Analysis of Retardation Properties of Highly Sorbing Nuclides from the Sorbed Amounts on the Fracture Surface

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1. Introduction

Migration experiments have been carried out for highly sorbing nuclides through a fractured granite. This work is one of the series experiments and described experimental results of sorption and migration for *THO*, ^{237}Np and ^{241}Am. The objectives of this study are as follows,

To investigate the applicability of retardation coefficient measured in batch test ,

To study the extent of matrix diffusion according to flow rates or contact times.

To validate migration processes by identifying the migration plumes between experiments and simulations for various cases, and

To study the applicability of the variable aperture channel model with the particle tracking method.

2. Experimental system

Granite cores were sampled at the boreholes in the Äspö HRL. Specific surface area of the grinded sample was about 1.1 m^2/g by BET test. The core was cut as the length of 150*mm* and the diameter of 52*mm* for the migration test. The *pH* was 7.5 and DOC was 42.3*mg/l* for the water. The *Eh* values were varied from +50 *mV* to -200 *mV*.

The experimental setup was designed like an autoclave type reactor to endure high pressure conditions. A core sample having a single fracture was placed into a cylindrical stainless steel sleeves. For the migration experiment, the actinide cocktail was injected for 10 days with a flow rate of 0.04 *ml/h*. Afterwards, natural groundwater of the drill hole was pumped. Afterwards, the core was recovered and opened.

In order to map the aperture distribution in the fracture core, the optical images of the rock slices and the images of X-ray tomography were analysed. The fracture of the core was described as a two dimensional geometric field in the simulation with a variable aperture channel model. From the scanning image of the fracture at each slice, the aperture value at each subsquare point was calculated. The fracture field is well developed and there is no local closed zone except both edges of the inlet face. By summing up the aperture value at each point, the pore volume in the fracture space was obtained as 2.33 cm^3. And the mean aperture value was calculated 0.031 *cm* by dividing the pore volume with the geometric surface area of the fracture plane.

The fluid flow through the fracture was then calculated for a constant flow rate as well as for constant pressure conditions. The volumetric flow rate, Q_{ij}, at a subsquare *i*may be written as:

$$Q_{ij} = C_{ij} (P_i - P_j) \qquad (1)$$

By rearranging the above equation for each node, it can be obtained a system of linear equations in the form,

$$[B]\,[P] = [E] \qquad (2)$$

where ***[B]*** is a coefficient matrix describing the flow conductance. The matrix ***[P]***is an array describing the pressure distribution and ***[E]***is an array describing net flow rates. Except for the nodes at the boundaries, the pressure at each node can be solved.

After obtaining flow vectors at all nodes, solute transport can be simulated in this flow field. The simulated pressure distributions in the fracture were shown at the bottom figure in Fig.1. The pressure drop between the inlet and the outlet is about 24 *dyne/cm²*when flow rate is *0.04 ml/h*. That is, there is only a little pressure drop along the main flow field. The distribution of the pressure shows almost symmetric to the center line of the fracture plane along the longitudinal axes.

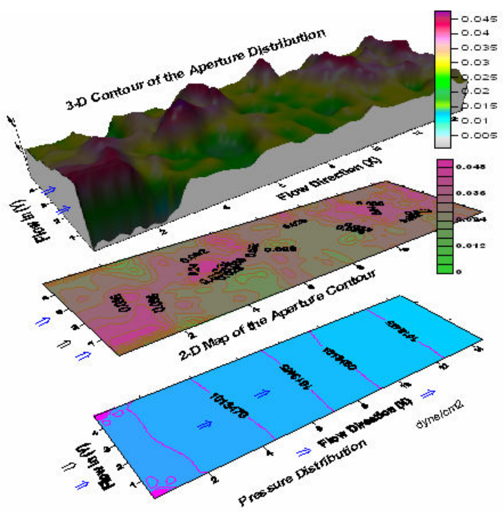

Fig.1 Aperture distribution of the granite core.

3. Migration of the Radionuclides

A two-dimensional random-walk particle tracking algorithm was used to simulate the solute transport through the flow fields. Four transport processes were considered : advection, longitudinal dispersion, diffusion into the rock mass, and sorption. Particle displacements in each time step consisted of an advective displacement based on local velocities calculated using the pressure field, random diffusive displacement, and retardation by sorption.

During the migration test, it was not observed any elution peaks of ^{237}Np. Thus, The remaining concentration of ^{237}Np in the fracture after migration test is determined by alpha-counting and dissolution of the slice. And an average concentration profile over the whole fracture is shown in Fig.2. This profile was obtained by taking into account that in total eluted water was about 210 *ml*. From the profile the retardation factor, *Rs*, is determined as 243.

In order to simulate the migration plumes of ^{237}Np, the following four cases were considered. The simulation was carried out with the particle tracking method when the water flow rate was *0.04 ml/h* and shown as three dimensional plumes as a function of eluted volume.

In the first case, the radionuclide retards only by sorption with *Rs* = 69. The nuclides flow out almost when the water eluted about 50 *ml*. The case 2 is added the matrix-diffusion process from the case 1. The proposed pore diffusivity is $1.5*10^{-10}$ m^2/s as same as the tritium. It shows much retarding effect than the case 1. The nuclide starts to come out from the water elution volume of 100*ml*. In the case 3, the radionuclide retards only by sorption as the case 1 but *Rs* = 243. The main migration plume comes out when the water eluted about 80 *ml*. The case 4 is added the matrix diffusion process from the case 3. It shows much retarding effect than the case 3. Its migration plume at 210 *ml* of the eluted volume shows very similar profile with the experimental one as shown in Fig.7. Thus matrix diffusion process gives a significant retardation and dispersion effect in the migration process comparing to the case of sorption only in this flow system. Therefore, when *Rs* = 243 and the pore diffusivity is $1.5*10^{-10}$ m^2/s. The migration plume fits very well to the experimental result.

The form factor for surface area includes deviations of the actual surface and void volume of the fracture from the calculated geometrical properties. Combining the *Np* sorption data with the information obtained from sorbed *Np* in the core, a form factor for the surface of 21.5 is calculated. From the batch *Ks* data, *Rs* value of ^{237}Np can also be obtained. It is very interesting topic to compare the values secured from the static batch and the dynamic migration test. If the determined values are adopted from eq.(2), such as $Ks = 0.16cm$ and $f = 21.5$, and d = 0.031 *cm*, then *Rs* of ^{237}Np is calculated as 111, which is lower value than the value of 243 obtained from the migration simulation. It may be caused by the difference of contacting time. In the batch test, radionuclides were contacted for 14 days. While in the migration test, they were contacted more than 50 days. The longer contact time gives much chance to diffuse into the pore and to sorb. Thus it will give larger value of *Rs* than 111. That is, *Rs* = 243 from the migration is seemed as a reasonable value.

4. Conclusion

The retardation properties of highly sorbing nuclides are successfully analyzed from the sorbed amounts on the fracture surface. The migration of ^{237}Np through a fractured granite core showed much retarded effect than expected from the static sorption test. It is interpreted due to difference of the contact time between nuclides and rock fracture. That is, much contact time in migration test shows much retarding phenomena than the batch test. The developed variable aperture channel model was successfully applied in describing the migration of radionuclides in the rock fracture. Results from the migration test and modelling of the transport show that diffusion into the interconnected micropore space in the rock mass has an effect on the retardation, especially for the case of tritium and ^{237}Np.

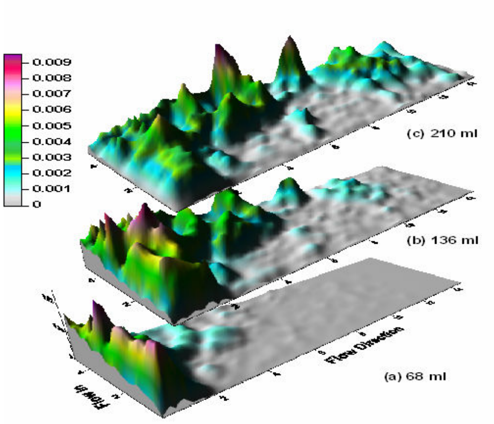

Fig.2 Simulation of the migration plumes of ^{237}Np

REFERENCES

[1] B.Kinzler, P.Vejmelka, Römer, J., Fanghänel, Th., P.Wilberg, M.Jansson, T.Eriksen ; Swedish-German actinide migration experiment at ÄSPÖ HRL., Journal of Contaminant Hydrology 61 (2003) 219-233

[2] J.Römer, Kienzler, B., Vejmelka, P. E.Soballa, A.Gortzen, M.Fuss, Actinide migration experiment in the HRL ÄSPÖ, Sweden: results of laboratory and in situ experiments (Part II). Wissenschaftliche Berichte, FZKA-6770 (Oktober 2002

[3] C.K.Park, T. T. Vandergraaf, D. J. Drew, and Hahn, P.S., Analysis of the Migration of nonsorbing tracers in a natural fractures in granite using a variable aperture channel model, J. of Cont. Hydrol.26, 97 (1997).

[4] J.Hadermann, The Grimsel Migration experiment : integrating field experiments laboratory investigations and modelling, J. of Cont. Hydrol. 21 (1996) 87-100

[5] C.K.Park, Ryu,B.H., and Hahn, P.S., Migration characteristics of some chemical species in a granite fracture according to their chemical properties, Kor. J. of Chem. Eng., 19(5) 765 (2002)