In-situ Experiments for Investigating the Retention Properties of Rock Matrix in ONKALO, Olkiluoto, Finland – 14258

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ABSTRACT

Spent nuclear fuel from the nuclear power plants owned by TVO (Teollisuuden Voima Oy) and Fortum, is planned to be disposed at a repository at a depth of more than 400 meters in the bedrock of Olkiluoto (Eurajoki, Finland). The repository system includes multiple release barriers: the nuclear fuel, copper canister with a cast iron insert, bentonite buffer around the canister and backfilling of the tunnels. Furthermore, the surrounding rock is the last barrier if the man-made barriers fail during the passage of time. Therefore, safe disposal of spent nuclear fuel requires information also about the radionuclide transport and retention properties within the porous and water-containing rock matrix along the water conducting flow paths.

To this end, three types of experiments are being performed and planned within ONKALO, the underground rock characterization facility in Olkiluoto, as part of the project “rock matrix REtention PROperties” (REPRO). The research site is located at a depth of 420 meters close to the repository site. The aim is to study the diffusion and sorption properties of nuclear compounds in the rock matrix under real in-situ conditions. The first in-situ experiment was performed during 2012 using HTO, Na-22, Cl-36 and I-125 as tracer nuclides. Breakthrough curves show retention and asymptotic behavior that are in-line with those caused by matrix diffusion and sorption were observed in their breakthrough curves. Weak sorption was also observed in the breakthrough curves of Na-22 and I-125. However, the determined distribution coefficients were lower than the ones obtained from laboratory batch sorption experiments. In addition, a detailed predictive modeling for two other experiments that are planned to execute is presented.

INTRODUCTION

It is known that elements migrating in water that flows in a fracture are transported at a lower speed than that of the water [1]. The retention is caused by diffusion of these elements away from the flowing water to stagnant water in the pores of the surrounding rock matrix, and by their chemical sorption on mineral surfaces of the rock matrix, fracture walls and fillings [2-3]. However, the relevance of retention in repository conditions has remained unclear, and the impact of retention varies from one nuclide to another.

In general, it is believed that conditions might be different in laboratory than in-situ due to, e.g., stress relaxation caused by sampling. In addition, sawing artifacts might change the characteristics of rock sample. Stress relaxation might cause increase of porosity, which causes

increase of diffusivity since it can open or widen pore throats and in that way enhance migration of elements by diffusion. Sawing artifacts might increase the porosity and furthermore specific surface area available for sorption, which would increase the value of distribution coefficients. The same effect is seen in laboratory batch sorption experiments when using crushed rock samples. In-situ diffusion experiments avoid some of the problems caused by sampling and stress relaxation. To this end, it is important to measure diffusion and sorption properties with in-situ experiments and compare their results with those from laboratory experiments. This comparison can be used to build confidence that assumptions applied in the safety case are in line with site evidence. In addition, in the performance assessment conditions most of the retention takes place in the vicinity of the deposition holes. This emphasizes the significance of careful quantification of the retention properties of the bedrock in the conditions present in the repository near-field.

The objective of the REPRO project is to perform experiments so as to investigate the retention properties of the rock matrix under realistic in-situ conditions. Three different types of experiment will be carried out to investigate these properties: 1. Water Phase matrix Diffusion Experiment (WPDE), 2. Through Diffusion Experiment (TDE), and 3. Gas Phase matrix Diffusion Experiment (GPDE). In WPDE a short concentrated pulse of selected radionuclides is injected into a water flow through an artificial fracture. This artificial fracture is formed on the circumference of a 2 m long packed-off section of drillhole by placing a cylindrical flow guide on center of the drillhole. In TDE a concentrated mixture of selected radionuclides is injected into a meter long packed-off section of a drillhole. Breakthrough of these radionuclides is followed in two observation drillholes about 10 cm away from the injection drillhole. GPDE is similar to WPDE, except that it will be performed in the gas phase, which requires continuous drying of the experimental section and the rock matrix around it. Performing the tracer experiments along artificial fractures in packed-off drillhole sections in WDPE and GPDE enables a better control of the flow field and a better recovery of tracers, and thus this experimental set-up allows a more precise characterization of the rock matrix properties. In addition, a large number of supporting experiments are being performed in the laboratory using rock samples from the experimental drillholes. These experiments provide parameters to be used for analyzing the in-situ experiments and they also give valuable information for sorting out differences between in-situ and laboratory data.

During past decades the retention properties of rock matrix using in-situ experiments have been studied in various underground laboratories. In general, these experiments as well as analysis of their results have been challenging due to the complexity of the conditions and the number of unknown parameters. Similar in-situ experiments like here have recently been performed at the Äspö Hard Rock Laboratory, Sweden, and at Grimsel test site, Switzerland [4-5]. Other in-situ tracer experiments have been successfully carried out in crystalline, sedimentary and clay rock in, e.g., Canada, Sweden and Switzerland [6-10].

The scope of this paper is to describe the investigation site and the experiments that are being performed or planned at that site. First results together with its future prospects are reported for the on-going WPDE experiment and predictive modeling is shown for experiments that are still under preparation (TDE and GPDE).

EXPERIMENTAL

Site description

The in-situ experiments are performed in ONKALO, the underground rock characterization facility in Olkiluoto, Finland. Preliminary plan of site selection and experiments were done by Aalto et al., 2009 [11]. The site was selected based on information from pilot holes, and aim then was to perform experiments at the depth of the final repository and away from the large water conducting fractures. After site selection an experimental niche was excavated at a depth of 420 m (see Fig. 1.).

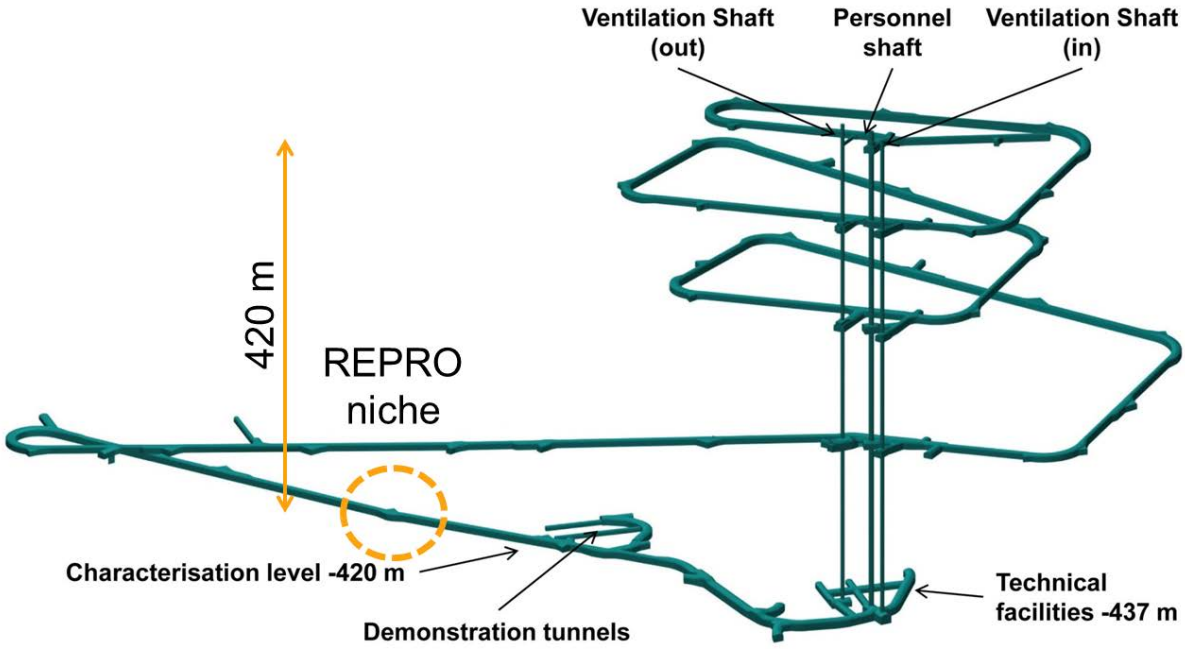

Fig. 1. REPRO research niche locates at a depth of 420 m in the underground rock characterization facility (ONKALO) in Olkiluoto, Finland.

After excavation of the REPRO research niche the aim was to find a 1-2 m section of unfractured rock that is located at least 7.5 m away from the tunnel wall. Fractures were avoided in order to investigate properties of the intact rock and the experimental section needed to be far enough from the tunnel wall to minimize the effects of niche and tunnel to the background stress field. Sections containing identified microfractures and sealed fractures were also avoided when the exact locations of experiments in the drillholes were selected. In addition, the orientation of foliation was also considered when selecting the drilling directions. In the case of TDE location the aim was to place one observation drillhole perpendicular and the other one parallel to foliation respect to injection drillhole. Special attention was paid to drilling in order to keep the drillholes as straight as possible and to avoid mechanical damage in the drillhole walls. Nine drillholes were drilled in the REPRO niche in order to find suitable places for all in-situ experiments [12]. Five of these holes are currently used by the planned experiments; three holes are used in TDE, one hole in WPDE and one hole in GPDE.

In general, the bedrock around the REPRO niche is pegmatitic granite and migmatitic gneiss (subgroup: veined gneiss). Pegmatitic granite is from light gray to pale colored with coarse

K-feldspar and commonly also cordierite and garnet. Migmatitic gneiss is a metamorphic, heterogeneous mixture of small grained mica gneiss and coarser leucosome veins of thickness that varies from several millimeters up to ten centimeters. Experiments were planned to be performed in veined gneiss. Foliation of veined gneiss in the experimental area is mainly weak to moderate banded foliation. Fracturing, fracture fillings and rock quality were also investigated from the drillcores. A more detailed geological description of the site is given by Toropainen, 2012 [12].

Water phase matrix diffusion experiments

A straightforward way to investigate properties of rock matrix under in-situ conditions is to carry out a water phase matrix diffusion experiment (WPDE). A special double-packer system was constructed for the experiment (see Fig. 2). The packer system forms a two-meter artificial flow channel along the perimeter of the drillhole. The volume and aperture of the flow channel are minimized by an impermeable cylindrical flow guide inside the packer system. The inlet and outlet positions of water are located at the opposite ends of the packed-off section. WPDE tracer tests are performed using slow flow rates that are generated using a piston pump. The experiment is executed using synthetic groundwater to carry to tracer solution. Composition of the synthetic groundwater was prepared based on a careful analysis of real fracture water in the REPRO site (see TABLE I).

TABLE I. Chemical composition of synthetic granitic groundwater used in WPDE.

Element	Concentration [mg/l]	Element	Concentration [mg/l]
Na	2700	Cl	5100
K	7.9	Br	33
Mg	35	F	1.5
Sr	0.5	SO_4	0.3
Ca	520	HCO_3	12

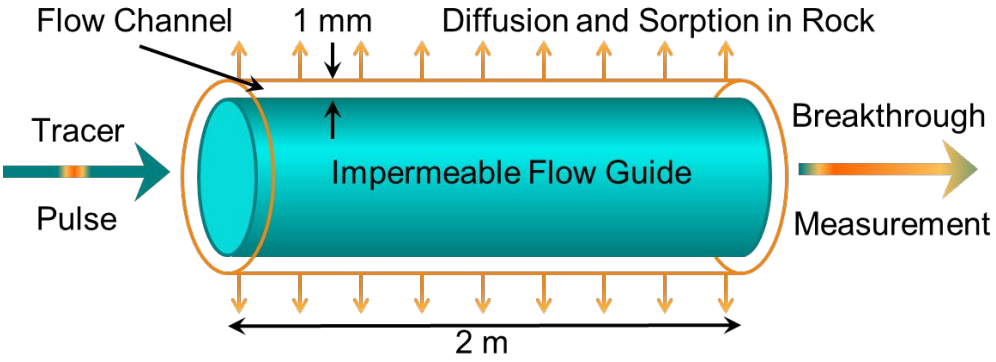

Fig. 2. WPDE experiment studied retention of HTO, Na-22, Cl-36 and I-125 by matrix diffusion for flow through an artificial fracture that is formed along a packed-off drillhole section with the help of an impermeable cylindrical flow guide.

A sharp pulse of tracer solution was injected into water flow which was then conducted to the inlet of the experimental drillhole section. Tracer molecules were diffused into the rock matrix and sorbed at the mineral surfaces during migration through flow channel. The early part of the breakthrough curve is dominated by the advection, diffusion and dispersion in the flow channel. However, matrix diffusion and sorption were expected govern the late part of the breakthrough curve, since after some time the advection-dispersion pulse had passed the channel and only the molecules that had undergone retardation by matrix diffusion (and sorption) remained in the system.

Retention observed in the experiment depends on the properties of the rock matrix, distribution of the flow in the flow channel and tracer properties. A special attention was paid to fix these parameters. A long section of the drillhole gives a stronger retarding effect on the breakthrough curve and small aperture reduces the total volume of system. On the other hand, the test section should not include any fractures, and the small aperture of the flow channel complicates installation of a very long instrumentation.

The measurements need to be carried out using low enough flow rates in order to see a measurable effect of the matrix diffusion, but also high enough so that the experiment time remains tolerable. Flow rates that were (and will be) used in the experiment were also selected so that diffusional mixing in the flow channel was strong enough, some extent, to even out variations in the solute mass flux caused by local changes in the channel aperture. Based on the predictive modeling, flow rates of 20, 10, and 5 µl/min were chosen since reliability of the experimental results requires tests with different flow rates. The first WPDE experiment has been performed with a flow rate of 20 µl/min and the second one with 10 µl/min is currently running.

In the first experiment HTO, Na-22, Cl-36, and I-125 were used as tracers. Tracers were selected such that the tracer cocktail contains a conservative (HTO), an anionic (Cl-36), and a weakly sorbing (Na-22). I-125, emitting low energy gammas, was included in the cocktail for online detection of the breakthrough.

The breakthrough of these radionuclides was followed by an online gamma detector and measured in the laboratory from collected samples of the outflowing water. In laboratory, the concentrations of Na-22 and I-125 were first measured by gamma spectroscopy (GS). After GS, chloride and part of iodide were precipitated from the solution using $AgNO_3$ because Na-22 interferes with measurement of Cl-36, which was measured by liquid scintillation counting (LSC) from dissolved precipitate. Both Na-22 and I-125 interfere with measurement of HTO which was measured from the supernatant by LSC. Na-22 and I-125 were measured after LSC counting once again with GS and their concentration was then subtracted from the HTO concentration. GS of Na-22 and I-125 were performed using a Canberra HPGe GXRS232 detector and LSC of HTO and Cl-36 using a Perkin-Elmer Liquid Scintillation Counter Tri-Carb 2910 TR.

Through diffusion experiment

The TDE will be executed by injecting a concentrated mixture of selected radionuclides into a meter long packed-off section of a drillhole and concentrations of radionuclides are followed in two observation drillholes about 10 cm away from the injection hole (see Fig. 3.). The observation drillholes are placed so that possible influence of foliation on the diffusion properties could be studied. One of the observation hole is perpendicular and the other parallel to the foliation in respect of the injection drillhole. Volume reducers will be used in all drillholes in order to raise the

tracer concentration in the injection hole and to improve the detection of the breakthrough in the observation holes. Use of a volume reducer increases also the sensitivity to detect concentration decrease in the injection hole, which offers further information about the properties of rock matrix. The drillhole equipment used in all holes will be similar as in WPDE. During the experiments pressures need to be controlled carefully so that pressure gradients are not created between the experimental drillholes.

During the experiment, solutions (about 400 ml) are circulated by pumps in the drillholes to maintain concentrations constant and to be able to perform reliable sampling of them. Sampling from the all drillholes is planned to be performed about once per month. In case of injection hole, sample volumes less than 0.5 ml are suitable due to the high concentration of radionuclides. In addition, each sampling decreases the strength of the source. In case of the observation drillholes, it is beneficial to collect larger samples (few ml) in order to increase their activity and reach the detection limit of measured radionuclides.

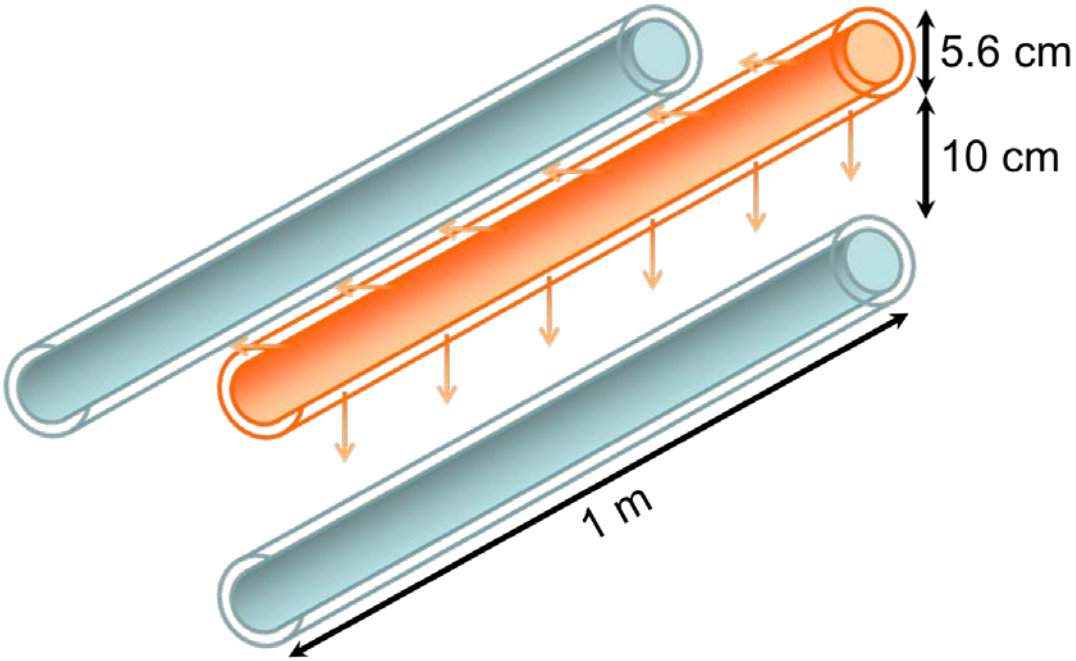

Fig. 3 In TDE the retention properties of rock matrix are studied in a system which contains one injection drillhole (orange) and two observation drillholes (blue).

In the current plan the tracer cocktail includes both non-sorbing and sorbing radionuclides: HTO, Na-22, Cl-36, Ba-133, and Cs-134. The strongly sorbing tracers included (Ba-133 and Cs-134) are not expected to be seen in the observation drillholes. However, their diffusion and sorption can be seen from the decreasing concentration in the injection hole. In addition, the diffusion and sorption profiles of these nuclides could be seen from overcored samples that will be taken after the experiment has been terminated. From the overcored samples it might be possible to study more closely the influence of foliation and anisotropy on diffusion and sorption of the tracers. Previously overcoring followed by laboratory analysis the samples has been performed in the Grimsel test site, Switzerland [13].

Gas phase matrix diffusion experiments

GPDE has the same idea and aims as WPDE, i.e., to evaluate the retention properties of the rock matrix from the breakthrough curves of tracer test (see Fig. 2.). Diffusion in the gas phase is about 10 000 times faster than in the water phase, which makes gas phase experiments considerably faster to perform. However, gas phase measurements require drying of the drillhole and the rock matrix around it, which, on the other hand, increases the amount of preparations needed. Drying needs to be continued also during the experiment. In addition, these measurements offer information only about matrix diffusion since only non-sorbing tracers will be used. Experimental setup will also be similar to those of WPDE, except that the breakthrough curve can also be measured in-situ using a mass spectrometer, and a tedious sample analysis can be avoided.

GPDE can be performed using larger flow rates than WPDE due to higher diffusion coefficients. Based on predictive modeling, typical experiments using flow rates of 5 ml/min to 50 ml/min will take from 10 min to a few hours. Suitable tracers for the experiment are, e.g., He, Ar, and Xe. Helium has been used previously in the gas phase diffusion experiments in the laboratory [14]. When selecting the tracer, its solubility in water has to be taken into account since it might affect the observed breakthrough curve. However, a relatively fast measurement time allows trying of multiple tracers without a considerable time loss.

A tracer in the gas phase diffuses only in the unsaturated part of the rock matrix. This means that rock matrix should be treated as limited in GPDE although it is in practice unlimited. This will be the case in particular if drying of rock will not reach deeper than the tracer molecules diffuse during the experiment. However, this phenomenon can be taken into account in the model [2]. Since the location of the experiment has to be dried, it will make in-situ PMMA impregnation possible as has been done previously at the Grimsel test site [13].

PREDICTIVE MODELLING

At the moment the TDE and GPDE experiments are being prepared. Hence, only predictive modeling of these experiments will be reported here.

Predictive modeling of TDE

Predictive modeling of TDE was done by solving diffusion equations in cylindrical coordinates with the appropriate initial and boundary conditions. Here the real dimensions of the system, a non-sorbing tracer, and infinitely long drillholes were used in the solution. In addition, the diffusion equation was first expressed in dimensionless form.

A modeled breakthrough curve in one observation hole (10 cm apart from the injection hole) is shown in Fig. 4 using the dimensionless time tD_p/r_1^2. Since dimensionless time includes the pore diffusion coefficient D_p, the breakthrough curve can be considered as a general solution for non-sorbing tracers.

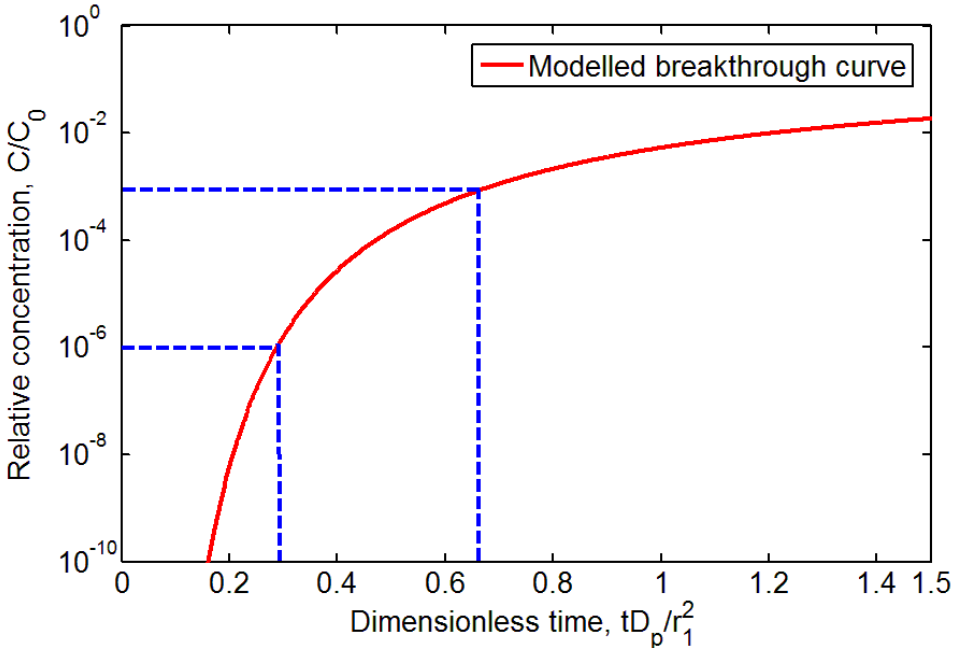

Fig. 4. Predictive modeling of TDE shows that a detectable amount of tracer will appear in the observation hole when tD_p/r_1^2 = 0.3 – 0.7.

Conservative estimates for detection limits (low and high) are also shown in Fig. 4. Based on limits detectable amounts of tracer will appear in the observation hole when the dimensionless time is 0.3 – 0.7. In TABLE II values for real times are determined based on the above interval of dimensionless time and conservative estimates for the pore diffusion coefficient. Upper and lower limits of D_p have been chosen based on laboratory experiments on samples taken from the planned TDE experimental section of drillhole. If D_p of 1×10^{-12} m^2/s is assumed, the breakthrough will be detected after 7 – 17 years. Respectively, if D_p of 3×10^{-11} m^2/s is assumed, the breakthrough will be detected after 0.25 – 0.6 years.

Let us assume that the volume of the injection and observation drillholes (including tubing) is 400 ml and that the total activity of the injected tracer is 200 MBq. This leads to an initial concentration of 0.5 MBq/ml. Let us also assume that the detection limit of radionuclides is 0.5 Bq/ml. This leads to the result that relative concentrations of 1×10^{-6} could be detected in the observation drillhole and that breakthrough will be detected after 0.25 – 7 years.

TABLE II. Predicted breakthrough times determined for dimensionless times between 0.3 (t_{min}) and 0.7 (t_{max}) and conservative estimates for D_p.

D_p [m^2/s]	t_{min} [y]	t_{max} [y]
1×10^{-12}	7	17
3×10^{-11}	0.25	0.6

Predictive modeling of GPDE

Predictive modeling of GPDE was done by solving the appropriate partial differential equations in cylindrical coordinates with the right initial and boundary conditions. The model takes into account advection, diffusion and dispersion in the flow channel and diffusion in the rock matrix. Here the real dimensions of the system, a helium tracer, and an infinite rock matrix around the drillhole were used. The model does not take into account the effect of tubing. It has been found that tubing will only delay the breakthrough and its effect on the shape of the curve is negligible.

A breakthrough curve of the predictive model is shown in Fig. 5. The early part of the breakthrough is dominated by advection, molecular diffusion and dispersion in the flow channel, whereas the effect of matrix diffusion can only be seen in the late time behavior. The modeled breakthrough curves in Fig. 5 are determined by assuming a flow rate of 50 ml/min, a porosity of 0.7 % and a pore diffusion coefficient of 1×10^{-8} m^2/s. These values were chosen based on the results of earlier laboratory experiments [15]. When the value of porosity or diffusion coefficient is higher, the tail of the breakthrough curve gets higher and it is observed earlier. When the flow rate is decreased, breakthrough is delayed and the advection dominated part of the curve gets wider and tilted towards late times as dispersion and diffusion in the flow channel get more dominant. Due to a wider peak, the effect of matrix diffusion gets stronger. However, since the peak gets tilted it will become more difficult to separate the effect of matrix diffusion from those of dispersion and diffusion in flow channel. Hence, the optimal range of parameters for this experiment can be found easily.

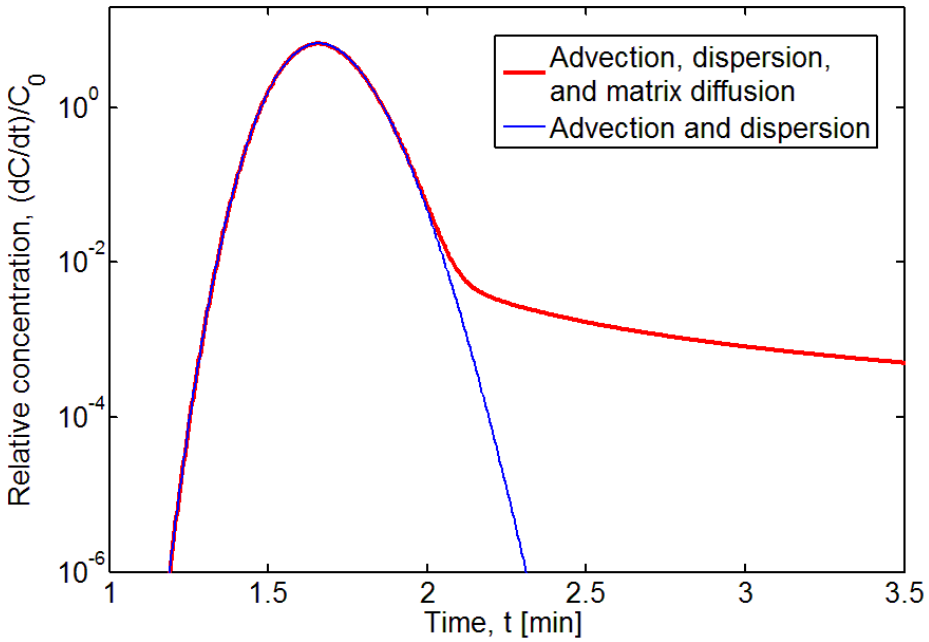

Fig. 5. Prediction for the result of GPDE using a flow rate of 50 cm^3/min. Results are shown separately for inclusion of advection and dispersion alone (blue) in the flow channel and for advection, dispersion, and matrix diffusion (red curve).

RESULTS OF WPDE

The first WPDE experiment using radionuclides was performed during the spring and summer of 2012. In this experiment water was pumped through the artificial flow channel with a constant flow rate of 20 µl/min and a mixture of HTO, Na-22, Cl-36, and I-125 was injected as a sharp pulse (0.5 ml) into the flowing water. Concentrations of these radionuclides as a function of time, i.e., their breakthrough curves, were measured by taking water samples from the outflowing water (see Fig. 6). Injected activities of all these tracers are shown in TABLE III.

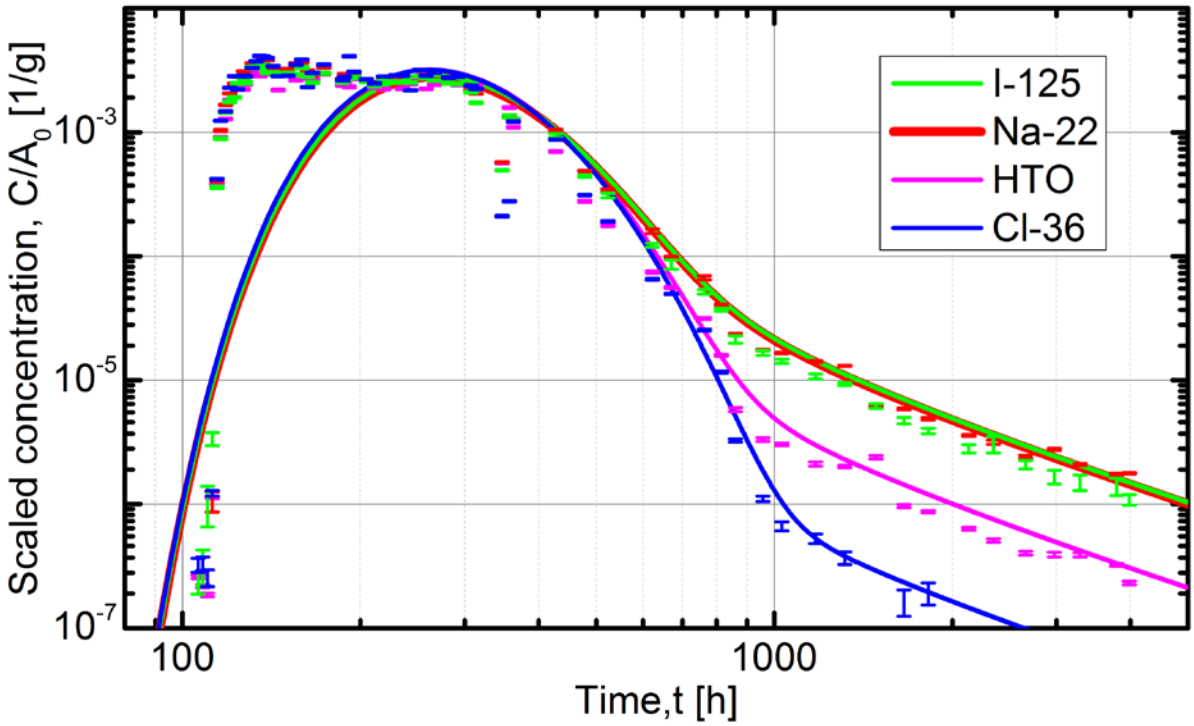

Fig. 6. Late time behavior of the breakthrough curves of HTO, Na-22, Cl-36, and I-125 show typical power-law behavior by the matrix diffusion and sorption in rock matrix.

TABLE III. The Injected activities, porosities, pore diffusion and distribution coefficients (A_0, ε, D_p and K_d) as determined from the experiment for HTO, Na-22, Cl-36 and I-125.

Nuclide	A_0 [MBq]	ε [%]	D_p [m^2/s]	K_d [m^3/kg]
HTO	17.1	0.7	7×10^{-12}	-
Na-22	1.4	0.7	7×10^{-12}	6×10^{-5}
Cl-36	1.3	0.3	$< 1\times10^{-12}$	-
I-125	1.4	0.7	7×10^{-12}	6×10^{-5}

The measured breakthrough curves were analyzed using a numerical model that takes into account advection, (Taylor) dispersion, and molecular diffusion in the flow channel, and matrix diffusion and sorption in the rock matrix. Values of related parameters, found by fitting the measured breakthrough curve by the model solution, are shown in TABLE III. As a result, a pore diffusion coefficient of 1×10^{-12} m^2/s was determined for Cl-36, and 7×10^{-12} m^2/s for HTO, Na-22 and I-125. Furthermore, for Na-22 and I-125 a distribution coefficient of 6×10^{-5} m^3/kg was determined which indicates weak sorption of them. The K_d of Na-22 given by the model was about one order of magnitude lower than the one from batch sorption experiments. The modeled breakthrough curves fail to explain completely the early part of the measured curve. The first breakthrough takes place earlier and gets higher than the model predicts. These issues create some uncertainty the errors in the results. However, clear power-law behavior was observed in the late part of breakthrough curve which is typical for matrix diffusion and sorption.

The analysis of breakthrough curves is not straightforward. Here the analysis was done by first assuming that K_d for HTO is equal to zero and the average porosity of laboratory measurements (0.7 %) was used in the model [15]. After fixing K_d and ε, D_p was adjusted so that good agreement with the experimental curve was found. In case of Na-22 and I-125, the same values for D_p and ε were used, and K_d was adjusted to find good agreement with experiment. It was impossible to explain the breakthrough curve of Cl-36 using the same values of D_p and ε as for HTO. Since it is unrealistic to decrease only D_p or ε, both were decreased so as to find good agreement with the experiment. It is possible that, as chloride is an anion, it is repelled by the negatively charged mineral surfaces and thus it can diffuse only in part of the pore space. This will cause also decrease of the diffusion coefficient. Similar results for Cl-36 have also been obtained in laboratory studies. This is only an assumption and it will be verified by the on-going experiment.

CONCLUSIONS

A series of in-situ experiments for investigating the in-situ diffusion and sorption properties of selected radionuclides was introduced. These experiments are and will be performed under conditions that are equivalent to those of the final repository for spent nuclear fuel in Finland. They will provide data on matrix diffusion and sorption of radionuclides relevant under in-situ conditions in the repository depth and valuable information for the safety analysis of the repository.

Predictive modeling of TDE and GPDE show promising results and it is likely that they will offer new information about the retention properties of the bedrock. However, some uncertainties remain: 1. In TDE the breakthrough time depends strongly on the diffusion coefficient and therefore duration of the experiment may be intolerable. 2. In GPDE it has been challenging to dry the rock matrix, which may have a considerable effect on the results, although a partly dried rock can be taken into account in the modeling as a finite depth of the rock matrix. If diffusion coefficient determined by WPDE (7×10^{-12} m^2/s) and the present estimate for the lower detection limit (1×10^{-6}) is used to predict the first breakthrough in TDE, a detectable amount of tracer will appear in the observation holes after one year.

In the first WPDE tracer test, the retarding effect caused by matrix diffusion and sorption were evident and preliminary analysis of the measured breakthrough curves were successfully performed. However, the advection dominated part of the curve was not fully explained by the model. The effects caused by an inhomogeneous flow field, a rock matrix of dual porosity, or a disturbance of rock matrix caused by drilling might be able to explain the early part of the breakthrough curve. At the moment the effects of these properties are being estimated.

The second WPDE tracer test with a lower flow rate (10 µl/min) and HTO, Na-22, Cl-36, Ba-133, and Sr-85 as the tracers is now running. The aim is to get information about diffusion and sorption for a wider range of radionuclides and at multiple time scales. It is believed that diffusion will more efficiently level out the possible unevenness of the flow field in this experiment. The early part of the breakthrough curve will then be easier to explain by the model, and more importantly, the uncertainties that are connected to the control of the matrix diffusion by the flow field should decrease. Since in the first WPDE the late part of the Cl-36 breakthrough curve was near to detection limit, its initial activity was increased in second experiment. This will raise the tailing of the breakthrough curve and enable more reliable determination of the rock matrix retention properties for Cl-36.

Parallel to these in-situ experiments, a number of laboratory experiments are being performed, some of which were referred to already here. Samples for the laboratory studies were taken from the drillcores of in-situ measurement intervals. In addition to matrix diffusion and through diffusion experiments, independent structural and mineralogical studies as well as porosity measurements are being performed on them.

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