Humic colloid-associated migration of radionuclides in an argillaceous formation

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It has been known for a long time now that natural humic colloids in the argillaceous Boom Clay formation (Belgium) modify the speciation and transport characteristics of several long-lived radionuclides [1]. However, universal formalisms to describe the complex suite of processes and mechanisms in such environment were lacking, resulting in 'operationally defined migration parameters' [2] that did not allow significant confidence building with regard to safety assessments.

In this paper we present results from long-running (>10 years) and short-term column migration experiments, and from batch experiments with several radionuclides (Cm, Pu, Np, Tc and Pa) that differ significantly in both valence state and inorganic speciation in Boom Clay. In spite of their differences, we observed strikingly similar features among these different elements, resulting from a general humic colloid-associated migration pathway that outweighs all other transport processes.

Using a classical bottom-up approach, a geochemical modelling concept relying on thermodynamic sorption models (TSMs) and colloidal transport, is presented that succeeds in describing both adsoption and migration data. The role of humic colloids as a transport vector is thoroughly discussed and evaluated within the larger framework of geological disposal of radioactive wastes.

[1] Henrion *et al*. (1985) *Eng. Geol.* **21**, 311–319. [2] SAFIR 2–Safety Assessment & Feasibility Interim Report 2 (2001) NIROND 2001–06 E, Brussels, Belgium

Bed-scale reaction transport modeling of dolomitization reveals the emergence of self-organizing patterns

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Outcrop samples taken at 30-cm spacings along 60 to 150 m lateral transects reveal spatial patterns in the porosity and geochemistry of Mississippian, Eocene, and Pliocene dolomites. Spatial variance in all attributes exhibits a large near-random component and spatial correlation over ranges of 5-12 m. Some attributes also exhibit a low magnitude pattern at wavelengths up to ~40 m. The Pliocene dolomite shows these patterns can form during dolomitization as they are absent in undolomitized limestone. The longer range pattern maybe due to weathering in the Eocene example; post-dolomitization burial diagenesis can be eliminated as the origin of patterning in all examples.

Reaction-transport models (RTM) coupled to textural evolution during dolomitization reveal insights to the formation of the lateral patterns. We used Sym.8, a continuum-based water-rock interaction simulator that uses conservation of elemental mass, diffusive and advective mass-transfer, equilibrium reactions among solutes, and kinetic reactions between aqueous solutes and minerals. Model domain is a 2 m-high bed with 10 x 30 cm cells. Calcite grains and dolomite crystals are modeled as spheres that dissolve or grow through the simulation. Permeability is dependent on evolving porosity and total grain surface area. Models tested the effect of variations in fluid Ca:Mg ratios and alkalinities, fluid-flow rates, bed height, and extent of initial porosity and textural heterogeneity.

The patterns formed in the RTM are identical to those observed in the ancient dolomites – a reduction in the amount of randomness, short-range correlation to 3-5 m, and a longer range cyclic pattern, although it is not ubiquitous. Pattern formation is weakly sensitive to textural heterogeneity, fluid-flow rate, and fluid chemistry. Critical to pattern formation are feedbacks between porosity heterogeneity, convergence and divergence of flow, and solute diffusion.