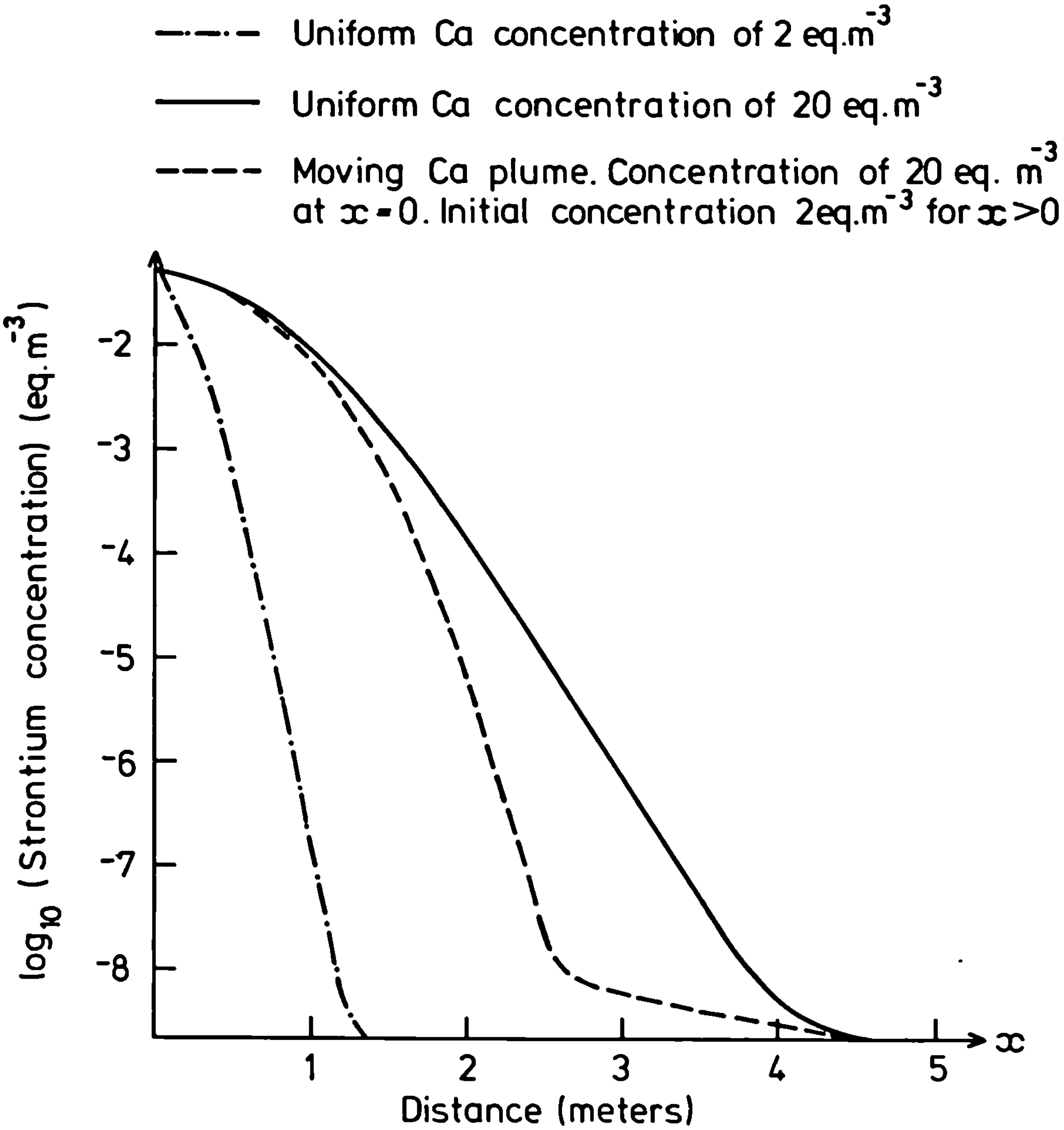

FIGURE 2: EFFECT OF CALCIUM CONCENTRATION ON TRANSPORT OF STRONTIUM.

Title:	Geochemical Modelling
Contractor:	Department of Chemistry, Risø National Laboratory, Denmark
Contract No.:	FIIW/OO79
Duration of contract:	August 1986 - July 1990
Period covered:	January 1989 - December 1989
Project leader:	Bror Skytte Jensen

A. OBJECTIVES AND SCOPE

To develop a versatile and userfriendly program for geochemical modelling. The program is intended for PC-use and should have a wide applicability and take advantage of new and recent insight in fundamental processes.

B. WORK PROGRAMME

2.1. To translate the earlier WHATIF-programs into PASCAL. The pc version Turbo Pascal 4.0 has been found very convenient.

2.1.1. The limitations of the earlier versions are removed simultaneously with the introduction of possibilities for handling new important reactions and processes.

2.1.2. The following phenomena are considered:

Free and floating pH and pE.
Total carbonate or fixed CO_2 pressure.
The SO_4/S redoxpair active or not.
Multielement ion-exchange.
Quantitative precipitation and dissolution reactions.
Solid solutions, co-precipitations.

2.2. Experimental investigations of adsorption phenomena on calcite are undertaken. The formation of solid-solutions 'in' surface layers is considered a possibility.

C. PROGRESS OF WORK AND OBTAINED RESULTS

2.1. 2.1.1. 2.1.2.

The translation of the WHATIF-programs into Turbo Pascal is finished. The core of the program with its associated database is working fast and reliably. To secure the userfriendliness of the program it has been extensively 'menuterised' both for database maintenance and for chosing calculation modes.

The program can handle multielement ion-exchange directly, with either fixed or floating modes in pH and pE exposed to fixed carbondioxide pressure or not. It allows the user to chose conditions where the SO_4/S redoxpair is active or not simultaneously with the other options.

An algorithm has been constructed which allows for the quantitative calculation of mass-transfers, when minerals precipitate or dissolve, simultaneously adjusting the composition of the aqueous phase to the new conditions. The computation is reasonably fast and with the choice of relevant 'generator functions', reaction path studies may be performed directly with the new program. Co-precipitation has been handled under certain simplified assumptions, which although they neglect non ideal interactions in the solid phases, might prove usefull for making estimates of trace element distributions among mineral phases.

Taking all the above improvement into account, it was felt that the new program deserves its own name. It has therefore been christened: JENSEN, a name which should be easily remembered.

2.2.

Numerous experimental data have been collected, which show the expected competition between Ca and Mg for carbonate. The adsorption of europium on calcite has, not unexpected, been proven to be proportional to the available surface. The first experiments indicated two types of adsorption, the one almost instantaneous the other irregulary slow. The first is interpreted as direct ion-exchange on the surface the second as the incorporation into a surface layer of finite thickness, a process displaying nucleation phenomena. Later experiments have not shown the two types of adsorption, probably because the laboratory environment now contains plenty of the nuclei for 'compound adsorption'.

Title:	Geochemical Databases
Contractor:	Department of Chemistry, Risø National Laboratory, Denmark
Subcontractor:	F.J. Pearson, GROUND WATER CHEMISTRY. 10700 Richmond Avenue, Suite 263 Houston, TX 77042
Contract No.:	FII/OO8O
Duration of contract:	August 1986 - July 1990
Period covered:	January 1989 - December 1989
Project leader:	Bror Skytte Jensen

A. OBJECTIVES AND SCOPE

Apart from the obvious demand that the geochemical problem to be solved shall be well defined and the calculations being done with sufficient precision, the nescessery input of thermodynamic data, usually taken from literature, may suffer from lack of internal consistency or may be too imprecise referring to a species which have not been identified with certainty. For this reason the geochemical calculations may be in error or screwed and should not be used without some hesitation.

The present project aims at improving existing databases, such that they may be used to handle a wider range of problems.

B. WORK PROGRAMME

2.1. To develop a database management system, PMATCH, which can be used to make available data internally consistent, i.e. to refer to the same set of fundamental standard data.

2.2. To investigate the possibilities of using Expert Systems in handling and evaluation of geochemical data.

2.3. To take part in the work in the CHEMVAL group.

C. PROGRESS OF WORK AND OBTAINED RESULTS

2.1. The PMATCH program, A Program to Manage ThermoCHemical Data, has been developed by J.F. Pearson and John D. Avis (Intera Technologies Ltd. Ottawa, Ontario), and will soon be published.

The program is written in Turbo Pascal intended for IBM compatibles.

The program is of great use for those who wish to compile and develop their own databases.
The program has been tested by an untrained user, BSJ, and been found easy to start and to use.

2.2. An expert-system, with the provisional name 'Little Joe', has been constructed by J.F. Pearson and Andreas Haug (Intera Technologies, Inc. , Austin, Texas). The program, which shall be looked at as an experiment in applying expert systems to geochemistry, is intended to supply expert judgement of the quality of analyses of ground water. The development has been presented and discussed at a meeting in The American Chemical Society evoking considerable interest.

2.3. The achievements of the CHEMVAL group is presented elsewhere.

MIGRATION OF RADIONUCLIDES BY HIGH-DENSITY BRINES: FINALISATION OF THE METROPOL CODE

Contractor: RIVM, Bilthoven, The Netherlands
Contract No.: FI 1W/0081
Duration of contract: October 1986 - October 1988
Period covered: January 1989 - December 1989
Project Leaders: P.Glasbergen, F.Sauter

A. OBJECTIVES AND SCOPE

Transport of radionuclides, possibly released from a salt dome repository in the surrounding aquifers, is a key problem in the safety analysis of waste disposal in salt. For the numerical solution of the set of equations (Darcy's, Fick's law and extensions of these laws for high density brines) which describe the transport, a family of computer codes, called METROPOL, has been developed.

So far METROPOL-1, 2 and 3 have been developed, dealing with, respectively, steady state 3D flow, transient 3D flow and transient 3D flow of fluid with high density differences.

The objective of the present contract is to complete the development of the computer code METROPOL-4, which simulates the transport of low concentration dissolved species, including such processes as adsorption and decay.

In order to make METROPOL easily accessible to other users documentation will have to be written, whereas for an easy interpretation of calculated results much effort will have to be put into the development of post-processing facilities.

B. WORK PROGRAMME

1. Inclusion of physico-chemical processes, such as dispersion, diffusion, adsorption, desorption, decay, dissolution/ precipitation of salt, chemical reactions, in the METROPOL-code.
2. Inclusion of thermal effects.
3. Code development. New numerical techniques should be studied for the solving of large sets of linear equations as well as for the integration in time (adaptive time integration scheme).
4. Testing and validating the METROPOL-3 code in the case of the flow of high-density brine.
5. Preparing user manuals for METROPOL.
6. Development of post-processing facilities such as particle tracking, contour plotting.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Although the contract beteen the RIVM and the CEC has ended, in the following a review of the work done in connection with the development of METROPOL in the year 1989 will be given. Summarising, the following aspects are treated below:

- the extension of METROPOL-4 (the transport module of METROPOL) to include matrix diffusion,
- the extension of METROPOL to include interaction between grondwater and surface water,
- the introduction of new spatial and time integration schemes in METROPOL-3 (the brine flow module of METROPOL),
- the inclusion of new terms in Darcy's law and Fick's law and of a consistent set of boundary conditions in METROPOL-3,
- the development of a preprocessing program which automatically assigns parameter values to a finite element mesh as used by METROPOL, taking input from digitalised maps with parameter distributions.

Up till now the following reports have been published /3/, /4/, /5/.

PROGRESS AND RESULTS

1. Inclusion of physico-chemical processes

Extension of METROPOL-4 to include matrix diffusion. In many natural geological media, two types of pores can be distinguished: micropores and macropores. Micropores are considered to consist of dead-end pores, cracks, stationary pockets and very tiny pore spaces. Although these are connected to the overall pore space, by virtue of their geometry they contribute very little to the macroscopic transport of fluids through the medium. Nevertheless, micropores play an important role in the retardation of species carried by water. One may assume that macroscopic transport of water and its components take place through macropores. Examples of media which can be described with such a micropore-macropore structure are clayey soil containing aggregates and fractured consolidated formation containing large pores separated by blocks of low permeability rock. The regions of soil containing the micropores are called "matrix" and therefore the process of exchange of mass between micropores and macropores is refered to as "matrix diffusion".

Obviously, to calculate the distribution of mass of solutes in macropores and micropores, two sets of mass conservation equations need to be solved (simultaneously).

The normal mass balance equation for the transport of a species by groundwater includes terms for advection, molecular diffusion, hydrodynamic dispersion, adsorption/desorption and decay/production. An exchange term, accounting for the micropore-macropore mass exchange, needs to be added to this equation of mass balance for macropore. The mass balance equation for solutes in the micropores contains the same mass exchange term; there are no advective or dispersive terms present here because we assume that the fluid in the matrix stays stagnant. In the extended version of METROPOL-4, a Picard iteration scheme is employed to solve the two equations.

Another process included in METROPOL is the interaction between groundwater and surface water. A non-linear relation between hydarulic head and re- or discharge to the surface water has been implemented. A Picard iteration combined with under-relaxation is employed to solve the non-linear problem.

3. Code development.

Because it was felt that, especially in the case of the salt mass balance (METROPOL-3); the spatial integration scheme used up till now was not accurate enough, an integration scheme using Gauss points was implemented. Furthermore the time integration scheme has been changed. Originally, a fully implicit scheme has been employed, where the non-linear equations (coupled fluid mass balance and salt mass balance) were solved using Picard iterations. A problem here is the time step selection, since a too large time step may result in a non-converging Picard iteration. The new scheme chosen is an impicit/explicit scheme (IMPES). The calculations within a time step are now as follows:

a) calculation of pressures from the fluid mass balance, fully implicit in pressure and explicit in salt mass fraction

b) calculation of salt mass fraction from the salt mass balance, fully implicit in pressure and explicit in salt mass fraction

c) time step selection based on the Courant number for each grid block.

Steps a) and b) both give a linear system of equations, so non-linear iterations are not neccessary anymore. The implementation of this new scheme is now in a test phase.

Extra terms in Darcy's law and Fick's law for high salt concentration (/1/, /2/) have been implemented in METROPOL-3. The inclusion of these terms is an option for the user; no superfluous work is done by METROPOL-3, when these terms are not to be included.

Seven different combinations of boundary conditions are now available for METROPOL-3. This means that no free choice can be made between a boundary condition for the pressure and one for the salt mas fraction in order to avoid physically unrealistic combinations which often cause numerical problems. For example, when both the pressure and the salt mass fraction are fixed on a boundary, this combination leads to problems when outflow takes place. In this case, the mass fraction level has to be maintained and negative mass fractions often occur in the model region.

6. Pre- and post processing

A certain flow problem, now run with METROPOL, involves 100000 grid blocks. In order to enable the user to handle such large amount of data, a preprocessing program, METROPRE, has been developed. This program takes input from digitalised maps with areas representing a certain parameter distribution. METROPRE takes care of assigning parameter values to the nodal points or blocks of a finite element mesh as used by METROPOL.

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STUDY OF THE COUPLED-HYDROMECHANICAL EFFECTS DUE TO A HLW REPOSITORY IN THE GRANITE GEOLOGICAL FORMATION

Contractor : COMMISSARIAT A L'ENERGIE ATOMIQUE
31/33 rue de la Fédération
F-75752 Paris
Contract No. : FI 1 W-0148
Duration of contract : October 1987 - May 1989
Period covered : January 89 - May 89
Project leaders : M. Durin, A. Millard, P. Oustriere, P. Raimbault

A. OBJECTIVES AND SCOPE

For the evaluation of the performance of radioactive waste geological storage, the description of hydraulic, thermal, chemical and mechanical phenomena is necessary in the near field and in the far field. The difficulty of this description tests, on the one hand in the definition of behaviour laws in accordance with time and space that we consider, on the other hand in the connection of phenomena.

The study already achieved on this subject in the different countries are concerned with either thermo-mechanical effects or thermo-hydraulic effects.

The main purpose of this study is to consider together the thermo-hydro-mechanical effects due to a HLW repository in a granite site. In order to do so two models available now are coupled : CASTEM (thermo-mechanical model) and TRIO (thermo-hydraulic and migration model). Studies of radionuclides migration in the granite rock are conducted with the coupled models in order to evaluate the influence of the different phenomena.

B. WORK PROGRAMME

1 - Thermal calculations
2 - Thermo-mechanical computations
3 - Hydraulic and migration calculations
    3.1. Reference calculation without thermal and mechanical coupling
    3.2. Coupling with thermal effects
    3.3. Full coupling (thermal and mechanical effects)

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The thermal calculations, thermo-hydraulic coupling and preliminary thermo-mechanical calculations were presented in the 1988 progress report. As for 1989, the full thermo-mechanical hydraulic coupling has been performed with trajectory and migration calculations which completed the study.

Progress and results

1. Thermo-mechanical computations

New calculations have been performed to obtain a better precision. The volume of rock modelled has been extended to 8 000 m x 2 500 m and the number of grid elements increased from 1 328 to 2 700 (figure 1).

The granite of the initial condition is considered as an homogeneous elastic material with limited traction resistance. Fissure opening occurs after the elastic limit is reached with a strain softening behaviour.

Fissure opening is considered homogeneous on each grid element and characterized by an increase in porosity and a general direction perpendicular to the main stress component.

Contour plots joining elements with the same increase of porosity are shown in figure 2 at 150 years which corresponds to the maximum amplitude of rock deformation and in figure 3 at 1 000 years.

The new conditions modify slightly the horizontal deformation of the repository and the fissure positions near the surface.

2. Hydraulic and migration calculation (full coupling)

The coupling between thermal effects and groundwater flow is considered both ways : through buoyancy effects and through the modification of permeability and transient hydraulic behaviour from mechanical rock deformations /1/.

The variations of permeability are calculated, assuming an initial density of fissures and fissure opening calculated from the initial porosity and permeability of the equivalent porous medium. The fissure opening is supposed to be modified uniformely, the fissure density remaining constant. The sensitivity of the result to this hypothesis has been considered. The values of hydraulic heads and velocity flow charts are shown on figures 4, 5, 6 and 7 at 150 years and 1 000 years.

Calculation of particle trajectories and transit times from the repository to the surface for different departure times, show that in the most pessimistic case the transit time is reduced by half for an early departure time. This decrease in transit time is reduced to 10% after 1 000 years (figures 8, 9 and 10).

Assuming a gaussian source release which maximum is at 25 000 years and standard deviation 12 500 years and assuming a retardation coefficient of 10, the difference in activity release without and with thermal effects (partial and full coupling) is shown in figure 11.

These results indicate that with the parameters and hypothesis which have been used, and which should be confirmed, it is justified to decouple phenomena associated to thermal effects from the migration of radionuclides in the geosphere related to the normal evolution scenario in crystalline rock.

/1/ A. Millard, A. Stietel, M. Durin, A. Bengaouer, JM Peres, P. Raimbault : Symposium on the Safety Assessment of Nuclear Waste Repositories - Paris - France - 9/12 Oct. 1989 - Paper P25.

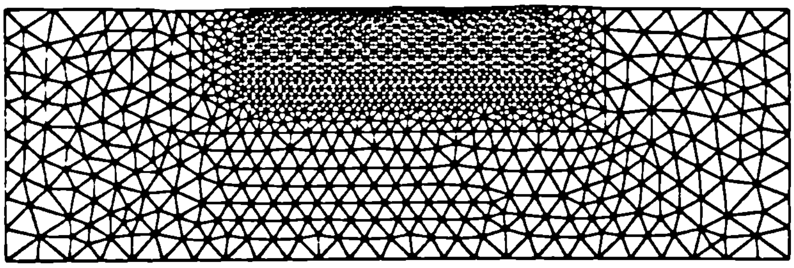

Figure 1 : Grid mesh for the mechanical calculations

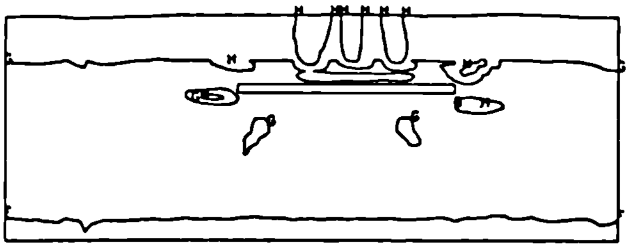

Figure 2 : Porosities at 150 years

UNITES
F = 0 1000000E- 3
G = 0 5000000E- 3
H = 0 1000000E- 2

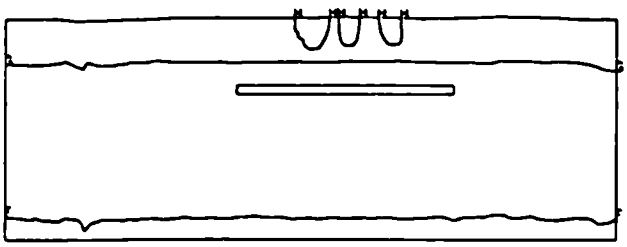

Figure 3 : Porosities at 1 000 years

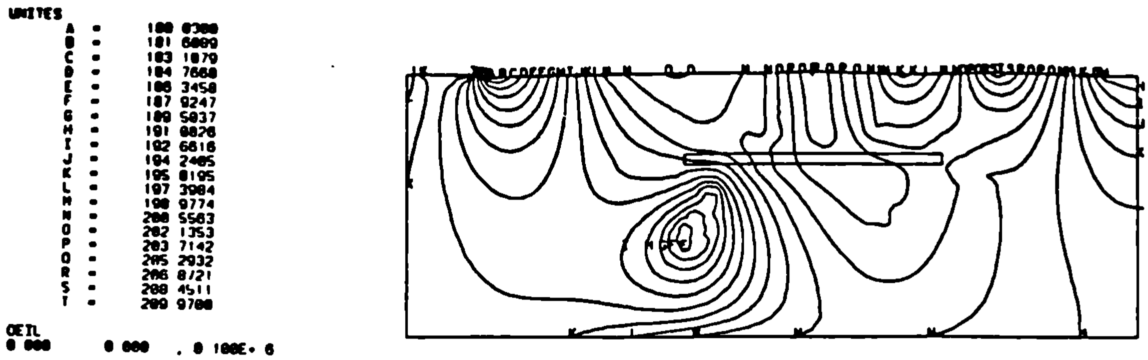

Figure 4 : Hydraulic heads at 150 years

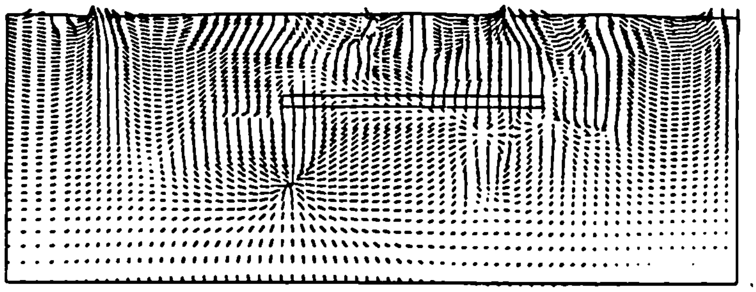

Figure 5 : Flow chart at 150 years

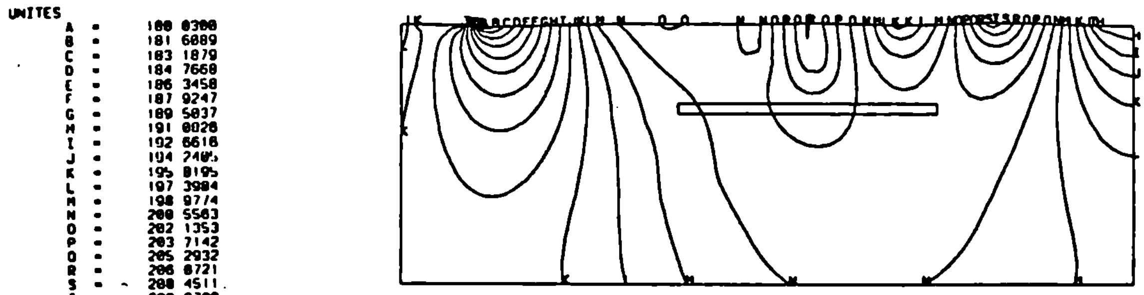

Figure 6 : Hydraulic heads at 1 000 years

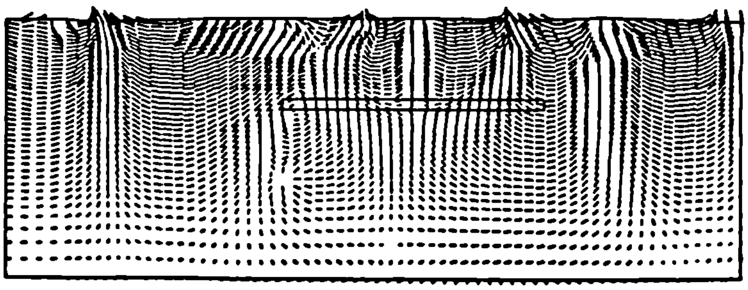

Figure 7 : Flow chart at 1 000 years

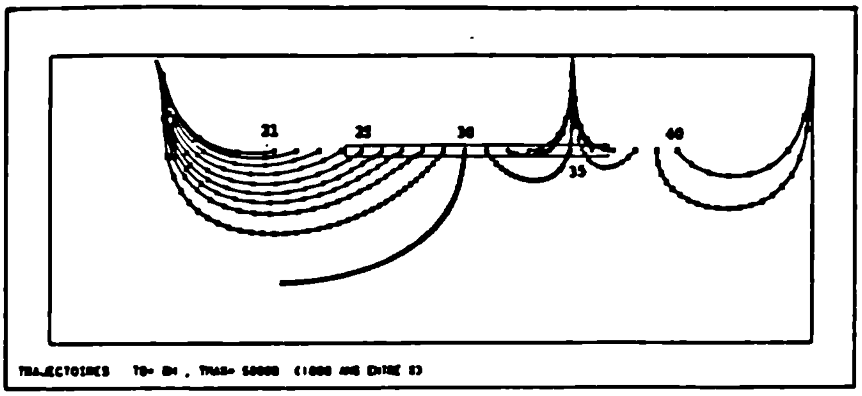

Figure 8 :
No thermal effects

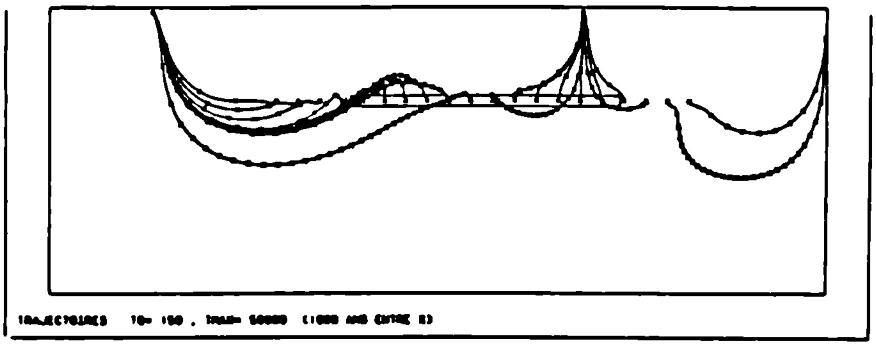

Figure 9 :
Thermal effects
T0 = 150 years

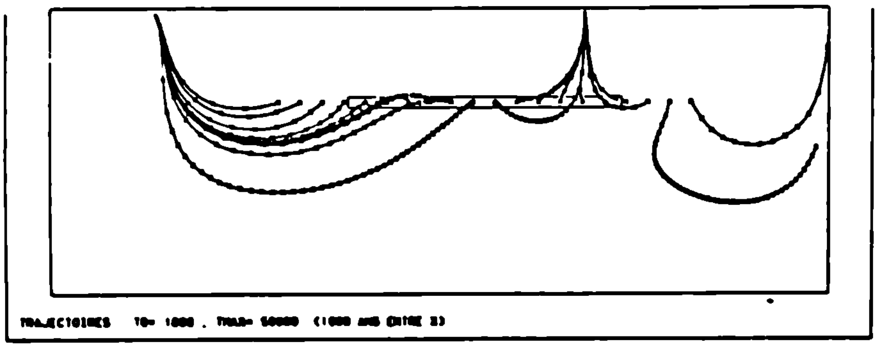

Figure 10 :
Thermal effects
T0 = 1 000 years

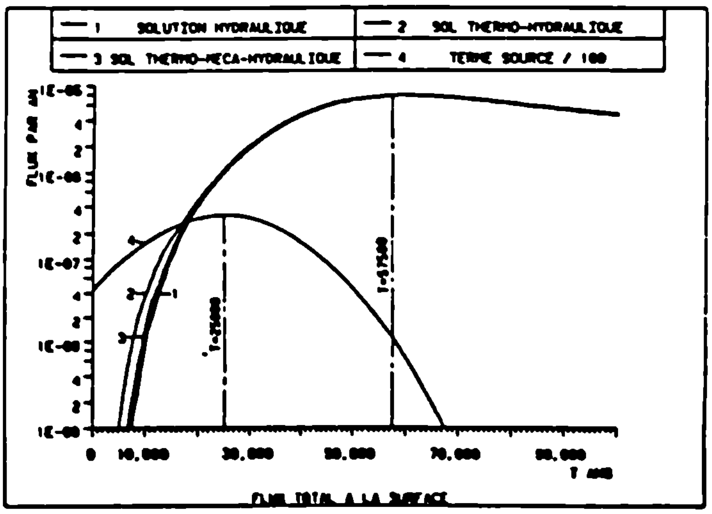

Figure 11 :
Activity at the geosphere surface and source term

TRANSFER MECHANISMS OF RADIONUCLIDES IN THE GEOSPHERE

Contractor : Centre d'Etudes Nucléaires de Fontenay-aux-Roses, CEA/IPSN/DAS/SAED - FRANCE

Contract : FI1W/0167

Duration of contract : October 1987 - March 1989

Period covered : January 1989 - March 1989

Project leader : A. CERNES

A. OBJECTIVES AND SCOPE

It is generally agreed that the main processes governing solute transport in the geosphere are : convection, hydrodynamic dispersion, molecular diffusion (Fick's law) and interaction with the rock matrix (adsorption - desorption). In most cases, when mathematical models are designed to take into account only these physical phenomena, or even just a few of them, they will give a true representation of reality.

In the science of non-equilibrium thermodynamics, which describes the evolution of the systems caused by thermodynamic forces, this method deals with non-diagonal couplings of the forces and fluxes. However, this simplification is only valid if one can be sure that the effects of the coupling is negligible. It is probable that the special conditions prevailing in the vicinity of radioactive waste, in particular the high temperature gradients, exclude the use of the simplified approach.

The objective of this study is therefore to investigate the effects of these physical phenomena and to express them mathematically as well as to evaluate the importance of their role in the aforementioned special conditions.

B. WORK PROGRAMME

1 - Theoretical research

This phase makes a review of the principal phenomena likely to play a role in the transfers occuring in natural media and to extract and adapt their mathematical expressions.

2 - Bibliographic study

The bibliography, as extensive as possible, is undertaken on these phenomena, in order mainly to have an idea of the scale of the parameters involved.

3 - Application

The results obtained will be applied to the radionuclides transfer in a geological medium, particularly to a porous medium. Calculations will be made (1D or 2D) in order to show the relative importance of the different transfers, especially regarding the thermal effect of the disposal.

C. PROGRESS OF WORK AND OBTAINED RESULTS

The two organizations conducting this study are the Paris School of Mines (Centre d'Informatique Géologique and Centre de Physique de l'Irréversibilité) and the Commissariat à l'Energie Atomique (IPSN/DAS).

While the effort during 1988 was devoted to the characterization of the potential influence of the Soret effect (cf. precedent progress report), which may be far from being negligible (cf. figure 1), the work in 1989 concerned the potential influence of the non dominant "electric" phenomena, due mainly to the presence of electrically loaded ions (like U^{+4}, UO_2^{++}, Pu^{+4}, Am^{+3}, $Th+^{4}$...) in the transported solution. These ions may be attracted or repulsed by the electric fields existing in the repository area.

Like for the other non dominant effects, a wide-ranging review of already published theories and articles was made. The formulation of the behavioral equations has been realized for the main categories of phenomena (telluric streams, electrokinetics, thermoelectrics, electrochemicals) but it was impossible to unify their description in a general model which should provide an integrated description of all these kind of phenomena and, in consequence, allow to assess the weight of each of them inside the global transport term, like what was made in 1988 for the Soret effect (cf. figure 1). It seems quite obvious that a much more substantial theoretical effort should be necessary in order to progress significantly in this way.

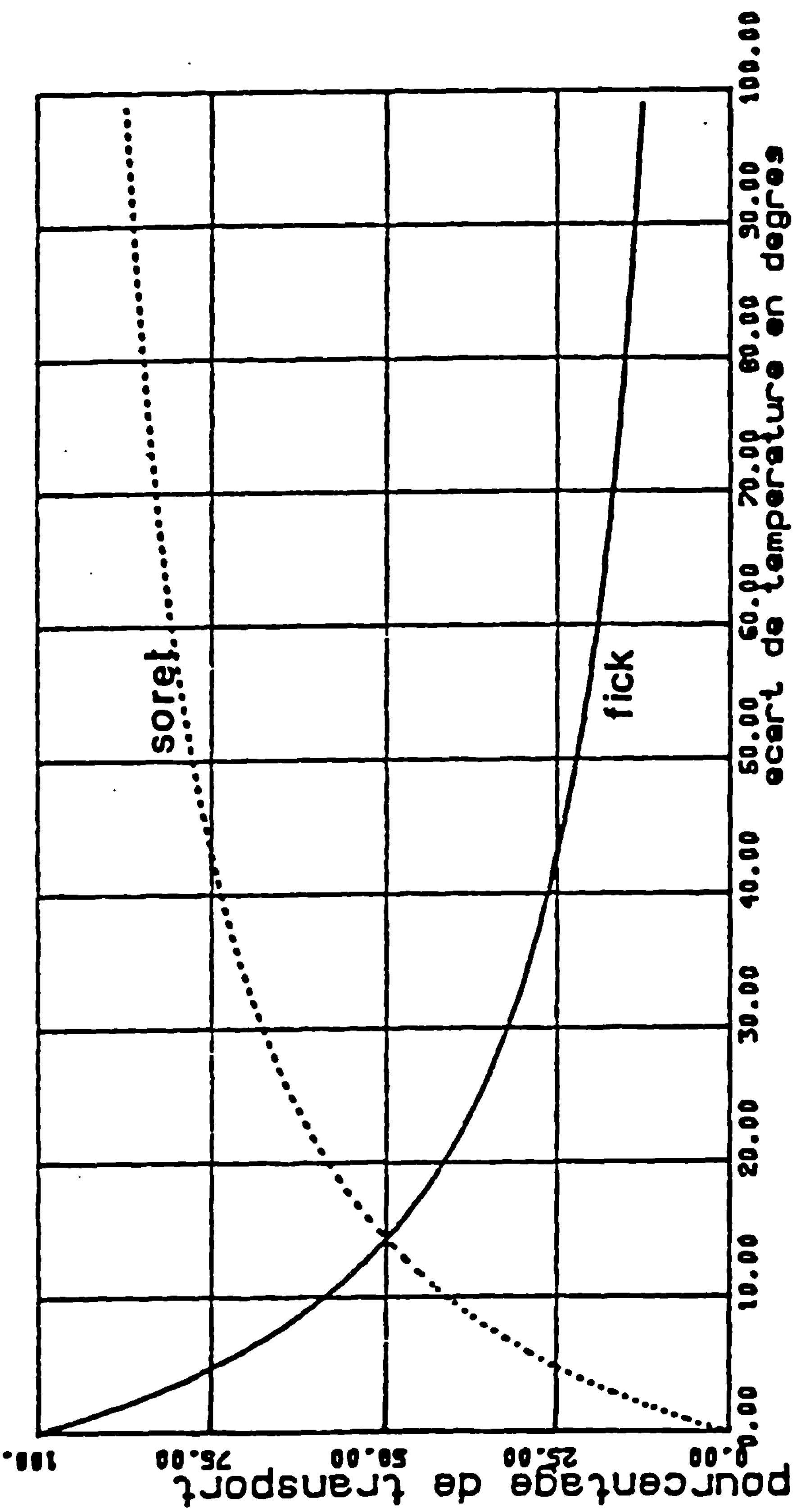

Fig. 1 - Comparison of the SORET and of the PICK (classical diffusion) effect

DEVELOPMENT OF THE CHEMTARD CODE

Contractor:	W.S. Atkins Engineering Sciences, Epsom, UK
Contract No:	FI1W/0232
Duration of Contract:	October 1988 - March 1990
Period Covered:	January 1989 - December 1989
Project Leaders:	D Read, S K Liew

A. OBJECTIVES AND SCOPE

In order to evaluate the long-term performance of a geological repository, predictions need to be made of the likely extent and rate of radionuclide transport following contact with groundwater. Several chemical transport simulators have been written for this purpose and during previous development work the Lawrence Berkeley code CHEMTRN was substantially modified and enhanced. The enhanced code is called CHEMTARD (Chemical Transport Adsorption Redox and Decay) and allows the treatment of aqueous complexation, transport by advection and/or diffusion, reversible precipitation-dissolution, radioactive decay and sorption by ion-exchange or surface complexation. It may be used either as a stand- alone coupled transport program, or as part of an integrated radiological assessment methodology.

As with other programs capable of simulating reactive chemical transport, numerical problems may be encountered with CHEMTARD owing to unfavourable combinations of parameter values. Such difficulties are compounded by the long run times required for all but the simplest simulations.

This contract aims to address the above in order that the code enter efficient routine use. The objectives are as follows:-

i) enhancement of numerical methods
ii) development and testing of component models
iii) improvement of code documentation.

B. WORK PROGRAMME

1. The modular structure of CHEMTARD is to be improved and a separate executive routine will be constructed. Means of increasing the efficiency of the existing Newton-Raphson procedure and reasons for convergence failure will be explored.

2. The functional scope of the code will be extended to cover transport by pure diffusion. Current methods of treating oxidation-reduction and mineral precipitation will be improved, the latter to permit supersaturation of solids.

3. Interactive software will be written to facilitate construction of input files together with a graphics package to display CHEMTARD output. Comprehensive documentation will accompany the developed code.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

Aspects 1 and 2 of the work programme, dealing with code modularisation and enhancements to the functional scope of CHEMTARD have been completed. New routines to allow differential diffusion rates for aqueous species and supersaturation of mineral solids have been added and shown to perform satisfactorily in tests to date. A menu-driven graphics package has been developed to display CHEMTARD output in the form of 2 or 3-D plots. Much of the code documentation has now been written and adheres to standards approved by an independent UK DoE-appointed Software Co-ordinator.

Progress and Results

1. Numerical Methods

i) Modularisation

As described in the previous report, CHEMTARD required major re-structuring in order to facilitate development of the numerical schemes. This modularisation is now complete. The most recent version of the code has one main calling routine, the CHEMTARD executive, which performs no numerical calculations but accesses independent subroutines for defined chemical, hydrologic or purely numerical functions. The new code structure is shown schematically in Figure 1.

ii) Efficiency and Stability

Convergence problems are frequently encountered in cases when steep changes in concentration profiles occur in temporal or physical space. Recent developments have enhanced the ability of CHEMTARD to resolve steep concentration gradients in time. Large concentration changes in space are more difficult to resolve mainly due to the need to fix the number, and the layout, of space nodes prior to dynamic calculations. To allow CHEMTARD to be run on a workstation size computer, a pragmatic approach has been adopted allowing the user to specify the layout of space nodes to cover the region(s) where large concentration gradients are judged most likely to occur. This approach has been found to work satisfactorily.

The Jacobian matrix solver in CHEMTARD has been assessed in detail with the aim of identifying redundancy and inefficiency in the software. As a result of this assessment, it was found that instability or divergence in the solutions could be trapped at an early stage of the calculations. A check has been incorporated in CHEMTARD to stop a test run where a residue of the Jacobian exceeds a given value.

2. Physical and Chemical Models

Two new features have been incorporated into CHEMTARD during the present phase of work:-

- differential diffusion of aqueous species
- supersaturation of solid phases

In the original version of CHEMTARD, a single "global" diffusion coefficient was applied to all aqueous components of the chemical system. It is well established, however, that, in the absence of significant advective flow, the rate at which individual species diffuse may vary substantially in response to a given concentration gradient. A new routine has been added to take account of this effect.

The simulation of mineral precipitation-dissolution during chemical transport is resource-intensive, owing to the need for re-equilibation at each time and space node. It is useful, therefore, for the user to know beforehand which minerals are likely to precipitate or dissolve as conditions change. For this reason, the precipitation routine in CHEMTARD has been amended to allow supersaturation of solids at any point within the time/space grid.

Finally, improvements have been made to the method used for treating oxidation - reduction reactions. Problems arise when either a reductant or oxidant precipitates and subsequently decays since this affects the mass balance of the electron. Numerical difficulties have been largely overcome but the conceptual validity of alternative approaches to coupling redox and decay is still under review.

3. Input/Output Facilities

The structure of the input data file has now been finalised with error traps introduced to identify potentially spurious values. Improvements made to both input and output format greatly facilitate information retrieval.

A graphics package has been developed and interfaced fully with the executive routine. Options include three-dimensional surface plots, contour plots and cross-sections in the X or Y plane. A menu-driven selection facility is available allowing the user choice of colours, annotation, scales and orientation.

4. Testing and Quality Assurance

All new software is being produced and documented according to guidelines laid down by the UK DoE Disposal Assessments Software Co-ordinator. The latter performs an independent quality assurance audit of the work at regular intervals.

Testing has been carried out in parallel with code development. At each stage of production, acceptance testing has been performed using a series of pre-defined cases covering almost all aspects of code operation. Additional verification has been provided via participation in the concurrent CHEMVAL/MIRAGE project (contract FI1W 0077 UK(H)). CHEMTARD was used in the formulation of coupled transport problems within CHEMVAL and comparisons have been made with similar programs employed elsewhere. The results of this work will be reported in Spring 1990, as part of the CHEMVAL series.

5. Documentation

The documents being produced under the current contract comprise a User's Guide, Programmer's Manual, Acceptance Test Plan, and an updated Technical Report. Of these, the User Guide has been written while both the Programmer's Manual and Acceptance Test Plan document are at an advanced stage.

Three papers describing CHEMTARD work have been presented at recent international conferences:-

1. S.K. Liew, C.S. Mawbey and D. Read. The Application and Testing of Chemical Transport Models used in Radiological Risk Assessment. IAEA/CEC/NEA Conference, Paris, October 1989.

2. S.K. Liew, A. Dawes and D. Read. The Application of a Coupled Chemical Transport Model in a Trial Assessment of Deep Disposal of Low and Intermediate Level Radioactive Waste. MRS, Boston, November 1989.

3. D. Read, P.J. Hooker, M. Ivanovich and A.E. Milodowski, A Natural Analogue Study of an Abandoned Uranium Mine in Cornwall England. Radiochim Acta (in press).

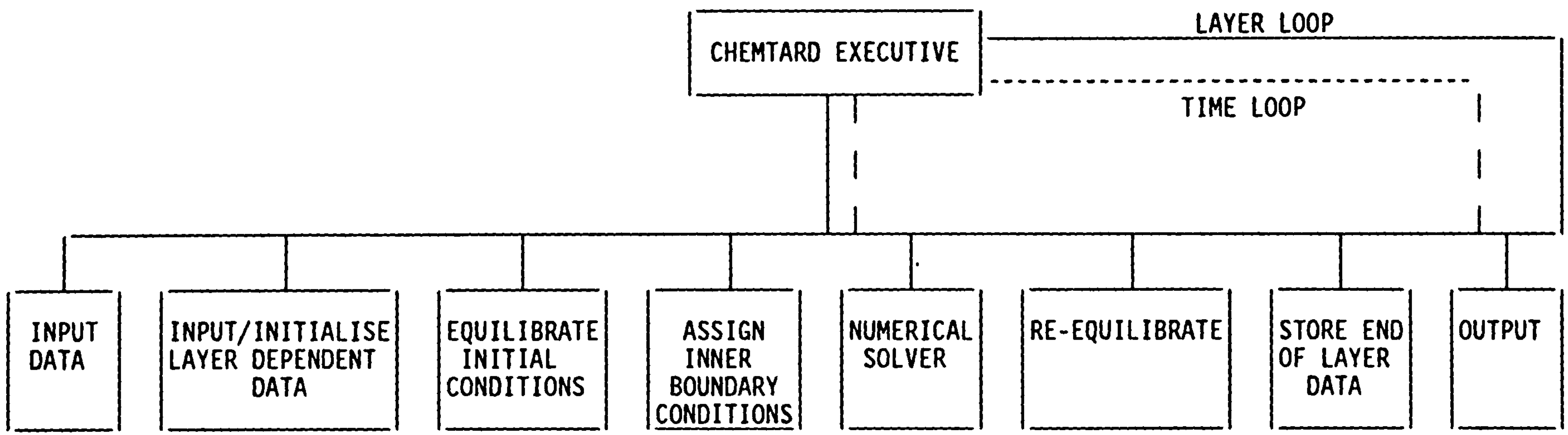

FIGURE 1 : SIMPLIFIED SOLUTION SEQUENCE FOR CHEMTARD

Investigation into the potential of fuzzy sets and related approaches for the treatment of uncertainty in migration models.

Contractor: Exploration Consultants Limited   Contract No: ETCC-0003
Duration of contract: from 1/11/88 to 31/8/89
Period covered: 1/11/88 to 31/8/89
Project Leader: Dr. William T. Shaw

A. OBJECTIVES AND SCOPE

A feature of many safety analyses is the presence of uncertainty. For those analyses involving a mathematical model, one standard method of treating such uncertainty is probabilistic risk assessment. Input to an analysis is represented probabilistically, it is propagated through a calculation according to the rules of probability, and the output is given in terms of a probabilistic measure such as an expectation value.

The aim of the project was to assess the suitability and feasibility of fuzzy set theory and related approaches as providing an alternative methodology for treating uncertainty. A set of criteria for the success of such an investigation is that fuzzy forms of the representation of inputs should exist, that there are fuzzy forms of arithmetic and calculus suitable for propagating uncertainties of the type encountered in safety analyses, and that there should be a means of representing uncertain output and making decisions involving such output. It is also highly desirable that should concepts should be capable of automation through a computer.

At the start of the project there were already existing ideas about the representation of input and output. The project had to clarify the application of these to the present context. Less well developed was the theory of fuzzy arithmetic and calculus, and these concepts were of central importance.

The project included a sub-contract to Dr. J.F. Baldwin and Dr. T.P. Martin of Bristol University, and Fril Systems Ltd.

B. WORK PROGRAMME

Task 1: Review of fuzzy sets and related approaches

1.1: General Framework: The aim is to elucidate those aspects of fuzzy set theory which may be relevant to the treatment of the types of uncertainty encountered in safety analysis.

1.2: Role of Expert Judgement: A potentially useful feature of fuzzy set theory is its ability to handle input based on human, and therefore possibly subjective and vague, judgement.

1.3: Mathematical and Computational Aspects: How does one represent uncertain parameters and variables in fuzzy form? How does fuzzy calculus work? Can one solve fuzzy differential equations? Can all of these approaches be handled on a computer?

1.4: Present Applications: The task is to survey present applications and to define possible links to ideas relevant to safety analyses.

Task 2: Application to a simplified problem

2.1: Specification

2.2: Solution

2.3: Discussion

The intention here is to define a problem or family of problems, related to radionuclide migration, and to compare the results of a probabilistic analysis with those of the corresponding fuzzy analysis.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

The contract was completed with all tasks accomplished. The highlights of the study included the selection and development of a methodology for defining and solving differential equations with fuzzy parameters and fuzzy initial values. In the course of this several existing systems of fuzzy calculus were investigated and only one /1/ was found to give a propagation of uncertainty in agreement with intuition. This approach was developed further.

A detailed comparison of fuzzy and probabilistic approaches to a simplified migration problem was carried out, based on the calculation of a transit time in a geological medium. The specific example chosen was very simplified, and was based on /2/. This example was able to illustrate the representation of uncertain parameters by fuzzy numbers, the manipulation of such quantities by fuzzy arithmetic, and the interpretation and use of fuzzy output. It also showed how one could perform fuzzy calculations by computer. The paradigm for the fuzzy analysis was a paper on rock slope stability /3/, in which the notion of a fuzzy stability factor was introduced. This notion is easily adapted to the notion of a "safety factor", rather unlike probabilistic measures, and based on the concept of "necessary support" for a quantitative fuzzy statement. It can also be described straightforwardly within a computing environment based on the Fuzzy Reasoning Inference Language FRIL /4/.

This study suggests that Fuzzy Set theory (sometimes called "possibility theory") provides a credible alternative to probabilistic risk assessment, and that further investigation, with a view to defining a detailed fuzzy safety assessment methodology, is highly desirable. The study found other existing applications of the theory /5/ which may also be of wider use.

Progress and Results

Task 1: Review of fuzzy sets and related approaches

1.1: General Framework.

The main result of this sub-task was to elucidate the nature of fuzzy set theory in a way which would be relevant to the Waste Management Community. Ordinary sets may be characterized by a "degree of membership" (DoM) which takes the values zero or one. It is a function on objects, and takes the value 1 if and only if an object is in the set. Fuzzy sets are obtained by allowing the DoM to attain values between zero and one. This concept is important since it admits the notion of a "fuzzy number". An example is illustrated by Figure 1. The set A is the set of numbers not differing significantly from three, and Figure 1 shows a plot of the DoM of numbers s plotted against s. For comparison, the DoM of an ordinary set of "Numbers exactly 3" is also shown.

It is important to realise that the shape shown is to be interpreted differently from a probability density function. It is best thought of as the "weight of evidence" that s is in the set. Such shapes could, for example, be defined by soliciting opinions from a group of experts. The DoM of s could be defined as the proportion of experts who agree that s is in the set. Such shapes are not normalised to unit area, as would be pdfs.

1.2: Role of Expert Judgement.

Where detailed experimental data is unavailable, it may be necessary to seek expert opinion as to the values of parameters, or the likelihoods of events. The resulting opinion may be no more precise than "the rock is very porous" or "the event is "rather improbable". It is possible to set up mappings between such statements and fuzzy numbers describing parameters or probabilities of events. In this way such input can be manipulated alongside other quantitative data.

1.3: Mathematical and Computational Aspects.

Some of this has been described in the overall statement of advancement. There exist systems for doing fuzzy arithmetic and calculus. However, not all yield results in accordance with intuition, and that of /1/ was selected out as being appropriate. An entirely different approach based on the propagation of membership functions down the trajectories of differential equations was developed. This was found to give results consistent with /1/, and had the further merit of being applicable to some types of partial differential equations.

The important outcome is that fuzzy uncertainty propagates through a calculation in a different way from probabilistic uncertainty. The rules for addition, subtraction, multiplication and division, as well as calculus, lead to a different mapping between input information and output information.

1.4: Present Applications.

Applications of fuzzy set theory have included Decision Making, Control, Learning, Diagnosis, AI and robots, Image Processing, Speech Recognition, Applied Operations Research, Economics, Sociology, Psychology and Linguistics. An application that may be of more specific interest is Reliability Analysis. Whereas mechanical equipment often fails according to probabilistic rules, the failure of systems involving humans is more difficult. One approach, as described in /6/, represents failure rates for humans as fuzzy numbers, and suggests that possible system failures such as the Chernobyl accident are best treated in fuzzy terms.

Task 2: Application to a simplified problem

2.1: Specification, 2.2: Solution, 2.3: Discussion

Several problems were considered, including the problem of fuzzy inventory evolution, and the solution of a fuzzy diffusion equation. For detailed consideration, a very simple problem was selected which had already been the subject of a SYVAC analysis, i.e. the probabilistic approach applied computationally. In the present study this was considered probabilistically, but analytically, and in fuzzy form, both analytically and computationally. Here the principle of the fuzzy calculation will be outlined. The problem considered was the calculation of a transit time T for groundwater flow in a sand medium. The arithmetical formula for the time T is given in terms of the hydraulic gradient I, the hydraulic conductivity K, the porosity P and the length of the medium L, by the formula: $T = L.P/(I.K)$.

In a probabilistic approach one calculates a pdf for T by suitable manipulations applied to the pdfs of L, P, I and K. In a fuzzy approach one represents these four parameters by fuzzy numbers. The simplest possible description is in terms of a triplet of values (lower bound, best estimate, upper bound), leading to a triangular fuzzy number. If the best estimate (which in fuzzy terms might be a value which many experts all agreed was a reasonable value for the parameters) extends over a range, a trapezoidal shape (such as are shown in Figure 2) may be obtained. Although the DoM might not necessarily be linear between these points where it starts or stops being zero or one, the behaviour of these particular points when fuzzy arithmetic is applied is quite straightforward. For example, if we consider the ratio P/K, the upper bound of this is just the upper bound of P divided by the lower bound of K, and such results come naturally from fuzzy arithmetic. The behaviour within arithmetic of intermediate points is more complicated, but for the sake of simplicity of presentation we may re-approximate the intermediate shapes by straight lines. (This was done within FRIL). The results of manipulations such as these is a fuzzy number, defined by a shape such as one of those shown in Figure 2. How do we use such a quantity? For simplicity, suppose that it is considered desirable that the transit time T should be bigger than some critical time T_{CRIT}. If we were doing a probabilistic calculation we would compare the expectation of T: <T> with T_{CRIT}, or give the confidence interval defined by T_{CRIT}. In a fuzzy calculation we do something rather different, and give values for the "Necessary and Possible Support" for the statement that $T > T_{CRIT}$. These numbers are represented by an ordered pair (n, p), and are shown graphically in Figure 2 for the various possible cases. Inspection of this Figure shows the nature of the necessary support for various cases. It may be interpreted in various ways, including as "our degree of certainty that $T > T_{CRIT}$." This degree of certainty has been computed by use of fuzzy arithmetic from input parameters represented as fuzzy numbers. These numbers may have been obtained from experiments, from expert opinion or a combination of both.

The results of this type of analysis suggest that it would be appropriate to consider a more substantial attempt at defining a fuzzy assessment methodology for treating uncertainties.

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Figure 1: An example of a Fuzzy Number

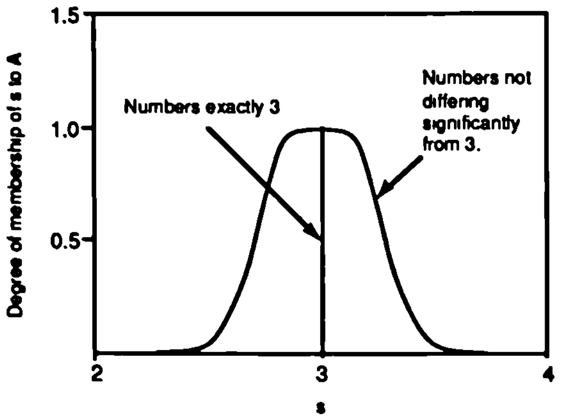
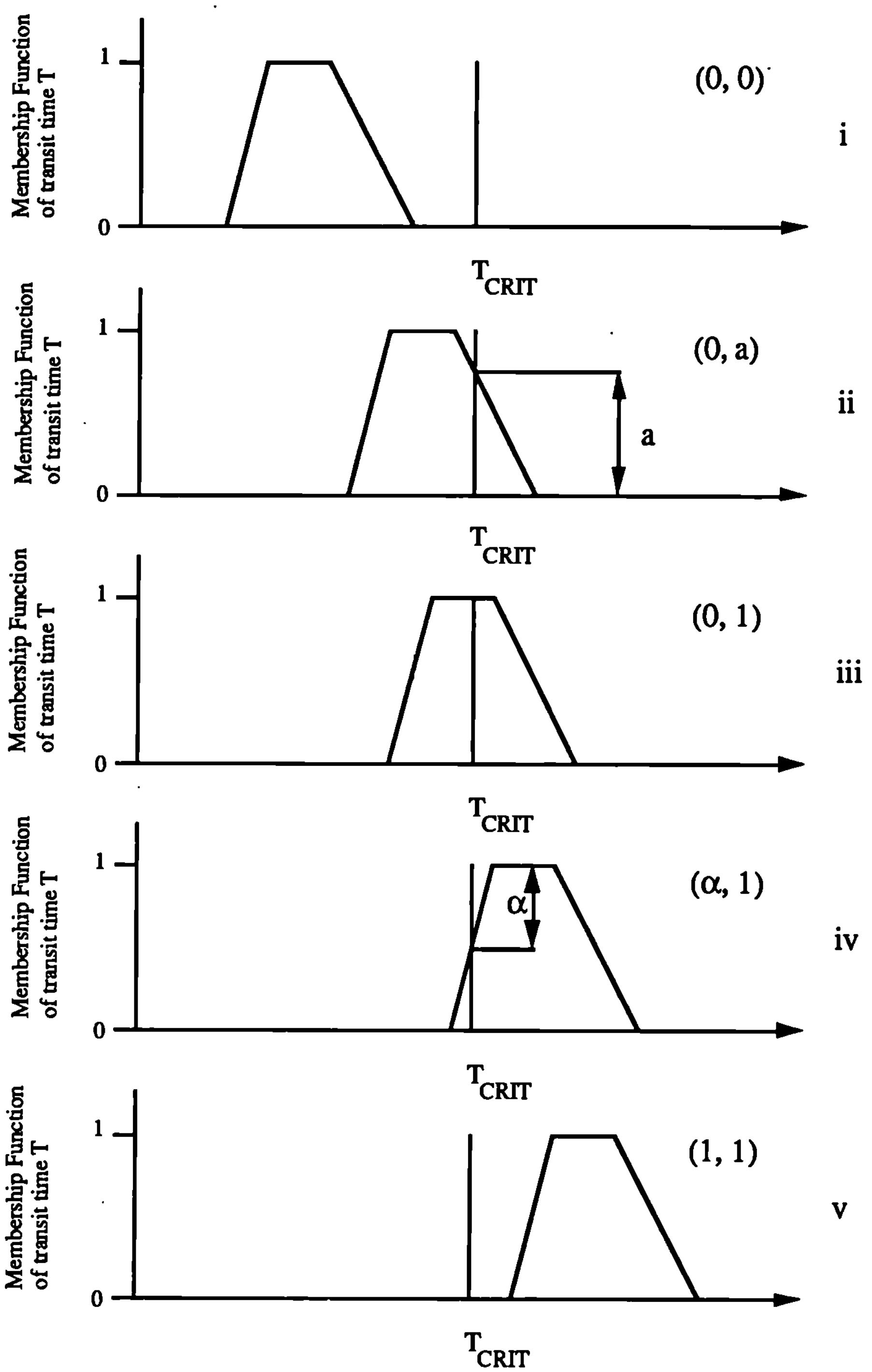


Figure 2: Linear Fuzzy Numbers, and the Definition of Support Pairs



Publication

Shaw, W.T. & Grindrod, P., Investigation of the Potential of Fuzzy Sets and Related Approaches for Treating Uncertainties in Radionuclide Transfer Predictions. Final Report on CEC Contract ETCC-0003. Preprint, to appear as CEC report no. 12499.

4.4. SHALLOW LAND BURIAL

IN-SITU STUDY OF THE EFFECT OF A PERMANENT ARTIFICIAL IMPERVIOUS SCREEN ON GROUNDWATER FLOW, IN A POROUS MEDIUM, IN ORDER TO IMPROVE THE SAFETY OF A RADIOACTIVE SHALLOW DISPOSAL

Contractor : COMMISSARIAT A L'ENERGIE ATOMIQUE
F-Fontenay-aux-Roses
Contract No. : FI 1W/0213
Duration of the contract : July 1988 - September 1989
Period covered : January 1989 - September 1989
Project leader : J. Cl. Gros

A. OBJECTIVES AND SCOPE

This technique should allow to improve shallow land burial quality by reducing velocities while increasing the groundwater transfer times without interrupting its flow.

Flow velocities and dispersion coefficients are modified by the screen.

Pollution is thus confined downstream in a zone where velocities have been considerably reduced.

The experimental device is made of (figure 1) :
- 1 injection well,
- 18 piezometers in a fan array, down flow from the injection well,
- 5 pumping wells about fourteen meters from the injection well.

The groundwater speed will be controlled and a parallel stream flow will be maintained in order to study the transverse dispersion.

A theoretical study has been carried out to determine the flow and the dimensions of the experimental device.

The grout diaphragm wall of cement-bentonit was realized by SOLETANCHE.

The theoretical study, the "in-situ" experiments and the calculations have been executed by the CEA/DERS/SERE of CEN/Cadarache (France).

B. WORK PROGRAMME

1. In-situ study of the effect of a permanent impervious screen on groundwater flow.
1.1. Dimensions of the screen.
1.2. Selection of the impervious "setting slurry".
1.3. Character of grout.
2. Hydrodispersive studies of the aquifer ground in disturbed flow by the impervious screen.
2.1. Checking for leaks of the screen and studies of the flows around and from of the screen.
2.2. Hydrodispersive studies properly speaking.
2.2.1. Experiments with tracer.
2.2.2. Calculation of longitudinal and transversal dispersion coefficients.
2.2.3. Determination of the geometry of the radioactive confined area from the impervious screen.

2.2.4. Evaluation of the velocity decrease and radioactive concentration decrease from the screen.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The dispersion study of the pollutant was executed with a radioactive tracer (I 131). The experiments have begun on January 1989.

The groundwater speed is controlled with a system of 5 pumping wells, and a parallel stream flow was maintained during the experiment.

An instantaneous punctual injection of I 131, (injection time ~ 5 to 10 s), has been carried out in a piezometer, from of the impervious screen.

Tracer concentrations were determined as a function of time and depth, with a γ probe in piezometers.

A calibration curve of the γ probe was determined for different concentrations of I 131 and for a same volume of ground.

Progress and results

1. Hydrodispersive studies of the aquifer ground in disturbed flow by the impervious screen.

1.1. Checking for leaks of the screen and studies of the flows around and from of the screen.
A tracer injection in piezometer F1 has proved the imperviousness of the screen.

1.2. Hydrodispersive studies properly speaking.

1.2.1. Experiments with tracer.
Two injection experiments have been carried out in piezometer P5 for two different velocities of the groundwater.

1.2.2. Calculation of longitudinal and transversal dispersion coefficients.
In this homogeneous porous medium, three mediums were found with different hydrodynamic parameters and different dispersive coefficients.

1.2.3. Determination of the geometry of the radioactive confined area from of the impervious screen.
The experiments have confirmed the presence of a radioactive confined area from of the screen, between piezometers P5 and P14, where the hydraulic gradient has decreased with a factor of 5.

1.2.4. Evaluation of the velocity decrease and radioactive concentration decrease from of the screen.
- Pollutant groundwater transfer times have increased with transfer tines in ratio of 2.
- Concentrations decrease with a factor of 10 after putting of the screen.
- Theoretical and experimental curves of concentrations as a function of time are represented in figures 2 and 3.

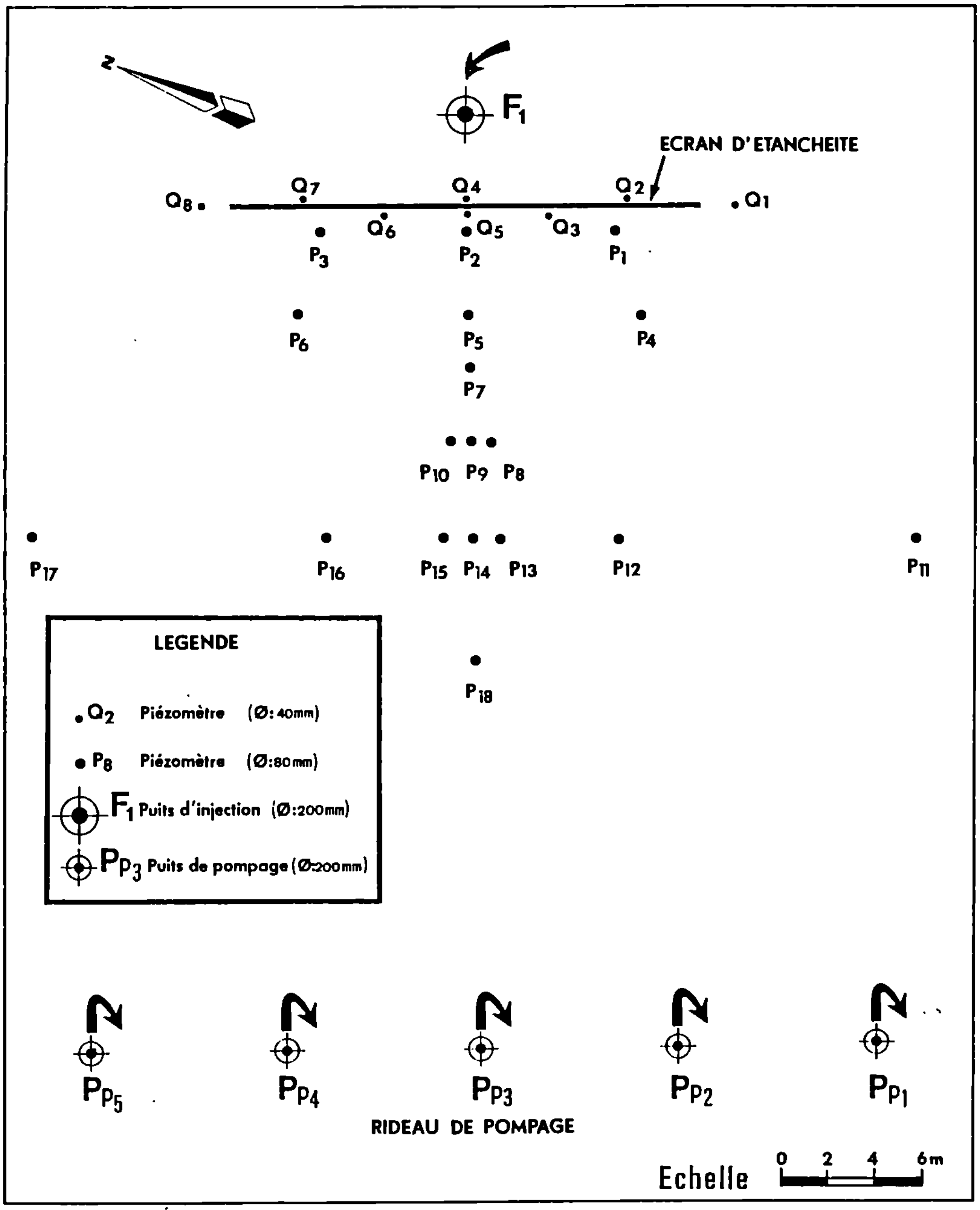


Figure 1 : Plan of the experimental site

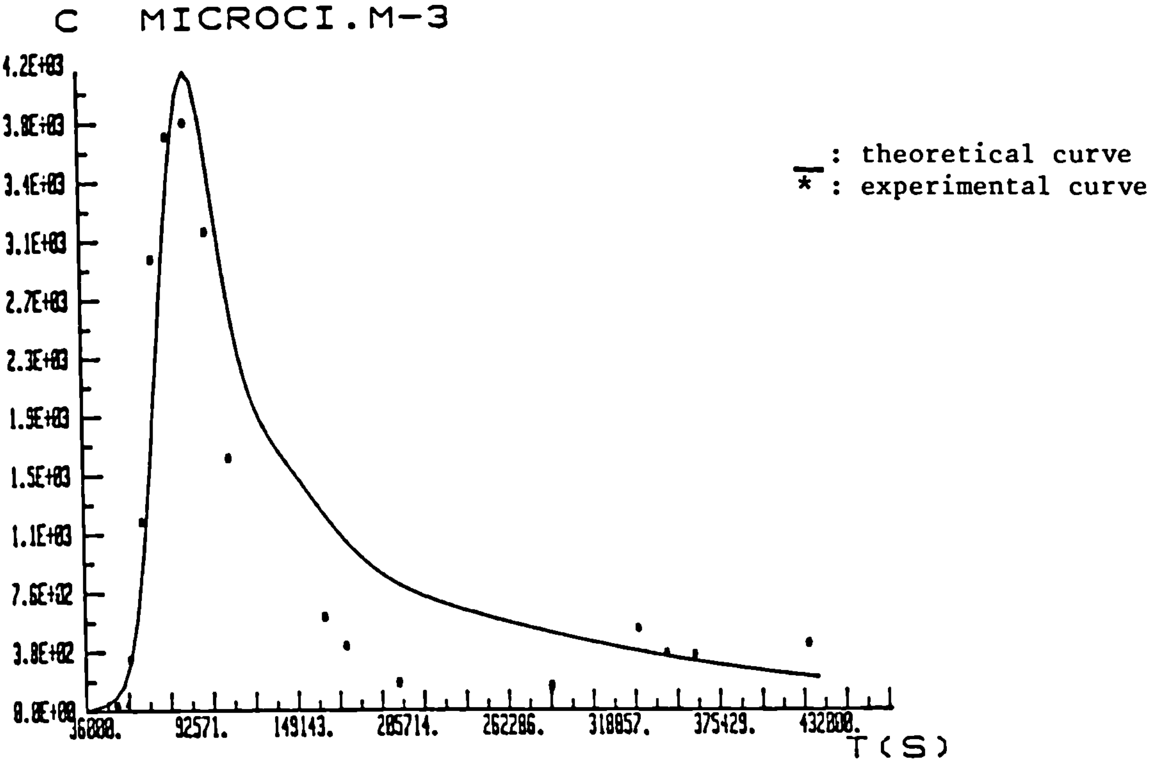


Figure 2 : Theoretical and experimental concentrations-time curves for piezometer P7 - Experiment with a high hydraulic gradient (1989)

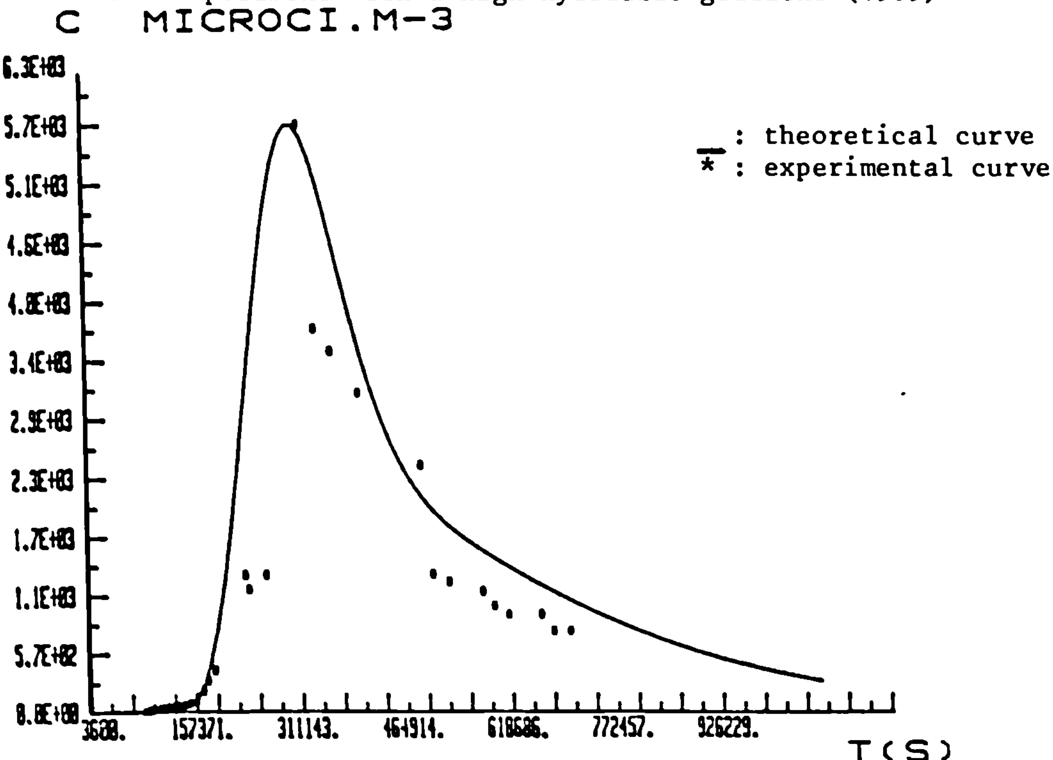


Figure 3 : Theoretical and experimental concentrations-time curves for piezometer P7 - Experiment with a low hydraulic gradient (1989)

TITLE: A Large Scale Laboratory Investigation into Gas and Water Permeability of Clay Barriers Exposed to the Environment.

Contractor: Taylor Woodrow Construction Ltd
Contract No: FI 1W/0215
Duration of Contract: from July 1988 to June 1990
Period Covered: 1989
Project Leader: T.P. Lees

A. OBJECTIVES AND SCOPE

To investigate the hydraulic and gaseous transmission properties of compacted clay caps constructed to probable site specifications on a laboratory scale and subjected to an accelerated weathering regime.

B. WORK PROGRAMME

1. Literature survey of recent work relevant to the proposal. Material Selection and Characterisation.
2. Design and Construction of the test beds. Preparation and compaction of clay samples.
3. Subject test beds to environmental conditioning. Commence periodic sampling and testing programme.
4. Analyse results of testing programme and identify change in physico chemical properties with depth due to environmental conditioning programme.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

Two clay types were chosen:

Two test beds were designed and constructed using heavy gauge steel plate to withstand the heavy stress applied during compaction.

The clays were bought and transported to the laboratory. Each clay was compacted into its test bed on top of a 250mm layer of coarse sand with geotextile material interviewing. The depth of compacted clay is 300-450mm thick.

Each clay has been subjected to three heating cycles to simulate direct sunlight but with a limitation on maximum temperature reached by the clay of 40°C. Each heating cycle has been followed by a wetting cycle to simulate rainfall.

Progress and Results

1. The two clays used were: a London Clay of high plasticity obtained from RAF Chattenden (Kent, England) and a Glacial Till of low plasticity obtained from RAF Cowden (Humberside, England). Both sites are well documented. The material used was taken from 1.5m or more below existing ground level so that the effect of weathering is minimum.

2. Two test beds were constructed from heavy gauge steel plate. Coarse concreting sand was compacted into the base of each test bed so that it was level with the internal partition. Two layers of a geotextile (TERAM 1000) were laid on the sand and the clays compacted on this to give a compacted layer 300-450mm thick.

   The clays were compacted in layers about 150mm thick using a double vibratory roller. This was not effective at the edges of the beds and these were compacted using a vibratory tamper. In general each layer was given 3-4 passes of the roller, it was not possible to use compaction which would encourage mixing of the layers (eg "sheep foot" or the like). The clay was used at the moisture content obtained at the time of excavation but some wetting of the layers was done to assist compaction.

   The Glacial Till material contained stones of up to 40mm size and it is expected that the presence of these may make measurement of permeability of samples difficult. However these stones were not removed during compaction.

3. Environmental canopies to fit on the test beds have been constructed. They use arrays of 150 watt light bulbs to raise the temperature of the test bed to 40°C $\pm$ 1°C. An accelerated heating cycle is 7 days at this controlled temperature. 40°C was chosen as a temperature which may on occasion be reached in UK and yet is not sufficient to cause mineralogical breakdown of the clays.

   The canopies are removed and a wetting cycle is the application of the equivalent of 7mm rain/day on each of 7 days. The 7mm rainfall is applied over a 3-4 hour period and represents an average heavy rain fall.

   Each clay bed has had 3 heating and 3 wetting cycles.

   Samples have been taken of the compacted clays before weathering. The falling head method of water permeability has been found to be unsatisfactory. It has given acceptable results for the plastic clay but the presence of stones in the low plasticity Glacial Till has made it impossible to seal the specimens in the measuring cell by normal methods. Alternative seals are being considered.

   However falling head permeametry is unsatisfacory for the research programme since it requires saturation of specimens for testing and this is not possible when the effects of weathering are being considered.

4. There has been no progress under this heading during the reporting period.

OPTIMIZATION STUDY ON THE CONCEPT OF CONCRETE STRUCTURES IN SURFACE OR SUB-SURFACE STORAGE FACILITIES WITH RESPECT TO THEIR LONG TERM INTEGRITY

Contractor: ENRESA, Spain
Contract No.: FI1W-0216-E
Duration of Contract: December 88 - April 90
Project Leader: Pablo Zuloaga

A. OBJECTIVES AND SCOPE

The main objective of this research project is to define certain parameters dealing with the design and construction of special parts of surface or sub-surface storage facilities, in order to maintain their long-term integrity, under extreme conditions, based on the characteristics of a specific site (El Cabril).

Analytical and experimental studies will be carried out on the properties of the structures, as a function of various deteriorating effects, such as water attack, temperature cycles and mechanical actions.

The research work is being done in co-operation with the French ANDRA, the INSTITUTO DE LA CONSTRUCCION EDUARDO TORROJA (ICCET) acting as a subcontractor of ENRESA.

B. WORK PROGRAMME

1. Analysis of the interaction of the water with the concrete.

2. Modelling and structural calculations.

3. Optimization study.

C. PROGRESS OF WORK AND OBTAINED RESULTS

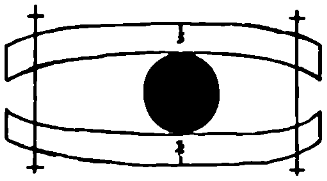
The first phase was directed to the preselection and characterization of the different componentes of the concrete to be used in the aging tests and to the fissuration modelling to be carried out in the following steps. Data and results obtained are presented in Tables 1 to 8.

The second phase is devoted to analyze the performance of cracked reinforced concrete, for which two kind of tests are being carried out.

1. Cracked beams subjected to permanent humidity.

2. Concrete prisms were cracked by means of a fatigue test up to the 80% of the maximum load.

For the first type of experiment, beams of 8x9x190 cm were fabricated with two types of cements: OPC (Ordinary Portland Cement) and FAC (Fly Ash Cement). The proportion of cement was of 400 Kg. per cubic meter. A central reinforcing bar of 10 mm in diameter was embedded with two covers of 2 and 4 cm. A set of beams was fabricated with galvanized rebars and cured during 28 days.



Cracks of ~ 0.1 mm. and ~ 0.5 mm. were introduced by loading two parallel beams as shown in the Figure.

When cracked, the beams were maintained in a dry environment during two months, in order to allow the cracks to be carbonated. When the beams were 3 months old, they were introduced in tanks with water up to 2 cm, in order to have the beams partially inmersed in the water.

The corrosion potential, Ecorr, of the rebars is being monitored every week and the corrosion intensity in the region of the crack was measured when the beams were 6 months old. Results collected up to now do not show any significant corrosion.

For the second experiment, 6 concrete prisms of 15x15x58 cm in size were also fabricated with the type of cement selected to be used (Atlántico). Carbonation tests were carried out in order to compare this cement with the other one in study, the results showing that carbonation progress and permeability to air were much lower in the selected one. A central bar of 20 mm. in diameter was embedded and the prisms were cured during 28 days.

After this period of curing, two prisms were held as blank test and the other four were subjected to the fatigue test. Two of these "fatigued" (cracked) are being subjected to 4 hour cycles, submerged in water at 50°C and 6 days drying with a heating fan.

The corrosion potential of the bars are being monitored and up to now, no signs of corrosion were noticed.

Table 1. Determination on solid samples

MUESTRAS	CLORUROS	SULFUROS	TRIOXIDO DE AZUFRE
ARENA	0,00	0,00	0,025
GRAVA	0,00	0,00	0,065

Table 2. Determination on underground and surface water samples

MUESTRA	CLORUROS	OXIDO DE CALCIO	TRIOXIDO DE AZUFRE	pH
SG-11bis	35,5	1,6	61,5	12,6
SG-16 bis	28,4	336,0	6,33	12.3
SG-4 bis	24,1	13,6	2.5	7,5
PRESA	45,4	23,2	20,16	9,5

Table 3. Potential reactivity of aggregates

MUESTRAS	SILICE SOLUBLE MILIMOLES/LITRO	REDUCCION DE LA ALCALINIDAD EN MILIMOLES/LITRO	REACTIVIDAD POTENCIAL
ARENA	15.984	2,71	No presenta
GRAVA	11.988	0,00	No presenta

Table 4 - Granulometric analysis of aggregates

ANALISIS GRANULOMETRICO DE ARIDOS																COMPOSICION DE ARIDOS PARA HORMIGON		
Tamices Luz de mallas	0-4			6-12			12-30											
	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene	Pasa	Curva Total
M M	g	%	%	g	%	%	g	%	%	g	%	%	g	%	%	%	%	Pasa %
									100									
32								1	99									100
25								5	94							1		99
16						100		58	36							16		83
12,5					13	87		23	13							10		73
8					43	44		13								16		57
6,3			100		16	28										5		52
4		5	95		28											11		41
2		16	79													8		33
1		24	55													10		23
0,5		34	21													4		9
0,25		14	7													6		3
0,125		6	1													3		
0,075		1																
Resto																		
Suma		100			100			100			100			100		100	100	

Table 5 - Granulometric analysis of aggregates

ANALISIS GRANULOMETRICO DE ARIDOS																COMPOSICION DE ARIDOS PARA HORMIGON		
Tamices Luz de mallas	0-4 Mach.			0-4 Natural			6-12			12-20								
	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene		Pasa	Retiene	Pasa	Curva Total
M M	g	%	%	g	%	%	g	%	%	g	%	%	g	%	%	%	%	Pasa %
25												100						100
16											58	42					16	84
12,5									100		30	12					8	76
8								54	46		12						20	56
6,3			100			100		20	26								6	50
4		1	99		5	95		22	4								8	42
2		28	71		16	79		4									10	32
1		21	50		24	55											10	22
0,5		14	36		34	21											11	11
0,25		7	29		14	7											5	6
0,125		9	20		6	1											3	3
0,075		8	12		1												1	2
Resto		12															2	
Suma		100			100			100			100			100		100	100	

Table 6. Chemical characteristics of cement

DETERMINACIONES EN TANTO POR CIENTO	CEMENTO		
	ATLANTICO	ASLAND	ASLAND
Pérdida de peso a 105°	0,30	0,19	0.19*
Pérdia al fuego	3,17	1,71	1.71
Residuo insoluble	1,89	26,00	0.12
Dióxido de Silicio	18,86	13,95	29.95
Trióxido de Aluminio	3,82	4,43	11.57
Trióxido de Hierro	3,88	2,22	3.38
Oxido de Calcio	63,32	45,42	46.27
Oxido de Magnesio	1,24	1,35	1.66
Trioxido de Azufre	2,91	3,43	3.18

Table 7. Physico-mechanical properties of cement

ENSAYO	CEMENTO					
	ATLANTICO			ASLAND		
Peso específico	3,14			2,78		
Finura de molido	Rdo 900-0,04%			Rdo 900-0,17%		
	" 4900-0,35%			" 4900-2,12%		
Agua de consistencia normal	126cc.(25,2%)			134cc (26,8%)		
Tiempo de Fraguado						
Principio de Fraguado	2h. 40 min.			2h. 45 min.		
Final de Fraguado	4h. 40 min.			4h. 35 min.		
Resistencia mecánica	3	7	28	3	7	28
Flexión (Kg/cm2)	33	52	74	34	45	65
Compresión (Kg/cm2)	167	264	432	172	259	389

Table 8. Mechanical strength of concrete

HORMIGON	Cono de Abrams	R7	R28
H-I	8	203	275
H-II	6	215	265
H-III	6	234	349
H-IV.a	6	279	350
H-IV.b	6	246	309

T A S K N o 5

SAFETY OF GEOLOGICAL DISPOSAL

TASK 5: SAFETY OF GEOLOGICAL DISPOSAL

A. Objective

Assessment of the performance of isolation systems for radioactive waste and of the corresponding radiological impact.

B. Research performed under the 1980-1984 programme

Initiation and implementation of the first phase of the PAGIS (Performance Assessment of Geological Isolation Systems) project, the purpose of which is to assess the capacities offered by the various geological disposal options (salt, clay, granite and marine sediments) for the containment of high level radioactive waste.

C. 1985-1989 Programme

a) - The PAGIS Information Day (Madrid, 30 June 1989) was organized in order to present the results of the project and discuss them with personalities from the political, scientific, industrial and ecological fields, as well as, representatives from IAEA, NEA/OECD and information media.

- An International Symposium on "Safety assessment of radwaste repositories" was jointly organized (CEC - IAEA - OECD/NEA, Paris, October 1989) to summarize the state-of-the-art and discuss future developments in this field.

The studies carried out under contract concern :

a) Continuation and completion of the PACOMA project (Performance Assessment of COnfinements for Medium-Level or Alpha bearing waste) : Safety assessment of disposal systems for alpha contaminated radioactive waste and for medium-level waste buried into geological formations (clay, granite and salt); evaluation of the corresponding radiological impact.

b) Support activities to the evaluation of safety of geological disposal of radioactive waste.

P A C O M A PROJECT

Title : Performance evaluation of confinement for alpha waste repository in granite formations (Pacoma project).
Contractor : Commissariat à l'Energie Atomique
31-33 rue de la Fédération F75752 PARIS
Contract n° : FI1W/0040-F
Duration : 1/04/87 - 30/06/89
Period covered : from 01/01/89 to 30/06/89
Project leader : J. LEWI

A - OBJECTIVES AND SCOPE

The general objective is the safety assessment of a deep repository in granitic formations for alpha wastes.
This work has consisted in a first time in the definition of a detailed waste inventory for alpha bearing waste and in the adaptation to the α case of the source-term model CONDIMENT developped for the PAGIS project (vitrified wastes).

B - WORK PROGRAMM

B.1.1 Basic data : a new waste inventory has to be provided. The sites are the same as in Pagis (Auriat, Barfleur, national Uk site). The repository design has to be defined for alpha-bearing wastes.
B.1.2 The methodology will be similar to what was done for the PAGIS project (deterministic, sensitivity and uncertainty analysis).
B.1.3 Model and calculations tools : as far as possible, the same as for PAGIS ; a new version of the source model (CONDIMENT) has to be realized.
B.1.4 Calculations : similar to what has been done for the PAGIS project.

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The PACOMA project is over since the 30/06/1989. Calculations have been performed for the following fission products : Se79, Tc99, Zr93, Pd107, I129, Sn126 and Cs135 and for the four radioactive actinide chains. We have studied the sensitivity of the results to the main parameters of source and geosphere and we have performed a study of the sealing shaft defect scenario.

Results

The main results of deterministic calculation, for the three sites are given in table 1.

In all cases, for normal scenario, we have found there is no significant release of radioactivity to the exutories before 100 000 years ; the maxima are reached after 10^5 years for iodine 129 and after 3.10^6 years for the actinides. The values of dose rates are more that 3 orders of magnitude lower than the 10^{-3} Sv/y ICPR limit. The most relevant radionuclides are I-129 for the 1 000 000 first years of repository and Np237 and Th229 after. The other fission products take part for less than 1 % to the total dose rate (figure 1).

The 2D sensitivity analysis shows that the most influent parameters on the Np237 calculations to the total dose rate are : the permeability of the geosphere, the retardation factor in granite, the solubility limit (for low soluble radionuclide as Np237).

The altered scenario calculations (access shaft sealing defect) show that the permeability increase causes a modification of local flow but that this modification is limited and the perburbation flow due to this altered scenario does not influence the behavior of radionuclides in the geological barrier.

D - REFERENCES

- C. BRUN-YABA, A. CERNES (CEA/IPSN), J.P. MANGIN (CEA/IRDI), P. GOBLET (ENSMP)
  Evaluation des performances des dépôts pour l'isolation des déchets α enfouis dans les formations géologiques profondes - Projet PACOMA, option granite
  International symposium on the safety assessment of radioactive waste repositories, CEC-IAEA-OECD/NEA-CEA, Paris (France) 9-13 octobre 1989

- J. LEWI, A. CERNES, M.J. MEJON-GOULA, C. BRUN-YABA
  Etude de sensibilité et évaluation des incertitudes affectant les calculs découlant des déchets associés à un stockage de déchets nucléaires en formation géologique profonde
  International symposium on the safety assessment of radioactive waste repositories, CEC-IAEA-OECD/NEA-CEA, Paris (France) 9-13 octobre 1989

CONCEPT DE STOCKAGE A

	AURIAT Les pallands	BARFLEUR Le Ruisseau	Site Britannique avec couverture (Ex2)	Site Britannique sans couverture (Ex2)
Maximum de l'équivalent de dose individuelle (Sv/an) totale	$2{,}02 \times 10^{-8}$	$2{,}39 \times 10^{-7}$	$1{,}76 \times 10^{-8}$	$1{,}58 \times 10^{-7}$
Date d'occurence (années)	5 000 000	7 000 000	4 600 000	3 370 000
Principaux radionucléides	Np 237 Th 229 U 233	Th 229 Np 237 U 233	Np 237 Th 229 U 233	Np 237 Th 229 U 233
Contribution radionucléides (%)	59 28 8	87 9 1	57 35 8	61 31 7

CONCEPT DE STOCKAGE B

Maximum de l'équivalent de dose individuelle (Sv/an) totale	$1{,}96 \times 10^{-8}$	$2{,}40 \times 10^{-7}$	$2{,}12 \times 10^{-8}$	$1{,}88 \times 10^{-7}$
Date d'occurence (années)	4 250 000	7 100 000	4 670 000	3 170 000
Principaux radionucléides	Np 237 Th 229 U 233	Th 229 Np 237 U 233	Np 237 Th 229 U 233	Np 237 Th 229 U 233
Contribution radionucléides (%)	59 30 8	90 9 1	57 35 8	63 29 7

Tableau 1 - PRINCIPAUX RESULTATS CONCERNANT LES CALCULS "DETERMINISTES" DU SCENARIO D'EVOLUTION NORMALE (FACTEURS DE DOSE CORRESPONDANT A LA CIPR 48)

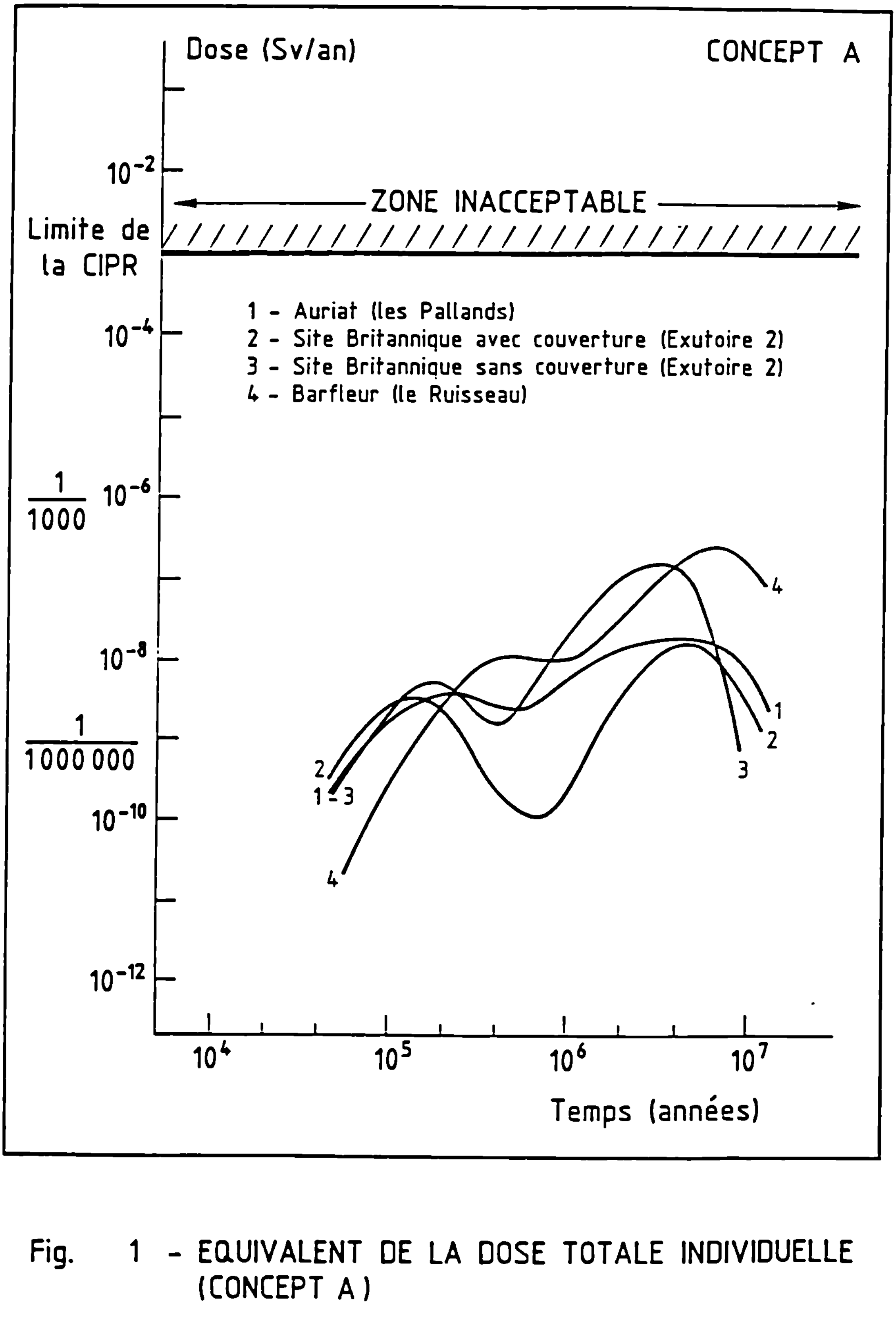


Fig. 1 - EQUIVALENT DE LA DOSE TOTALE INDIVIDUELLE (CONCEPT A)

Performance Assessment of Confinements for MLW and Alpha Waste (PACOMA Project) Clay option

Contractor: NRPB, Chilton, UK
Contract No.: FI1W/0041-UK(H)
Duration of contract: from October 1986 to December 1989
Period covered: January 1989 - December 1989
Project Leader: Ms M D Hill

A. OBJECTIVES AND SCOPE

The overall objectives of the UK contribution to the PACOMA Project are to develop and demonstrate procedures for assessing the radiological impact of disposal of intermediate level waste in a deep repository located in a clay formation. The hypothetical repository considered is assumed to be at Harwell in Oxfordshire. The research is co-ordinated by the UK Department of Environment and is being carried out by four organisations:

National Radiological Protection Board (NRPB)
Theoretical Physics Division, UKAEA Harwell Laboratory
Electrowatt Engineering Ltd
CAP Scientific Ltd

The NRPB work is in two phases. The objectives in Phase I are to establish a detailed methodology for the assessment, to collect data for biosphere modelling and to carry out preliminary calculations. In Phase II the aim is to carry out the assessment, using information provided by other UK contractors and in consultation with other participants in PACOMA, particular CEN/SCK.

B. WORK PROGRAMME

B.1 Adaptation of PAGIS methodology for use in the assessment of intermediate level waste disposal, identification of radionuclide release scenarios.

B.2 Review of available biosphere data, and preliminary calculations for typical releases.

B.3 Detailed planning of calculations to be carried out in the full assessment, finalising biosphere data and assumptions for each scenario.

B.4 Best estimates of doses and risks to individuals and populations for each scenario.

B.5 Sensitivity and uncertainty calculations.

B.6 Co-ordination of joint report by the four UK contractors.

C. PROGRESS OF WORK AND OBTAINED RESULTS

STATE OF ADVANCEMENT

The project has been completed and a draft version of the final report has been sent to CEC for comment. Calculations of the radiological impact of disposal of intermediate level waste in a deep repository located in a clay formation were performed for a number of scenarios using the methodology adopted for the PAGIS project. This involved best estimate calculations, using the models and data from Electrowatt Engineering Ltd, UKAEA Harwell and NRPB together with uncertainty and sensitivity analyses performed using NRPB models. Data for the geosphere were collected by CAP Scientific Ltd and they also compared the geosphere models used for the best estimate analysis and the uncertainty analysis.

Progress and Results

B.1 Assessment methodology and release scenarios

The PAGIS methodology was adopted as the basis for the PACOMA project hence best estimate calculations of peak individual doses and risks, and of integrated collective doses, were performed for several scenarios. These were supplemented by uncertainty and sensitivity analysis for the most probable scenario. The scenarios considered related to different assumptions for the biosphere and were the normal scenario, intrusion by drilling an exploratory borehole, intrusion by abstracting water from a well and two future climate states, a periglacial biosphere and a "greenhouse" biosphere.

B.2 Review of biosphere data and preliminary calculations

This task was completed in the previous reporting period.

B.3 Planning and finalisation of data and assumptions

The only aspect of this task that was not completed in the previous reporting period was the finalisation of the data for the best estimate calculations for the "greenhouse" biosphere. This has now been completed.

B.4 Best estimates of doses and risks to individuals and populations for each scenario

Best estimates of the doses and risks to individuals and populations were calculated for the scenarios described in B.1. The results for the normal scenario were obtained by running the NRPB biosphere model BIOS using the flux histories generated by UKAEA. The results are summarised in Table I. The peak individual dose from the normal scenario is $2\ 10^{-5}$ Sv a^{-1}. The results for the intrusion scenarios were obtained by running the NRPB models BOREHOLE and WELLS and using concentration histories in the geosphere generated by UKAEA. The risks (see Table I) were obtained by considering the probability of intrusion occurring as well as the probability of the dose giving rise to a health effect. The results for the two future climate states were obtained by running modified versions of BIOS and using the best estimate flux histories generated by UKAEA.

One interesting aspect of the project is the existence of two essentially different pathways from the repository to the biosphere. These were termed the upper and lower pathways respectively. In the former, the radionuclides diffuse upwards against a very small groundwater velocity and enter the chalk aquifer overlying the clay formation in which the repository is located. In the latter, the radionuclides migrate downwards towards the underlying aquifer, continue along this aquifer and finally upwell into the biosphere. Although the peak doses from the upper pathway arise earlier than from the lower pathway, it is the lower pathway that gives rise to the higher peak individual doses and integrated population doses.

The predicted peak individual risks are all below the ICRP limit of 10^{-5} a^{-1}. The integrated collective dose from the normal scenario is zero for the first 10^4 years and rises to 3 10^5 man Sv after 10^8 years. For perspective the collective dose delivered to the UK population as a result of natural background radiation is 10^5 man Sv every year.

B.5 Sensitivity and uncertainty analyses

Two types of sensitivity analyses were performed: single parameter variations using the best estimate biosphere model BIOS and simultaneous variations of many parameters using the uncertainty analysis models and database. The former type, termed local sensitivity analysis, showed that the results were sensitive to the proportion of the release that entered the deep soil or the river water and to the assumptions used for future climate states. Variations of about a factor of 1000 were seen. Variations in groundwater velocity and soil k_d caused smaller changes : typically a factor of three. In the latter type, termed global sensitivity analysis, a total of 33 parameters were varied for each nuclide and these parameters were chosen from the near-field, far-field and biosphere models. Overall, the most sensitive parameters were found to be the soil k_d of ^{135}Cs, the groundwater velocity in clay and the delay before leaching starts.

The uncertainty analysis was performed using a simple near-field model which was fitted to the best estimate model, the geosphere model TROUGH-1D and the biosphere model MINIBIOS. MINIBIOS is a simplified version of BIOS developed for use in the PACOMA uncertainty analysis. Ranges and distribution functions were derived for 33 parameters for each of the 6 radionuclides and the analysis was performed by running a total of 1000 cases using a latin hypercube sampling scheme. The resulting distributions of individual doses at each time were characterised by their means and various percentiles and these are summarised in Table II. The fluxes from the geosphere for each of these 1000 cases were also provided to CAP Scientific for their model intercomparison work.

B.6 Co-ordination of joint report by the four UK contractors

Regular meetings were held with all the UK contractors to discuss the format and content of the final report. A draft version of the complete final report has been produced.

Table I Summary of best estimate results

Scenario	Peak individual dose ($Sv\ a^{-1}$)	Peak individual risk (a^{-1})	Time of peak (a)	Integrated collective dose (man Sv) at 10^4 years	10^8 years
Normal scenario	$2\ 10^{-5}$	$3\ 10^{-7}$*	$3\ 10^{6}$	0	$3\ 10^{5}$
Intrusion by borehole after 300 years (close inspection of core)	$1\ 10^{-1}$	$9\ 10^{-7}$	$3\ 10^{2}$	-	-
Intrusion by abstraction of well water	$8\ 10^{-5}$	$9\ 10^{-8}$	$3\ 10^{5}$	0	$8\ 10^{2}$
Greenhouse climate	$2\ 10^{-5}$	$3\ 10^{-7}$*	$3\ 10^{6}$	0	$3\ 10^{5}$
Periglacial climate	$7\ 10^{-6}$	$1\ 10^{-7}$*	$3\ 10^{6}$	0	$3\ 10^{7}$

*Assuming a probability of occurrance of this scenario of unity

Table II Summary of uncertainty analysis results - individual dose ($Sv\ a^{-1}$)

Time	Mean	50th percentile	95th percentile
10^2	$2\ 10^{-8}$	$2\ 10^{-12}$	$8\ 10^{-9}$
10^3	$6\ 10^{-8}$	$5\ 10^{-11}$	$6\ 10^{-8}$
10^4	$2\ 10^{-6}$	$1\ 10^{-8}$	$6\ 10^{-6}$
10^5	$9\ 10^{-7}$	$1\ 10^{-8}$	$4\ 10^{-6}$
10^6	$9\ 10^{-6}$	$3\ 10^{-7}$	$4\ 10^{-5}$
10^7	$1\ 10^{-6}$	$1\ 10^{-7}$	$5\ 10^{-6}$
10^8	$2\ 10^{-9}$	$1\ 10^{-14}$	$4\ 10^{-9}$

ACQUISITION OF SUBJECTIVE DATA FOR USE IN MODELS FOR

WASTE SITE ASSESSMENTS (PACOMA PROJECT)

Contractor : PRINCIPIA MECHANICA LTD

Contract No : FI1W/0042-UK

Duration of contract : August 1986 to June 1989

Working Period : January 1989 - June 1989

Project Leader : J M Bealby

A. Objectives and Scope

In the modelling of radionuclide movement from a repository, a compromise must be achieved between accuracy and cost. For probabilistic site assessment using Monte Carlo simulation, one dimensional models are used due to their small computational cost whereas for deterministic modelling more detailed two- and three-dimensional models are used. The objectives of this programme of work are to develop and demonstrate consistent data acquisition and preparation techniques for probabilistic site assessment and for detailed deterministic modelling of radionuclide movement from a repository for intermediate level waste under the Harwell site. A comparison of radionuclide risks derived from both assessment procedures will be made and any inconsistencies in the input data and resulting inconsistencies in the risk estimates will be investigated.

B. Work Programme

B.1 Research Programme: development of methodology for comparing probabilistic site assessment codes and detailed deterministic models; planning for the data acquisition exercise.

B.2 Data Acquisition: the use of expert opinion to acquire data for 5 to 10 parameters.

B.3 Model Comparison: comparison of input data and assessment procedures.

C. Programme of Work and Results Obtained

Summary

The model comparison exercise was completed in April - June 1989.

The results of the NAMMU 2D deterministic code which was run by UKAEA Harwell, were compared with the results from the TROUGH 1D code, performed by NRPB.

A report describing the work, has been issued [3].

Progress and Results

B.1 Research Programme

Task B1 is complete and a report on the work has been issued /1/.

B.2 Data Acquisition

Task B2 is complete and a report on the work has been issued /2/.

B.3 Model Comparison

Task B.3 is complete and a report on the work has been issued /3/, (sub-reports 2 & 3).

The objectives of the model comparison work, were to compare the results of a detailed model with those of a simpler probabilistic risk assessment (PRA) code. One approach to meet these objectives is to compare results from a simpler 1D model with those from a detailed 2D model when both models are run in probabilistic mode. However in the PRA approach, a compromise must be achieved between accuracy and cost. Although greater accuracy may be achieved by using 2D or 3D models rather than 1D models, excessive computer time is required to conduct a full PRA on higher dimensional models.

In PACOMA, the results of the NAMMU 2D deterministic code which was run by UKAEA Harwell, were compared with the results from the TROUGH 1D code, performed by NRPB. Firstly TROUGH was run in deterministic mode with the same input data as NAMMU and the results from both models compared. This was followed by a comparison of the 1D model results using best estimate inputs against results when input values were sampled (i.e. in probabilistic mode). Finally, a number of sensitivity studies were performed using the 1D model in order to verify the reasons thought to account for differences in the results from both models. It was found that the best estimate results from the two models agree reasonably well for I_{129} when the input data is adjusted to be used in a consistent way. Recommendations for future work have been made (/3/, Report 3 Section 10.5).

References

/1/ Laundy, R.S. Data requirements for a comparison of one and two dimensional models using consistent data acquisition procedures. CAP Scientific draft report 3409/TR.1 February 1987.

/2/ Dalrymple, G.J. and Phillips, L.D. Using a structured approach to the acquisition of probabilistic data from expert opinion. CAP Scientific draft report. 3409/TR.2 July 1987.

/3/ Bealby, J. 'Acquisition of Subjective Data for Use in Models for Waste Site Assessments (PACOMA Project)' Final Report PM1008/TR-STH/40.

PERFORMANCE ASSESSMENT STUDIES FOR MEDIUM LEVEL WASTE DISPOSAL AT HARWELL SITE (PACOMA PROJECT)

Contractor: Harwell Laboratory, United Kingdom

Contract No: FI1W/0043 - UK (H)

Duration of Contract: December 1986 - June 1989

Period covered: January 1989 - June 1989

Project leader: Dr K.H. Winters

A. OBJECTIVES AND SCOPE

We shall simulate the transport to the surface of radionuclides released from a hypothetical repository in a clay layer beneath Harwell Laboratory. The rates of release from such a repository, for various waste inventories, have been calculated by Electrowatt Engineering. CAP Scientific have derived probability distributions for the values of parameters involved in the relevant groundwater-flow and solute-transport calculations, by structured questioning of groups of experts. Radiological risks due to the migration of radionuclides from the repository will be calculated by the National Radiological Protection Board. The final stage of the project involves a comparison by CAP Scientific of radiological risks derived by probabilistic risk assessment and from deterministic simulation. This analysis should identify any significant differences in the approaches to safety assessment adopted by UK Nirex Ltd. and by the United Kingdom Department of the Environment.

B. WORK PROGRAMME

1. Review , and if necessary revise, existing calculations of groundwater-flow in the vicinity of the Harwell site, so as to establish numerical accuracy and to examine sensitivity to modelling assumptions.

2. Carry out radionuclide-transport calculations and determine how results depend upon uncertainties in the modelling assumptions and in the values of physical parameters.

3. Assist CAP Scientific Ltd. to compare safety assessments based upon probabilistic and deterministic simulations.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of Advancement

We have completed the work programme described in section B. In particular we have carried out during the period of this report two-dimensional calculations of radionuclide transport from a hypothetical repository located in the Clay layer beneath the Harwell site, using as a source term the rate of release derived by Electrowatt Engineering from the VERMIN near-field model. The flux of nuclide through the water table was predicted for ten significant nuclides, including two nuclide chains. These computed fluxes were provided to NRPB as input to their biosphere model BIOS.

A local sensitivity analysis was carried out for the values of two key parameters, the permeability and the nuclide sorption. The nuclide fluxes at the surface showed a significant sensitivity to these two parameters, whose variation altered both the transit time and the distribution of nuclide between the two major pathways from the repository to the surface. The final report is in preparation.

Progress and Results

We have computed using the finite-element code NAMMU the transport of radionuclide through the far field, taking as nuclide source terms the release rates derived by Electrowatt from the near-field VERMIN model. These release rates were supplied mostly as values of the magnitude of the flux out of the near field at specific times, for each nuclide in the PACOMA inventory. In NAMMU the source flux is defined by a piecewise-constant function, constant for the duration of a time step and discontinuous at the beginning and end of each step. We ensured that the total flux released up to a given time, as predicted by VERMIN, was identical to the total flux input to NAMMU. The transport equations in NAMMU take into account the effects of advection, diffusion, dispersion, sorption and radioactive decay on the nuclide migration through the geosphere.

The same three-stratum model described in the previous progress reports was used for the far-field calculation and consists of a vertical two-dimensional section through the Chalk, Clay and Corallian layers. The calculations reveal two principal transport pathways through the geosphere to the surface: (i) diffusion upwards through the layer of Clay above the repository into the Chalk stratum, and (ii) advection downwards through the Clay into the Corallian stratum, emerging at the surface where the Clay outcrops. Extensive verification was carried out of the numerical methods for deriving the source fluxes from the supplied data; for computing the time-dependence of the nuclide concentration in the geosphere; and for the spatial integration of nuclide flux along the line of the water table. The simulation of the transport of a single nuclide typically involved the solution of 4352 simultaneous equations at each time, and calculations with up to 30 time steps were performed.

Figure 1 shows typical results for the variation with time of the fluxes through the water table, in units of Becquerels per annum. Each flux is shown as two separate contributions corresponding to the two pathways leading to release through the surface Chalk and the surface Clay. The results highlight the disparity in behaviour between different nuclides, arising from the wide range in the values of the half life and sorption coefficients pertaining to the different nuclides. We also note the long times required for the fluxes to attain their peak values, due to the particular disposition and thicknesses of the strata and the particular direction of groundwater flow for this site.

A local sensitivity analysis was carried out in which the values of the permeability and sorption were changed by an order of magnitude from their best-estimate values. Altering the permeability of the Clay layer had a pronounced effect on both the travel time to the surface and the magnitude of the surface fluxes for each pathway. Altering the values of the sorption coefficients in the three layers had a similar pronounced effect.

The main conclusions of this study are:

1. Accurate groundwater flow calculations require a grid that will resolve abrupt changes in gradients of pressure head.

2. The migration of nuclides from the repository to the surface occurs along multiple pathways.

3. Two-dimensional calculations of radionuclide transport can be predicted for a wide range of nuclides using NAMMU, provided attention is paid to the appropriate choice of time step to resolve the differing time scales associated with sorption, decay and leaching.

4. The nuclide fluxes emerging at the water table show a significant sensitivity to the adopted values of sorption and permeability.

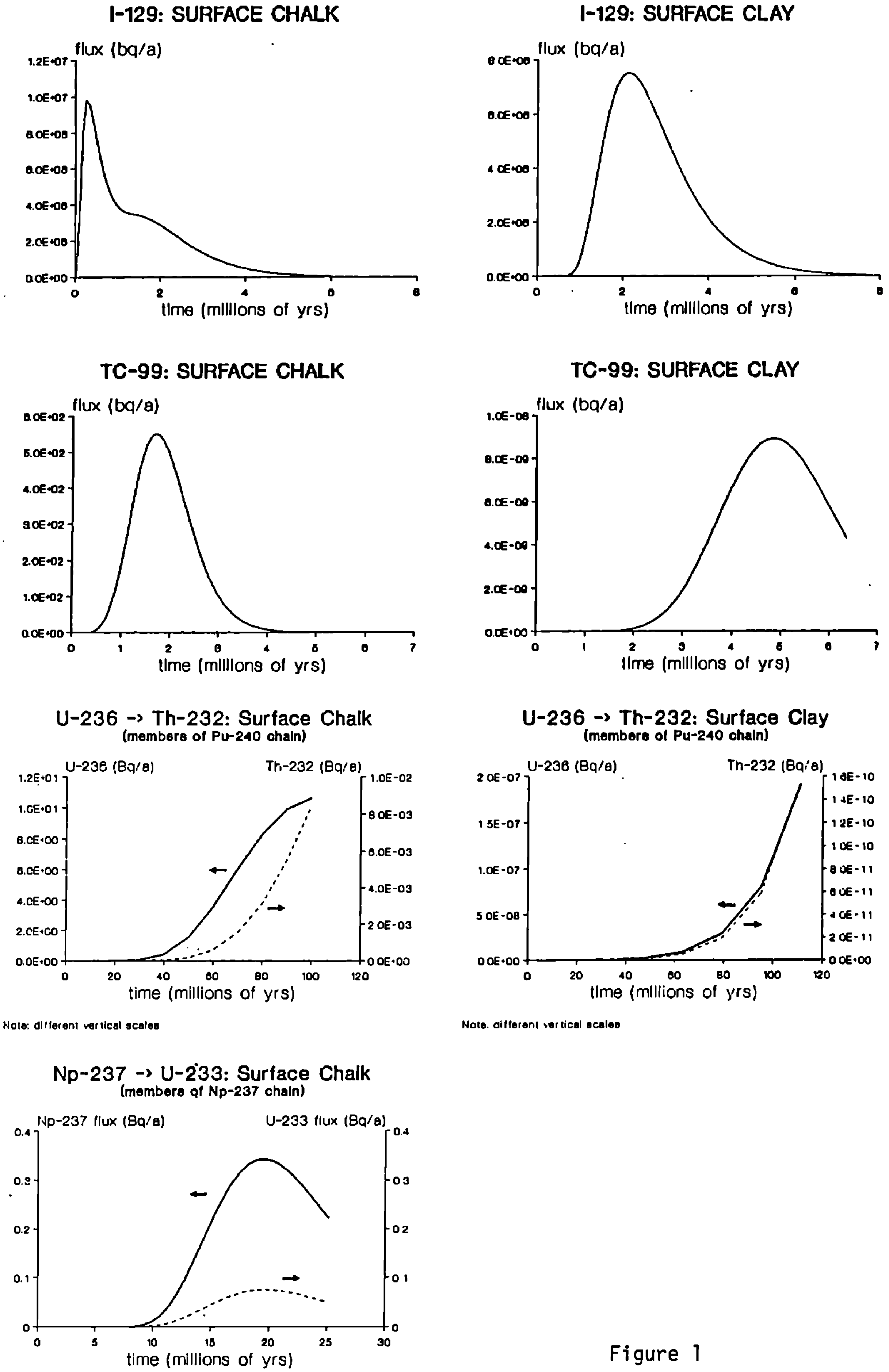

I-129: SURFACE CHALK
flux (bq/a)
time (millions of yrs)
I-129: SURFACE CLAY
flux (bq/a)
time (millions of yrs)
TC-99: SURFACE CHALK
flux (bq/a)
time (millions of yrs)
TC-99: SURFACE CLAY
flux (bq/a)
time (millions of yrs)
U-236 -> Th-232: Surface Chalk
(members of Pu-240 chain)
U-236 (Bq/a)
Th-232 (Bq/a)
time (millions of yrs)
Note: different vertical scales
U-236 -> Th-232: Surface Clay
(members of Pu-240 chain)
U-236 (Bq/a)
Th-232 (Bq/a)
time (millions of yrs)
Note. different vertical scales
Np-237 -> U-233: Surface Chalk
(members of Np-237 chain)
Np-237 flux (Bq/a)
U-233 flux (Bq/a)
time (millions of yrs)

Figure 1

Safety Evaluation of Geological Disposal Concepts for Low and Medium Level Wastes in Rock Salt - PACOMA Project

Contractor:	GSF - IFT Braunschweig, FRG
Contract No.:	FI1W/0044-D
Duration of Contract:	April 1987 - December 1989
Period covered:	January 1989 - December 1989
Project leader:	R. Storck

A. OBJECTIVES AND SCOPE

Tho overal objectives of the GSF contribution to the PACOMA project are to develop and demonstrate procedures for the radiological safety assessment of a deep repository for α-bearing and medium-level radioactive waste in salt rocks.

The research covers the disposal in a repository mined in a salt dome. The reference repository design will be taken as a basis for the calculations. Design variants and alternative disposal concepts are investigated. The results are given in terms of release rates to geosphere and in terms of doses to the individuals and populations.

B. WORK PROGRAMME

B.1. Disposal site
B.2. Repository design
B.3. Scenarios
B.4. Models
B.5. Calculations

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The final repository design was established taking the plans of the national authorities as a guideline and reducing the repository to LLW and MLW disposal locations for the reference case.

The scenarios are the same as for PAGIS, i.e. a normal evolution scenario and the two altered evolution scenarios, the accidental brine intrusion and the human intrusion.

For the calculation with best estimate values of parameters the PACOMA reference inventory and the German variant have been used. For comparison with results obtained by ECN, the Netherlands, the PACOMA reference inventory but without heat producing waste is used. Local and global sensitivity studies were performed on the reference repository with the PACOMA reference inventory only.

Progress and results

B.1. Disposal site

In PACOMA only one disposal site is considered which is identical to the reference disposal site of the salt option used in PAGIS. All geological and geochemical data are taken over with no changes, supplemented by sorption data for the additional elements C, Cl, and Ca. For a parameter variation study Kd-values of the Dutch site have been used.

B.2. Repository design

As a guideline for the repository design the repository of the PSE study is used /1/, disregarding the HLW wing. The size of the residual repository then is reduced according to the smaller number of MLW canisters compared to PSE which are disposed in the PACOMA case.

Design variants are additional dams and sealings on the chamber wing of the repository and variants due to the different disposal concepts which have been investigated. These alternative concepts are disposal of Iodine filters into boreholes and/or disposal of LLW-α into boreholes. Additionally, a repository including boleholes with HLW has been investigated as a variant. For comparison with results obtained by ECN a repository variant with no HLW and MLW boreholes is studied.

B.3. Scenarios

A normal evolution scenario and two altered evolution scenarios are investigated. The normal evolution scenario is the diapirism of the salt dome with subrosion at the top of the salt dome. The same model assumptions are used as for PAGIS. The altered evolution scenarios which have been considered are the following two scenarios:

- the combined intrusion scenario with a brine intrusion via the main anhydrite plus intrusion from undetected brine pockets into MLW boreholes,
- the human intrusion scenario, where a storage cavern is leached in a former repository area. The diameter of a cavern is approximately 50 m. Since the distance between MLW boreholes is 15 m (57 m between HLW boreholes in the PAGIS exercise) the canisters of about 4 to 9 boreholes may be laid open.

B.4. Models

All models used for PACOMA have been established during 1988.

B.5. Calculations

Comprehensive final best estimate calculations and local sensitivity studies have been performed during the first half of 1989. Parameters selected for local sensitivity studies are

- the time of brine intrusion,
- the volumes of brine pockets,
- the convergence rate,
- the permeability of dams,
- the solubility limits of Uranium,
- the Kd-values.

With the best estimate values of parameters there is a release of radionuclides only from those borehole where an intrusion from brine pockets has taken place. Although a much greater part of the chamber wing of the repository is inundated with brine compared to the borehole wing, there is no release of radionuclides from chambers.

Compared to results of preliminary calculations, given in the first topical report, the dose rate of I-129 is drastically reduced, since most of the Iodine is disposed of into chambers.

Similar to PAGIS the release of radionuclides is from those boreholes only, which are very close to the central field and have contact with brine pockets. Since most of the MLW Cs- and Np-nuclides are in feed sludges and cladding wastes, which are disposed of into boreholes, the results are comparable to those of the PAGIS study. The Cs-135 and Np-237 inventory is about two orders of magnitude smaller than that of a HLW-borehole in PAGIS, the same factor occurs comparing the dose rate maxima of the PACOMA and the PAGIS results.

A draft of the final report covering waste description and radionuclide inventory, sketch of the disposal concept, site selection and repository design, scenario description, input data and modelling procedures and results of the deterministic calculation has been written. Part of this report has been distributed as topical report.

In order to complete the final calculations for the PACOMA project a meeting was held in November at ECN in Petten, the Netherlands, for comparing the performance assessments and the results obtained by ECN and RIVM, the Netherlands, and by GSF on the other hand. The discussion was about discrepancies of the release rates from the repositories. The different solubility limits used for Uranium could be identified to be responsible for the different results. In addition, discrepancies were found for individual doses, whose reasons are the different pathways into the biosphere, i.e. the well on one side and the river on the other side.

References

/1/ STORCK, R., et al.: PSE Abschlußbericht, Vol. 16, Projektleitung HMI, Berlin 1985

SAFETY EVALUATION OF GEOLOGICAL DISPOSAL CONCEPTS FOR LOW AND MEDIUM LEVEL WASTES IN ROCK SALT (PACOMA-PROJECT)

Contractor : ECN, Petten, The Netherlands
Contract No. : FI1W/0045-NL
Duration of contract : April 1987 - June 1989
Period covered : January 1989 - June 1989
Project leader : J. Prij

A. OBJECTIVES AND SCOPE

The research covers the disposal of all types of low and medium level waste in salt formations. The aim of the study is the comparison of the safety of different repository designs and disposal sites. The methodology applied is the same as used in PAGIS. The work will be performed in cooperation with GSF in Braunschweig (FRG). A part of the work will be executed by the RIVM in Bilthoven (NL).

B. WORKPROGRAMME

1. Basic data

Data will be collected of all types of waste, including the radio nuclide inventories and the mobilization and transport models. Also the data defining the geosphere and biosphere will be collected.

2. Disposal site

Three types of generic formation will be studied. A salt dome with the top at a depth of 230 m; a small salt pillow at a depth of 800 m and a salt layer at a depth of 1200 m.

3. Repository design

Three types of design will be considered. Mined rooms and two types of solution mined caverns. In the first type of cavern the waste is dumped in the cavern which is still filled with brine while in the second type the brine is removed before the waste is disposed off.

4. Scenario's

The main scenario is of the water intrusion / extrusion type. Some attention will be paid to a normal evolution scenario, some alternative altered evolution scenario's and scenario's for human intrusion.

5. Models

In principle, the computer models to be used will be the same as used as for PAGIS. Where it is appropriate GSF and ECN will use the same models.

6. Calculations

Best estimate calculations of the dose rates to individuals will be performed for the different concepts of disposal. Collective dose rates and integrated collective doses will be calculated also. The sensitivity of the results will be investigated and a final assessment will be made.

C. PROGRESS OF WORK AND RESULTS

State of advancement

During this period the calculations have been executed and reported to the Commission. The final report of the ECN / RIVM contribution to the PACOMA project consist of a summary report and four annexes.

Progress and results

The most important parameters for the different salt formations are selected and given in table I, and II. The scenario selected to analyze in detail is the water intrusion / extrusion type of scenario.
The radio nuclide release from the waste containers and the transport through the salt formations has been calculated with the **EMOS** code developed by GSF. The main results are given in table III.
Nuclide transport calculations have been carried out for seven different cases with the computer program **METROPOL** developed at RIVM. The cases P1, P2, P3 and P4 have been analyzed for a travel path with a conservative travel time equal to .62 10^4 years. In the cases P5, P6 and P7 the conservative travel time was 0.89 10^5 years. In case P8 the nuclide transport calculation has not been carried out due to the fact that no travel path from the salt formation reaching the surface. In table IV an overview is given of the maximum release rates in the seven cases for all nuclides giving a maximum release larger than 1 Bq/a .
The migration of the nuclides in the biosphere and the exposure of human beings was calculated with the **BIOS** programme for seven cases. Calculated were the maximum individual dose rate (for three professional groups: arable farmers, cattle farmers and sea fishermen), the maximum collective dose rate and the maximum integrated collective dose. The analyses of the exposure were performed for such a period that the maxima of these three quantities were obtained. The maximum individual dose rate for each of the cases are summarized in table V. The evolution of these dose rates time is given in figure 1. The highest value amounts to 3.7 10^{-7} Sv/a for case P4 with the reference inventory R. The most important pathway is through arable products. The contamination by resuspended soil is negligible. For those nuclides which dominate the exposure, the nuclides from the Np-237 and U-238 series, root uptake is the dominant pathway.

Apart from the results for the water intrusion/extrusion scenario some estimates also are given for the exposure due to some other scenarios. For a normal evolution of a repository in a salt formation diapirism and subrosion can result in a release of nuclides. From conservatively chosen time averaged values for the diapirism rate and the subrosion rate (0.25 mm/a and 0.15 mm/a respectively) the maximum individual dose rate resulting from a repository with MLW and LLW only is estimated to be 4 10^{-8} Sv/a. It is indicated that there might be combinations of diapirism rates and subrosion rates giving a higher dose rate.

For the disposal concepts in a deep formation an alternative altered evolution scenario has been considered briefly. In this scenario the release from the salt formation is induced by water intrusion and the geosphere transport is modified by assuming leakage behind the casing of the boreholes in the overburden. Due to this leakage contaminated brine can move upward untill it reaches aquifers containing fresh groundwater. Human exposure in the biosphere is supposed to occur through water abstraction for public use. The estimate of the maximum individual dose rate amounts to 10^{-5} Sv/a.

With respect to the scenario's for human intrusion it has been argued that the only realistic scenario is formed by a future reconnaissance drilling into the repository. Doses are estimated for drillings at three different moments in future (100, 250 and 1000 years after closing of the repository) and for a routine or a very intensive inspection of the contaminated cores. The probability of this scenario is estimated from an annual drilling frequency of 4 $10^{-9}/m^2$. The maximum risk related to this scenario is estimated to be 1 10^{-9}/a.

Conclusions

From the work performed in PACOMA the following has been concluded.

* The influence of the repository design on the final results (eg. doses) is rather limited. The results of the analyses of disposal in caverns show that both removing the water before disposal and filling up the cavern with salt after disposal is certainly benificial. The results of the analyses of disposal in a mine are disappointing because they are not better than wet operated caverns! This, however, is caused by the fact that the main difference between caverns and mine disposal, viz. the dams, are analyzed with a very high permeability for those dams. [k = 2.25 10^{-15} m^2]. A more realistic permeability of 10^{-18} m^2 has been analyzed giving a zero release for a disposal mine.

* The exposure can be reduced if the disposal concept is designed such that the total release from the salt formation is as small as possible. This can be achieved by backfilling with salt so the amount of water which can intrude into the repository will be limited.

* The results show that the ultimate human exposure in the biosphere is proportional with the total released quantity of nuclides from the repository and independent of the rate of this release, which can be explained by the fact that the real travel time of the nuclides in the geosphere is much larger than the duration of the extrusion from the salt formation. Consequence also will be the degradation period of the containers and the mobilization time of the nuclides do not have such a large influence.

* The computerprogrammes used for the analyses are proven successfully for the assessment of the safety consequences of differences in the design of the repository.

* The results of the computerprogrammes are determined by the set of parameters and by basic assumptions regarding:
  . The release path through the saltformation and geosphere
  . The type of modelling of the geosphere transport
  . The contamination pathway in the biosphere
  The results show that these basic assumptions have the largest influence and therefore cause the largest uncertainty.

* Some simple estimates have been derived for the released mass from a salt formation and for the release rates from the formation and the geosphere. These relations are useful in explaining trends in the computed results and in checking the results of the computerprogrammes used.

Table I Parameters for different salt formations

Salt formation		Saltdome	Saltpillow	Bedded salt
Pathway of brine	in	Anhydrite	Anhydrite	Brine pocket
	out	Anhydrite	Anhydrite	Sealing
Disposal depth	[m]	700	900	1400
Lithostatic press.	[MPa]	16	19.5	31
Brine pressure	[MPa]	8.5	11	17
Salt temperature	[K]	308	317	330
Convergence rate	[%/a]	1	2	28

Table II Solubility limits K_{sp} [mol/l] and distribution coefficients K_d [m^3/kg] used in the nuclide migration analysis

Element	K_{sp}	K_d	Element	K_{sp}	K_d	Element	K_{sp}	K_d
C	$1\ 10^{-3}$	$2\ 10^{-3}$	Mo	$1\ 10^{-2}$	$25\ 10^{-2}$	Eu	$1\ 10^{-1}$	*
Cl	1	0	Nb	$1\ 10^{-3}$	$5\ 10^{-1}$	Cm	$1\ 10^{-6}$	1
Ca	1	$2\ 10^{-3}$	Tc	$1\ 10^{-1}$	$1\ 10^{-3}$	U	$1\ 10^{-2}$	$1\ 10^{-2}$
Co	$1\ 10^{-1}$	*	Pd	$1\ 10^{-4}$	1	Pu	$1\ 10^{-6}$	1
Ni	$1\ 10^{-2}$	2	Cs	$1\ 10^{-1}$	$1\ 10^{-3}$	Th	$1\ 10^{-6}$	$15\ 10^{-2}$
Se	$1\ 10^{-1}$	$1\ 10^{-2}$	Sn	$1\ 10^{-1}$	$2\ 10^{-2}$	Am	$1\ 10^{-6}$	1
Rb	$1\ 10^{-1}$	$5\ 10^{-2}$	I	$5\ 10^{-6}$	$1\ 10^{-3}$	Np	$1\ 10^{-5}$	$1\ 10^{-2}$
Sr	$1\ 10^{-1}$	*	AgI	$1\ 10^{-1}$	$1\ 10^{-3}$	Ra	$1\ 10^{-1}$	$5\ 10^{-3}$
Zr	$1\ 10^{-5}$	1	Sm	$1\ 10^{-1}$	10	Pa	$1\ 10^{-5}$	10

Table III Some general results of the migration of radionuclides in salt formations

case	salt-formation	repository design	initial state	nuclide inventory	end of release [a]	released quantity activity [Bq]	mass [kg]
P1	dome	cavern	dry	NL	$1.4\ 10^4$	$2.0\ 10^{15}$	$1.2\ 10^3$
P2	dome	cavern	wet	NL	$1.5\ 10^4$	$1.9\ 10^{15}$	$2.8\ 10^3$
P3	dome	room	dry	NL	$3.1\ 10^4$	$1.6\ 10^{14}$	$3.8\ 10^3$
P4	dome	room	dry	R	$2.9\ 10^4$	$8.9\ 10^{13}$	$1.4\ 10^4$
P5	pillow	cavern	dry	NL	$9.9\ 10^3$	$2.5\ 10^{15}$	$1.5\ 10^3$
P6	pillow	cavern	wet	NL	$1.7\ 10^4$	$1.8\ 10^{15}$	$3.7\ 10^3$
P7	pillow	room	dry	NL	$1.9\ 10^4$	$1.8\ 10^{14}$	$3.4\ 10^3$
P8	layer	cavern	wet	NL	$2.7\ 10^3$	$5.3\ 10^{15}$	$1.5\ 10^3$

Table IV Max. release rate [Bq/a] from the geosphere for the seven calculated cases. * Release rate < 1 Bq/a

Nuclide	P1	P2	P3	P4	P5	P6	P7
Cl- 36	*	*	*	3.0	*	*	*
Se- 79	$2.3\ 10^3$	$2.1\ 10^3$	$1.7\ 10^3$	$2.9\ 10^4$	$2.1\ 10^3$	$2.1\ 10^3$	$2.1\ 10^3$
Tc- 99	$2.8\ 10^6$	$2.6\ 10^6$	$2.1\ 10^6$	$3.3\ 10^7$	$2.8\ 10^4$	$2.8\ 10^4$	$2.2\ 10^4$
Pd-107	$1.1\ 10^2$	$1.1\ 10^2$	$0.9\ 10^2$	$1.0\ 10^2$	*	*	*
Sn-126	$2.6\ 10^4$	$2.4\ 10^4$	$2.1\ 10^4$	$1.6\ 10^4$	*	*	*
I -129	$2.4\ 10^4$	$2.2\ 10^4$	$1.9\ 10^4$	$1.0\ 10^6$	$8.1\ 10^2$	$8.2\ 10^2$	$6.6\ 10^2$
Cs-135	$3.0\ 10^5$	$2.7\ 10^5$	$2.3\ 10^5$	$1.7\ 10^4$	$9.2\ 10^3$	$9.4\ 10^3$	$7.4\ 10^3$
Pu-244	1.9	4.1	3.7	*	*	*	*
Pu-240	2.1	4.6	4.1	*	*	*	*
U -236	$4.0\ 10^4$	$8.6\ 10^4$	$9.9\ 10^4$	$4.4\ 10^5$	$1.6\ 10^3$	$3.7\ 10^3$	$2.9\ 10^3$
Np-237	$4.1\ 10^5$	$3.8\ 10^5$	$3.2\ 10^5$	$1.6\ 10^6$	$5.1\ 10^3$	$5.1\ 10^3$	$3.5\ 10^3$
U- 233	$2.6\ 10^5$	$2.3\ 10^5$	$2.0\ 10^5$	$1.6\ 10^6$	$5.7\ 10^3$	$5.8\ 10^3$	$4.1\ 10^3$
Th-229	$1.7\ 10^4$	$1.5\ 10^4$	$1.3\ 10^4$	$6.6\ 10^4$	$3.9\ 10^2$	$3.9\ 10^2$	$2.9\ 10^2$
Pu-242	1.4	3.1	*	*	*	*	*
U -238	$3.1\ 10^4$	$6.8\ 10^4$	$9.5\ 10^4$	$3.5\ 10^5$	$1.4\ 10^3$	$3.5\ 10^3$	$3.2\ 10^3$
U -234	$1.3\ 10^5$	$2.8\ 10^5$	$4.0\ 10^5$	$1.7\ 10^6$	$1.4\ 10^3$	$3.6\ 10^3$	$3.4\ 10^3$
Th-230	$6.6\ 10^3$	$1.4\ 10^4$	$1.9\ 10^4$	$8.2\ 10^4$	$1.0\ 10^2$	$2.5\ 10^2$	$2.4\ 10^2$
Ra-226	$1.9\ 10^5$	$4.1\ 10^5$	$5.5\ 10^5$	$2.4\ 10^6$	$2.8\ 10^3$	$7.1\ 10^3$	$6.7\ 10^3$
U -235	$3.4\ 10^3$	$7.4\ 10^3$	$7.3\ 10^3$	$2.3\ 10^4$	$1.5\ 10^2$	$3.6\ 10^2$	$2.4\ 10^2$
Pa-231	3.0	6.4	6.4	$2.0\ 10^1$	*	*	*

Table V Maximum individual dose rate [Sv/a]

case	t_{max} [Ma]	arable farmer	cattle farmer	sea fisherman
P1	0.4	$3.4\ 10^{-8}$	$3.3\ 10^{-8}$	$3.0\ 10^{-10}$
P2	0.4	$6.5\ 10^{-8}$	$6.3\ 10^{-8}$	$4.3\ 10^{-10}$
P3	0.6	$8.6\ 10^{-8}$	$8.5\ 10^{-8}$	$5.1\ 10^{-10}$
P4	0.6	$3.7\ 10^{-7}$	$3.6\ 10^{-7}$	$2.1\ 10^{-9}$
P5	4.0	$5.5\ 10^{-10}$	$5.5\ 10^{-10}$	$4.5\ 10^{-12}$
P6	4.0	$1.2\ 10^{-9}$	$1.1\ 10^{-9}$	$7.9\ 10^{-12}$
P7	4.0	$1.1\ 10^{-9}$	$1.0\ 10^{-9}$	$6.9\ 10^{-12}$

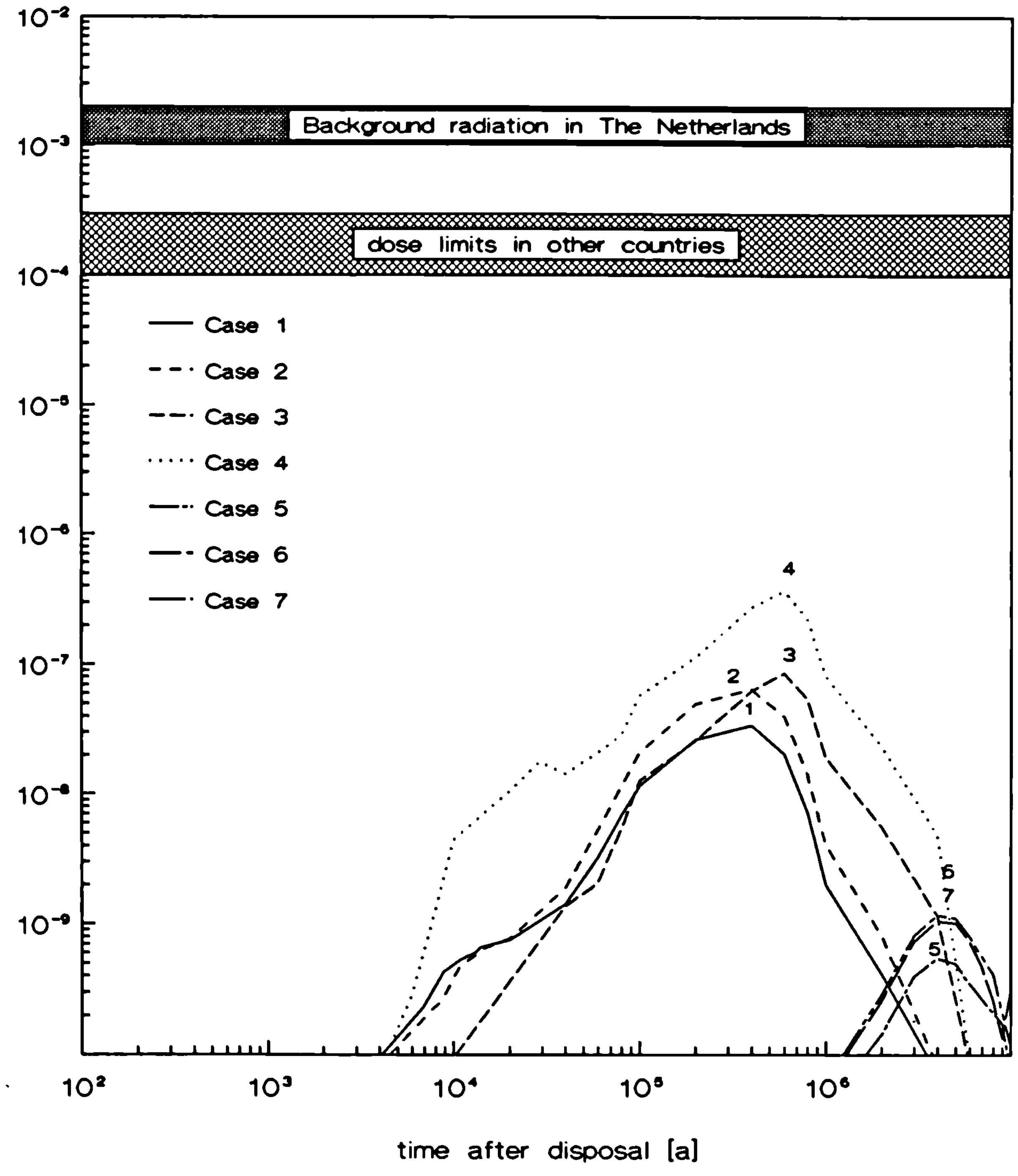


Figure 1. Maximum individual dose rates [Sv/a] for each case of the PACOMA-study. (water intrusion / extrusion scenario)

ASSESSMENT OF RADIOLOGICAL CONSEQUENCES AND RISK ASSOCIATED WITH THE GEOLOGICAL DISPOSAL OF MLW AND ALPHA-WASTE IN CLAY FORMATIONS - PACOMA PROJECT

Contractor : SCK/CEN, Mol, Belgium
Contract No : FI1W/0046
Duration of contract : from March 1987 through June 1990
Period covered : January 1989 - June 1989
Project leader : A. Bonne
Report by : J. Marivoet

A. OBJECTIVE AND SCOPE

The aim of the PACOMA project is to carry out, as an extension of the PAGIS action on HLW, a comprehensive assessment of the radiological consequences and risks associated with geological disposal of alpha-bearing and medium level wastes. The methodology used in the performance assessment is the one that has been developed during the PAGIS exercise. The main objectives of the study are :

- best estimates of the dose rates and risks to individuals and populations for the normal evolution and the relevant altered scenarios ;
- sensitivity studies of the results to variations of data bases, parameters and models ;
- analysis of the uncertainties.

B. WORK PROGRAMME

B.1. Data collection.
B.2. Adaptations of the computer codes
B.3. Best estimate calculations.
B.4. Sensitivity study.
B.5. Uncertainty analysis.
B.6. Final assessment.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Deterministic calculations of individual dose rates have been elaborated for three selected scenarios and various geosphere-biosphere interfaces have been considered. Earlier calculations had to be updated because the PACOMA reference inventories have been changed at the end of 1988. Collective doses have been estimated in the case of the normal evolution scenario.

The computer codes used to perform the stochastic calculations had to be adapted in order to include some model refinements in the clay migration module. The stochastic calculations are finished and their results have been used to perform sensitivity and uncertainty analyses.

The final conclusions of the PACOMA calculations for the Mol site have now been drawn.

Progress and results

Three scenarios are analysed with the deterministic calculational approach :

- the normal evolution scenario ;
- a climatic change scenario which assumes a reduction of the net precipitation ;
- a faulting scenario in which the occurrence of a tectonic fault through the repository site is assumed.

The individual dose rates that may occur via three geosphere-biosphere interfaces are evaluated ; the considered interfaces are :

- the discharge of contaminated groundwater into rivers ;
- the direct contamination of agricultural soils by groundwater ;
- the pumping of water from a well sunk in the aquifer overlying the repository.

The dose rates calculated in the case of the normal evolution scenario via the water well pathway are given in Fig. 1. The total dose rate results from contributions of I-129, C-14, Se-79, Tc-99, Np-237 and its daughter nuclides and of radionuclides in the U-238 decay chain. The highest calculated dose rate, which is mainly due to I-129, is 1 µSv/y and it occurs at about 50,000 years after disposal.

An overview of the main results of the various combinations of scenarios and pathways that are analysed is given in Figure 2. The highest individual dose rate is calculated in the case of the climatic change scenario in combination with the water well pathway. The maximum dose rate is equal to 6 µSv/y. The increase of the individual dose rates in the case of a dryer climate can be explained by the higher demand for well water for agricultural applications and by the reduction of the radionuclide dilution in the aquifer.

For the two pathways which do not assume a human intervention in the aquifer, the soil pathway yields dose rates which are some orders of magnitude higher than the dose rates calculated for the river pathway. However the soil dose rates are about a factor ten lower than the corresponding water well dose rates.

The faulting scenario leads to an acceleration of the radionuclide migration through the clay layer. Because of the small thickness of the fault plane, only a small fraction of the disposed radionuclides takes part in the accelerated transport. The resulting dose rate is of the same order of magnitude as the dose rate calculated in the case of the normal evolution scenario, but it occurs earlier.

The collective doses are evaluated for the river pathway in the case of the normal evolution scenario. The calculated collective dose commitments are given in Figure 3. The total collective dose commitment is as low as 12 man.Sv after 100 million years.

The risk associated with the analysed scenario is one of the most important output parameters that are calculated in the uncertainty analysis. An overview of the calculated risks is given in Figure 4. For the stochastic calculations all scenarios which do not affect the integrity of the host clay barrier, e.g. normal evolution scenario, climatic changes, glaciation effects in the sand layers overlying the host clay layer, sea level variations, river erosions, are combined into the extended normal evolution scenario.

A risk as high as 5 E-6 per year is calculated for the water well pathway in the case of the extended normal evolution scenario. This risk is mainly due to high C-14 dose rates that are calculated in the Monte Carlo simulation for combinations of conservative parameter values. The sensitivity analysis reveals that the C-14 dose rates are extremely sensitive to the clay migration parameters. A physical explanation for the high sensitivity is given by the fact that the C-14 half-life time, which is 5,730 years, and the break-through time of a non-retarded ion or molecule through the clay barrier, which is about 10,000 years, are of the same order of magnitude. It is expected that the maximum calculated risk may decrease one or two orders of magnitude when more accurate data sets, giving smaller ranges of possible parameter values, for the C-14 model parameters will become available from ongoing and future research programmes.

The risk associated with the faulting scenario is very low because of the low occurrence probability of this scenario.

<u>List of publications</u>

/1/ SALTELLI, A. and MARIVOET, J., Nonparametric statistics in sensitivity analysis for model output : a comparison of selected techniques, Reliability Engineering ans System Safety, (in press).

/2/ BONNE, A., The clay option : PAGIS results, PAGIS Information Day, Madrid, 30.06.1989.

/3/ BONNE, A. and MARIVOET, J., Preliminary safety assessment of a HLW-repository in a stratiform argillaceous formation underlying the nuclear site of Mol-Dessel in Belgium, Int. Symp. on Safety Assessment of Radioactive Waste Repositories, Paris, 09-13 October 1989.

/4/ MARIVOET, J. and BONNE, A., Deterministic performance assessment of a medium-level and alpha-bearing waste repository in clay, (same symposium).

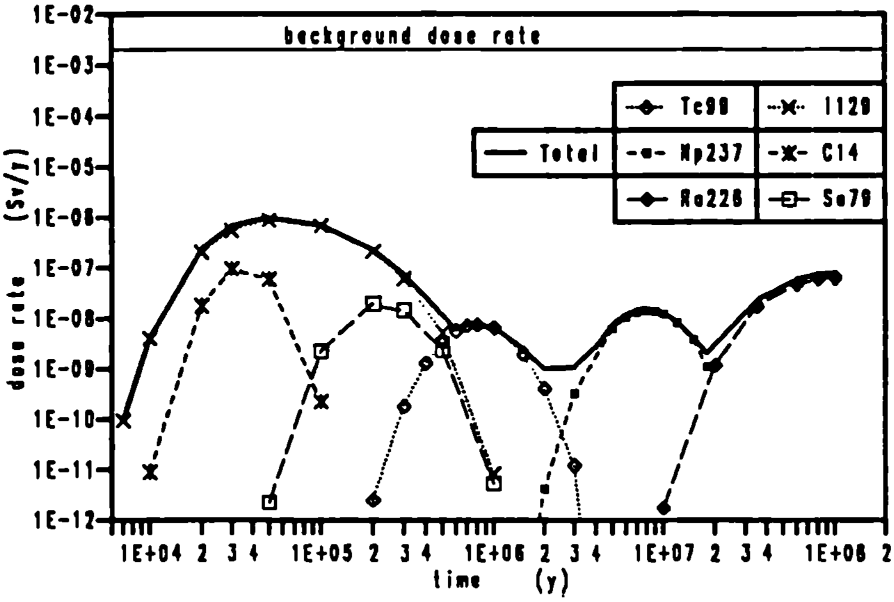


Figure 1. Dose rates calculated for the water well pathway in the case of the normal evolution scenario

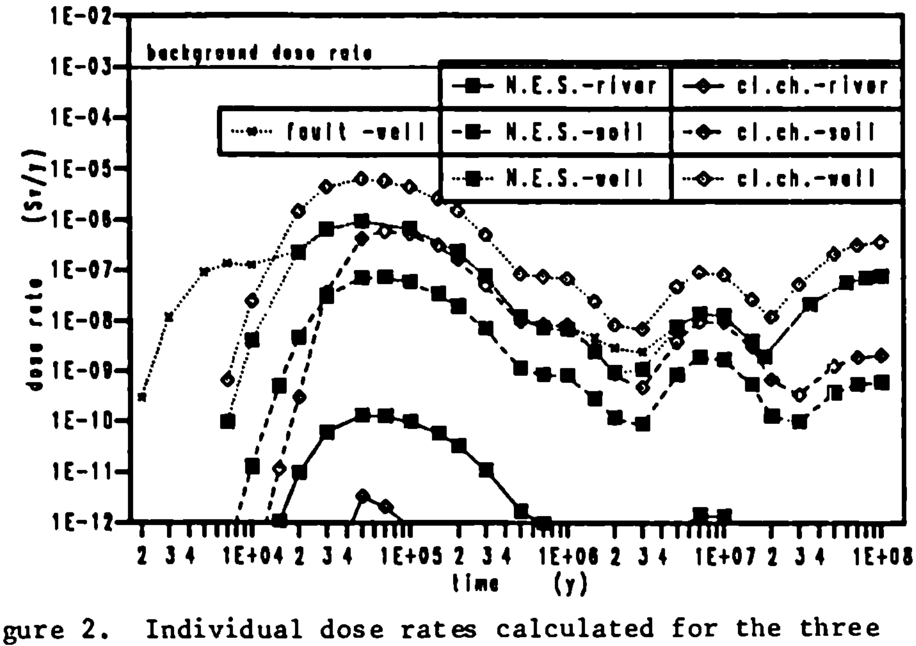


Figure 2. Individual dose rates calculated for the three considered scenarios (N.E.S. : normal evolution scenario ; cl.ch. : climatic change scenario)

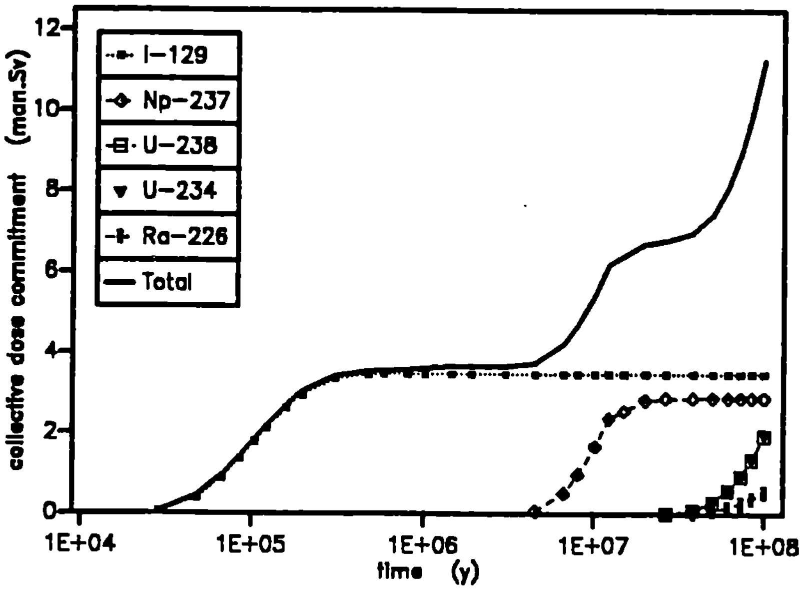


Figure 3. Collective dose commitments calculated for the rivers pathway in the case of the normal evolution scenario

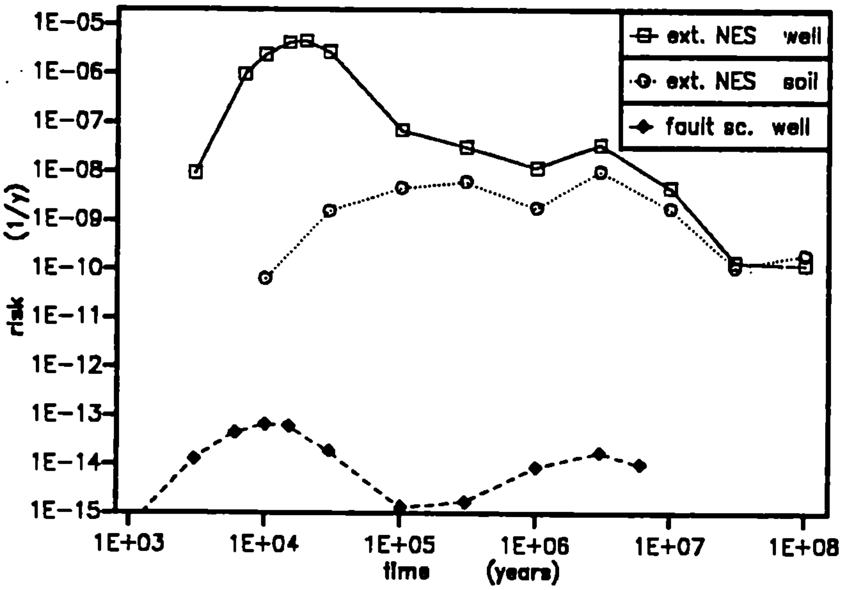


Figure 4. Risks calculated for the considered pathways and scenarios

SUPPORT STUDIES

Definition of standards of quality assurance related to the development of disposal facilities for radioactive waste.

Contractor:	Cedar Design Systems Ltd, London, U.K.
Contract No:	CEC - FI1W/0039 - UK (H1)
	DoE (UK) - PECD 7/9/384
	UKAEA - H2C 618390 T

Duration of contract:	January 1987 - March 1989
Project Leader:	I.E. Hill
Date:	January 1990

A. Objectives and Scope

The research project will produce a definition of an acceptable and appropriate set of procedures for quality assurance, verification, and validation. These procedures would form the basis for establishing uniform standards for risk assessment software, which could be implemented throughout the European radioactive waste disposal programme.

The value and effectiveness of the procedures will be demonstrated by their practical implementation as part of the work of other contractors on the CEC PACOMA projects. Included in this practical implementation will be the introduction of software tools for automating quality assurance procedures, where such tools are available or can be developed.

The work will be subdivided into 4 stages covering respectively, analysis of existing procedures and definition of requirements; a case study of the use of existing procedures in the UK Department of the Environment; definition of procedures; and implementation of procedures.

In order to ensure that the work done is appropriate to the development of risk assessment codes, reference will be made to the other contractors for the CEC PACOMA project.

B. Work Programme

B.1. Review of existing software quality assurance standards and definition of requirements

B.2. Case study of existing UK DoE procedures

B.3 Definition of procedures

B.4. Implementation of procedures

C. <u>Progress of work and obtained results</u>

State of advancement

Work on this project was completed in November 1989.

Progress and results
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1. Review of existing software quality assurance standards
   This stage was completed in 1988.
2. UK DoE case study
   During 1989 the DoE work concentrated on verification of the probabilistic risk assessment code, VANDAL, and the work carried out as part of this project consisted of reviews of the verification test specifications, test record keeping systems, and of the test results as they became available.
   The test procedures have been described and used as examples in the final project report [1].
3. Definition of Procedures
   The work done in 1989 concentrated on the documentation of the procedures recommended in the requirements report submitted in 1988.
   The procedures for development and use of software are documented in the final report [1] under
   - management
   - software development
   - documentation
   - configuration management
   - use of software;

   and recommendations for verification and validation include
   - reviews
   - code examination
   - consistency tests
   - validation of full programs
   - comparison with physical systems
   - evidence of verification and validation
4. Implementation
   The trial implementation of procedures was completed in 1988.

List of publications
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[1] HILL, I.E., MAYER, J., Software quality assurance procedures for radioactive waste risk assessment codes. (October 1989)

[2] HILL, I., UNSWORTH, M., CADELLI, N., THOMPSON, B., Quality Assurance for Safety Assessment of Radioactive Waste Repositories. Presented at the International Symposium on the Safety Assessment of Radioactive Waste Repositories, Paris, October 1989.

MODELLING THE LONG TERM EVOLUTION OF GEOLOGICAL RADWASTE DISPOSAL FACILITIES

Contractor:	Dames & Moore, Twickenham, United Kingdom
Contract No.:	FI1W/0169-UK
Duration of contract:	November 1987 - July 1989
Period covered:	January 1989 - July 1989
Project Leader:	P.S. Ringrose

A. OBJECTIVES AND SCOPE

The primary objective of the study is to allow more realistic modelling of the effects of ice sheet advances and retreats on deep underground disposal facilities for radioactive wastes and their environments. This is to be undertaken by advancing the status of fundamental research in this area. In Northern Europe glacial conditions are expected to return within the next 20,000 to 30,000 years and cycles of ice sheet advance and retreat to continue for the next million years at least. The influences on disposal sites could be significant. It is expected that algorithms will be developed from the work which, if used in a suitable computer code, will allow realistic modelling of ice sheet erosion and deposition and the effects of ice sheet advance/retreat cycles (including the associated ground freezing/thawing) on groundwater flow.

The project is being carried out in collaboration with researchers at the Grant Institute of Geology, University of Edinburgh, United Kingdom.

B. WORK PROGRAMME

B.1 Research status review: a brief review of the status of research into the effects of long-term environmental changes on deep disposal facilities.

B.2 Background Research: this second task is the major aspect of the work programme and incorporates research into aspects of glacial processes and in particular the growth of permafrost.

B.3 Specification for the incorporation of site evolution modelling into performance assessment of geological disposal of radioactive wastes, with reference to three suitable sites in Europe.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

All tasks in the work programme have been completed including a second phase of research work on permafrost. Some changes in the direction of research occurred during the project. More attention was focused on modelling permafrost and intended modelling of geomechanical effects of ice was not achieved. The work has been reported in a final report for the project /1/.

Progress and results

C.1 Research status review.

This is given in Volume I of the final report /1/. The review covers the following aspects:

- A summary of the nature of environmental change and the processes which are significant for deep disposal.
- A review of worldwide research into environmental change.
- A summary of environmental change modelling work at Dames & Moore.
- A comparison of approaches to the incorporation of environmental change into performance assessment.

C.2 Background Research.

A research programme lasting two years has resulted in the development of a fundamental model of the growth and effects of permafrost layers which accompany glaciation. This model involves the application of the continuum theory of mixtures to a three-phase medium (water/ice/rock) and at the scale of interest (depths of hundreds of metres and timescales of thousands of years).

The theoretical work, reported in Volume II of the final report /1/, is accompanied by various numerical illustrations. An assessment of the growth of permafrost under different rates of surface cooling is made and it is concluded that for relatively slow cooling rates (less than 1^{o} per 100 years) a steady-state calculation of permafrost depths for given temperature conditions is valid. Using a steady-state approximation an illustration of the likely variation in depth of permafrost in the United Kingdom over the last 750,000 years has been derived (Figure 1).

Such illustrations show the likely depth-range of permafrost and can be used to assess the implications for long-term storage of radioactive waste facilities deep underground. The work also forms a basis for assessing the effects of permafrost within models of long-term site evolution, as has been done within the model TIME4.

The new research work on permafrost complements existing work (at the University of Edinburgh) on glaciers and their effects, with the result that most aspects of glaciation can be assessed on a theoretical basis for use in performance assessment studies.

C.3 Incorporation of results into performance assessments.
This third aspect of the work involves specification for the use of site-evolution models in the various performance assessment situations within the EEC, and is reported in Volume I of the final report /1/. The development of site-evolution modelling with particular reference to TIME4 is also described elsewhere /2/.

List of publications

/1/ DAMES & MOORE, Modelling the long-term evolution of geological radwaste disposal facilities. Technical Report TR-D&M-12 (in two parts). To be published by the CEC and the UK DOE.

/2/ RINGROSE, P.S. et alia. Probabilistic simulation of the long-term evolution of radioactive waste disposal sites. In: "Safety Assessment of Radioactive Waste Repositories", Paris, October 1989, OECD (in press).

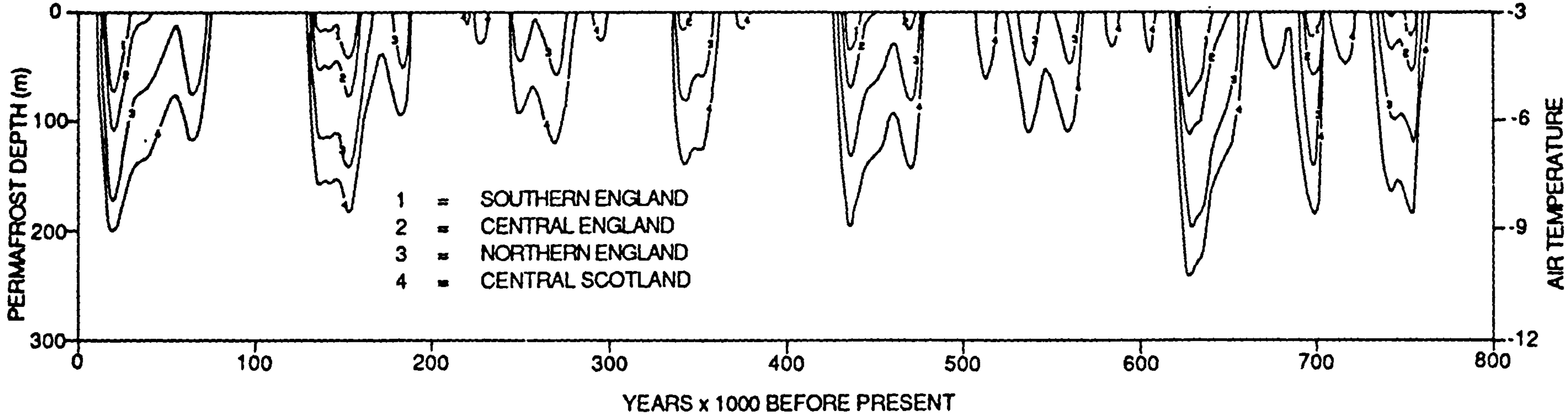

FIGURE 1
RECONSTRUCTED DEPTH OF PERMAFROST IN BRITAIN SINCE 750Ka BEFORE PRESENT

HUMAN INTRUSION INTO UNDERGROUND REPOSITORIES FOR RADIOACTIVE WASTE

Contractor:	Associated Nuclear Services, UK
Contract No.:	FI1W/0170-UK
Duration of contract:	October 1987 to March 1989
Period covered:	January 1989 to March 1989
Project Leader:	D.J. Nancarrow

A. OBJECTIVES AND SCOPE

The objectives of the work are;

- to establish a methodology for the assessment of risks associated with human intrusion events applicable to the types of underground repository studied in the PAGIS and PACOMA projects;

- to delineate the application of the methodology to the alternative disposal concepts, taking into account the characteristics of the host rocks (clay, granite, salt and sub-seabed sediments) and repository designs;

- to produce an authoritative source of reference (database and methodology) for use in safety assessment of HLW and ILW disposal facilities.

B. WORK PROGRAMME

1. The various human activities which might result in an intrusion into a repository, or disruption of the host geology leading to enhanced radionuclide migration, will be identified. Relevant information necessary to define and quantify each mode will be collated.

2. The radiological consequences, to potential intruders and to others, from defined intrusion modes will be reviewed and appropriate calculation schemes defined.

3. The probabilities of intrusive events, identified as significant, will be estimated for reference circumstances. Factors affecting these probabilities will be identified and quantified as far as possible.

4. From the analysis made above, a general philosophy for assessment of risk from human intrusions will be developed. The application of the methodology will be outlined for the particular host geologies and repositories considered in PAGIS and PACOMA, with the aid of examples.

5. The capacity for reduction of risks from intrusion by measures to reduce the probability or consequences of intrusion will be examined. This will include an examination of possible anti-intrusion measures.

C. PROGRESS OF WORK AND OBTAINED RESULTS

STATE OF ADVANCEMENT

The subcontract work on the characterisation of mineral and other resource exploitation (Mott, Hay and Anderson) and on assessing probabilities of human intrusion (London School of Economics & Political Science) have been completed and their reports have been attached as appendices to the main report. An additional appendix on the characterisation of groundwater abstraction activities has been completed. The main report plus appendices was issued in draft in May 1989. After receipt of comments from the CEC (and from the UK Department of the Environment and the National Radiological Protection Board) the report was issued in final in December 1989 to the CEC /1/.

PROGRESS AND RESULTS

1. Modes of intrusion

For each of the geological formations identified as being suitable to host a deep underground repository, human activities which would constitute an intrusion have been identified and are presented in Table I. This table shows the intrusion modes which are relevant to each host strata and indicates whether an assessment has been performed or should be performed when producing a safety assessment. With regard to intrusion by water abstraction from the host or surrounding stata the depth, yield, life expectancy and the frequency of occurrence of boreholes in appropriate lithologies and climates have been collated where possible. Detailed characteristics of a range of intrusion activities (other than for water abstraction) have been presented.

2. Radiological consequences

The exposure modes relevant to workers making the intrusion, and subsequently to nearby members of the public, associated with an intrusion into a deep underground repository or its contaminated environs has been reviewed. Schemes and equations to calculate the radiological impact to both groups of persons have been presented with associated data. The equations are presented for unit concentration of activity in the materials excavated; and for a single intrusion event.

3. Probabilities of intrusion

Probabilities of intrusion (events per year per km^2) have been derived for various forms of geological investigation for each of the host media and are presented in Table II. These probabilities were derived from available data in the UK and are subject to the following caveats:

- considerable problems of data availability were found; for example the national borehole register was of limited use because commercial secrecy restricts the publication of some borehole locations and hence information on the type of stratigraphy is unknown;

- activity rates are presented on an annual average basis; however, each geological situation is unique; world shortage in a particular commodity may override a cautious approach or the discovery of a mineral at a nearby location could engender a rapid increase in exploration activity within the area;
- un-notified drilling may mean rates are underestimated;
- activity rates are necessarily a reflection of past activity only and do not allow for marked technological changes or resources of no current value but which could become important.

4. Philisophy for risk assessment

Approaches to the assessment of human intrusion may differ in a number of respects and for a number of reasons. The following list of differences provides a useful means of characterising a given approach:

- the individual being assessed (intruder or person subsequently exposed);
- the impact criterion (dose, conditional risk or overall risk);
- the degree of pessimism adopted in defining a basis for intrusion assessments;
- the type of analysis (deterministic or probabilistic).

Each of these has been considered in turn in the context of estimating the radiological impact to an individual.

5. Capacity for risk reduction

Reduction of risk associated with human intrusion can be obtained by either reduction of the probability of intrusion or reduction of the consequences. The approach is summarised in Figure 1.

LIST OF PUBLICATIONS

/1/ NANCARROW, D.J., LITTLE, R.H., ASHTON, J., STAUNTON, G.M. The assessment of human intrusion into underground repositories for radioactive waste, EUR report in course of preparation.

TABLE I

INTRUSION MODES CONSIDERED FOR EACH SITE TYPE

Intrusion mode	Host strata			
	Crystalline rocks	Clays	Salt	Sub-seabed sediments
Deep drilling				
- exploration, e.g mineralogical, geotechnical	1	1	1	1
- exploitation e.g. hydrocarbons	1	1	1	1
Water abstraction	2	2*	1*	
Underground mining	2		1	
Sub-surface construction	1	1	1	
Cavern leaching			2	
Salt solution mining			2	
Geothermal exploitation	1			
Mining manganese nodules				2

Notes: 1. Relevant - in the opinion of the authors of this report these should be considered for a comprehensive safety assessment
2. Relevant - some assessment has already been performed
* From adjacent aquifers

TABLE II

INTRUSION RATES FOR EACH SITE TYPE

Medium	Type of event	Rate y^{-1} km^{-2}
Clay	Mineral drilling	2.3×10^{-3}
	Hydrocarbons	1.2×10^{-3}
	Water	8.0×10^{-5}
	Scientific (1)	5.3×10^{-5}
		$\sim 4 \times 10^{-3}$
Salt	Mineral drilling	2.3×10^{-3}
	Mine development	2.9×10^{-5}
	Hydrocarbons	1.2×10^{-4}
	Scientific (1)	5.3×10^{-5}
		$\sim 3 \times 10^{-3}$
Granite	Mine development	2.9×10^{-5}
	Scientific (1)	5.3×10^{-5}
		$\sim 8 \times 10^{-5}$
Seabed	Laying cable	5×10^{-10}
	Trawling for wreckage	4×10^{-8}
	Submersible wreckage study	2×10^{-9}
	Scientific (1)	1×10^{-8}
		5×10^{-8}

Note: 1. Includes boreholes drilled for geological research and geothermal exploration.

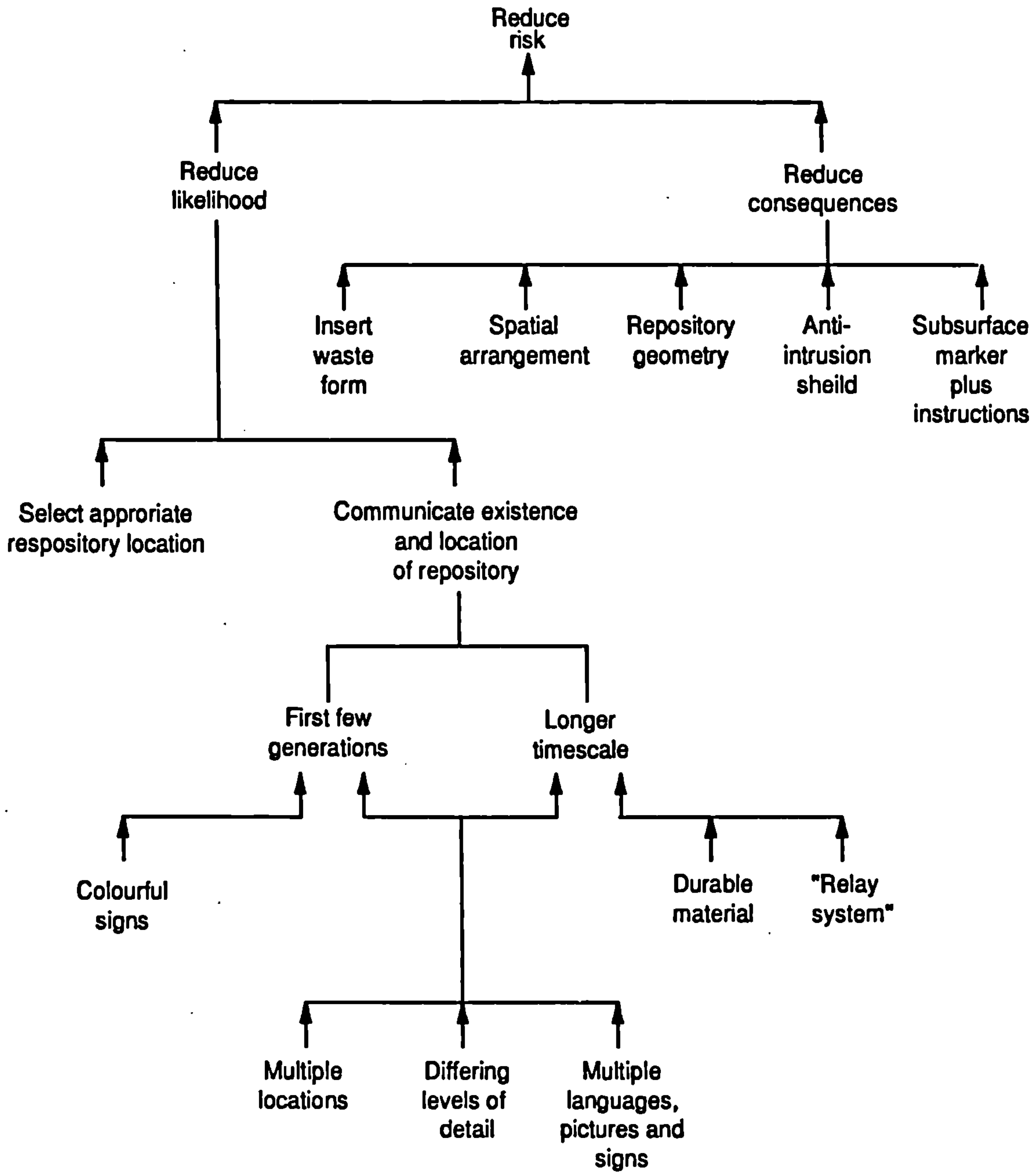

FIGURE 1 SCHEME FOR RISK REDUCTION

T A S K N o 6

JOINT ELABORATION OF RADIOACTIVE WASTE MANAGEMENT POLICIES

TASK No. 6: JOINT ELABORATION OF RADIOACTIVE WASTE MANAGEMENT POLICIES

A. Objective

Joint elaboration of waste management and disposal criteria.
Evaluation of possible approaches, at Community scale, for waste disposal.

B. Research topics dealt with under 1980-1984 programme

These research topics were not included in the 1980-1984 programme.

C. 1985-1989 programme

The following activities have been defined:

a. development and harmonization of acceptance criteria for radioactive waste conditioning, with respect to their handling and final disposal;
b. development of radiological criteria for disposal, especially for the periods of time involved in geological disposal;
c. elaboration of recommendations concerning the satisfactory execution, taking into account the safety and environmental protection standards, of the various operations involved in the management and disposal of radioactive waste;
d. study of "de minimis" criteria, with regard to alpha/non-alpha and radioactive/non-radioactive waste;
e. multi-national dimensions of waste management; influences on its optimization; regional disposal.

D. Programme implementation

A working group of national experts has been set up to deal with Task 6 activities. Topics of first priority have been identified as items b) and d) of paragraph C above. Work on radiological criteria for disposal has been performed through cooperation in an ad hoc group of national experts and Commission staff. The group produced the report on "Objectives, standards and criteria for radioactive waste disposal in the European Community", which has been published (Report EUR 12570, available at the office for Official Publications of the EC, Luxembourg).
In the field of the development of rules for exemption of specific radioactive waste streams from regulatory control, the group has selected radioactive waste not linked to the nuclear fuel cycle (hospital, research and other radioactive waste), as the first waste stream to analyse. Five contracts were concluded in order to produce input for the group's work.

THE MANAGEMENT OF RADIOACTIVE MATERIALS ON NON-NUCLEAR SITES IN THE UK

Contractor:	WS Atkins Engineering Sciences Ltd, Warrington, England
Contract No:	FI1W-0236
Duration of contract:	from 1 September 1988 to 31 January 1990
Period Covered:	from 1 January 1989 to 31 December 1989
Project leader:	Dr B J Tymons

A. OBJECTIVES AND SCOPE

The majority of the effort in the area of the management of radioactive materials has been concerned with the large reprocessing plant, commercial power reactors and waste disposal sites. There are, however, a large number of "non-nuclear" sites, such as hospitals, laboratories and industrial organisations, where radioactive substances are produced, used or waste arisings disposed of. This study aims to report on the current practices regarding the use and storage of radioactive materials and the production or disposal of radioactive wastes at non-nuclear sites in the UK.

The objectives of the study are:

a) to identify the current methods of monitoring the use of radioactive materials at UK non-nuclear sites,

b) to carry out a detailed study into the use of radioactive materials and the production and disposal of radioactive waste at a sample of sites in the UK,

c) to examine other sources of information on these arisings, for example, the suppliers of radioactive materials and the National Disposal Service (NDS).

B. WORK PROGRAMME

B.1. Contacting the various organisations involved in the management of radioactive materials at non-nuclear sites to obtain their co-operation in the project and to obtain any relevant information for the study.

B.2. Compiling information on the management of radioactive materials and the production of radioactive waste by visiting a representative sample of non-nuclear sites to observe the procedures used.

B.3. Analysis of the information gathered as above.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Section B.I, was accomplished at the beginning of 1989. The information received in this phase of the study led to a slight change of emphasis and sections B.1. and B.3. were combined to produce an extended phase 1 report which gave a full explanation of the legislation etc which governed the management of radioactive materials at non-nuclear sites in the UK.

This revised section B.1. was completed by mid 1989 and a draft report was issued. The site visits necessary for section B.2. were completed by the end of 1989.

Progress and results

1. AN OVERVIEW OF THE ACTIVITIES AND LEGISLATION INVOLVING IONISING RADIATIONS AS APPLICABLE TO NON-NUCLEAR SITES

A report was produced which reviews all aspects of the management of radioactive materials on non-nuclear sites, including a survey of the legislation and associated codes of practice controlling the handling of radioactive materials and the protection of personnel, the production of waste and the options open for its disposal at non-nuclear sites in the UK. The procedures are summarised below.

1.1 UK Government policy

Government policy is set out in a government white paper "Nuclear Power and the Environment" which requires that:

a) the creation of waste from nuclear activity should be minimised,
b) problems of waste management are dealt with before any large nuclear programme is started,
c) waste is handled and treated in an environmentally conscious way,
d) waste accumulated at nuclear sites is disposed of in a programmed way,
e) there is adequate research and development into disposal methods,
f) waste is disposed of properly.

Other objectives based on the International Commission for Radiological Protection (ICRP) system of dose limitation, given in ICRP publication 26 have been added. These are:

a) all practices producing radioactive waste must be justified in terms of its overall benefit or detriment,
b) radiation exposure should be reduced to levels As Low As Reasonably Achievable,
c) the mean effective dose equivalent from all sources excluding background radiation and medical exposure, to the critical group should be less than 5mSv/year.

1.2 UK legislation

The Radioactive Substances Act 1960

The Act regulates the keeping and usage of radioactive materials and the accumulation and disposal of radioactive waste in the UK. It requires that premises where radioactive materials are held or used, and mobile sources of radioactivity, are registered. Sites accumulating or disposing of radioactive waste must do so in accordance to a government authorisation which sets out the terms of accumulation and restrictions on disposals for each site in a certificate. Exempt from all or part of the Act are nuclear licenced sites, premises operated by the UK Atomic Energy Authority and Crown Property. There are also a number of exemption orders that can be applied.

The Ionising radiations Regulations 1985 and the Approved Code of Practice

These regulations were issued under the Health and Safety at Work Act 1974, they bring into British law the majority of the provisions in the European Commission Directives 80/836 and 84/467 Euratom. They are designed to regulate the protection of those exposed to ionising radiation by dose limitation techniques, including:

a) The production of a written system of work and "local rules" that are specific to each site. The local rules set out procedures to be followed for both routine and emergency handling of radioactive materials at the site and working procedures that do not lead to unnecessary exposure of workers or others. They are intended as an adequate guide for the ordinary user in the safe use of radioactive materials.
b) The designation of restricted access areas (known as controlled areas) where work with ionising radiation or source materials is done. Entry into these areas is limited to workers who are certified as medically fit and for whom a record of radiation exposure and health is maintained. These workers are known as classified persons.
c) Each site must have a person who is; knowledgeable about the properties of ionising radiation, has experience in practical radiation protection, and is familiar with the type of work done at the site to act as a Radiation Protection Advisor (RPA). In the case of a large user more than one person may be appointed to this role, forming a corporate body RPA.
d) Records must be maintained of the nature and location of all sources held on the site and the date and method of disposal of radioactive wastes.
e) Each site must provide an assessment of the hazard and environmental impact of its activities, part of this assessment must be an emergency plan, ie procedures that would be applied in the event of conceivable accidents at the site.

1.3 Options available for disposal

Solid waste from non-nuclear sites can be disposed of as follows:
a) In domestic refuse - VLLW is exempt from control under the Radioactive Substances Act and can be treated as uncontaminated refuse. Sites are often authorised to dispose of small volumes of LLW with domestic refuse. This is known as "dustbin disposal"
b) Incineration - The burning of limited activities of solid LLW was found to be widespread, in most cases the site authorisation specifies a disposal route for ashes.
c) Special precautions burial - Waste that is too active for dustbin disposal can be sent for burial at authorised landfill sites with uncontaminated refuse subject to given precautions.
d) Burial at a special site - British Nuclear Fuels plc operate a dedicated LLW burial site at Drigg in West Cumbria. More active waste can be sent by special arrangement to this site for disposal. The acceptance limits placed on waste by Drigg are generally regarded as an upper limit on the definition of LLW.
e) The National Disposal Service (NDS) - In some cases waste is consigned to Drigg through the NDS, in cases where the waste is too active it may be held by the NDS at its site in Harwell awaiting the final disposal in a national repository.

Liquid LLW - Many sites are authorised to discharge regulated volumes and activities of water soluble liquid waste directly into the drainage system, on a "dilute and disperse" basis.

Gaseous LLW - Sites are usually authorised to discharge regulated volumes and activities of gases directly into the atmosphere via a flue to ensure adequate dispersion.

2. VISITS TO A REPRESENTATIVE SAMPLE OF UK SITES

A major hospital, university, minor hospital, polytechnic, research establishment, radioactive source producer and an industrial site were visited to ascertain the procedures used in the management of radioactive materials.

Determination of activity levels and recommendations for the exemption of radioactive wastes from installations non including basic nuclear installations

Contractor : Commissariat à l'Energie Atomique, Fontenay-aux-Roses, FRANCE
Contract n° : FI.1W.237
Working Period : January 1989 - December 1989
Project Leader : A.M. CHAPUIS, J.M. ASSELINEAU

A survey of the French production of radioactive waste from hospitals, university, research and industry was a first step of this study.

Information was collected in order to better know uses and main applications of radionuclides.

Visits in "small producers" installations have allowed a better knowledge of the waste problems : management, collection, sorting, storage etc.

A generic study of practices connected with ordinary waste burial was realized. It was then possible to describe scenarios corresponding to these practices.

Investigation of ordinary and toxic waste treatments, especially incineration, is under way. Household refuse incineration is a well know treatment procedure, it is not the case for toxic and chemical waste (laboratory refuses) and for biological waste produced in hospital units (bandage, anatomical waste). Information collected during these investigations will allow the description of different radionuclides pathways between waste and man in reasonably pessimistic scenarios.

PRACTICE OF TREATMENT OF RADIOACTIVE WASTES ARISING OUTSIDE THE NUCLEAR FUEL CYCLE IN THE FEDERAL REPUBLIC OF GERMANY

Contractor : GRS, Köln, Federal Republic of Germany
Contract No : FI1W/0238
Duration of contract : December 1988 - March 1990
Period covered : January 1989 - December 1989
Project leader : W. Müller

A. Objectives and scope

It is intended to provide an overview on the classification, treatment and, where appropriate, the exemption from regulatory control of radioactive wastes arising outside the nuclear fuel cycle. The investigations are based on an exemplary evaluation of the current situation.

B. Work programme

1. Analysis of waste streams for main sources, activities and amounts

2. Investigation of the organisation of waste collection and treatment, especially in the areas

   - criteria for classification
   - licensing situation
   - kind of measurement
   - organisation of responsibilities
   - available conditioning procedures in place

3. Evaluation of results and possibilities for improvements

C. Progress of work

Information has been collected from a number of typical waste producing organisations and institutes. Particular attention has been paid to radionuclides presenting difficulties in treatment and disposal, as Tritium, C-14, Radon and J-129.
Waste streams have been analysed, and proposals for improvement of means for conditioning, measuring and disposing of waste are being prepared.

EVALUATION OF THE MANAGEMENT PRACTICES IN BELGIUM FOR RADIOACTIVE WASTE NOT LINKED TO THE NUCLEAR FUEL CYCLE

Contractor : Belgatom S.A., Brussels, Belgium
Contract No. : FI1W/0239
Working Period : November 1988 - March 1989
Period Covered : January 1989 - March 1989
Project Leader : P. Dardenne

A. Objectives and scope

The objective is the investigation of the situation of the management of radioactive waste outside the nuclear fuel cycle, which is called AMIRI in Belgium (Applications médicales et industrielles des radio-isotopes).

The study is limited to the Belgian territory. The aim is to set up a "state-of-the-art" report based on information supplied by the agency responsible for the management of this type of waste (ONDRAF/NIRAS) and by a number of waste producers providing voluntarily the requested data. The practice for storage, treatment and transport of this matter will be compared to repository procedures, set up in particular by the national agency. Information received is to be presented in a statistical (anonymous) manner.

B. Work programme

2.1. Evaluation of types and quantities of radioactive waste based on a questionnaire to be sent to about 60 producers.

2.2. Analysis of the existing practice in Belgium, concerning treatment, storage and transport of this type of radioactive waste in Belgium, and comarison to national regulations.

C. Progress of work

In Belgium, waste production from small producers amounts to about 350 to 400 m^3/year. The total activity corresponds to about $3.7.10^{12}$ Bq (sealed sources excluded).

In a final report, the legal situation in Belgium has been analysed. Waste from small producers has to be taken over by ONDRAF/NIRAS, the national waste management agency; financially, collection and treatment of the small quantities of waste represents a heavy burden both for the producers and the agency, and schemes for more rational management are needed; improvements are proposed.

Elements of the final report are to be published later together with results from the other Task 6 contractors.

EXEMPTION OF RADIOACTIVE WASTES PRODUCED OUTSIDE THE NUCLEAR FUEL CYCLE IN SPAIN

Contractor : Empresarios Agrupados, Madrid, Spain
Contract No. : FI1W/0250
Duration of contract : September 1989 - March 1990
Period covered : September 1989 - December 1989
Project leader : R.A. Trujillano

A. Objective

The objective is to characterise the very low level radioactive wastes produced by Spanish medical centres and research institutions and to perform a radiological evaluation of the impact of its unregulated disposal as domestic or urban waste.

The final aim of this work is to perform a generic assessment of the feasibility of exemption as a starting point to plan a detailed program of actions, studies and decisions on the management and disposal of this type of waste.

B. Main milestones and progress

The first step was the selection of a representative sample from the producers of these wastes (both liquids and solids). Sealed sources were not in the scope of the study. There are 275 installations in Spain which are potential producers.

ENRESA, the Spanish radioactive waste management agency, collected the waste from 98 of those installations, whose productions cover 75% of the total.

A detailed questionnaire was sent to 64 installations (37 medical centres and 27 research centres and laboratories), being considered a reliable sample of the production of the ENRESA suppliers.

As a second step the collected data was filed, analysed and treated to characterize and quantify the produced radwaste.

Both steps were finished by December 1989. The remaining activities, planned for the first two months of 1990 are:

- Calculation of the radiological impact of the disposal of two streams of waste, taking into account different scenarios and practices.
- Comparison of results with the recommended individual dose limits and the collective dose targets.

C. Results : radwaste production and management

Fig. 1 and 2 show the production of the 98 installations whose wastes are collected by ENRESA.

The producers deliver to ENRESA 40% of their radwaste mass production. These wastes contain long-lived nuclides: C-14, H-3, Cl-36, Ca-45 and also some I-125. Most of them are mixed solids and organic liquids. The balance of the production is self-managed

by the producers. These wastes contain short-lived radionuclides: Tc-99m, I-125, I-131, S-35, Fe-59, Cr-51, P-32, Co-57, Co-58, Xe-133, Ga-67, Tl-201, In-111 and Y-90. The solids are disposed as urban waste (in the dustbin) after they have decayed for a certain period of time and the liquids (mainly biologicals) are diluted to obtain a concentration lower than the maximum allowable value and sent to the sewer.
Two streams were selected to perform the radiological assessment. These were biological solids (60kg) and organic liquids (15.570kg).

D. Conclusion

The main conclusions reached after reviewing the information of the questionnaires and the visits to some production centres, are the following:

- small producers annually generate a significant volume of radwastes of low radioactivity, consisting mainly of short-lived radionuclides;

- each production centre has its own segregation and management criteria developped during all the years they have used isotopes;

- ENRESA and the producers are working together in the definition of a methodology to standardize the handling of radwaste generated in their facilities;

- it has been noticed that small producers are generally inclined to let solids with short-lived isotopes decay below 100 Bq/g and them dump them as conventional waste;

- regarding liquid waste soluble in water, the practice is to retain them for decay and then dilute them below the limits of permitted activity and dump them in the sewer;

- solid waste with long-lived isotopes, liquids insoluble in water and organic scintillation liquids, are usually collected by ENRESA;

- it is necessary to continue to improve the definition and application of criteria on the activity limits to manage solid radwaste (application of the 100 Bq/g criterion or other equivalent practices);

- small producers consider that it would be convenient to provide them with more technical assistance to resolve and accelerate the management of their wastes;

- the total mass of radwaste generated annually by 98 radioactive material handling facilities that are ENRESA customers, is 47.347kg, of which 38% is generated as Mixed Solids, 2% as Biological Solids, 21% as Biological Liquids, 26% as Organic Liquids and 13% as Aqueous Liquids. These figures, when extended to include all 275 facilities that can generate radwaste, amount to 59,200kg of radwaste in total.

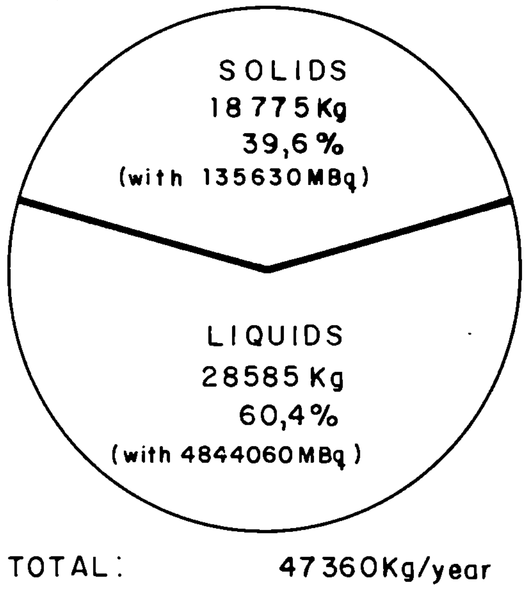

FIG. IV-5 RADWASTE ANNUAL PRODUCTION
(THE ACTIVITY VALUES ARE REFERED TO THE ORIGINAL ACTIVITY OF WASTE)

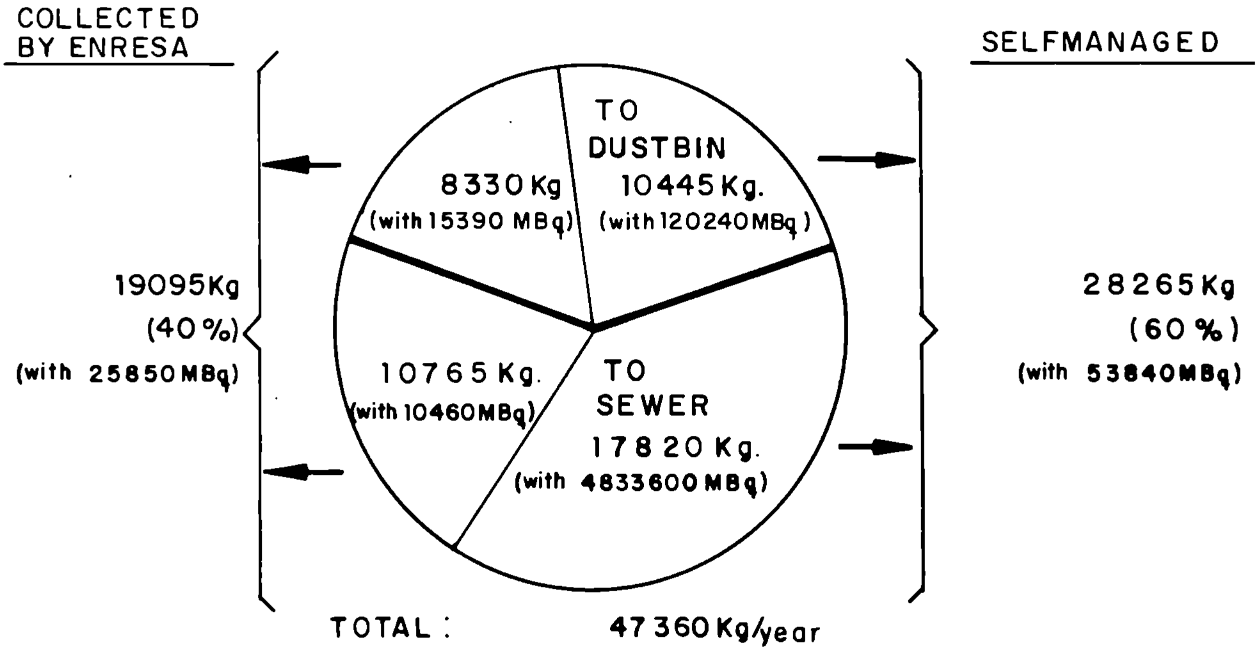

FIG. IV-6 RADWASTE DISPOSAL
(THE ACTIVITY VALUES ARE REFERED TO THE ORIGINAL ACTIVITY OF WASTE)

PART B

CONSTRUCTION AND/OR OPERATION OF UNDERGROUND EXPERIMENTAL FACILITIES OPEN TO COMMUNITY JOINT ACTIVITIES

The HAW Project at Asse

THE HAW PROJECT: DEMONSTRATION FACILITY FOR HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL IN THE ASSE SALT MINE

Contractor: GSF-IfT, Braunschweig, Federal Republic of Germany
Contract No.: FI1W/0003/D
Duration of Contract: from January 1985 to December 1989
Period covered: January 1989 - December 1989
Project Leader: T. Rothfuchs

A. OBJECTIVES AND SCOPE

Since 1968 the GSF has been carrying out research and development programmes for the final disposal of high-level radioactive waste (HAW) in salt formations. The heat producing waste has been simulated so far by means of electrical heaters and also cobalt-60-sources. In order to improve the final concept for HAW disposal in salt formations the complete technical system of an underground repository is to be tested in a one-to-one scale test facility.

To satisfy the test objectives thirty highly radioactive canisters containing the radionuclides Cs-137 and Sr-90 will be emplaced in six boreholes located in two test galleries at the 800 m-level in the Asse Salt Mine. The duration of testing will be approximately five years.

For handling of the radioactive canisters and their emplacement into the boreholes a system consisting of a transport cask, a transport vehicle, a disposal machine, and of a borehole slider will be developed and tested. The actual scientific investigation programme is based on the estimation and observation of the interaction between the radioactive canisters and the rock salt. This programme includes measurement of thermally and radiolytically induced water and gas release from the rock salt and the radiolytical decompositon of salt minerals. Also the thermally induced stress and deformation fields in the surrounding rock mass will be investigated carefully.

The project is funded by the BMFT and the CEC and carried out in close cooperation with the Netherlands Research Foundation (ECN). Additionally the French Agence Nationale pour la Gestion des Déchets Radioactifs (ANDRA) is participating in the field of dose and doserate measurements and of laboratory irradiation experiments and in situ inclinometer measurements and the Spanish Empresa Nacional de Residuos Radioactivos (ENRESA) is participating in the salt irradiation programme.

B. WORK PROGRAMME

B.1. Elaboration of the test plan and the supporting documents for the licensing procedure.
B.2. Development and procurement of the technical components for handling and emplacement of the radioactive canisters.
B.3. Procurement and installation of the data collection system.
B.4. Mining of the test field, drilling of the boreholes, installation of the measuring equipment and preparation of the emplacement of the canisters.
B.5. Test disposal including operation of two electrical reference tests and assessment of the technical components.
B.6. In situ-measurements of released water and gas from the salt, of thermally induced stress and deformation in the rock mass, and performance of seismic and ultrasonic measurements.
B.7. Accompanying and complementary laboratory investigations to ensure the transferability of the results to other salt sites.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The test disposal of highly radioactive canisters in the Asse salt mine will be performed in view of the planning, design and licensing procedure for a HAW repository in the FRG. In 1989 most emphasis has been given to the installation of technical components and the so called "Cold Training" of the canister handling procedure. Since late 1988 the two preceeding electrical reference tests in the heater boreholes A1 and B1 are in operation. Due to a delay in the licensing procedure emplacement of the radioactive canisters could not be performed and has been postponed to 1990/91.

Progress and results

1. Elaboration of the test plan

The test plan was prepared in 1984/1985 and the final version was issued in December 1985. It contains a detailed description of the scope, issues, and objectives and also of the test programme.

2. Development and procurement of the technical components

The installation of the handling system for the radioactive canisters was terminated in 1989 and handling of the canisters is now being exercised. The GNS-12 transport cask was licensed by the Bundesamt für Strahlenschutz (BfS). The Castor transport cask was to be modified to meet the requirements for the above ground interim storage of the radioactive canisters at the Gorleben site after termination of the tests at the Asse Mine.

The installation of both the canister guiding system (CGS) as well as the gap monitoring system (GMS) has been successfully completed for all boreholes. On September 13, 1989 the systems were approved by the licensing authority Bergamt and its consultant TÜV. With this approval the installation of the ECN equipment in the test field was finished and a period of intensive work was completed.

3. Procurement and installation of the data collection system

The data collection system (DCS) hardware components together with the software have been installed in the Asse mine. The system was approved by the authorities and is successfully in operation since November 1988.

4. Mining and preparation of the test field

The underground test field, consisting of two parallel galleries, each 60 m long, 10,5 m wide, and 8 m high, was completely mined in 1985. Until the end of August 1989 the liners in the emplacement boreholes were installed together with the necessary measuring equipment.

5. Test Disposal

The emplacement of the radioactive canisters was again delayed in 1989. One of the main reasons was the detection of traces of fissile material in the canisters which led to a revision of the licensing procedure. It is now envisaged to start the transportation of the canisters in late 1990.

6. In situ measurements

Already at the end of 1987 most of the instrumentation for monitoring stresses, stress changes, rock deformation, temperatures and gas release was installed. The set of instruments at the heater boreholes A1 and B1 was connected to the automatic DCS in summer 1988, and the instruments at the remaining emplacement boreholes in summer 1989. From then on the complete data set is continuously being monitored.

The major and the minor secondary principal stresses in the pillar between the two test galleries as determined from pressure readings are 20 MPa (approx. vertically oriented) and 4 MPa (approx. horizontally oriented) respectively.

Radial measuring pressure cells installed 12.3 m below the mine floor adjancent to the boreholes A1 and B1 reacted immediately on the start-up of heating and increased within 60 days after start-up of heating by about 6 MPa (see Figure 1).

The resulting horizontal creep rates in the test field above the heated test sites A1 and B1 increased from 24 mm/year before start-up of heating to 36 mm/year after start-up of heating but remained at 22mm/year at the non-heated test site A4 and B4.

The preliminary acoustic measurements for crack detection near the heater borehole B1 and in the pillar were completed. The heating activated the micro-cataclastic plastic flow of the salt considerably because of the gap between the salt and the borehole liner. This resulted in a decompaction of the salt and hence a decrease of the velocity of the ultrasonic waves. After the salt reached the liner a recompaction occured resulting in an increase of the velocity. This is shown in Figure 2.

Gas samples from the 46 boreholes available in the test field have been regularly taken. The boreholes located at the heated test sites A1 and B1 have been sampled every two weeks. Boreholes which are kept at ambient temperature did not show significant gas release. The gases CO_2 and CH_4 were detected at fairly low concentrations (below 400 vpm and 50 vpm, respectively).

In the heated boreholes A1 and B1, however, gases are detected at much higher concentrations. The equilibrium of the gas phase concentrations have already been reached in these boreholes. The detected levels are about 10000 vpm CO_2 and 800 vpm CH_4. The concentration of H_2 is low (<500vpm) but still slowly increasing. The hydrogen is attributed to a slow corrosion process. The gas-phase concentration of CO_2 depends strongly on the temperature of the surrounding rock salt and decreases with increasing distance of the measuring borehole from the heater borehole.

The salt samples which will be irradited inside the dummy canister have all been prepared. These samples are now stored at ambient temperature awaiting the start of the radiation source emplacement.

7. Accompanying and complementary laboratory investigations

Investigation of the gas release from salt samples of the pilot drillings of the emplacement boreholes and of other Asse salt samples are not yet finished.

The development of a computer-code for the modelling of the diffusion and migration of gas through rock salt due to the influence of gamma-radiation and heating was finished at the Rheinisch-Westfälische Technische Hochschule in Aachen. In order to calculate the behaviour of these gases in the vicinity of an emplacement borehole, data on the porosity, permeability, diffusity and the adsorption/desorption characteristics as a function of temperature and gas pressure are required. Such investigations of rock salt samples of different mineralogical compostion are part of the future programme.

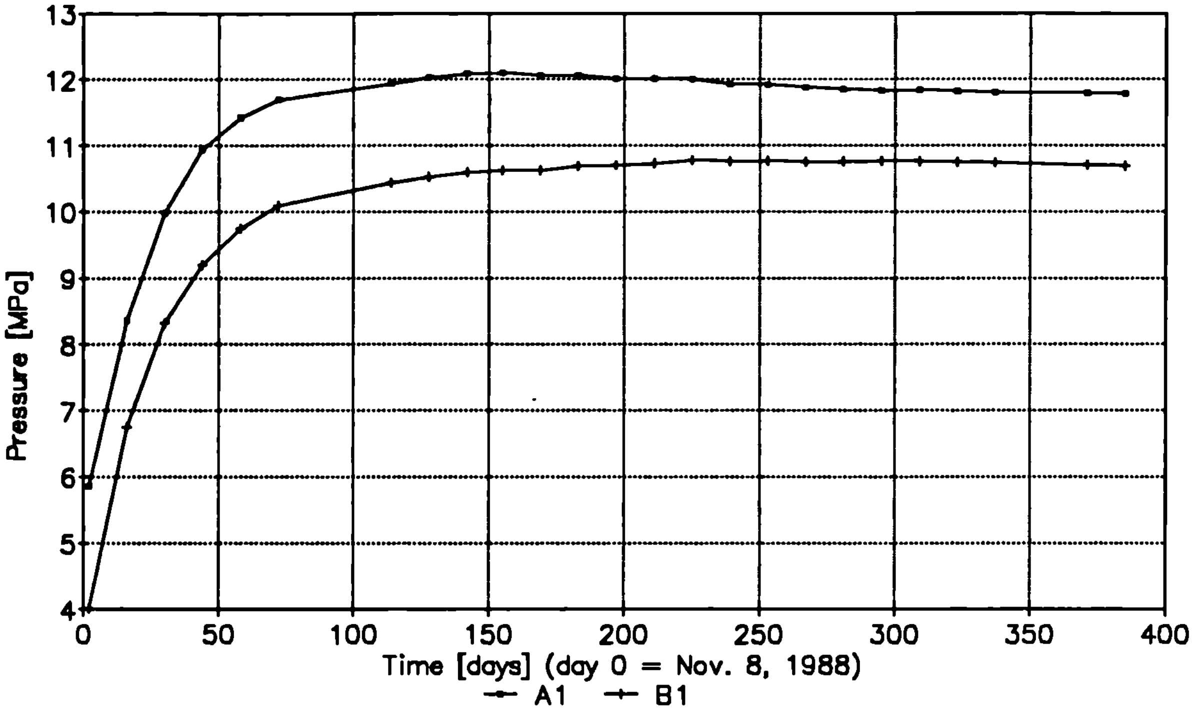

Figure 1: Increase of radial pressure at 7.5 m radius at heater boreholes A1 and B1

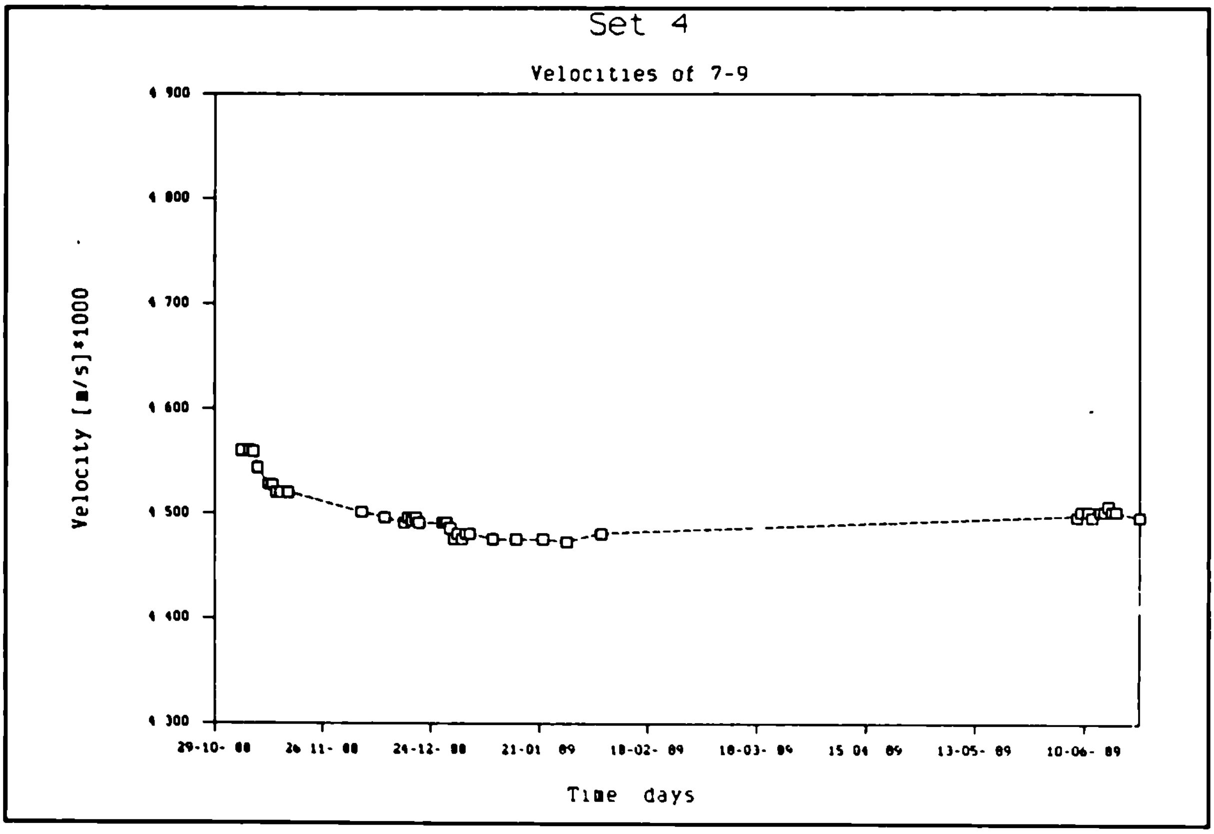

Figure 2: Changes of the ultrasonic wave velocities close to borehole B1

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IRRADIATION EFFECTS ON ROCKSALT

Contractor : ANDRA
Contract n° : FI1W/0199
Duration of contract : from Jan 88 to Dec 89
Period covered : from Jan 89 to Dec 89
Project leader : RAYNAL

A - OBJECTIVES AND SCOPES

ANDRA is involved in the Research and Development programme for the final disposal of radioactive wastes (HAW project) carried out by GSF-IfT in the Asse salt mine near Braunschweig (FRG). ECN (Netherlands) and ENRESA (Spain) are also part of this project which receive funds from the CEC. The ANDRA's participation consists of several topics :

- gamma-ray dose assessment (prediction and measurement),
- study of the radiolysis of salt,
- study of heat effects on the rockmass.

These experiments include:
- design and construction of special equipment adapted to the very demanding test conditions,
- in-situ measurements (dosimetry, inclinometry),
- laboratory investigations (irradiations, analysis, calculations).

ANDRA's subcontractors are CEA (Commissariat à l'Energie Atomique), IPG (Institut de Physique du Globe), LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique).

B - WORK PROGRAMME

1. Gamma-ray measurements

Gamma doses and dose rates are recorded in various location of the HAW test field or handling equipment using both solid-state dosimeters and ionization chambers systems.

2. Calculation of dose distribution

Theoretical calculations of gamma flux in the rocksalt surrounding the sources are performed using different computer models which take into account all mechanical, chemical and isotopic data of the test.

3. Parametric study of the radiolysis of salt

Salt samples from the Asse mine are irradiated in a French facility at different dose rates, integrated doses, grain size, temperatures, and under various initial gases. The products released are then analyzed.

4. Inclinometry measurements

High sensitivity measuring devices are installed around the test field, in a way to assess the displacements due to heating and to know the long-term mechanical behaviour of the rockmass.

C - PROGRESS WORK AND OBTAINED RESULTS

State of advancement

The recording systems for ionization chamber measurements are completed and partly installed. Thermoluminescent products for solid-state dosimetry are selected and calibrated, and the complete measuring devices are being manufactured. The calculation of gamma dose distribution started and the results of the first step will be available in January 90. Concerning the analysis of radiolytic gases, the complete procedure is now determined, the irradiation programme started for ambient temperature, and firts results are now available. The inclinometry measurement system, consisting of 3 groups of 4 tiltmeters + 1 temperature recorder, was installed in holes A.1 and B.1 in July 1988, before the heat starts. It operated first with a stand-alone data recording system, and was connected to the main DCS in November 89.

Progress and results

1. Gamma ray dosimetry
1.1. Ionization chamber measurements

Such measurements are performed at the surface level in transfer machine and at the test site in the guide tube and boreholes (- 800 m level), so two equipments were required:

- Surface level (transfert machine)

This equipment aims to record 2 profiles of activity for each glass canister when it is transferred from the transport cask to the mine cask. The ionization chambers were specially designed for the test conditions (size and temperature). This equipment was installed at the Asse in November.

- 800 m level (test site)

The recording equipment and the winch are mounted on a trolley. Three different ionization chambers are used, each with a separate cable and spool for the winch : in the guide tube both chambers (for radioactivity and for background) are used with a Teleflex type cable which allows to push the chambers into the tubes. In the boreholes a larger chamber is used with an ordinary twin-lead shielded cable .

1.2 Solid state dosimetry

Those dosemeters use the properties of certains crystals of emitting light after having been irradiated : the radio-thermoluminescence (RTL).
Selected products are Al_2O_3 (corrundum) and $CaSO_4$(Dy), they could be pre-dosed, in order to estimate the influence of temperature during irradiation. These dosemeters will be used :

- in the dummy canister above the sources,
- in the guide tubes and boreholes (mounted on a chain),
- in the transfer machine to measure the total activity of each source.

2. Calculation of dose distribution

The calculation started with some delay due to late modifications of the goals. Now, the first step of the works consist in calculate the energy deposited in salt using the MERCURE-4 model. It will provide an axial and radial mapping around the sources with a 5-cm mesh. The second step, based on TRIPOLI II model will give information on the gamma-energy spectrum at the location of the different measurements and inside the dummy canister. Final works will be devoted to interpretation of in situ dose measurements.

3. Radiolysis of salt

was devoted to test and qualify experimental procedures and to determine the irradiation programme. Samples consist of 200 g of salt sealed under controlled atmosphere in glass ampoules and irradiated by spent fuel elements. Gas analysis procedure includes :

- mass spectrometry (H_2, O_2, N_2),
- gas chromatography (CO, CO_2, N_2O, CH_4),
- ionic chromatography (Cl, SO_4, NO_2, NO_3)
- FTIR (Fourier Transform Infra-Red) only for some samples.

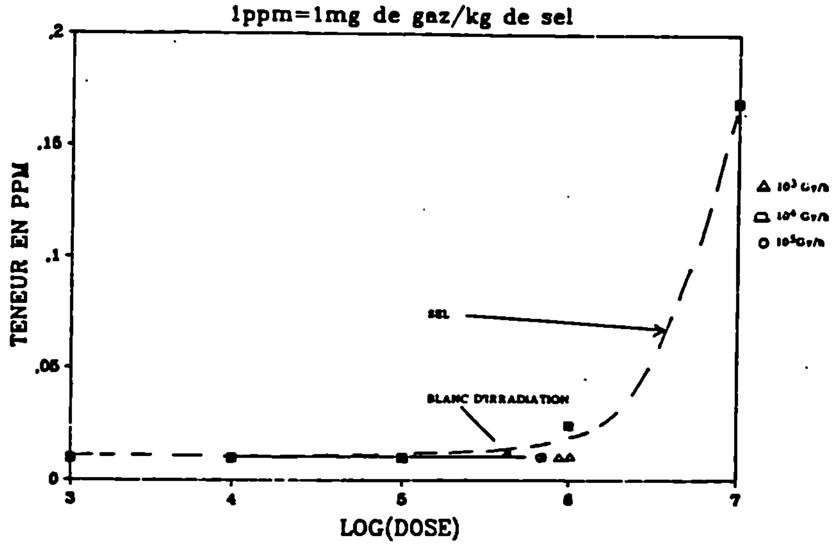
The results available by the end of 1989 (see figures attached) lead to the following conclusions :
- gas production is not depending upon dose rate, but is related to the total dose,
- under neutral atmosphere (N_2, H_2, Ar), the gas production due to radiolysis is very poor,
- with synthetic air, the main gases are CO_2 and N_2O. Their amount increases as the dose increases. These gases are not significantly produced when oxygen is not available,
- at a constant dose, the gas production increases with temperature,
- The gas production decreases as the grain size increases.

4. Inclinometry

This part of the contract, signed later in August , aims to measure the movements of the deep rocksalt submitted to heating, using high resolution tiltmeters. Twelve of such tiltmeters were installed in three locations surrounding the test field. They were emplaced three months prior to start the heaters in holes A.1 and B.1, in a way to observe the behaviour of the rockmass without external stress. Since November 1989 they are connected to the DCS and are monitored as any other sensor in the test field. The curve shows that after the heating started the creep rate increased strongly and remains constant after one year of measurements.

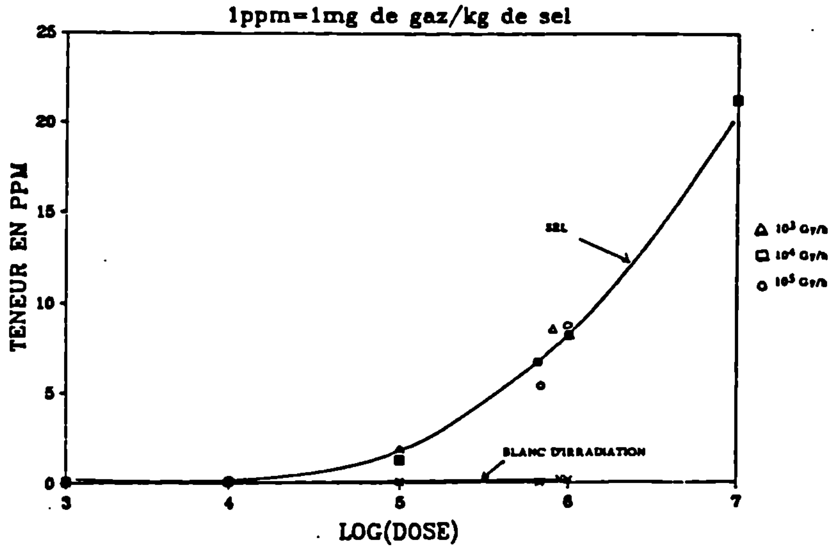
GAZ DE RADIOLYSE: SO_2

1ppm=1mg de gaz/kg de sel



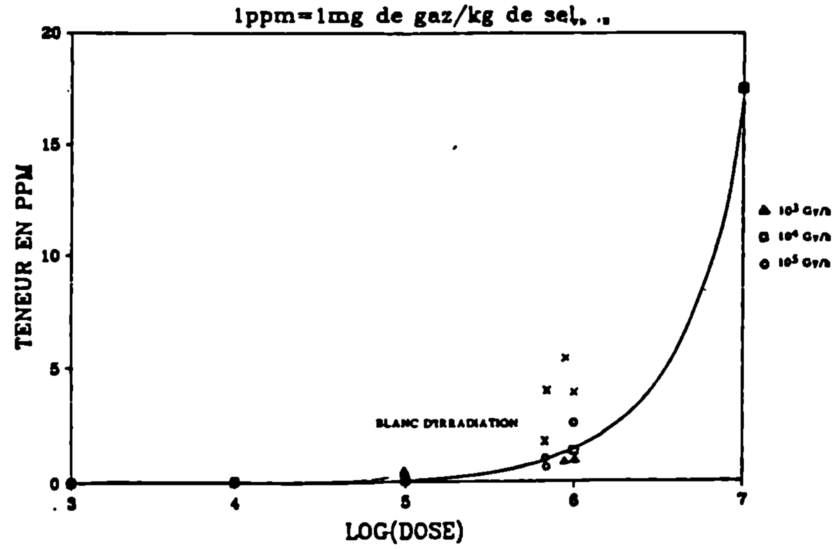
GAZ DE RADIOLYSE : CO_2

1ppm=1mg de gaz/kg de sel



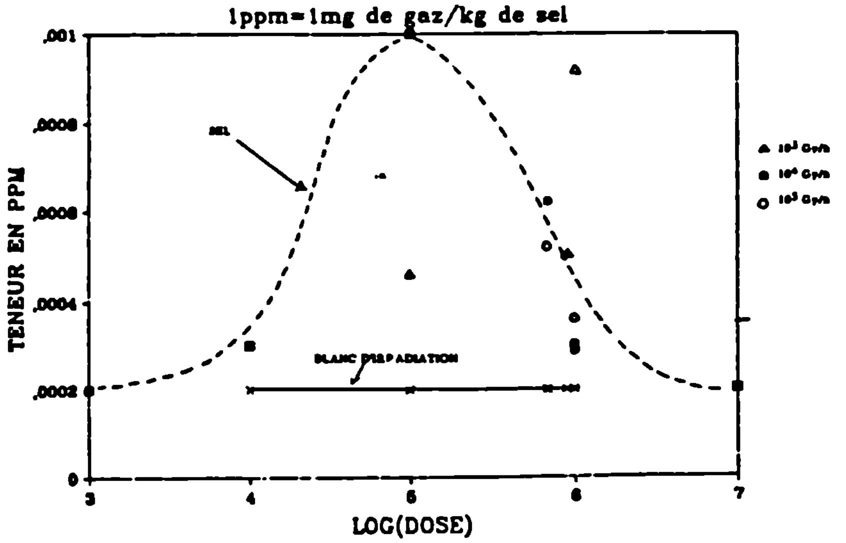
GAZ DE RADIOLYSE: N_2O

1ppm=1mg de gaz/kg de sel



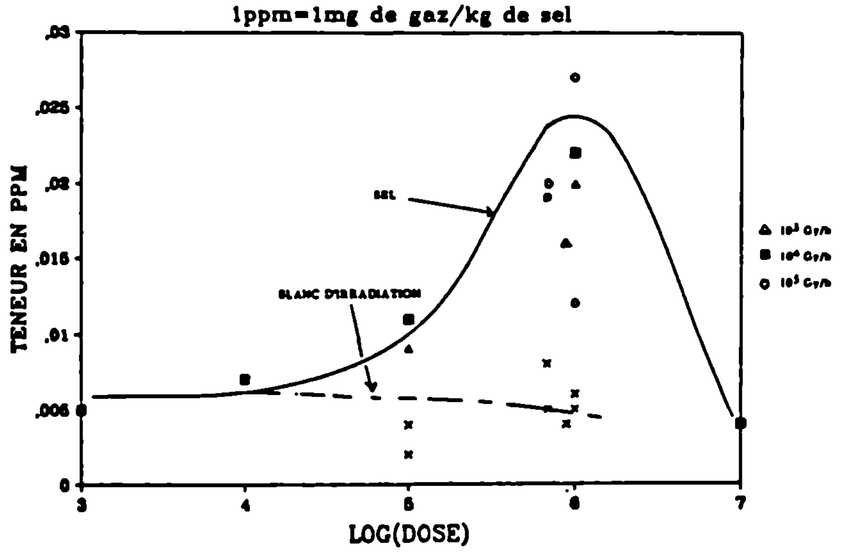
GAZ DE RADIOLYSE:CH4

1ppm=1mg de gaz/kg de sel



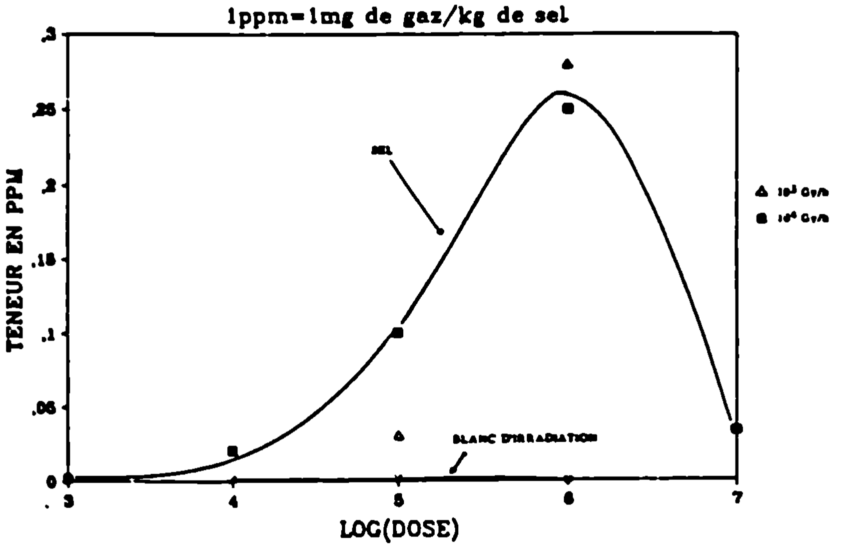
GAZ DE RADIOLYSE:H2

1ppm=1mg de gaz/kg de sel



GAZ DE RADIOLYSE:CO

1ppm=1mg de gaz/kg de sel



TEXTURAL AND FLUID PHASE ANALYSIS OF ROCK SALT SUBJECTED TO THE COMBINED EFFECTS OF PRESSURE, HEAT AND GAMMA RADIATION

Contractor: ENRESA, Spain
Contract No.: FI1W-0235-E
Duration of Contract: November 88-June 90
Project Leader: Fernando Huertas

A. OBJECTIVES AND SCOPE

Previous studies have shown that gamma irradiation of salt creates defects in the crystal lattice, primarily, the generation of F and H centers; under appropriate conditions, these defects can, eventually, give rise to the formation of colloidal sodium and trapped molecular chlorine. The formation of colloidal sodium gives rise to stored energy which could be suddenly released, originating a significant temperature increase. This is a main concern of irradiation damage with respect to the safety aspects of nuclear waste disposal in salt formations.

Due to differences in potential energy between neighbouring crystals, grain boundaries tend to move at the cost of the most energetic crystals; this recrystallization process, enhanced by the presence of brines, clears the crystal from irradiation damage.

The main goal of the project is to check the hypothesis that fluid assisted recrystallization can occur in gamma irradiated natural rock salt, and to evaluate the phenomenon, under different total dose, dose-rate, temperature and time of gamma irradiation.

The research work is being done in cooperation with the Netherlands Energy Research Foundation (ECN), the University of Utrecht and the University of Barcelona. The salt irradiation will be conducted at the HAW test field at Asse (FRG).

B. WORK PROGRAMME

1. Sampling of salt specimens from spanish sites and the HAW test field; preparation of synthetic polycrystalline and Harshaw samples.
2. Chemical, petrological and mineralogical characterization of the salt samples. Solid and fluid phase determination, chemical evolution from step heating and microstructural analysis.
3. Textural analysis of undisturbed samples.
4. Preparation of samples and pressurized containers for the irradiation phase.
5. Irradiation of 180 samples in the HAW test field at Asse.
   Samples will be irradiated from 6 months to 5 years, at different total dose, dose-rate and temperature, in pressurized containers, with different brine contents.
6. Irradiation of salt specimens at the High Flux Reactor (HFR) (Petten).
7. Post-irradiation analysis to investigate the amount of stored energy and the variation in chemical composition, water content and microstructure.
8. Theoretical calculations, on the basis of the Jain-Lidiard model, of the damage which could be expected by irradiation and heating.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

Salt samples from the Sallent mine (Spain) have been obtained by airflushed rotary drilling.

The preparation of both natural and synthetic salt samples is almost completed. Pressurized containers are already available at ECN.

The drilling fines and core samples from the Sallent mine have been geochemically characterized.

The irradiation system of the HFR facility (Petten) has been modified for a better temperature control of the samples.

Progress and results

Core samples had been analyzed for Cl, $SO_4^=$, Ca, Mg, K, Sr, Br, and Li, by AAS, ICP, XRD and volumetric methods. Water content has been analyzed by thermogravimetry. Salt geochemistry and water content data are summarized in Table I.

Brine composition has been determined using two methods:

a) Overall method by substracting Mg, K and Br contents of two aliquots. One aliquot was dry milled and the other was milled in alcohol. The difference, after this procedure, was related to the water content, obtaining the brine composition.

b) Analysis of frozen fluid inclusions by SEM-EDX

Brine composition ranges, determined by overall analysis, are:

12.9 to 85 gr/kg, for Mg
3.2 to 43.1 gr/kg, for K
0.3 to 8.8 gr/kg, for Br

Chlorine and $SO_4^=$ can not be determined by this method

Brine composition ranges, from individual fluid inclusions, are:

4.41 to 5.52%, Cl
1.56 to 2.18%, Na
0.56 to 0.78%, Mg
0.38 to 0.64%, K
up to 0.20%, $SO_4^=$

Gas contents have been analyzed both in the laboratory and in situ. Laboratory analysis gave the following results:

872 l/m^3 of rock, for O_2
5 l/m^3 of rock, for CO_2
0.12 l/m^3 of rock, for CH_4

In situ gas determinations, of the rock mass, show the presence of hidrogen, ethane and propane, besides the aforementioned gases, their content being, nevertheless, below the method quantification limit.

Natural salt samples have been machined and/or polished to perfect fit the pressurized sample holders after been wrapped in silver foil for isolation purpose. Each pressurized holder will contain one 40 mm long natural salt sample and a 10 mm long Harshaw crystal. Thin sections of the samples are made for microstructural analysis.

Stored energy measurements in the Sallent mine salt samples have been performed, prior to gamma irradiation, by Differential Thermal Analysis (DTA) in a SETARAM calorimeter. Based on the results obtained, it can be stated that the stored energy values, on the Sallent salt light material (more pure), ranges from 0.1 to 1.6 J/gr, with a brine content ranging from 0 to 0.4% (in weight); in the dark material the stored energy value found ranges from 2.8 to 9 J/gr, with a brine content ranging from 0.3 to 0.9% (in weight).

Four irradiation experiments have been performed at the HFR facility of Petten. Due to some difficulties in holding the temperature at the 100ºC set and with container contamination, it was decided to modify the system. A new system is already available for this irradiation back up experiment. Nevertheless, stored energy measurements on the irradiated samples have been performed, by the same method as the one described above for methodology calibration.

The same irradiated samples have been used for microprobe observations to see whether gross chemical composition differences could be found No difference in composition has been found, either between differently damaged sample parts nor between irradiated and non-irradiated samples. It has to be noted that microprobe resolution, in the way which has been applied, is only in the 100 ppm range.

Recrystallization experiments in Sallent irradiated salt samples has been initiated.

TABLE I: GEOCHEMICAL COMPOSITION AND WATER CONTENT

	Cl (%)	SO4 (%)	Ca (%)	Br (ppm)	K (ppm)	Mg (ppm)	Sr (ppm)	Li (ppb)	Water (%)
MEAN VAL.	57,95	1,96	0,96	60	112	428	230	720	0,32
ST. DEV	1,51	1,32	0,60	5	32	99	162	280	0,20

The HADES Project at Mol

THE HADES PROJECT : A PILOT FACILITY IN THE ARGILLACEOUS LAYER BENEATH THE NUCLEAR SITE AT MOL

Contractor : SCK/CEN, Mol, Belgium
Contract No. : FI1W/004/B
Duration of contract : from January 1985 through June 1990
Period covered : January 1989 - December 1989
Project leader : A.A. Bonne

A. OBJECTIVE AND SCOPE

In 1974 SCK/CEN launched a R&D-programme concerning the possibilities for disposal of high level solidified and alpha-bearing radioactive wastes in a continental stratiform clay formation (Boom clay) situated below its own site. Site investigations, safety studies, repository design, conceptualisations and in situ research confirm progressively the favourable characteristics of the host rock and the site for disposal of radioactive wastes.

Many particular areas require further studies and technological tests on a larger scale and in situ demonstrations under realistic conditions. These technological tests, studies and demonstrations will contribute to increase the confidence in the technical practicability, the economical feasibility and the safety of the disposal option in deep clay.

The direct demonstrations deal with the constructibility of real scale galleries without particular conditioning of the rock, the choice and dimensioning of a realistic lining and support system, the interaction between the underground structures and the immediate geological environment (e.g. the influence of heat and radiation), the handling of hot and radioactive canisters, the backfilling and its behaviour in time, the performance of various system components during the operational phase and of monitoring systems.

Within the HADES project a technological test related to a gallery lining technique according to the convergence-confinement principle is performed by ANDRA (France) (see contract FI1W/0112) and geotechnical interpretation is made by GCG (see contract FI1W/0200).

B. WORK PROGRAMME

The demonstration/pilot phase of the HADES project is scheduled in two phases, which are complementary to each other and may be developed in parallel.

B.1. Phase I : the construction and operation of a test drift (TD) with tests related to :

B.1.1. Mining technology (digging, lining, extrados backfilling, rheology).

B.1.2. Radioactive waste disposal (experimental emplacement, backfilling, degradation of waste matrices and migration of radionuclides, in situ irradiation of clay, thermo-mechanical behaviour of clay and gallery structures, monitoring and auscultation systems).

B.2. Phase II :

B.2.1. The construction of a pilot facility with a new shaft and extended gallery, connecting chamber and utility structures.

B.2.2. Tests and observations on handling, emplacement, backfilling and retrieval of dummies and finally of acutal radioactive wastes.

(The performance of B.2. is scheduled beyond the present contract period).

C. STATE OF PROGRESS AND RESULTS OBTAINED

After having installed the CERBERUS-test equipment and several safety related items in the Test Drift in 1988 the principal facts related to 1989 concern the loading of CERBERUS with the Co-sources and starting of the heating.

The general auscultation of the time dependent behaviour of the clay host rock around the Test Drift and its lining were continued and confirmed the excellent geotechnical behaviour of the whole structure.

Validation exercises of the rheological models for simulating the geotechnical behaviour of the Boom clay and the lining during tunnel construction and thereafter were continued. Further, design efforts and parametric studies were made for the planned large scale gallery heating test (GHT, a separate or combined test).

Progress and achievements

1.1. Mine-by test

Systematic measurements were going on for providing information about the time dependent behaviour of the whole structure of the Test Drift and its immediate surroundings. No major changes occurred as far as deformation and pore water pressure in clay are concerned.

The average diametrical convergence versus time of the 7 measuring sections of the concrete lining is presented on figure 1. The rings 15 to 52 behave similarly with diametrical variations of 48 to 53 mm after 24 months. The evolution of the convergence of the rings 71 to 105 is similar in spite of remaining at a lower level.

The "convergence-confinement" of the lining was followed and the maximal convergence remains close to 1 %. A stabilisation of the pressure seems to be near. The extensometric measurements in the end front of the Test Drift indicate a failure in the clay between 2 and 4 metres behind the front cask.

1.2. CERBERUS test

The start of the Cerberus test previously foreseen in January 1989, was postponed until October 1989. This decision was based on the slow dissipation of the perturbations caused by drilling and emplacement of Cerberus equipment. The calculation tool MPGSTN was adapted to estimate the pore pressure evolution around the borehole. A consolidation coefficient of 10 m^2/y was taken into account instead of 1 m^2/y obtained by the labo-tests and which cannot explain the pore pressure observations around digging and drilling works in the Boom clay. The results indicate that one year is almost necessary to reach 98 % of the equilibrium. This waiting period was used to perform controls of the data acquisition system, in situ calibration tests and training exercises for the transport of the Co-60 sources and its loading into the Cerberus device.

The control of the data acquisition system has shown several failures of the Glötzl system for pressure measurements (e.g. printer fault and build-up of the zero pressure point of the transmitter placed in the MFA unit). This equipment could only be replaced during the month of September which obliged in the mean time to perform manual measurements for the follow-up of the in situ ground pressure build-up. Up to now, two cells did not function properly, probably due to the failure of the valve or the presence of an air bubble into the cells which could not be flushed away by the pump.

Preliminary tests with a Co-60 source and an Am-Be neutron source were carried out to get more information about density, homogeneity of the backfill material and dosimetry measurements.

The transport procedures were tested out as well as the loading of the Co-60 source in the Hot-cell of BR2 to fit the safety regulations and to allow the operators to be familiar with the equipment. The unloading in the underground facility was tested and followed by audio and videomonitors from the surface. The carrier of the container was adapted, container and elevator painted and foreseen with marks for smooth handling and good positioning.

The first Co-60 source was placed on 17.10.1989 and the following five the week after at 1 source/day. The transport and emplacement was very successful. A measuring campaign was performed after emplacement of the source and before heating which started on the 27th of November. The total activity reached 397 TBq distributed homogeneously ($\pm$ 10 %) into a cylinder with an active height of 0.540 m and an active diameter of 0.02 m.

1.3. Gallery Heating Test

Within the present contract it is aimed at to define the test, as a separate or combined test, to settle construction specifications (if construction of a new gallery section is required), measuring devices, emplacement procedures, perform mathematical predictions and establish an appropriate planning.

Two emplacements would be suitable for the test in the present configuration of the underground facility : the first option being the existing first 20 m of the Test Drift and the second option a lengthening of the small exploratory gallery, build in 1984. Advantages and inconveniences of both emplacements (technically, planning and manpower) were examined.

It was concluded that, from a technical point of view, the best emplacement for the test would be the latter option, its advantages being significant : construction and instrumentation being appropriate in function of the test, interferences with other tests limited and mathematical modelling easier.

Thermal computations of the GHT for the latter option were performed, in order to choose the different measuring devices, by making use of two different codes and with the assumptions on material properties based on the actual state of our knowledge. Temperatures up to 90 °C may be expected, as well as significative variations in pore water pressure ; deviatoric effective stresses, on the other hand, should be limited, so that failure in clay should not occur.

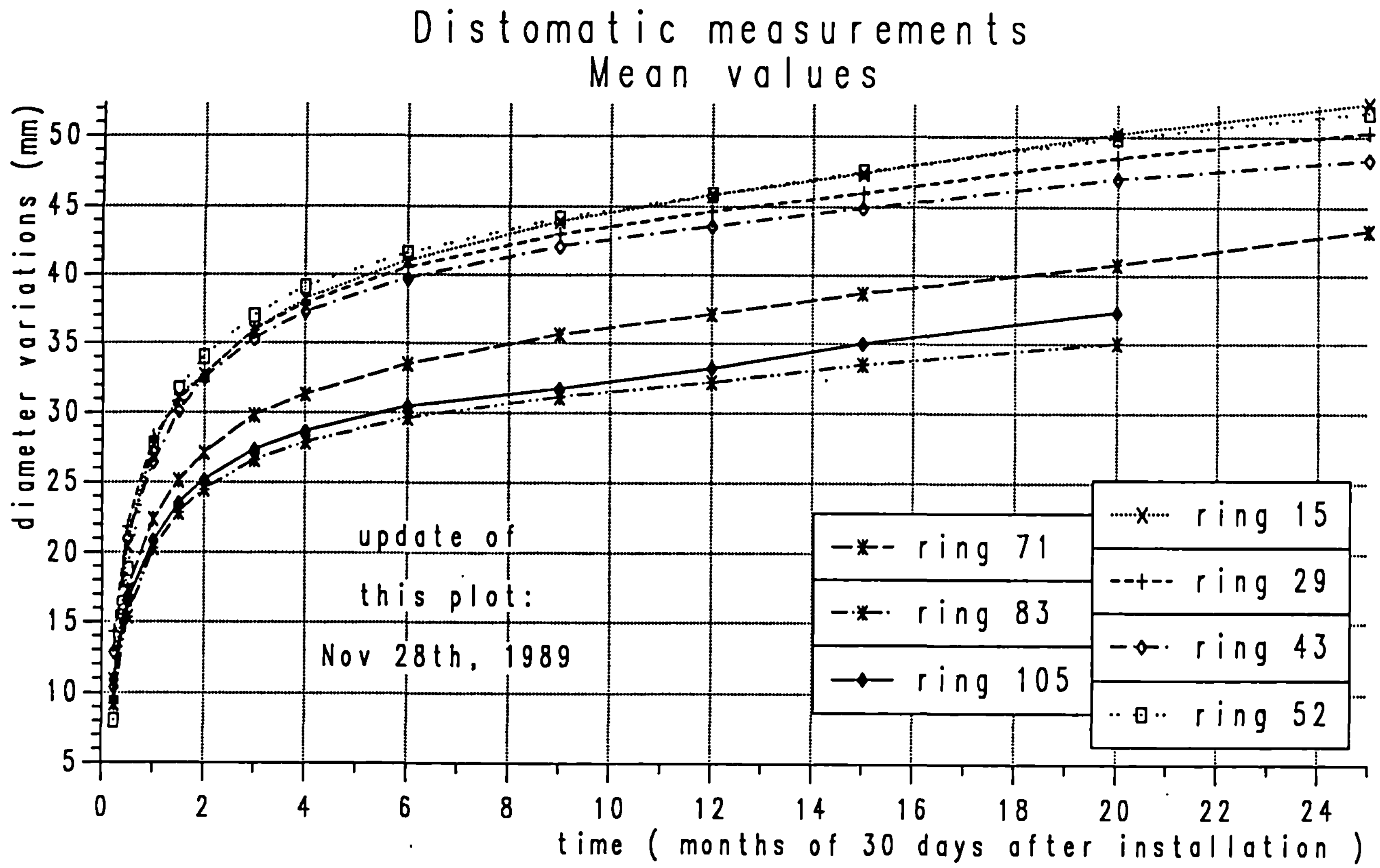


Fig. 1. Convergence of the lining versus time

DIMENSIONING OF LINING OF GALLERIES EXCAVATED IN DEEP CLAY FORMATIONS

Contractor : ANDRA, PARIS, FRANCE
Contract n° : F I1W/0112
Duration of contract : January 87 - December 89
Period covered : January 89 - December 89
Project leader : M. RAYNAL

A - OBJECTIVES AND SCOPE

The dimensioning of lining of galleries in deep clay formations depends directly on the long term stress undergone by the support.

The mechanical characteristics of the clay formation could be used to optimize the lining. It seems possible to minimize the pressure in the support, by allowing the excavation wall to converge enough with time. Different factors can be considered to reach this objective, as the time lag for the rock to come to contact with the support or the flexibility of the lining.

The objective of this work is to implement a lining wich answers this criterium. It is made of steel ribs showing a significant stiffness with sliding devices adapted at the joint elements, allowing the convergence. THis configuration will test the closure - confinement concept : the convergence of the wall prevents the confinement to rise above a certain threshold. Moreover this thin lining presents practical advantages i.e. the handiness and ease of transport, the rapidity for building and the small volume of material to dig and dispose.

The conception of the project and the study of the rockmass-lining behaviour are conducted by LMS (Laboratoire de Mécanique des Solides de l'Ecole Polytechnique). Engineering department of SIMECSOL was responsible for the instrumentation and measurements during construction, collaboration of CEN/SCK for surveying was since requested.

B - WORK PROGRAMME

1. Build up of a 12,5 m long test gallery in the continuation of a concrete drift made by the CEN/SCK. A transition zone of 2 m with concrete archstones prevent any perturbation of the CEN gallery.

2. Measurements of :

   - closure of implemented rings,
   - sliding of the ribs,
   - rib strain,
   - pressure on the outer face lining,

- displacement inisde the clay formation,
  during and after excavation, for 24 months at least.

3. Interpretation of results

C - PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The excavation, lining and instrumentation of the gallery were performed from 10/20/87 to 12/03/87. A buffer zone was set up at the end of the drift. It consists of a portion of a circular gallery 2 m long lined with concrete archstones. Its end is completed by a hemispheric front stabilized with gunite (figure 1). The measurement started during the digging and is still underway.

Progress and results

2. Measurements

   Some measures are already automatically taken. The frequency of the others was daily during the excavation phase, weekly during the first month after the completion of the work, bi-monthly during the 6 next months (1st semester 1988), then monthly. It will be quarterly at 01/01/90.

   . Closure of complemented rings : a little ovalization in the vertical direction can be observed in the four ribs (figure 3).

   . Sliding of the ribs : in a first time after digging (3 months) slidings are importants, then the phenomena's evolution is quite moderate. The sliding of the ribs are depending on their distance of the stabilized front (figure 2). Nine months after the gallery's construction the maximal convergence reaches 2,1 %, and today, 21 months after, 2,4 %.

   . Rib strain : KOVARI's theory is used to estimate the normal load and the bendings moments, related to the strains by an elastoplastic model of steel behaviour. The critic normal force for the first sliding has a value of 500 kN the same that predicted value.
   The normal force is about 1 050 kN and the bending moments about 50 kN. The maximum values equal 50 % steel plastic limits.

   . Pressure on the outer face lining : it is measured by Gloetzl cells disposed on the interface clay-backfilling. The pressure increases quickly as soon as the rib has been set and during one month, then its average value is quite constant and less than 1 MPa. These values seem however underestimated ones, as those derived from rib strains give 1.6 MPa.

   . Displacement inside the clay formation with extensometer : they increase in accordance with pressure measured by Gloetzl cells.

They are the smaller than the excavation is away (at 10 meters there is no displacement).

3. Interpretation of results

- The lithostatic stress can ben considered almost isotropic.

- The average confinement pressure is about the third of this stress after 21 months. The experimental curve of confinement (figure 4) pointed out that pressure and closure become in a long run less and less important, but the stabilization is not yet reached.

- The Gloetzl cells don't give exactly this confinement pressure (the measures are 66 % of the value calculed by ribs strains).

The process permits rock mass movements without appearence of too large plastic strain (the lenght of the plastic zone reaches 5 meters from gallery's wall).

REFERENCE

/1/ BUBLITZ D. "Essai de soutènement à convergence contrôlée - Compte-rendu de mise en oeuvre - Interprétation des mesures (november 87 - July 89)" CCE half yearly report (contrat n° FI1W/0112) December 1989

/2/ ROUSSET G. "Comportement mécanique des argiles profondes - application au stockage des déchets radioactifs" - Thesis ENPC July 1988

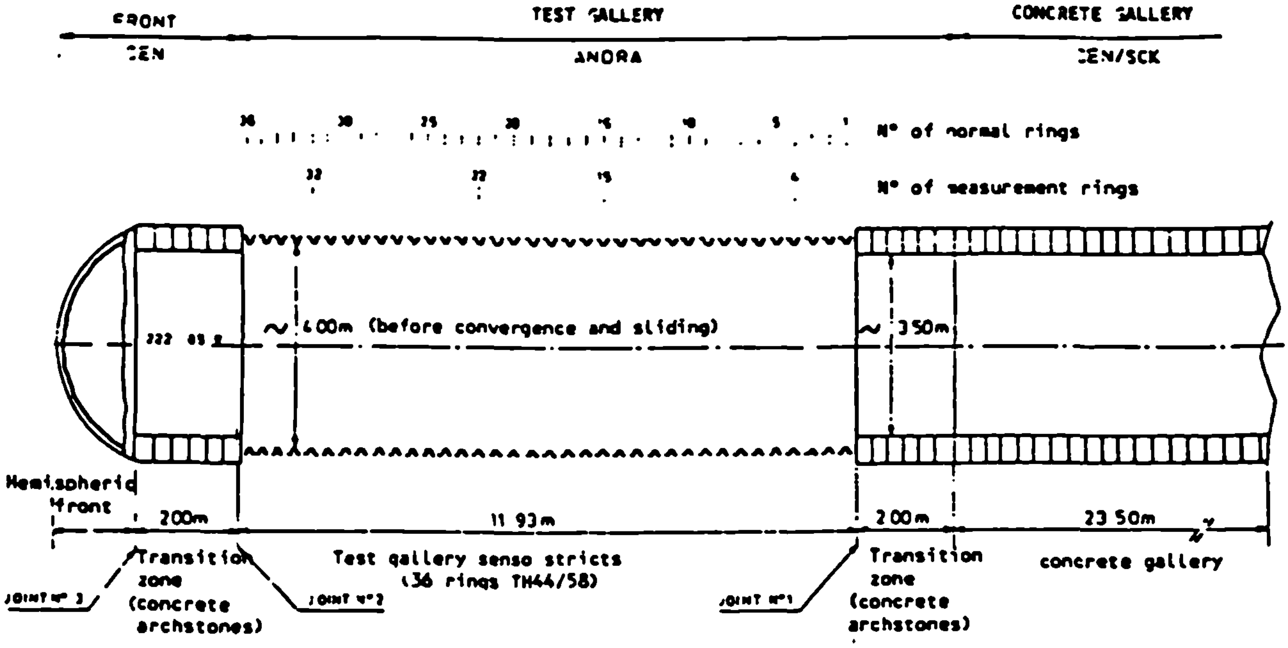


Fig. 1 : ANDRA TEST GALLERY

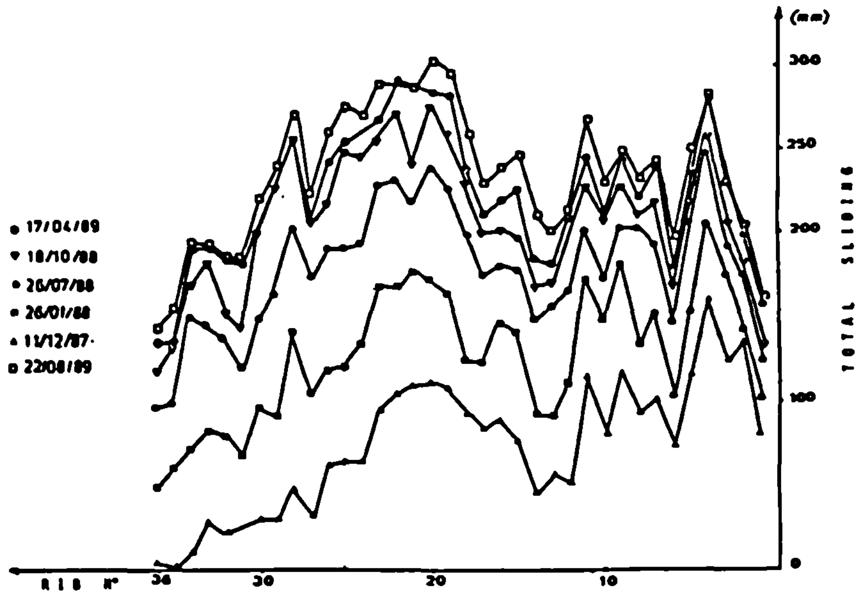


Fig. 2 : AVERAGE SLIDING

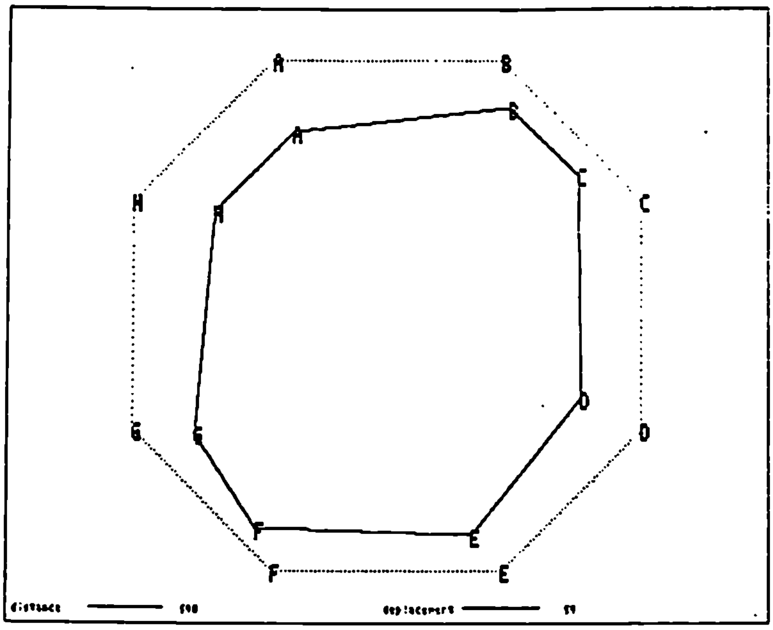


Figure 3 - Ovalization rib n° 15

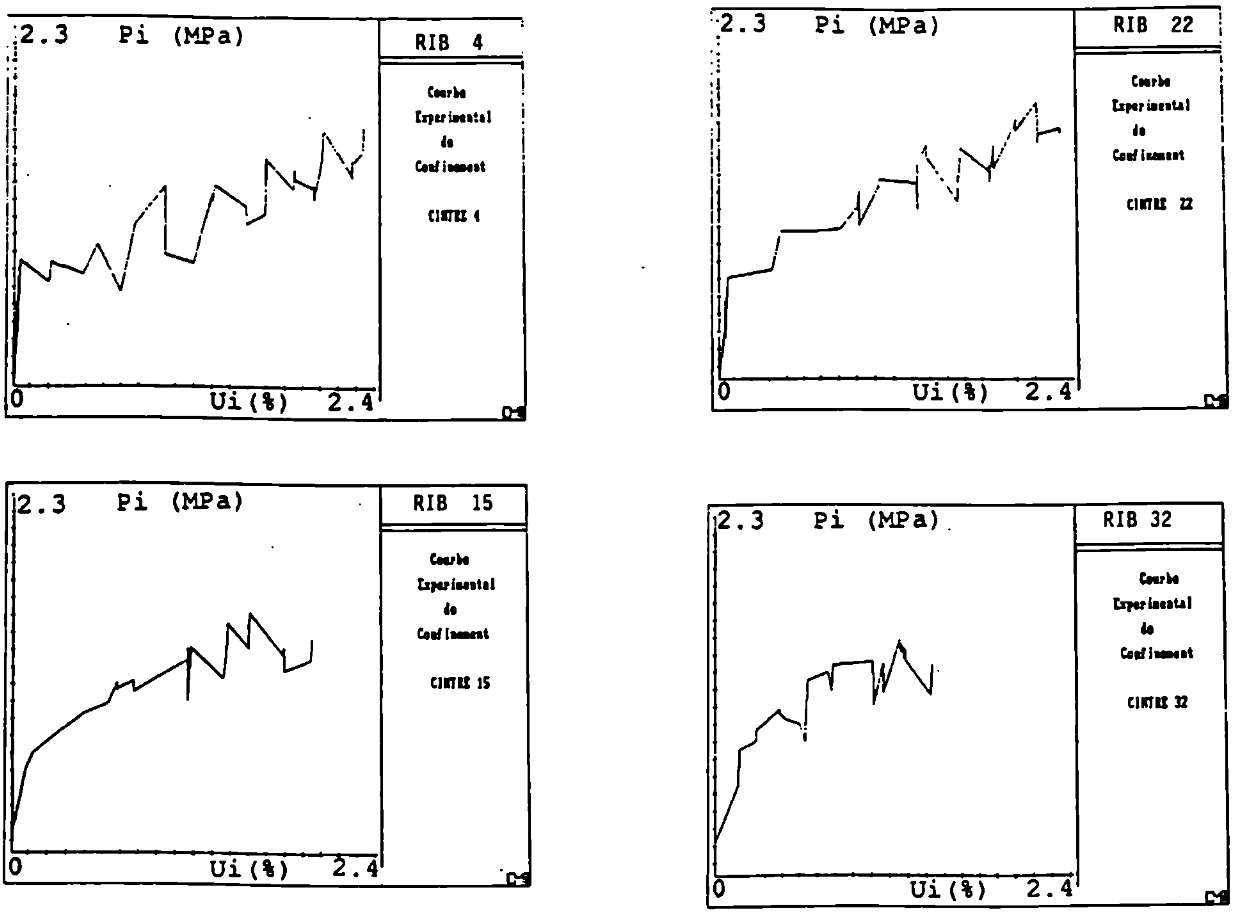


Figure 4 - Experimental confinement curves

REPOSITORY TUNNEL CONSTRUCTION IN DEEP CLAY FORMATIONS

Contractor: Geotechnical Consulting Group, London, UK
Contract No.: F1lW/0200
Duration of contract: February 1988 - January 1991
Period covered: January 1989 - December 1989
Project Leader: Dr R J Mair

A. OBJECTIVES AND SCOPE

At Mol, Belgium, the feasibility of constructing a repository at large depth in a clay formation is being investigated (HADES project). The most recent construction has been a test drift of a length of 40 m and an internal diameter of 3.5 m; 60 cm thick concrete block segments were used for linings. As an extension to the test drift, a 12 m long gallery has been constructed using sliding steel ribs as linings. Extensive measurements of lining loads, pore-pressures and ground movements have been made in the test drift.

The objectives of this project are as follows:

(i) Analyse and interpret available measurements made at the HADES project at Mol in respect of tunnel construction.

(ii) Carry out laboratory stress path testing on samples of Boom clay taken during the construction of the test drift at Mol.

(iii) Research and develop a self-boring retracting pressuremeter (SBRP) as an in-situ testing device for use in hard clays, with specific reference to tunnel lining design.

B. WORK PROGRAMME

The work programme consists of the following activities:

1. Data Analysis and Interpretation: (a) synthesis of data (b) plasticity calculations (c) finite element analyses (d) interpretation and generalization of data in respect of possible deep repository construction in clay formations.

2. Laboratory testing: (a) development of a high pressure computer controlled stress path triaxial testing apparatus (b) testing samples of Boom clay.

3. In-situ Testing:

3.1 Develop a prototype self-boring retracting pressuremeter (SBRP).

3.2 Carry out field tests in the UK with the SBRP in appropriate geological materials such as mudstones.

3.3 Depending on results, carry out tests from the gallery or test drift at Mol.

C. PROGRESS OF WORK AND OBTAINED RESULTS

State of advancement

The further available measurements made at Mol on lining loads, pore-pressure and ground movements have been synthesized and interpreted. Plasticity calculations and finite element analyses have been undertaken of an advancing tunnel heading at great depth in a clay formation corresponding to the test drift construction at Mol. Plasticity solutions have been used to interpret stiffness of the clay from measurements of ground movements beneath the experimental shaft and drift at Mol. They have also demonstrated the sensitivity of the predicted ground deformations around the test drift to the assumed values of strength and stiffness. Further finite element analyses have been undertaken with effective stress soil models to investigate the effects of different soil behaviour, lining permeability and the long-term build-up of radial stress on the linings.

The high pressure computer controlled stress path triaxial testing apparatus developed for the project has been used to perform a number of compression and extension tests on high quality samples of Boom clay. Detailed stress-strain measurements have been made.

The SBRP has been designed, working drawings produced and the first prototype instrument manufactured. Extensive calibration tests have been undertaken in the laboratory and a first series of field tests has been carried out. Further field tests will be undertaken in appropriate geological materials in the UK, such as mudstones, but not at Mol as originally considered.

Progress and results

1. Further plasticity calculations and finite element analyses of the test drift have been performed, aimed at interpreting the behaviour of an advancing tunnel heading in clay /1/. The plasticity calculations have been based on the simplifying assumption that a tunnel heading can be approximated as one half of a thick sphere; cylindrical symmetry is used for conditions remote from the tunnel face. For the finite element analyses, axisymmetric conditions were assumed and the actual test drift construction sequence was modelled as closely as possible. The soil parameters adopted were based on an assessment of the currently available laboratory test results on Boom clay, including the latest data from the special tests at The City University.

The new finite element analyses of the test drift construction have used effective stress soil models, which allow long term conditions to be assessed. Predicted lining pressures and convergence developed in the short term are in reasonably good agreement with the field measurements. Predicted long term lining pressures are significantly lower for permeable linings than for impermeable linings; in both cases, the pressures are considerably lower than the total overburden pressure. The analyses indicate that no significant increase of lining pressure with time will be observed in the test drift at Mol.

Finite element predictions of the development of radial ground movement as the test drift advances are compared with measurements in Figure 1. Agreement between the predictions and measurements at a radius of 3.2 R_e is reasonably close, but there is less good agreement between the predictions and measurements at a radius of 2.8 R_e.

As part of the analysis and interpretation, there has been participation in the pilot benchmark exercise INTERCLAY organized by the CEC; this was to compare predictions of calculations using different soil models for well-defined problems related to excavation in Boom clay at Mol. For this exercise, effective stress soil models were used.

2. A high pressure computer controlled stress path triaxial testing apparatus has been designed and assembled at The City University. A number of compression and extension tests have been performed on high quality block samples of Boom clay taken from the face of the test drift. The stress-strain curves from these tests are reproduced in Figure 2. Detailed behaviour of the Boom clay has been obtained, including measurements of stiffness at small strain levels. The parameters measured in these special laboratory tests have been used in the more recent finite element analyses.

3. The design and construction of the first prototype SBRP has been completed at the University of Newcastle Upon Tyne. Its purpose is to measure in-situ stiffness and ground response of soil and weak rock when they are unloaded from in-situ stress conditions, with specific reference to shaft or tunnel lining design. To achieve this the instrument should be installed with minimum disturbance to the surrounding ground by a self-boring process; it then must be capable of being retracted under controlled conditions so that measurements can be taken of the change in stress and associated displacement.

A prototype instrument has been calibrated in an extensive series of laboratory experiments. Preliminary tests have been carried out in the field in an overconsolidated stiff clay. These have been successful in proving the retracting mechanism of the device to operate satisfactorily. A high pressure instrument has been designed and will shortly be constructed.

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Preliminary analysis of construction of the test drift in Boom clay at Mol using plasticity solutions and finite elements.
CEC Technical Session, Brussels, 1-2 December 1988, Proceedings (CEC Report EUR 12027 EN/FR, 1989) pp 127-139.

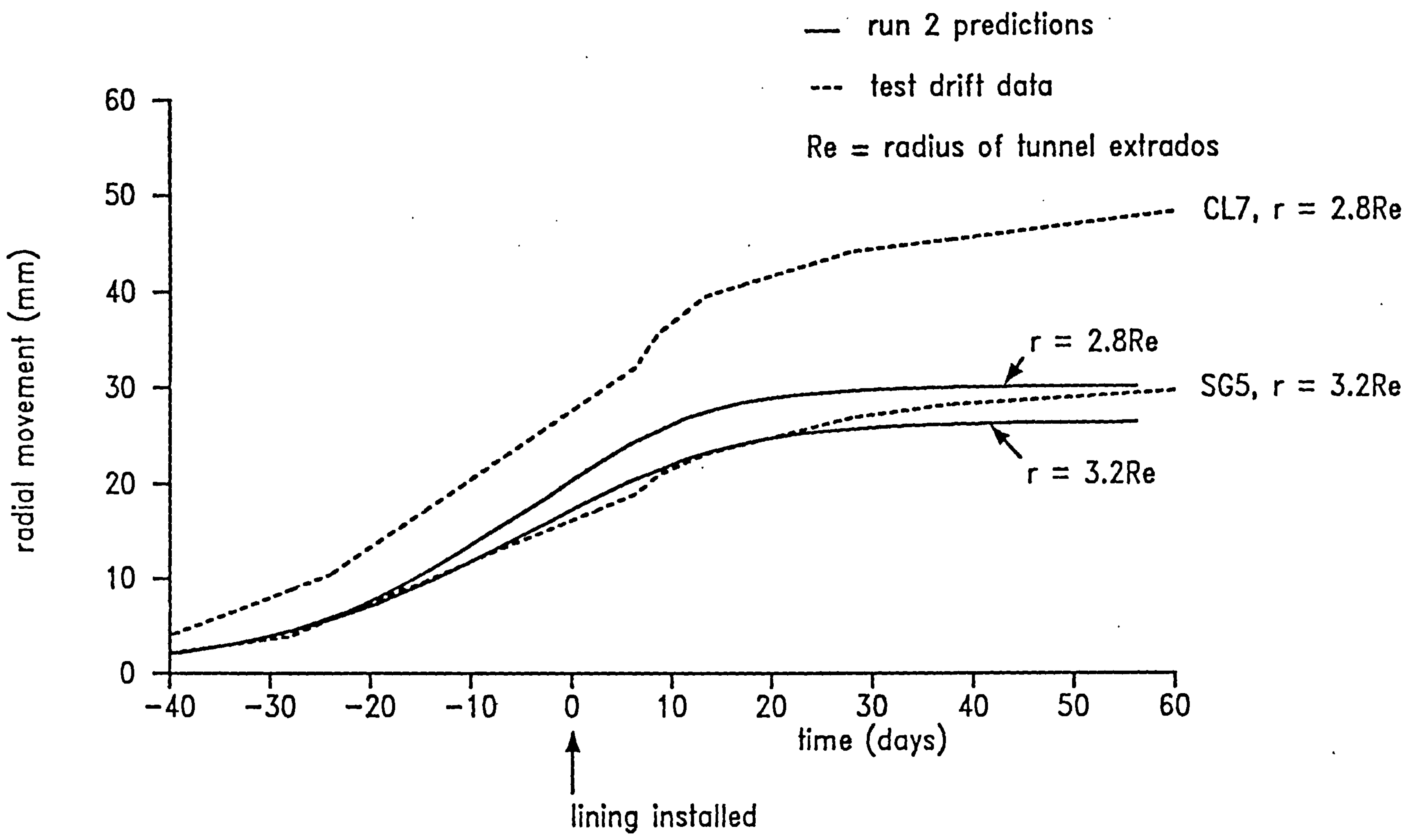

Figure 1 Comparison of predicted soil movements with measurements during construction of the test drift at Mol

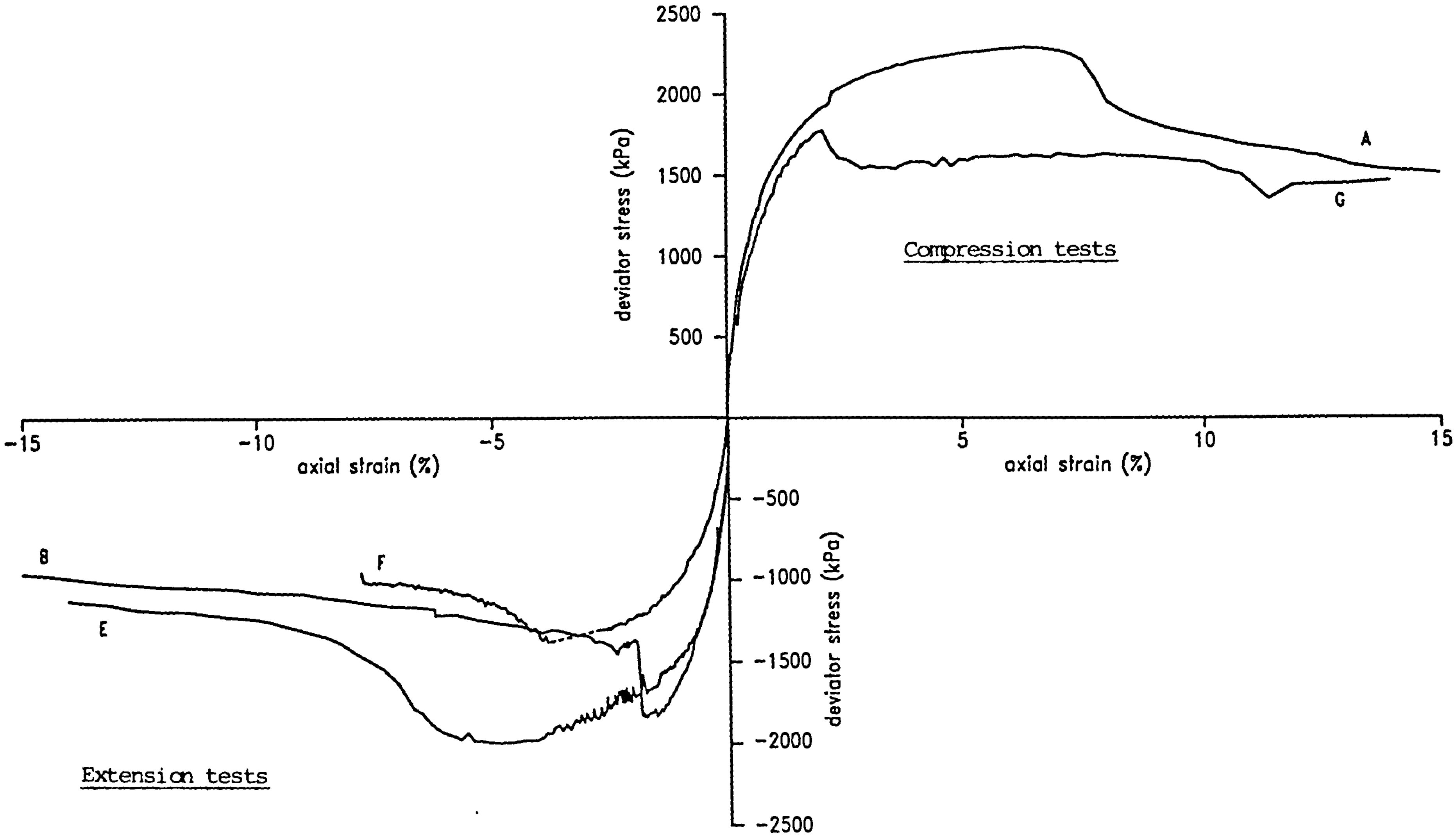

Figure 2 Stress-strain data from the triaxial stress path tests undertaken at high pressure on block samples taken during construction of the test drift at Mol

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