The diffusion of tritiated water and chloride through granodiorite

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Background

Spent nuclear fuel from Finnish power plants will be disposed of deep in the crystalline bedrock in western Finland. When they are possibly released into the bedrock, radionuclides will be transported by advection along water conducting fractures. Retardation can occur by molecular diffusion from the fractures into the stagnant pore water and/or by immobilisation onto mineral surfaces in the rock matrix. Estimating the transport behaviour of radionuclides in groundwater is important in assessing the risk to health due to radionuclide release at the waste disposal site.

The Swiss National Cooperative for Disposal of Radioactive Waste (Nagra) have been conducting extensive in-situ experiments at the Grimsel test site (GTS) in the field of radionuclide transport and retention. The second Long Term Diffusion (LTD) experiment started in autumn 2013 using radionuclides H-3, Na-22, Cs-134, Cl-36 and Ba- 133 as well as nonradioactive element selenium. In this work, diffusion of H-3 and Cl-36 are studied in the laboratory scale to be compared with the results of on going in-situ experiment.

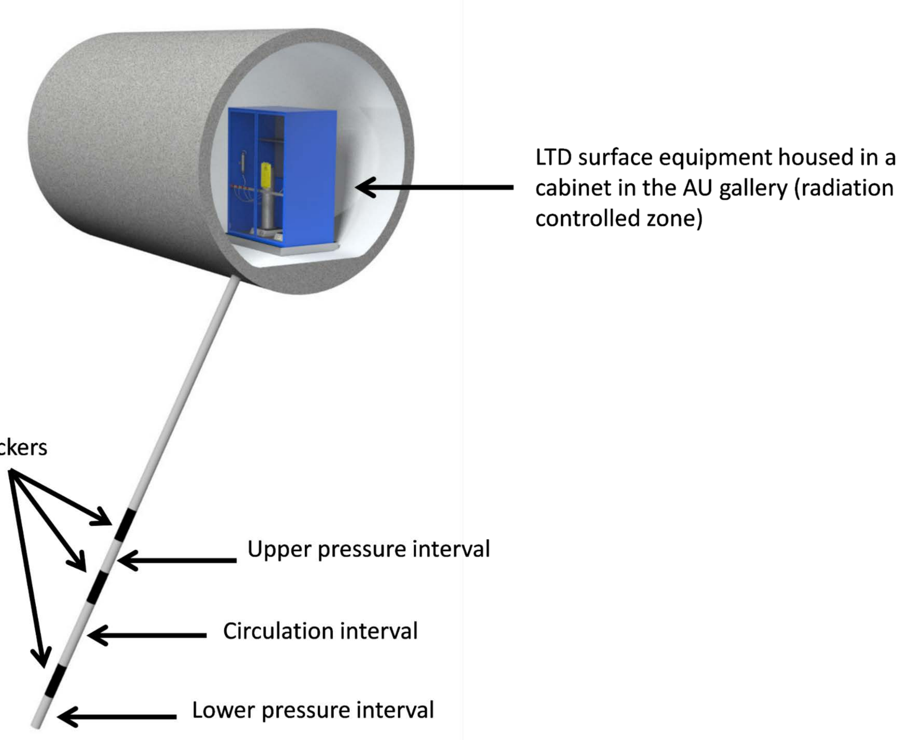

Fig. 1. Schematic illustration of the LTD in situ experiment consisting of an 8 m borehole

Experiments

The effective diffusion coefficients of H-3 and Cl-36, which do not interact significantly with the matrix pore walls, were obtained by in- and through diffusion experiments in a Grimsel granodiorite (GG) rock block (length 20 cm and diameter 30 cm, see Fig 2). Changes of the H-3 and Cl-36 activity in the inlet and observation bore holes was measured using liquid scintillation counting.

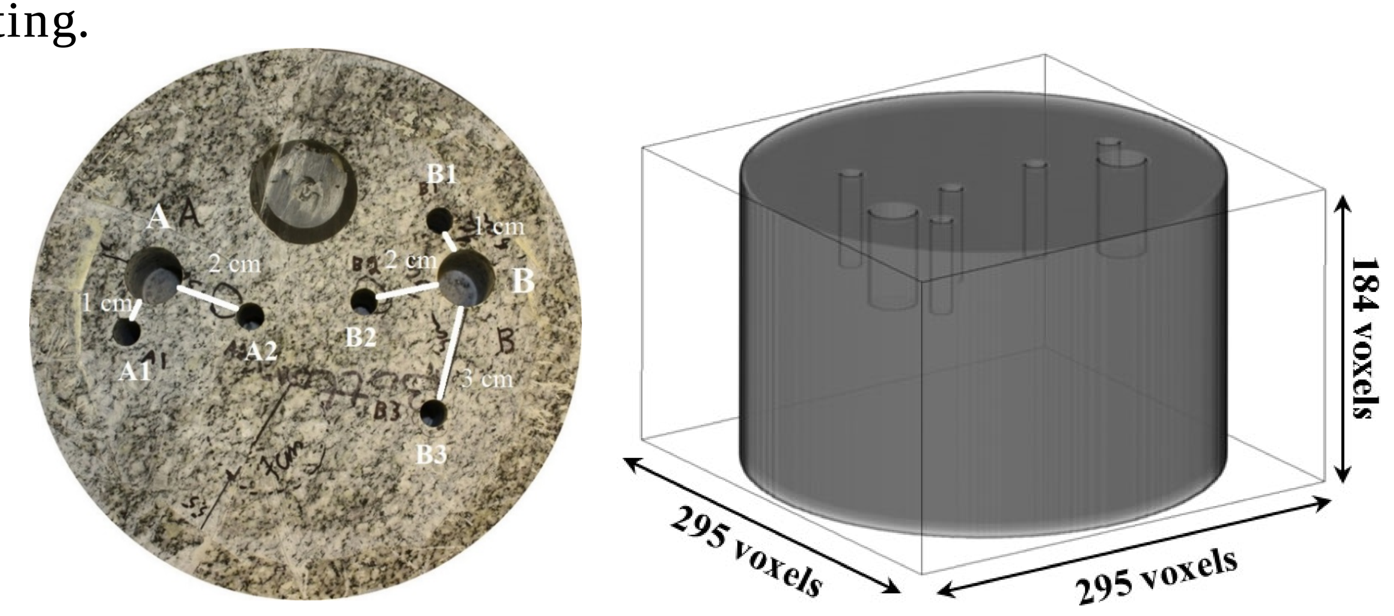

Fig. 2. Grimsel granodiorite rock block used in- and through diffusion experiment (left) and schematic drawing of experimental setup used in simulations (right).

The measured breakthrough curves were modelled using Time Domain Random Walk (TDRW) modelling which has been found to be powerful tool when for analyzing results of in-situ and laboratory experiments especially when initial or boundary conditions are complicated [1, 3]. The modelling grid was created according dimensions of the GG sample (see Fig 2.) and the measured breakthrough curves of H-3 and Cl-36 to five observation holes (A1, A2, B1-B3) were modelled by varying porosity and effective diffusion coefficient (D_e). In practice, the porosities were determined from the early parts and D_e from the late parts of the curves.

Results

The average effective diffusion coefficients (D_e), that were determined from breakthrough to observation holes (Fig 3.) using concentration decrease of inlet holes as a source term, were $(3.5\pm0.8)\times10^{-12}$ m^2/s for H-3 and $(2.6\pm0.8)\times10^{-12}$ m^2/s for Cl-36. Average porosity determined by TDRW modelling was 0.010±0.001. Porosities and diffusion coefficients from individual observation holes are presented in table 1. There are only slight variations in the results except in case of A1. Higher D_e is possibly caused by feature with higher water conductivity in rock matrix of this area. D_e of H-3 is slightly higher than D_e of Cl-36. This is caused by anion exclusion. The effect is not large in Grimsel granodiorite because of the relatively large pore openings of this type of rock matrix. Results are in line with the D_e of selenium $(2.5\pm1.5)\times10^{-12}$ m^2/s determined from similar diffusion experiment in the same GG rock block [1]. D_e of H-3 is also in line with the one determined from the in-situ LTD experiment 2.0×10^{-12} m^2/s [2, 3].

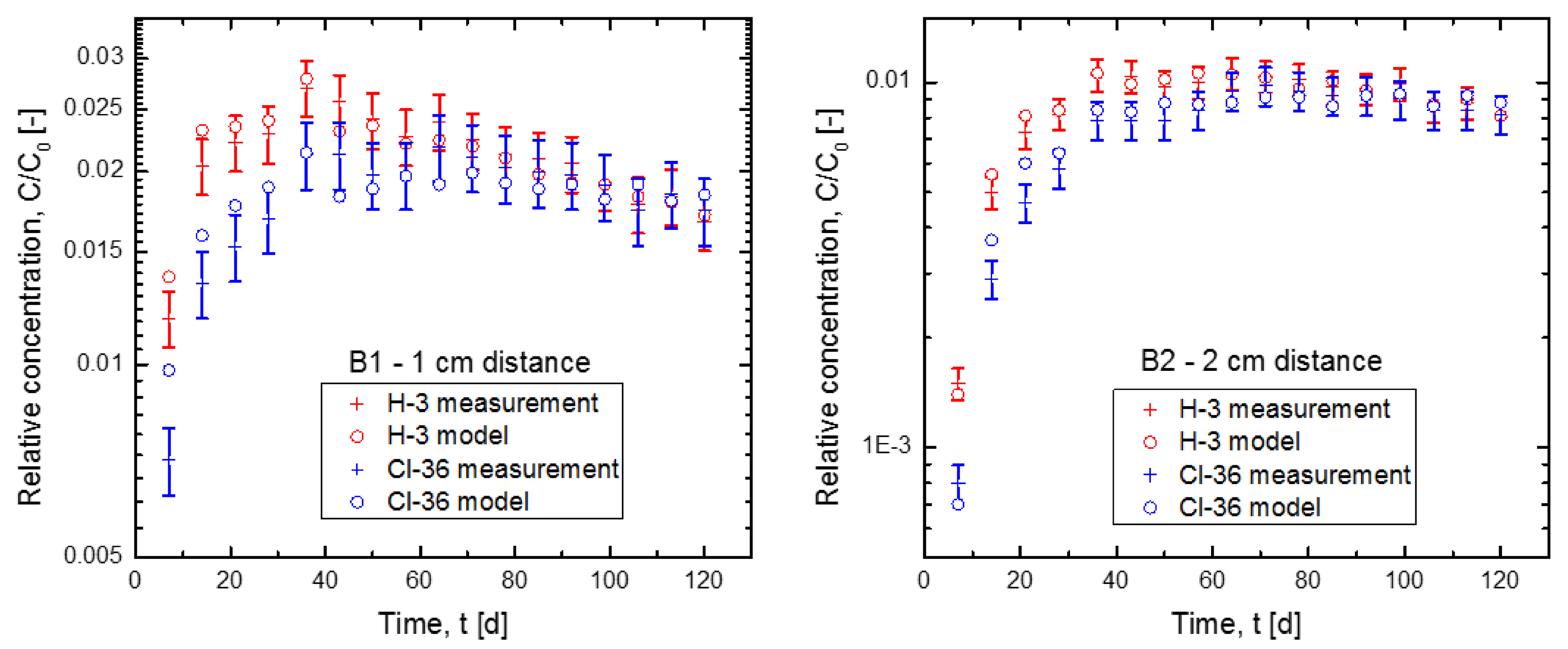

Fig 3. Measured and modelled breakthrough curves for H-3 (red) and Cl-36 (blue) to observation holes B1 (left) and B2 (right).

Table 1. Effective diffusion coefficients and porosities determined from individual observation holes using TDRW modelling.

	HTO		Cl-36	
	D_e [m^2/s]	Porosity [-]	D_e [m^2/s]	Porosity [-]
A1	$(5.5 \pm 1.0)\times10^{-12}$	0.008 ± 0.001	$(5.0 \pm 1.0)\times10^{-12}$	0.008 ± 0.001
A2	$(3.8 \pm 0.8)\times10^{-12}$	0.012 ± 0.001	$(2.8 \pm 0.8)\times10^{-12}$	0.011 ± 0.001
B1	$(3.2 \pm 0.8)\times10^{-12}$	0.014 ± 0.001	$(2.4 \pm 0.8)\times10^{-12}$	0.013 ± 0.001
B2	$(3.6 \pm 0.8)\times10^{-12}$	0.010 ± 0.001	$(2.8 \pm 0.8)\times10^{-12}$	0.009 ± 0.001
B3	$(3.2 \pm 0.8)\times10^{-12}$	0.008 ± 0.001	$(2.2 \pm 0.8)\times10^{-12}$	0.007 ± 0.001

Conclusions

- Slight variation in De values can be explained by structural heterogeneities of the pore structure.
- The results indicate only small anion exclusion effect in Grimsel granodiorite.
- D_e of H-3 and Cl-36 are in line with D_e of selenium determined from similar diffusion experiment in the same GG rock block.
- It is observed that difference between in-situ and laboratory conditions in block scale do not affect considerably to D_e of H-3.

References

[1] Ikonen, J., Voutilainen, M., Söderlund, M., Jokelainen, L., Siitari-Kauppi, M., Martin, A., 2016. Sorption and diffusion of selenium oxyanions in granitic rock J. Contam. Hydrol. 192, (203-211).

[2] Ikonen, J., Sardini, P., Jokelainen, L., Siitari-Kauppi, M., Martin, A., Eikenberg, J., 2016. The tritiated water and iodine migration in situ in Grimsel granodiorite. Part I: determination of the diffusion profiles. J. Radioanal. Nucl. Chem. http://dx.doi.org/10.1007/ s10967-016-4890-6.

[3] Ikonen, J., Sardini, P., Siitari-Kauppi, M., Martin, A., 2016. In-situ migration of tritiated water and iodine in Grimsel granodiorite, Part II: Evaluation of the diffusion coefficients by TDD modelling. submitted.