Geophysical Research Abstracts,
Vol. 11, EGU2009-8827, 2009
EGU General Assembly 2009

Radionuclide migration in clayrock host formations for deep geological disposal of radioactive waste: advances in process understanding and up-scaling methods resulting from the EC integrated project 'Funmig'

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One of the 'pillars' supporting Safety Cases for deep geological disposal of radioactive waste in clayrock formations is the knowledge base regarding radionuclide (Rn) retention by sorption and diffusion-driven transport which is why the EC integrated project 'Funmig' focused a major part of its effort on advancing understanding of these two macroscopic phenomena. This talk presents some of the main results of this four year effort (2005-2008).

One of the keys to understanding diffusion-driven transport of anionic and cationic radionuclide species in clayrocks lies in a detailed understanding of the phenomena governing Rn total concentration and speciation (dissolved, adsorbed) in the different types of pore spaces present in highly-compacted masses of permanently charged clay minerals. Work carried out on a specifically synthesized montmorillonite (a model for the clay mineral fraction in clayrocks) led to development, and preliminary experimental validation, of a conceptually coherent set of theoretical models (molecular dynamics, electrostatic double layer, thermodynamic) describing dissolved ion and water solvent behavior in this material. This work, complemented by the existing state of the art, provides a sound theoretical basis for explaining such important phenomena as anion exclusion, cation exchange and the diffusion behavior of anions, weakly sorbing cations and water tracers.

Concerning the behavior of strongly sorbing and/or redox-reactive radionuclides in clay systems, project research improved understanding of the nature of sorption reactions and sorbed species structure for key radioelements, or analogues (U, Se, Eu, Sm, Yb, Nd) on the basal surfaces and in the interlayers of synthetic or purified clay minerals. A probable mechanism for Se(IV) retention by reduction to Se° in Fe2+-containing clays was brought to light; this same process was also studied on the Callovo-Oxfordien clayrock targeted by the French radwaste management program.

The migration of most radionuclides in clayrocks, in particular the actinides, is limited by their strong sorption on rock mineral surfaces. Much effort was devoted in Funmig to improving understanding of this process on the clayrocks being studied in the Swiss, Belgian and French radwaste management programs. Specific attention was focused on (i) elucidating the effect of dissolved organic matter on Am(III), Th(IV), Eu(III) sorption on clayrock surfaces and (ii) determining the link between Kd measured on dispersed rock systems and the Kd operant in intact rock volumes, i.e. during diffusion. Regarding the latter question, results indicate that Kd values for 'dispersed' and 'intact' materials are quite similar for certain elements (Na, Sr, Cs, Co). On the other hand, Kd values obtained by modeling results of diffusion experiments involving strongly sorbing elements as Cs, Co and Eu were always significantly smaller than those predicted based on sorption data measured in corresponding batch systems. This is an area where additional research is being planned.

A major effort was devoted to improving understanding of the effects of small-scale ([U+F06D]m to cm) clayrock structure and large-scale (dm to hm) mineralogical composition on radionuclide diffusion-retention. The program focusing on the small-scale produced a method for simulating the results of tracer diffusion in an

intact rock based on the actual rock microstructure of the rock sample to be used in the diffusion experiment. This model was used to predict / inverse model the spatial distribution of highly sorbing tracers (Eu, Cu). This overall approach is also being used to understand how changes in mineralogical composition can affect the values of macroscopic diffusion parameters (De, tortuosity, anisiotropy). At a much larger scale, the results of (i) a geostatistical analysis of clayrock mineralogical variability and (ii) measurements of De and Kd dependence on mineralogy for Cs and Cl, were combined to create models of parameter variability at the formation scale. These models were used to evaluate the effects of formation scale heterogeneity on predictive modeling of radionuclide migration. Measurements and modeling of natural tracer profiles were also carried out in order to evaluate the diffusion characteristics at geological time and space scales.