Contribution ID: **378** Type: **Invited**

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Tuesday, 13 May 2014 08:30 (30 minutes)

Humic colloid-associated migration of trivalent radionuclides in an argillaceous formation

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It is generally accepted 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 a comprehensive overview of the latest achievements that have been obtained in the understanding, interpretation and description of trivalent radionuclide retention and migration in the Boom Clay. Using a classical bottom-up approach, a geochemical modelling concept relying on thermodynamic sorption models (TSMs) is presented. With this approach, we have succeeded in describing adsorption of Eu(III) and Am(III) in batch suspensions of increasing complexity, from relatively straightforward mixtures of illite and dissolved organic matter (DOM) [3], to Boom Clay suspensions. The results of these adsorption experiments demonstrate that trivalent radionuclides are strongly bound to the solid phases present in Boom Clay, but that their solid-liquid distribution is heavily influenced by DOM.

Hereafter we discuss results from short-term and long-running (>10 years) column migration experiments with Am(III) on Boom Clay cores. These experiments clearly indicate the existence of a kinetically-controlled colloidal transport pathway that determines the flux of Am(III) through the clay core. A modified kinetic colloid transport model has been developed [4], based partly on the aforementioned adsorption models, that allows for the quantitative description of all experimental data obtained thus far. In this model, it is considered that the radionuclide is predominantly transported as an organic matter complex/colloid that slowly dissociates, and both the colloid as well as the dissolved radionuclide ions are capable of sorbing to the solid phase.

The role of humic colloids as a transport vector is thoroughly discussed and evaluated within the larger framework of geological disposal of radioactive wastes.

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Session Classification: Chemistry of Nuclear Fuel Cycle 2

Track Classification: Chemistry of Nuclear Fuel Cycle / 1st ASGARD International Workshop