INTERMEDIATE RESULTS FROM THE LABORATORY SCALE MODELING OF THE FINNISH REACTOR WASTE DISPOSAL SYSTEMS

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ABSTRACT

In Finland the reactor waste from four nuclear power plants will be disposed of in two repositories to be built in the granitic bedrock at the two power plant sites. The aim of this work is to model experimentally the inner structures and materials of two reactor waste repositories. The experimental modeling of the multibarrier system is conducted on laboratory scale by using the same principal materials as are employed in the Finnish reactor waste disposal concepts. Migration of radionuclides is then studied in two or more consecutive layers of these materials. The laboratory arrangements include the following test materials: bituminized resin and cemented ion-exchange resin, concrete, crushed rock, and water corresponding to the materials in the planned disposal systems. Cs-137, Co-60, Sr-85, and Sr-90 are used as tracers, with which the ion-exchange resin, water or crushed rock is labeled depending on the specimen type. Three to nine activity profiles have been determined for each cylindrical gamma-active sample. So far clear diffusion is observed in the profiles for cesium and strontium in crushed rock. Diffusion of Cs-137 in concrete and Co-60 in crushed rock is evident, but not so clear. Some activity profiles have been measured for beta active Sr-90 doped samples, after splitting them. Also two beta-active cubic samples have been splitted and measured.

INTRODUCTION

A necessary part in the development of numerical models is validation. This document is a description of the experimental scale modeling of the inner structures and materials of two reactor waste repositories for validation of a numerical near field release model, REPCOM (1). These two repositories will be used for the disposal of the reactor waste from the four Finnish nuclear power plants, and they will be constructed by the two Finnish nuclear power companies. The repositories will be constructed at the power plant sites at a depth of 50 m to 120 m.

Migration of radioactive tracers is being studied by using small samples which contain materials present in the planned reactor waste repositories. The basic specimen geometries are cylindrical and cubic. In the cylindrical geometry the test materials were placed in sequence into PVC-tubes. The gamma-active nuclides in the cylindrical samples were measured non-destructively by scanning to determine the activity profile of each specimen. The beta-emitting Sr-90 in separate samples will be measured after splitting the samples.

The main results presented in this report consist of the activity profiles. Both gamma- and beta-active samples were measured. Some profiles are also fitted for analytical solution of diffusion equation in porous media.

CONSTRUCTION MATERIALS

In the Ref. (2) and (3) construction of the samples and the materials used in the samples were thoroughly described. In the samples either the waste form or crushed rock or water was labeled with radioactive nuclides. The tracers used were carefully chosen according to their significance in the waste forms, diffusion coefficients, half-lives, and ease of analyzing (2). The chosen tracers were Co-60, Cs-137, Sr-85, and Sr-90 of which only Sr-90 is beta-active, and it is used because the half-live of Sr-85 is too short. The principal properties of the tracers are listed in Table I.

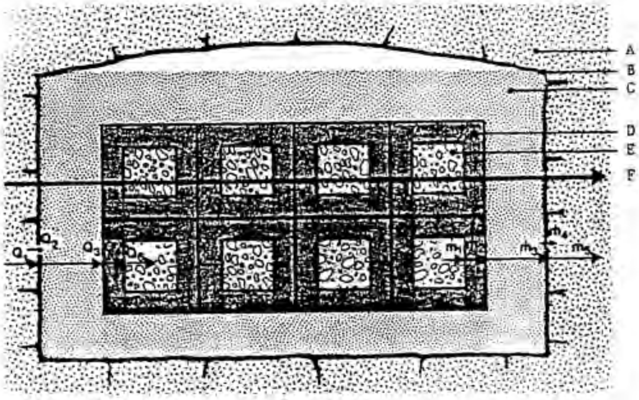
TABLE I
Properties of the Tracer Elements

Isotope	Half-live [a]	Activity type	Photon energies [MeV]
Co-60	5.26	gamma	1.17, 1.33
Cs-137	30	gamma	0.662
Sr-85	0.175	gamma	0.514
Sr-90	28	beta	-

The repository for reactor wastes at the Loviisa power plant site (IVO) will be located at a depth of 120 m. It consists of separate tunnels for dry waste and a cavern for cemented waste. The main part of the activity will be found in the cavern.

The numerical release model, REPCOM, describes the repository as shown in Fig. 1. The materials chosen for sample construction were simulated cemented ion-exchange resin, concrete, and crushed rock and natural groundwater from Hästholmen (Loviisa power plant site) equilibrated for one week with crushed concrete.

The repository at the Olkiluoto nuclear power plant site (TVO) will be constructed at a depth from 50 to 100 m and will consist of a waste vault with two silos. Bituminized ion-exchange resin cast into steel drums will be placed into one silo, which is provided with an extra barrier consisting



A Bedrock
B Injected and shotcreted concrete
C Crushed rock
D Concrete container
E Cemented waste
F Groundwater flow through the repository

Fig. 1. Sketch picture of IVO's repository (1).

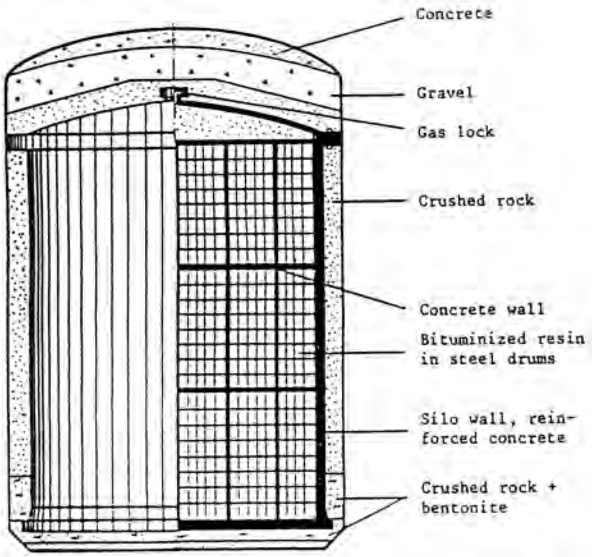


Fig. 2. Sketch picture of TVO's repository (4).

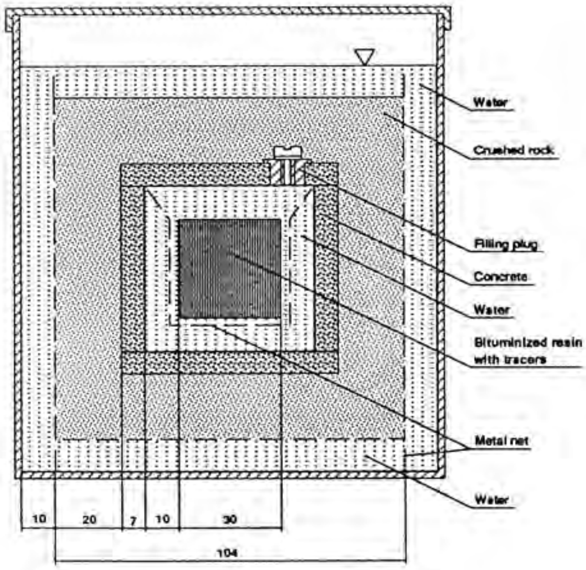
of a 500 mm thick reinforced concrete wall, and dry solid waste will be placed into the other silo.

The modeled repository is illustrated in Fig. 2. The materials chosen for sample construction were simulated bituminized ion-exchange resin, concrete, crushed rock, steel (steel drums) and simulated Allard groundwater (5) equilibrated for one week with crushed concrete.

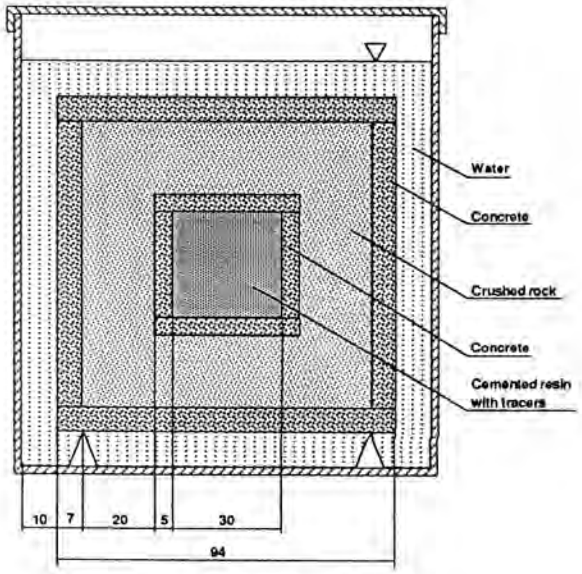
SAMPLES

Altogether 60 samples were constructed. Half of them contain one beta-active tracer and half of them contain three gamma-active tracers. Totally there are 11 cylindrical types (four samples of each type) and two cubical types (eight samples of each type).

Specimen types I1, I3, I4, I6, I7 and I8 are simple cylindrical samples with two or three material layers. Type I2 is a more complicated cylindrical sample with five layers. Type I5 is a complicated cubic sample. Type T2 is identical with I4, and type T3 is identical with I6. Types T4 and T5 are even more complicated than I2. The figures of specimen types are sketched in Fig. 3, 4, and 5.



Specimen type I5



Specimen type T1

Fig. 3. Cubical specimen types.

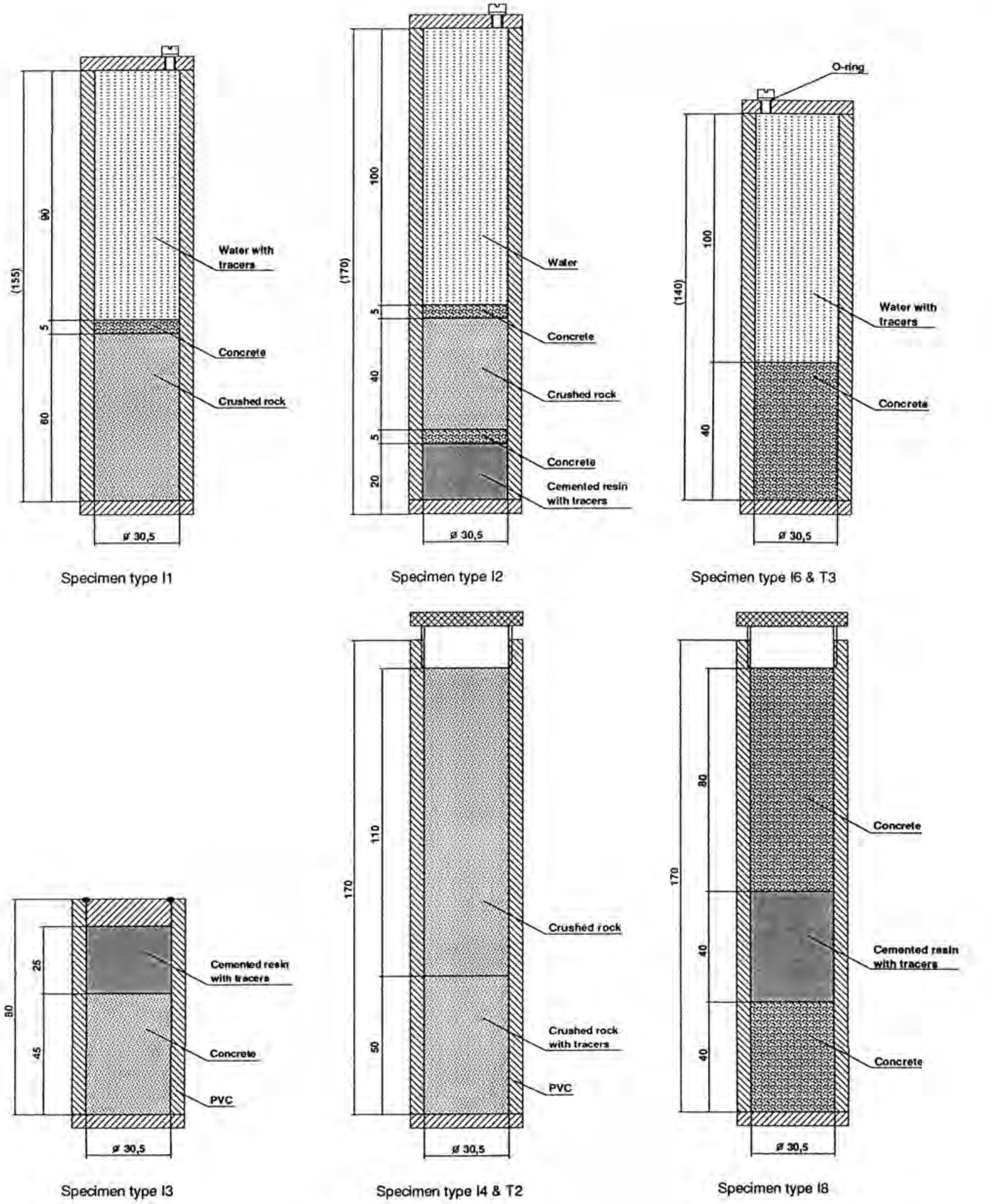


Fig. 4. Cylindrical specimen types.

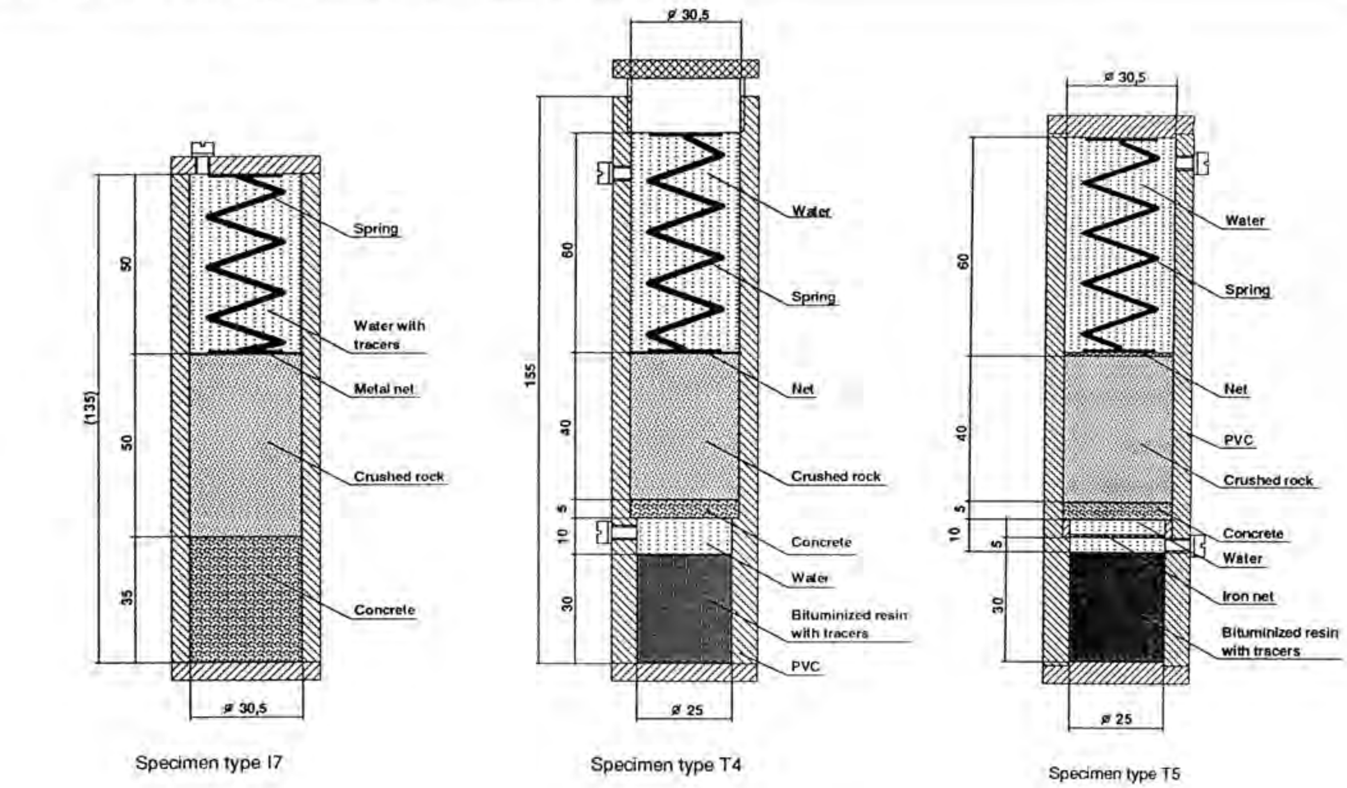


Fig. 5. Cylindrical specimen types.

MEASUREMENT SYSTEMS

Gamma Measurement System

All the gamma-activity profiles were measured using a set of equipment illustrated in Fig. 6 (2). The smallest positioning step of the scanner is about 0.003 mm. Opening of the slit of the collimator used in these measurements was 1.0 mm.

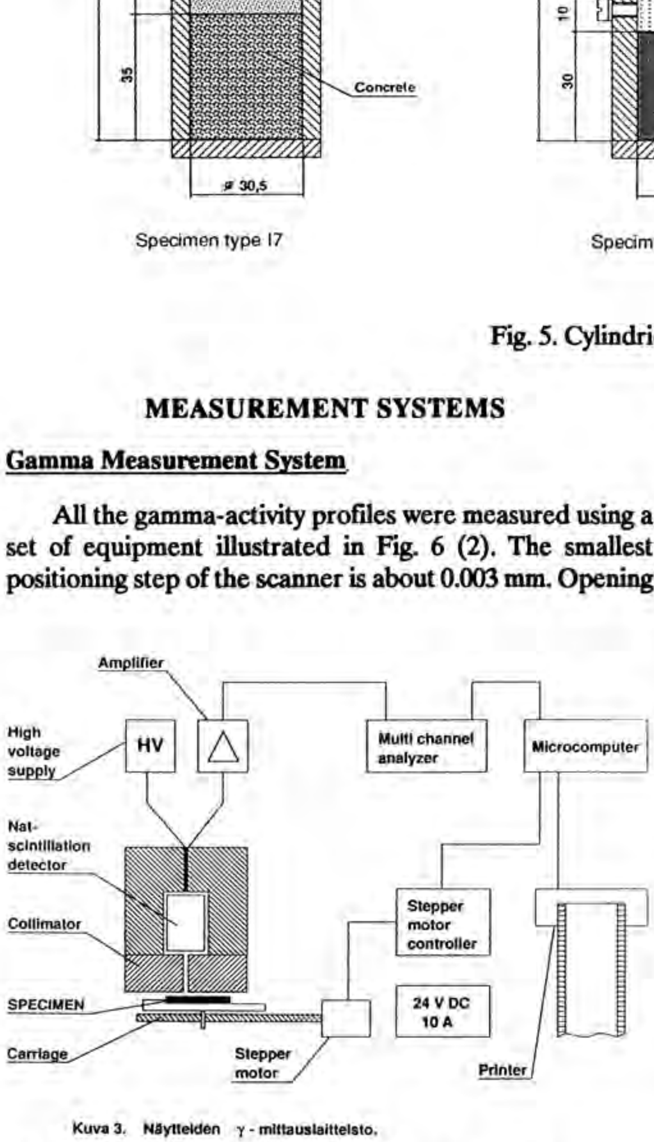


Fig. 6. A schematic diagram of the activity profile measurement system (2).

All cylindrical samples were measured with scanner at least three times, but some even nine times. In a typical measurement about twenty positions were analyzed with about one hours' analyzing time, thus taking about one day to measure. In addition some measurements were also performed using longer analyzing times for comparison purposes.

Beta Measurements System

To obtain satisfactory results the beta-active samples had to be split before measuring the profile. After trying several methods for splitting the cylindrical samples, simply use of saw and drill was chosen, because of the low risk of contaminating the splitting system. First the PVC-coverage is split in two pieces with the saw, the concrete samples are then subsampled with a small diamond-pointed hand drill and crushed rock with a small pipe. Subsampling is performed to obtain a profile. With these methods the achieved accuracy of the profiling was 1 mm in concrete and about 3 mm in crushed rock.

Sr-90 is a beta-active nuclide the daughter nuclide of which is Y-90. This decays emitting an electron to stable Zr-90. The equilibrium between Sr-90 and its daughter Y-90 is attained in 26 days (99.9 %). The maximum energy in the beta-emission of Sr-90 is 540 keV and in the beta-emission of Y-90 over 2 MeV. Activity of Y-90 was analyzed by measuring the photons coming from Cherenkov-radiation caused by electrons moving faster than light in that media. The photons are then observed with photomultiplier tubes. The measurements were carried out using 20 ml glass-vials with Wallac's Spectral-scintillation counter.

THEORY

Diffusion in porous media follows slightly modified Fick's second law (which takes into account the porosity and sorption)

$$\frac{\partial c}{\partial t} = D_a \frac{\partial^2 c}{\partial x^2}$$

where D_a is apparent diffusion coefficient

$$D_a = \frac{\varepsilon G D_w}{\alpha}$$

where D_w is diffusion coefficient in water, α is capacity factor, ε is porosity and G is geometrical factor.

Two kinds of solutions were used, one for solid source (specimen types I4 and T2) and one for source in water (specimen type I7). These solutions are well known eg Ref. (6).

MEASUREMENT RESULTS

Both gamma- and beta-activity profiles were measured. Because of the large number of measured profiles only a selected collection is presented and described in this report. The collection consists of those specimens, where diffusion has been clear. In most excluded specimens the diffusion is hardly visible.

All the cylindrical gamma-active samples have been measured at least three times, some of them even nine times (7,8).

In the activity profiles the relative net count rate versus the axial distance from the bottom of the specimen is plotted. The vertical lines in the profiles indicate the approximate position of each boundary between different materials. The measured points are simply connected with lines.

The zero point of the sample age was determined at the moment when diffusion started. At that moment the labeled material was put into contact with the first inactive medium.

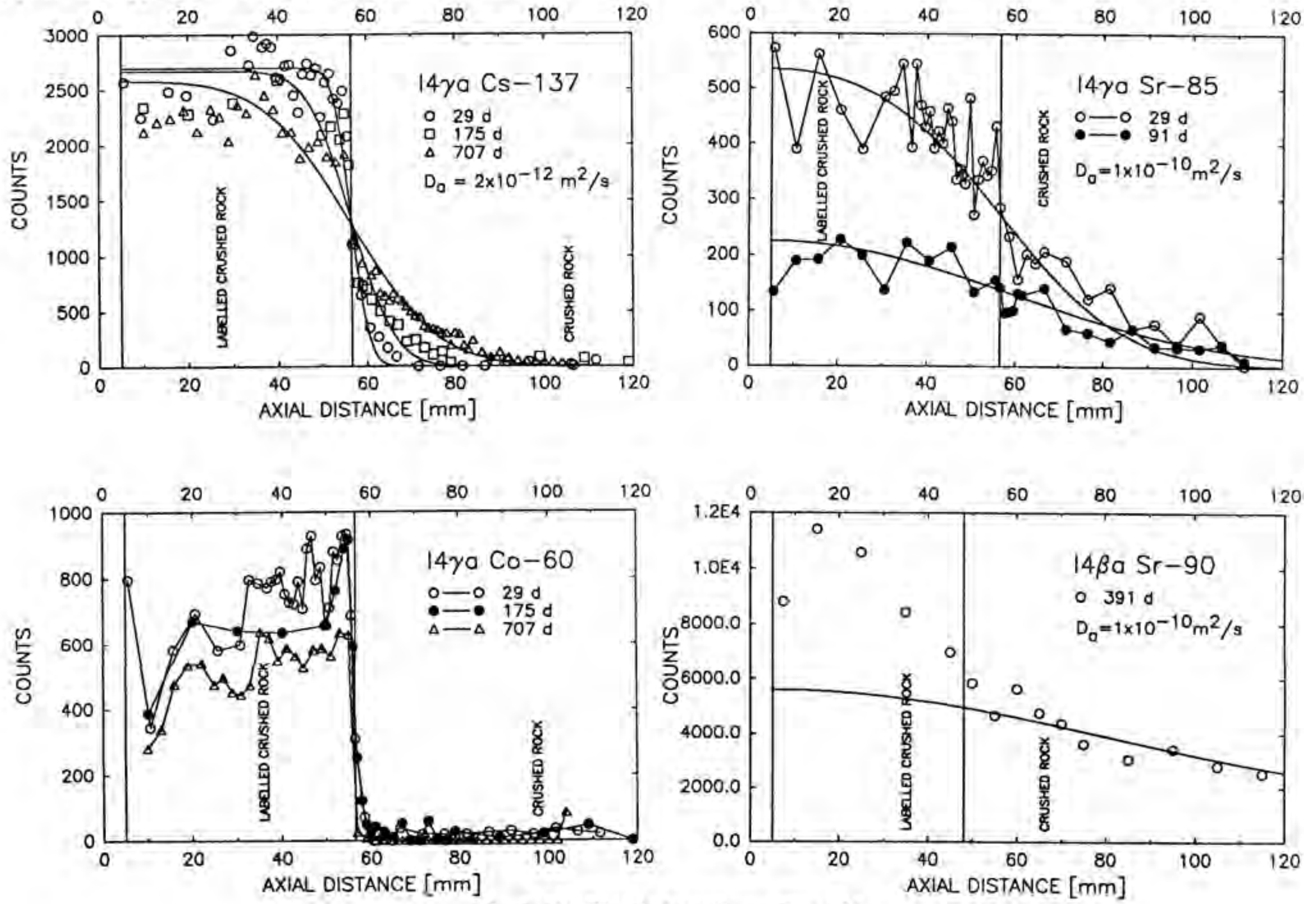


Fig. 7. Profiles in Specimen Type I4.

Specimen Type I4

During the first 707 d the tail in Cs-137 profile has observably moved in crushed rock about 35 mm (Fig. 7). The diffusion coefficient used for fitting was $2x10^{-12}$ m^2/s, which gives rather good results. Co-60 did not indicate any distinct diffusion behavior.

Sr-85 moved in crushed rock 50 mm during the first 91 d (Fig. 7). The beta-analysis done after 391 d (note: this is not the same sample as used in gamma-analysis) confirms the large movement of strontium in crushed rock. This was anticipated because strontium has a low sorption factor and high diffusivity in crushed rock. The diffusion coefficient used for fitting was $1x10^{-10}$ m^2/s, which is good for short times but indicates also that part of the strontium is immovable.

Specimen Type I7

Cs-137 has moved in crushed rock about 40 mm during the first 580 days (Fig. 8), which is in good accordance with the profiles of sample type I4. In model calculations diffusion constant was $2x10^{-12}$ m^2/s, which is same as in type I4, and capacity factor was 2400.

Sr-90 has moved about 40 mm during 302 d. Model diffusion constant was $4x10^{-12}$ m^2/s and capacity factor 1.9. This diffusion constant is much lower than that used in specimen type I4.

Specimen Type T2

Cs-137 and Co-60 indicated no noticeable movement.

Sr-85 has moved about 20 mm during the first 43 d. In 395 d the movement has increased to 60 mm (beta active sample). Model diffusion constant $2x10^{-11}$ m^2/s 1.9 was used. Also the phenomena of immovable strontium occurs.

There is an evident difference between crushed tonalite and rapakivigranite in the comparable specimen types I4 and T2. Both Cs-137 and Sr-85 have penetrated much further into rapakivigranite.

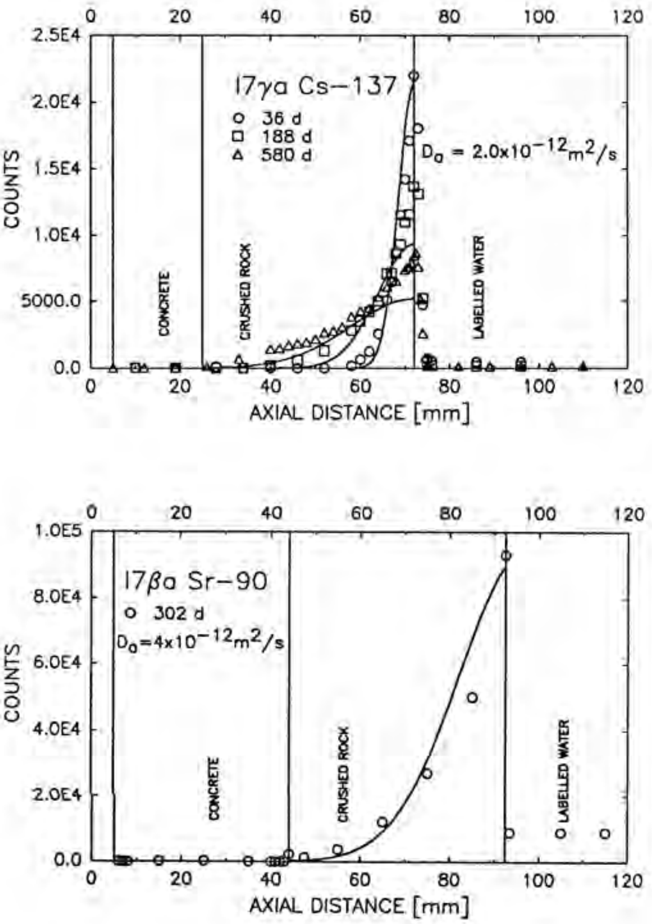


Fig. 8. Profiles in specimen type I7.

DISCUSSION

The scanning measurements of gamma-active cylindrical samples have proved to be successful. The beta-measurement method is laborious needing plenty of hand work, but the results are quite satisfactory.

Analytical calculations give good enough results on those specimen types where diffusion has been evident. These results are same of the kind as those of REPCOM calculations (9). On the basis of analytical calculations part of strontium seems to be immovable, which may be caused by matrix diffusion.

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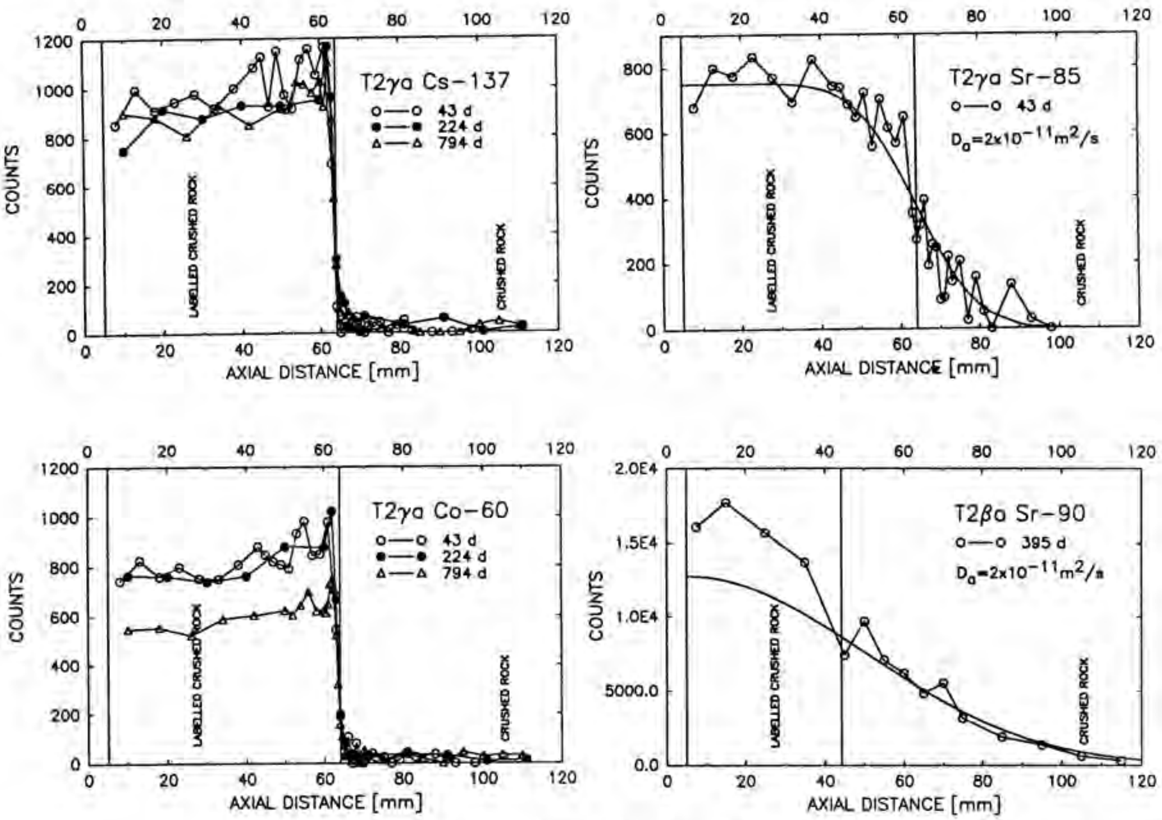

Fig. 9. Profiles in specimen type T2.

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