GROUPING OF RADIONUCLIDES IN THE BELGIAN GEOLOGICAL DISPOSAL CONCEPT FOR RADIOACTIVE WASTE

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Introduction

ONDRAF/NIRAS is entrusted by law with developing a coherent policy for safe management of all radioactive waste on Belgian territory and, subject to parliamentary and governmental review. For the safe long-term management of the high-level and/or long-lived radioactive waste in Belgium, ONDRAF/NIRAS proposes deep geological disposal in poorly indurated clays (Boom Clay or Ypresian clays) taking into account international accepted safety management requirements (NIROND, 2009). Research on the behaviour of radionuclides and heavy metals is crucial because it provides the necessary understanding of the system. Based on this understanding abstracted models and associated parameters are derived to be used in Safety Assessments (SA). These SA's should give proof, based on scientific elicitation using multiple lines of evidence, developed models and calculations, that radiotoxic and chemically toxic substances contained within the waste are not reaching the biosphere at harmful concentration levels. The previous Safety Assessment and Feasibility Interim Report (SAFIR 2) (NIROND, 2001) was released in 2001. Now, a first Safety and Feasibility Case (SFC 1) is being developed including new state-of-the-art knowledge to assess a geological disposal in poorly indurated clays.

Methods

A 4 group system for the radionuclides has been proposed by SCK•CEN for the SFC-1 (NIROND, 2013). Each group consists of a group 'representative' and 'other' group elements. The elements are grouped based on thermodynamic considerations (inorganic speciation in Boom Clay pore water), experimental observations (sorption, solubility in presence of organic matter, migration behaviour) and scientific literature (environmental conditions). The group representatives are selected based on the amount of available experimental and literature data to support understanding, description and prediction of its retention/migration behaviour under Boom Clay conditions. Subsequently, phenomenological models are developed that should fit all solutes of a given group. The four groups are (with their group representative):1) Reference non-retarded tracer (HTO), 2) Solutes subjected to anion exclusion (I), 3) Solutes influenced by cation-exchange (Cs,Sr), and 4) Solutes interacting with natural organic matter (Tc).

The developed models for the different groups are then tested to analyse their applicability in SA's. Certain conditions which are important factors in phenomenological models may not necessarily be important factors in the long-term SA.

Results

Specifically for the solutes of group 4, Maes et al. (2011) developed a phenomenological transport model for transition metals, actinides and lanthanides which interact with the natural organic matter present in the Boom Clay. In this model radionuclides are present in solution as either 'free inorganic' radionuclides ($[RN_{inorg}]_{liquid}$) or as a mobile complex with the organic matter (RN-OM). The relationship between both is described by a complexation constant or the ratio between the association and dissociation kinetics

(k_{comp}/k_{decomp}). ($[RN_{inorg}]_{liquid}$ and RN-OM can interact with the solid phase and this is described by a retardation factor R_{RN} and $R_{RN\text{-}OM}$ respectively. Dissolved organic matter is poorly retarded within the Boom Clay and therefore $R_{RN\text{-}OM}$ is only of secondary importance to describe RN-coupled transport at the scale of the geological formation. The kinetically controlled complexation reaction (k_{comp}/k_{decomp}) can thus enhance the transport of radionuclides (e.g. Tc and U). When these elements associate with the dissolved organic matter, this increases the concentration of the element in solution and decreases its sorption onto Boom Clay solid phases.

The above described mathematical model, used to fit the radionuclide migration experiments, consists of 9 variables and is therefore relatively complex to be used in SA calculations. Based on this model and the experimental results of Bruggeman et al. (2012), Govaerts and Weetjens (2015) derived a reduced, more practical, model which can be used in SA calculations. The reduced model doesn't take into account the kinetic rate of the (de)complexation reaction (k_{decomp} and k_{comp}) as its effect is negligible when dealing with space and time scales relevant for geological disposal (i.e., upscaling).

Conclusion

In the context of the SFC-1, phenomenological transport models are developed for each of the group representatives. These models are based on an extensive experimental program of more than 30 years and describe the current understanding of the processes (and associated parameters) affecting radionuclides transport in the Boom Clay. Nevertheless, these models, mostly based on "short-term" experiments and in a limited set of conditions, can include processes which may eventually have a limited effect when used to estimate radionuclides spread on large time scales and over the thickness of the geological barrier. ONDRAF/NIRAS will therefore keep on performing research in order to confirm the applicability of the 4-group system to the expected conditions within the host rock for the period of concern and the strategy followed to develop these phenomenological models. As a matter of course these models should be thoroughly re-evaluated before being used in performance assessment calculations.

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