LABORATORY SCALE MODELLING
OF THE FINNISH REACTOR WASTE DISPOSAL SYSTEMS

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ABSTRACT

The aim of the work is to model experimentally the inner constructions of two reactor waste repositories. The modelling of the multibarrier systems was started on a laboratory scale using the same principal materials as in the Finnish reactor waste disposal concepts. Migration of cesium, cobalt and strontium was studied in various combinations of the following materials: bituminized resin, cemented resin, concrete, crushed rock and water. Sample preparation is described and activity profiles of some samples are demonstrated in the paper. The maximum sample age was 224 d. Distinct diffusion was observed with strontium in crushed tonalite and rapakivigranite, and with cesium in crushed rapakivigranite and in concrete. Cobalt showed no mobility.

INTRODUCTION

A necessary part in the development of numerical models is validation. This document is a description of the initial experimental phase in the validation of REPCOM (1), a release model for underground reactor waste repositories.

The repositories are going to be constructed by the two Finnish nuclear power companies, Imatra Power Company (IVO) and Industrial Power Company Ltd (TVO) (2). The repositories will be constructed at the power plant sites, to be commissioned druing the year 1992.

Migration of radioactive tracers is being studied by using small samples which contain materials present in the planned reactor waste repositories.

The main results presented consist of the first activity profiles. The validation computations will start in the year 1987.

SPECIMEN MATERIALS

The principal construction and filling materials in the reactor waste repositories will be concrete and local crushed rock. The waste forms in this study are bituminized resin (TVO) and cemented resin (IVO). The resins are granular. The water types are simulated and natural groundwaters which were balanced with concrete. The tracer elements are cobalt, cesium, and strontium.

As a mutual factor, all the barrier materials, and cemented resin were wetted as perfectly as possible - by using evacuation when necessary. The maximum grain size in every material was limited to 2 mm.

The repository for IVO will be located at the level -120 m. It will consist of separate tunnels for dry waste and a cavern for cemented waste.

The numerical release model, REPCOM, describes IVO's repository as shown in Fig. 1. Cemented resin will be molded into concrete containers which will be disposed in the cavern. The void volume between the containers and the bedrock will be filled by using local crushed rapakivigranite. The walls of the cavern will be injected and shotcreted. The repositories are described in greater detail in another paper in the proceedings beginning on page xxx (Härkönen & Tusa, "LLW Management by Finnish Power Utilities).

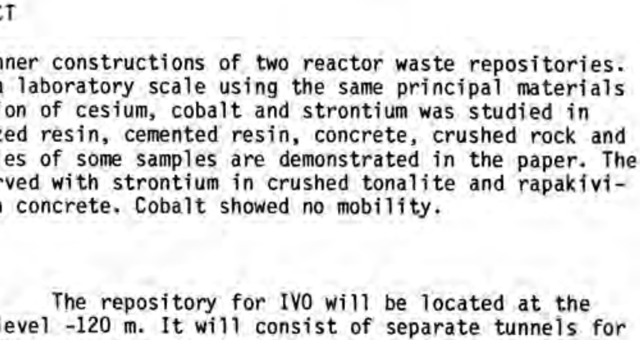


Fig. 1. IVO's reactor waste repository concept as described in REPCOM-model.

The following composition was used for the simulated container wall and the shotcreted concrete:

30 w-% low temperature cement; Kolari LH
54 w-% crushed granitic rock;
16 w-% water.

The porosity of the backfill material, crushed rapakivigranite, was 26 %, which indicates a broad grain size distribution. The loosely bound tracers were washed away when labelling the crushed rock. The water content of crushed rapakivi was 17 w-% or 32 v-% after washing. Washing removed about 4 w-% of rock material.

The repository for TVO will be located between the levels -50 m and -100 m. It will consist of a waste vault with two silos. A scheme of the vault can be seen in Fig. 2. Bituminized resin in steel drums will be emplaced in one silo and dry waste in the other. The silo for bituminized waste will be provided with an extra barrier consisting of a 500 mm thick reinforced concrete wall. When sealing the repository, all the empty space around the concrete structures will be filled with crushed tonalite. The void volume between the drums will be allowed to become filled with water.

Steel and bedrock are excluded in the samples. Water serves as an activity sink in the study instead of the bedrock.

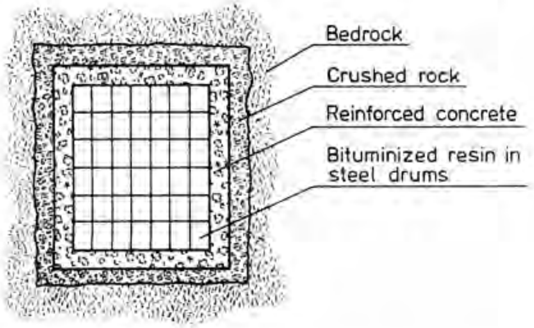


Fig. 2. A simplified scheme of TVO's underground silo for bituminized waste.

The composition of the simulated silo wall concrete was as follows:

26 w-% cement; low temperature Kolari LH
64 w-% crushed granitic rock
10 w-% water
0.1 w-% ligninesulfonate

The porosity of the backfill material, crushed tonalite, was 40 %, which indicates a narrow grain size distribution. The loosely bound tracers were washed away after labeling the crushed rock. The water content of wet crushed tonalite was 20 w-% or 39 v-%. The washing did not remove any significant amount of rock material.

Waste forms

Cemented resin represents IVO's waste form in this work. Cemented waste was simulated using the following composition:

19.6 w-% wet resin; water and resin surfaces even.
Volume ratio between the anion and cation exchange resins:1/1
Anion exchange resin : Duolite ARA
Cation exchange resin : Duolite ARC 9351
0.6 w-% boric acid
0.0 w-% tracers Cs-137, Co-60, Sr-85, and Sr-90
2.4 w-% calcium hydroxide
60.5 w-% blast furnace cement
16.9 w-% water

Bituminized resin represents TVO's waste form in this study. Bituminized waste was simulated using:

33.5 w-% dried cation exchange resin Duolite ARC 9351 in H^+ form
16.5 w-% dried anion exchange resin Duolite ARA 9366 in OH^- form
50.0 w-% bitumen BIT-45

Wet resins were first mixed and then labelled with Co-60, Cs-137, Sr-85, and Sr-90. They were dried at 140°C for 16 hours. Dried resin and bitumen were mixed together and molded at 110°C.

Tracers

Not only the waste products themselves, but also crushed rock and water were labelled with radioactive nuclides. The chosen tracers were Co-60, Cs-137, Sr-85, and Sr-90. Only Sr-90 is beta active. It was considered suitable because the half life of gamma-active Sr-85 is too short for long term testing.

Tracer solutions had to be concentrated enough to avoid excessive amounts of carrier compounds. Thus it was easier to minimize the precipitation of tracers, because the maximum solubilities were not exceeded as rapidly. The basic properties of the tracer solutions are given in Table I.

TABLE I

Properties of the tracer solutions.

Isotope	Carrier	Solution	Activity conc. in May 1986
			(MBq/ml)
Co-60	$CoCl$	0.5 M HCl	37
Cs-137	$CsCl$	0.1 M HCl	37
Sr-85	$SrCl_2$	9 mg $NaCl$/ml	37
Sr-90	$Sr(NO_3)_2$	1 M HNO_3	3.5

The doses of each γ-nuclide were between 2...8 MBq/specimen. Sr-90 doses were 0.2...0.8 MBq/specimen.

When labelling the crushed rock the final activity depended strongly on sorption. Tracer solution was added to the mixture of crushed rock and deionized water. An initial test showed that crushed rock shifted the pH of deionized water to 7.6 with tonalite and to 9.0 with rapakivigranite. Tracers were in contact with the crushed rock for one week. Finally the rock was rinsed with water.

Resins for the bituminized waste and for the cemented waste were labelled by injecting the tracers directly into the wet resin. The resins were not rinsed.

The tracers were directly injected to the concrete waters in those cylindrical samples which contain labelled water in their own sections.

Waters

The waters, except the water for making concrete, were in contact with crushed concrete for one week. The crushed concrete to water weight ratio was 0.1.

Natural groundwater from Hästholmen, IVO's site, was used for IVO's specimens. Allard water, a synthetic groundwater (3) was used for TVO's specimens (Table II). The water compositions before equilibrating with concrete are given in Table II.

TABLE II

WATER COMOSITIONS (mg/l)

Component	Natural groundwater	Synthetic groundwater
Bicarbonate	80	120
Calcium	670	18
Chloride	5000	70
Fluoride	1.5	-
Iron	0.2	2...3
Magnesium	280	4.3
Potassium	25	3.9
Silica	10	12
Sodium	2300	65
Sulphate	610	9.6

Other materials

The casings for the cylindrical samples were made of hard PVC.

Concrete blocks were either glued or molded into PVC-tubes. The walls of the concrete boxes for cubic samples were assembled by glueing them together. Two epoxy resins, Araldit Standard and Araldit Rapid (reg. trademarks of Ciba Geigy AG, Switzerland), were chosen for that purpose. Initial tests showed no obvious degradation of resins after being immersed in equilibrated water for over eight months.

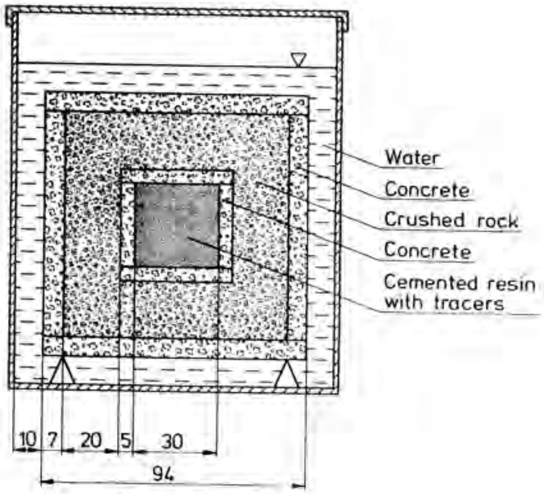
Nets of phosphoric bronze and polyamide were used for immobilizing the crushed rock in the water. Some nets were kept inplace with springs made of stainless steel.

SPECIMEN TYPES

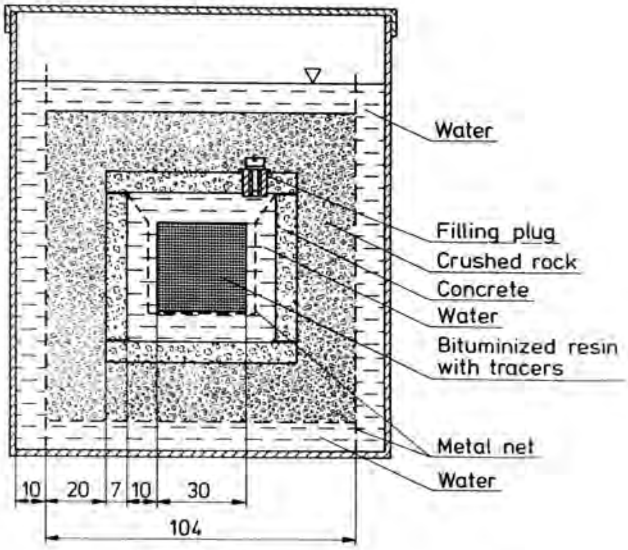
The main specimen categories were:
1) cylindrical specimens for one-dimensional numerical modelling and
2) cubic specimens for three-dimensional modelling.

The specimen types are presented in Figs. 3 and 4.

In the case of the cylindrical specimens (Fig. 3) all the materials were packed sequentially into PVC-tubes. Concrete blocks were either molded or glued into the tubes. The glued blocks were first cut with a diamond saw from bigger concrete blocks. They were wet by keeping them first in vacuum and by allowing water to penetrate into the pores. All the waste form sections were molded directly into the tubes. Bulk waters could be removed through the plugs when it was needed to monitore them separately.

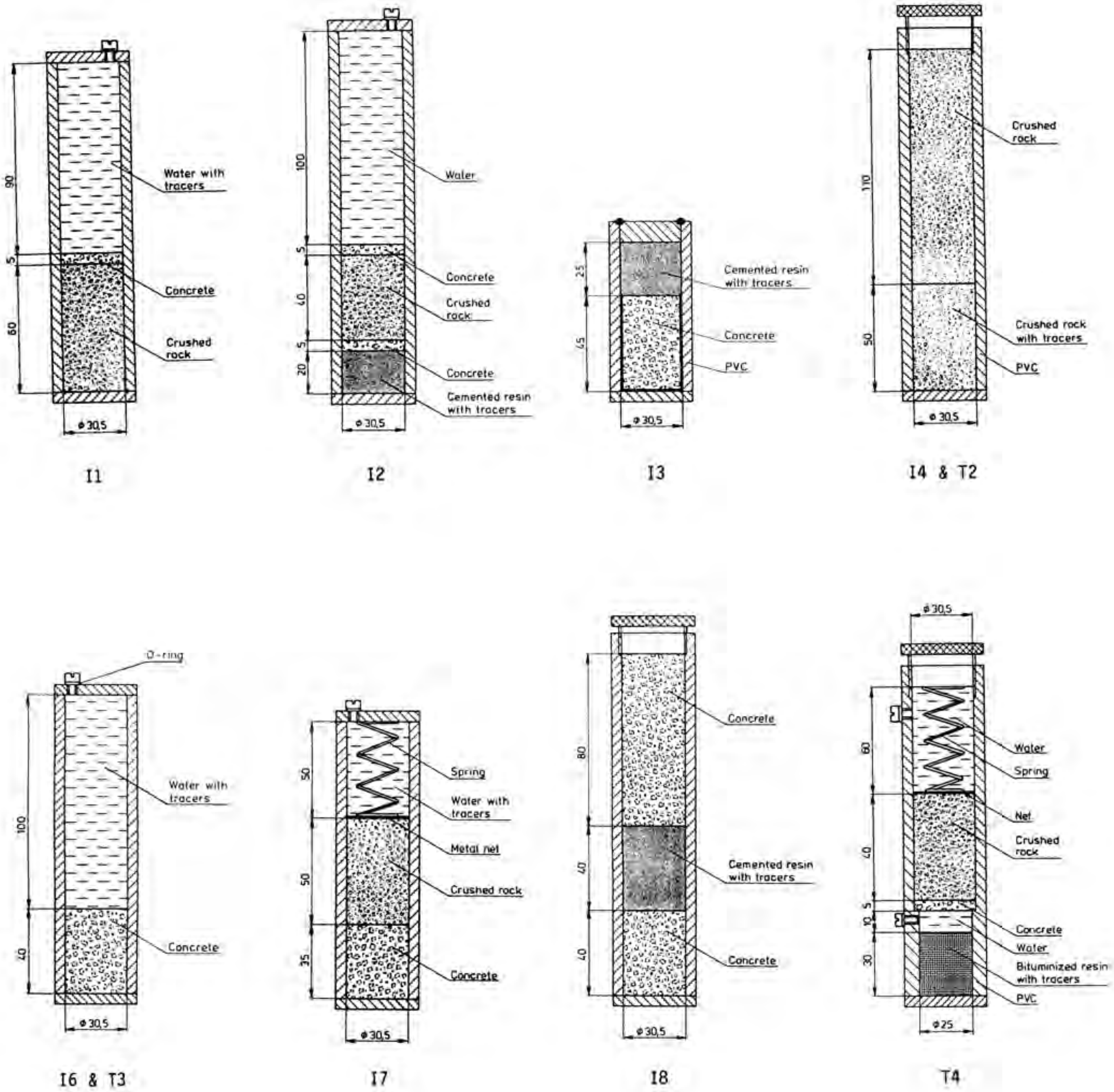


Sample I5



Sample T1

Fig. 3. Cubic specimens.


I1 I2 I3 I4 & T2

I6 & T3 I7 I8 T4

Fig. 4. Cylindrical sample types. The dimensions given are considered as averages. The axial distances in the activity profiles were measured from the outer bottom of the sample tubes.

In the cubic geometry (Fig. 4) the specimens consist of material layers inside the other. Concrete boxes were assembled by glueing concrete plates together.

The material thicknesses were chosen on the basis of analytical computations.

MEASUREMENT METHODS

The activity profile of each tracer isotope in every sample was measured periodically. There were different measurement methods for gamma and beta active specimens. The cylindrical gamma-active samples could be measured through the container wall with a scanner (Fig. 5). The sample had to be destroyed in order to determine beta-activity profiles.

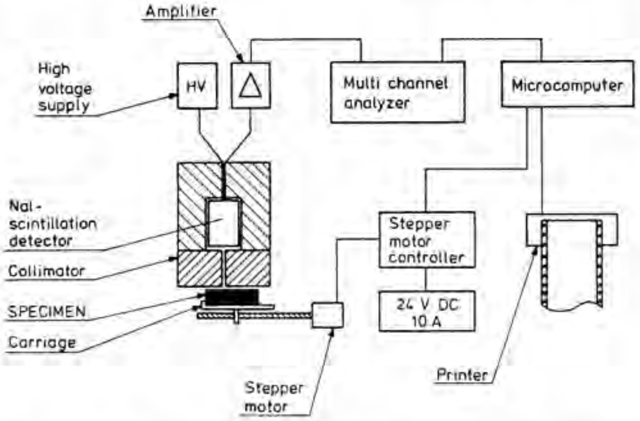


Fig. 5. The activity profile measurement system for gamma-active tracers. The sample can be positioned with a good accuracy in front of the collimator. The collimator opening was 1 mm.

No beta-activity profiles were measured before the WM'87 meeting.

MEASUREMENT RESULTS

The main information was received in the gamma-activity profiles. Because of the large number of measured profiles only a selected collection was presented and described in this paper. A larger collection can be found in Ref. (4). The comments are based on visual examination of the activity profiles.

All the cylindrical gamma-active specimens were measured at least once. Some activity profiles are shown in Figs. 6 through 10. The relative or absolute net count rate versus the axial distance from the bottom of the specimen is plotted. The vertical lines in some profiles represent the boundaries between the different materials. The measured points were simply connected with lines. Thus the given profiles do not always provide the precise activity values at areas of abrupt change.

Besides the fluctuation typical to radioactive decay, there can also be seen fluctuation caused by the uneven tracer distribution in barrier materials (cobalt in Fig. 7) or in waste forms. Deviation in Sr-85 profiles increased rapidly with time because of the short half-life, 64 d.

Cesium penetrated the first concrete layer of the sample I2 as shown in Fig. 6. The profile was measured 64 days after the molding of the 5 mm thick concrete layer. It is not yet possible to see if cesium would be noticeably more mobile during the setting of concrete than later. Cobalt and strontium did not seemingly move in concrete in any sample.

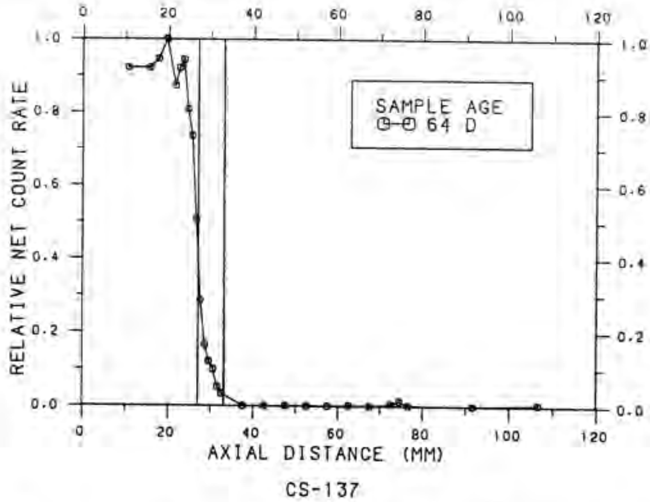


Fig. 6. Activity profile of cesium in sample I2. The boundaries of the lower 5 mm thick concrete layer are marked with two vertical lines. Cemented resin appears in the section 5...27 mm.

The first clear diffusion profiles were detected in the sample I4 in crushed rapakivigranite (Fig. 7). The boundary between labelled and unlabelled crushed rock is marked with a vertical line. Cesium moved 20 mm during 175 d. Cobalt displayed no mobility. One can see inhomogeneity in the distribution of cobalt on the labelled side of the specimen. In principle, the numerical model, REPCOM, accepts arbitrary initial profiles. Strontium moved 20...30 mm during the first 29 d. It has high diffusivity and a low sorption factor.

Migration in crushed tonalite was remarkably slower than in crushed rapakivi, as can be seen when comparing Fig. 8 with Fig. 7. There are no clear movements of cesium and cobalt in the sample T2 between the ages 43 d and 224 d. Strontium moved approx. 20 mm during the first 43 d, which is also less than in the case of rapakivi.

During the first 108 d no migration of any tracer was detected in the sample I3, which contains glued concrete and molded cemented resin. Fig. 9 shows two activity profiles of cesium. Within the error limits one cannot distinguish between the profiles. The same observation holds true for cobalt and strontium.

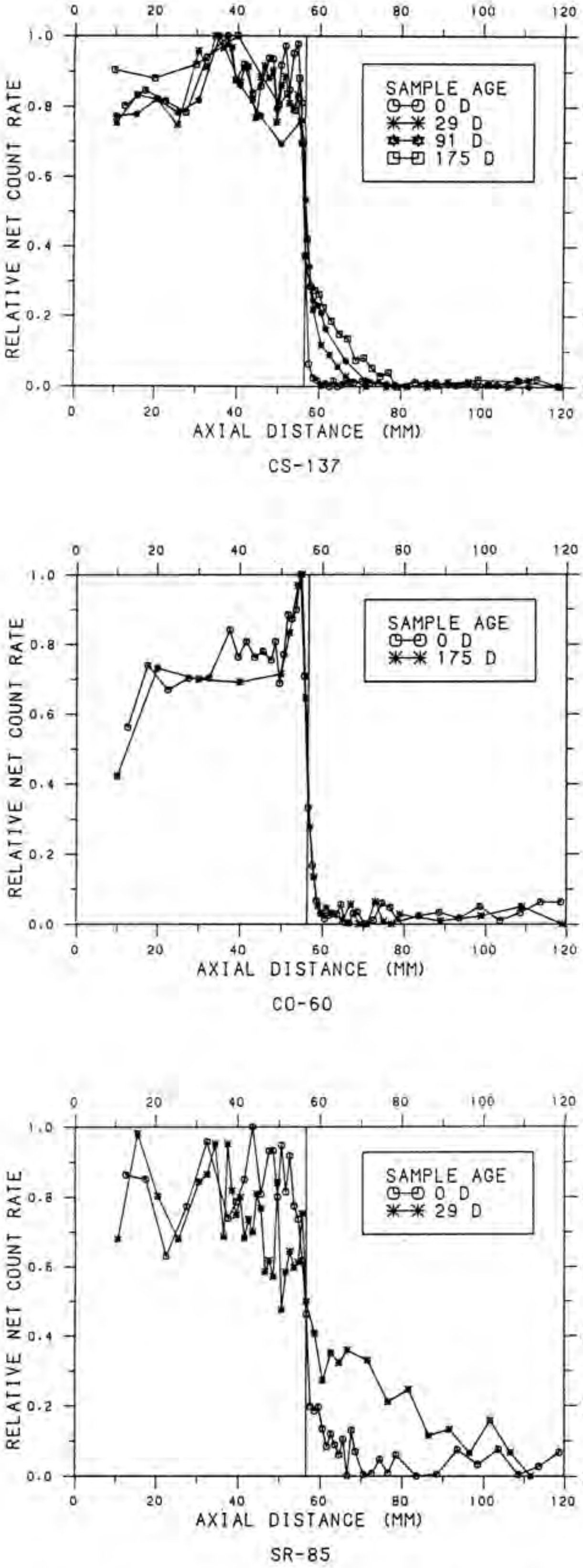


Fig. 7. Activity profiles of Cs, Co and Sr in sample I4.

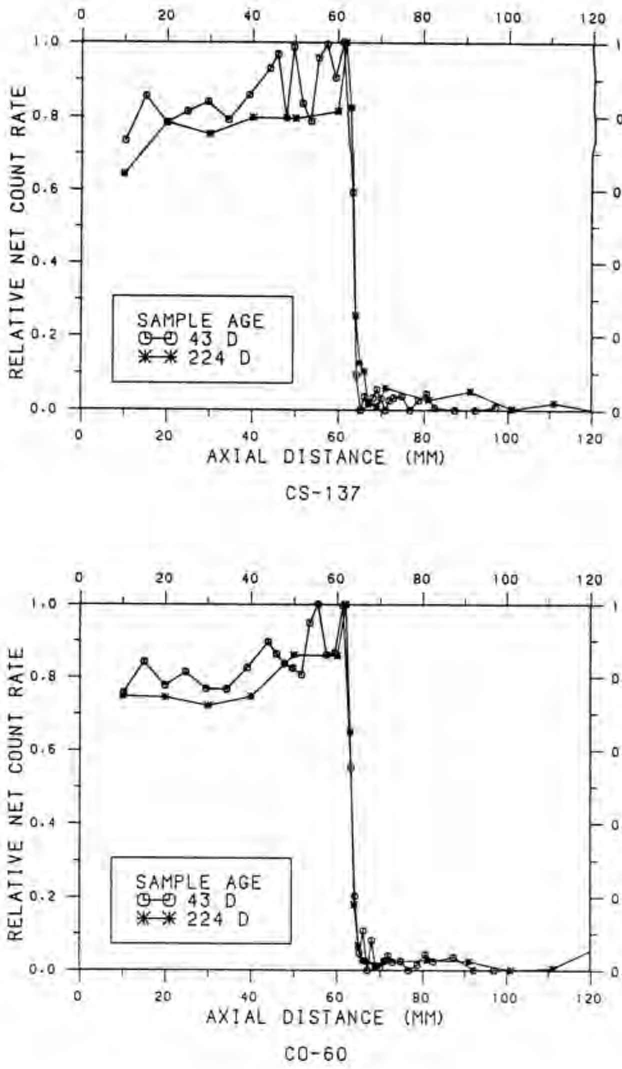


Fig. 8. Activity profiles of Cs and Co in sample T2.

In samples such as I1, I6, I7 and T3 are labelled waters in own compartments. It is important to know how the tracers are distributed between the waters and the walls. In addition, the ratio between loosely precipitated compounds and sorbed compounds must be known.

In the cesium profiles in sample I6 (Fig. 10), one can notice sharp peaks at the boundary between the labelled water and the concrete. The nuclides were sorbed on this boundary, but also on the inner wall of the PVC-tube. The activity distribution was studied in greater detail as described in the following.

Initially, the profiles in I6 were measured before shaking. Secondly, I6 was measured after shaking in order to find out if there was any loose precipitation. Finally, the water was removed from the sample and the profiles were measured without water in order to measure the sorbed activity on the wall of the PVC-tube and on the concrete surface. In

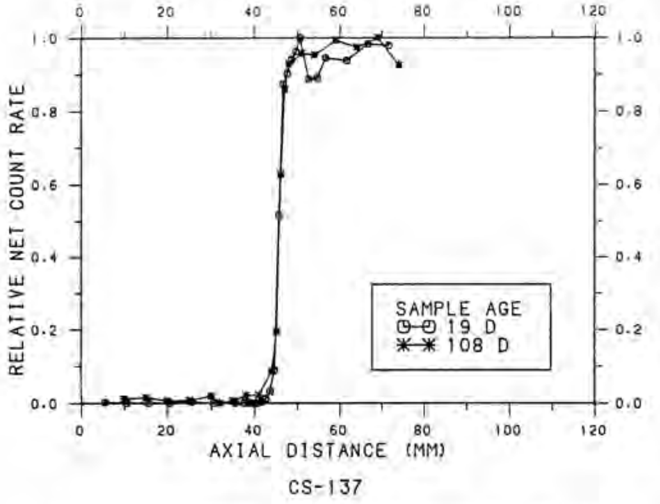


Fig. 9. Activity profiles of Cs in Sample I3.

addition, both the total activity of the water and the specimen were determined.

In every case, shaking had no clear effect on the activity profiles. Thus there were no loose precipitations.

Only 4 % of the total cesium remained in the sample after removing the water. Thus 96 % of the cesium seems to be in a soluble or colloidal form. The surface concentration of cesium on the concrete disk was higher by a factor of 10 than on the tube wall.

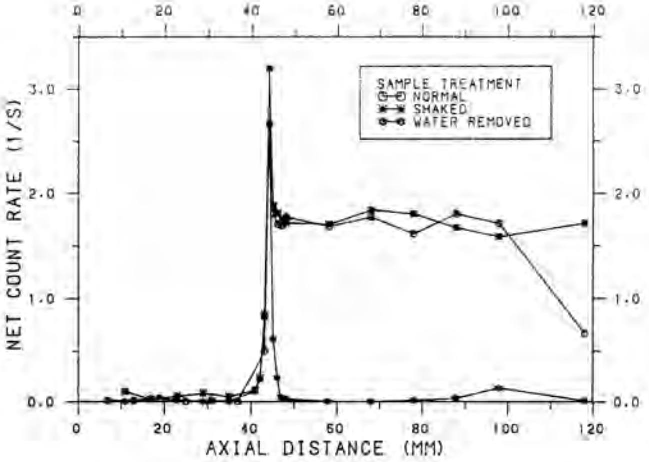


Fig. 10. Cesium distribution in the sample I6 between labelled water and the inner surfaces of the water containing sections. The sample was measured after various treatments in horizontal position at ages 23...47 d.

Also 4 % of strontium remained in the sample after removing the water. In this case the surface concentration of strontium was at the same level on the tube wall as on the concrete disk. This implies that strontium was dispersed homogeneously on the surfaces around the compartment filled by water and not concentrated on the concrete surface.

The fraction of soluble or colloidal cobalt in water was only 54 % of the total. The surface concentrations on the concrete and on the PVC- wall were at the same level.

DISCUSSION

On the grounds of the first measurements it looks as if the developed samples have performed satisfactorily.

Presently, there are already some extensive activity profiles which could be used for the first validation computations and for determination of diffusion coefficients. Such is the case with strontium in crushed rapakivigranite and in crushed tonalite, and cesium in crushed rapakivigranite and in concrete. For cobalt it would be possible to give only the upper limits of diffusivity, because practically no mobility was detected.

PLANS FOR THE FUTURE

The activity profile measurements will be emphasized in the year 1987. Development work for the measurement of the beta active samples with a liquid scintillation detector will be continued.

Sorption experiments which are needed for the validation work are being conducted at the University of Helsinki's Department of Radiochemistry.

The model computations will be made at the Nuclear Engineering Laboratory of the Technical Research Centre of Finland. The main part of the calculations will be performed using the compartment model REPCOM.

REFERENCES

1. H. NORDMAN, Ydinjätteiden loppusijoitustilan kompartmenttimalli (REPCOM) / A compartment release model (REPCOM) for nuclear waste final repositories, (in Finnish). Nuclear Engineering Laboratory, Technical Research Centre of Finland, Technical Report TUMA-12/86, Helsinki, 48 + 8 p, (1986)

2. E.H. TUSA et al., Final Disposal of Low and Intermediate Level Waste from Finnish Nuclear Power Plants. IAEA-SM-289/18, Proc. of the International Symposium on the Siting, Design and Construction of Underground Repositories for Radioactive Wastes, Hannover, (1986).

3. B. ALLARD et al., "Minerals and Precipitates in Fractures and their Effects on the Retention of Radionuclides in Crystalline Rocks", Proc. Near Field Phenomena in Geologic Repositories for Radioactive Waste, p. 93, Seattle (1981).

4. M. NYKYRI, "Validation of a Numerical Release Model (REPCOM) for the Finnish Reactor Waste Disposal Systems - Part I: Start Up of the Laboratory Experiments". Nuclear Waste Commission of Finnish Power Companies. To be published, (1987).