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Modelling the radionuclide transfer from bedrock to surface systems at Forsmark site (Sweden)

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Abstract. Quaternary sediments and soils at the Forsmark site (Sweden) would constitute the last barrier for radionuclide migration interposed between a deep repository of high level nuclear wastes (HLNW) and surface ecological systems. The retention capacity of these sediments is evaluated by means of reactive transport simulations. Two different scenarios have been modelled: (1) Inflow of deep fluids carrying radionuclides into shallow aquifers hosted in the carbonate glacial till, and (2) Inflow of deep fluids carrying radionuclides into organic matter-bearing clays. The model results predict that caesium is very efficiently retained in both scenarios for periods longer than 3000 years due to the strong affinity of this ion with the *frayed edge* sites of the illite layers. Strontium is also retained via cation exchange in illite and, in a lesser extent, via co-precipitation in calcite. The retention efficiency for strontium, however, quickly decreases to 0 after 1000 years. Uranium is retained in both scenarios with retention efficiencies higher than 40% at long term (>3000 years) due to precipitation of amorphous uraninite and adsorption on Fe(III) oxyhydroxides. Radium is only retained in the till aquifer since co-precipitation with barium sulphate does not occur in the clay porewater.

1. INTRODUCTION

The concepts of deep geological disposal of high level nuclear waste (HLNW) include a number of natural and engineered barriers designed to prevent radionuclide migration to surface systems in case of isolation failure of the waste canisters [1]. The Swedish concept of HLNW storage is based on the deep disposal in a granite environment, and two candidate sites, Forsmark and Laxemar, are currently under investigation by the Swedish Nuclear Fuel and Waste Management Company (SKB). In both areas, Quaternary regoliths overlying the granite bedrock would constitute the last natural barrier for the retention of radionuclides before reaching the surface systems. The retention capacity of these deposits depends on the chemical behaviour of the radionuclides and the geochemical features of the sediments (mineralogy, porewaters, organic matter content).

Deterministic calculations of radionuclide releases from the geosphere (granite) [1] show that Ra-226, Cs-135 and I-129 would be the main dose contributors to the surface systems at long term in case of canister failure. The determination of the hydrogeochemical behaviour of these radionuclides when in contact with Quaternary soils is, therefore, very relevant. The behaviour of radium and caesium, along with other radionuclides of interest such as uranium and strontium, is predicted in this work by using 2D reactive transport calculations in which retention is simulated under two different hydrodynamic environments in the Forsmark area. Iodine has not been modelled at this stage since the thermodynamic database for this element is still not well developed. The important outcome of the project is the categorization of the retention mechanisms considered.

2. CONCEPTUAL MODEL AND NUMERICAL IMPLEMENTATION

2.1 Surface geology and groundwaters at Forsmark

The transfer of radionuclides from geosphere to surface systems may take place in different geological environments at Forsmark (Fig. 1): (1) direct contact between crystalline bedrocks and biosphere-atmosphere (13% of areal extension), (2) Quaternary carbonate-rich till, which is the most abundant outcropping Quaternary deposit (~75% of surface extension), and (3) Quaternary lake sediments and wetlands. In the latter case, lake sediments commonly overlie glacial till. The thickness of Quaternary deposits is highly variable, usually from 0 to 2–3 m, although in some localities can reach over 10 m.

Glacial till is basically a mixture of calcite (20–30 wt%) and clay minerals (mainly illite). In addition, elemental analysis suggests the presence of minor amounts of other minerals such as Fe(III) oxyhydroxides [2]. On the other hand, the lake sediments consist of anoxic glacial clays and biogenic-derived sediments (*gyttja*), with abundant organic matter. The extensive microbial activity expected in this environment can eventually lead to the precipitation of iron sulphides, as observed in similar environments [3].

Shallow groundwaters flowing through the till show a fairly constant chemistry, with a predominance of Ca and HCO_3 (ionic strength, I $\sim 1.0 \times 10^{-2}\,\mathrm{mol \cdot L^{-1}}$) and neutral pH that reflects the equilibrium with the host rock. Glacial clay porewater is believed to be more diluted (I $\sim 6.0 \times 10^{-3}\,\mathrm{mol \cdot L^{-1}}$) with high content of organic compounds either dissolved or as suspended particulate ([DOC] $\sim 1.5 \times 10^{-3}\,\mathrm{mol \cdot L^{-1}}$). Finally, deep groundwaters, which are expected to be the potential radionuclide carrier in case of repository failure, are of Na-Cl type and much more saline (I $\sim 1.3 \times 10^{-1}\,\mathrm{mol \cdot L^{-1}}$).

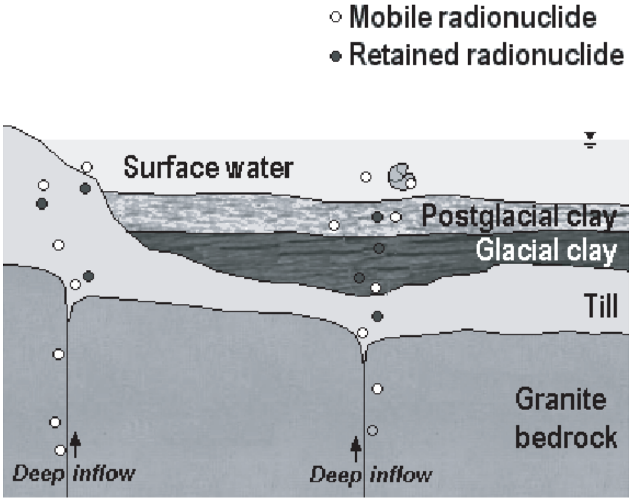

Figure 1. Section showing the interface between granite basement and surface geological systems in the Forsmark area (Sweden).

2.2 Retention processes

The analysis of the potential retention mechanisms indicates that illite charged surfaces are capable of effectively retaining via cation exchange both caesium and strontium in the till and glacial clay environments in the Forsmark area. In the case of caesium, the presence of frayed edge sites (FES) in illite is a powerful sink for this element [4, 5]. Co-precipitation of strontium with calcite is also a relevant retention mechanism in the till environment. The formation of $(Ba,Ra)SO_4$ solid solutions is known to be the solubility limiting process for radium [6]. At Forsmark, near-surface groundwaters are in equilibrium with barite so that radium co-precipitation can be favoured. For uranium, sorption on Fe(III) oxyhydroxide charged surfaces is the most efficient mechanism in relatively oxidising conditions [7], where these solids are thermodynamically stable, as those found in the till. On the other hand, the precipitation of amorphous uraninite ($UO_2 \cdot 2H_2O_{(am)}$) is considered the main process for uranium retention in the anoxic glacial clays.

2.3 Numerical implementation

2.3.1 Modelled cases

The retention capacity of the Quaternary deposits for the selected radionuclides is evaluated from 2D reactive transport simulations in two distinct domains using the PHAST code [8] The first case (Fig. 2, top) simulates a Quaternary till deposit overlying a granite bedrock. The bottom boundary of the modelled domain represents the contact between the till and the granite. Deep-seated, radionuclide-bearing fluids are assumed to migrate upwards through a fracture in the granite rock that contacts with the bottom of Quaternary sediments. The last 20 m of the top boundary represent the discharge area of the modelled domain (theoretically it could be a river, a lake or the Baltic Sea, Fig. 2). The second case (Fig. 2, bottom) simulates a clay layer at the bottom of the lake sediments, and is overlying the till deposit. The bottom boundary is assumed to be in contact with a higher permeability area of the till deposit, that allows the transfer of radionuclide-bearing fluids flowing up from the deep repository through a fracture in the underlying granite bedrock. The clay layer is assumed to have a subvertical flux that discharges into the lake.

2.3.2 Geochemical reactions

The geochemical reactions considered in the modelling exercises are (1) aqueous speciation, including complexation of calcium and uranium with organic acids in the clay environment, (2) cation exchange (Na^+, Ca^{2+}, Mg^{2+}, K^+, Sr^{2+}, NH_4^+, Cs^+) in three different sites in illite (planar, type-II and FES), (3) surface sorption/desorption of uranium and carbonate in Fe(III) oxyhydroxides, (4) equilibrium with pure minerals (Fe(III) oxyhydroxides in case 1 and pyrite in case 2) and (5) equilibrium with solid solutions ($(Ba,Ra)SO_4$, $(Ca,Sr)CO_3$)). The formation of siderite and uranium pure phases (e.g., amorphous uraninite) is also allowed.

In the till system, the redox state of the porewater is relatively oxidizing ($Eh = 0.03$ V) due to the equilibrium with Fe(III) oxyhydroxides. In contrast, redox potential in the glacial clay porewater is controlled by the equilibrium with pyrite, leading to reducing conditions ($Eh = -0.145$ V).

Initially, both systems were simulated with their natural radionuclide content to reach a hydrogeochemical stationary state prior to the inflow of radionuclides derived from the repository. This approach is done to reproduce the natural conditions that occur nowadays at Forsmark. After this stage, repository failure is simulated considering $[U] = 1.7 \times 10^{-8}$ M, $[Sr] = 8.4 \times 10^{-4}$ M, $[Cs] = 3.5 \times 10^{-7}$ M and $[Ra] = 9.2 \times 10^{-11}$ M in the deep groundwater, which interacts with the surface sediments for 2700 years.

3. RESULTS

The reactive transport calculations predict that radionuclides are effectively retained in the till system. In a conservative behaviour (not reactive), the radionuclides would reach the discharge area (see Fig. 1) in just 5 years. Considering reactive transport, Cs is readily retained via cation exchange in the illite surface, and at the end of the simulation (2700 years) the [Cs] is still almost 4 orders of magnitude lower than in the conservative case. In a similar way, at the end of the reactive simulation Ra and U concentrations are still significantly lower than in the conservative case. Ra is retained via formation of $(Ba,Ra)SO_4$ solid solution, and U is adsorbed on the Fe(III) oxyhydroxides. The release of Sr from the repository in the discharge area is retarded ~800 years due to cation exchange and formation of $(Ca,Sr)CO_3$ solid solution.

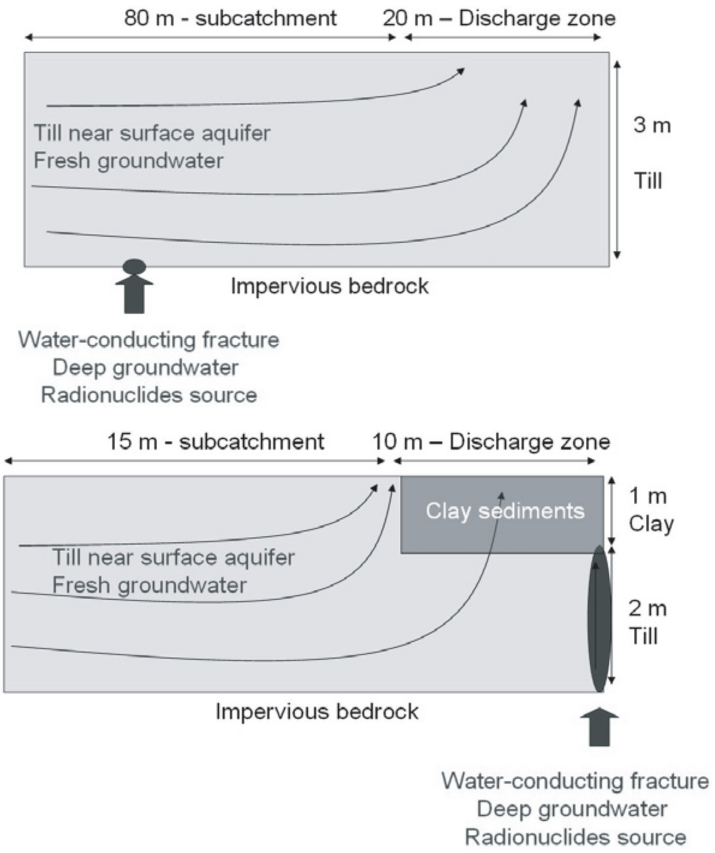

Figure 2. Conceptual model of the simulated domains: glacial till system (top), clay system (bottom).

Similar results have been obtained in the clay system for Sr, Cs and U · Sr and Cs are retained via the same geochemical processes as in the till system and U is retained via precipitation of $UO_2 \cdot 2H_2O_{(am)}$. Ra is not retained in the clay system since barite saturation is not attained along the simulation period.

For the evaluation of the retention capacity of a radionuclide at Forsmark, the parameter E has been defined as follows (1):

$$E = 100 \times \left(1 - \frac{C_R^t}{C_C^t}\right) \tag{1}$$

where C_R^t is the concentration of the radionuclide at some point of the domain and at time t considering it as a reactive solute, and C_C^t is the concentration at the same time and point considering the radionuclide as a conservative solute.

The predicted evolution of retention efficiency in the discharge area (see Fig. 2) for the radionuclides selected in this study is shown in Fig. 3. Caesium from the repository is retained very effectively in both scenarios since E is close to 100% for the whole simulation period (>3000 years). In contrast, Sr efficiency dramatically drops to 0% after 800 years in the glacial till scenario. In the clay environment, the retention efficiency also decreases but at lower rate. For uranium, the retention efficiencies are similar in both scenarios after 2700 years (∼30–40%) but the processes responsible for the retention are different. In the till porewater, uranium is mainly found as U(VI) and it is well adsorbed onto

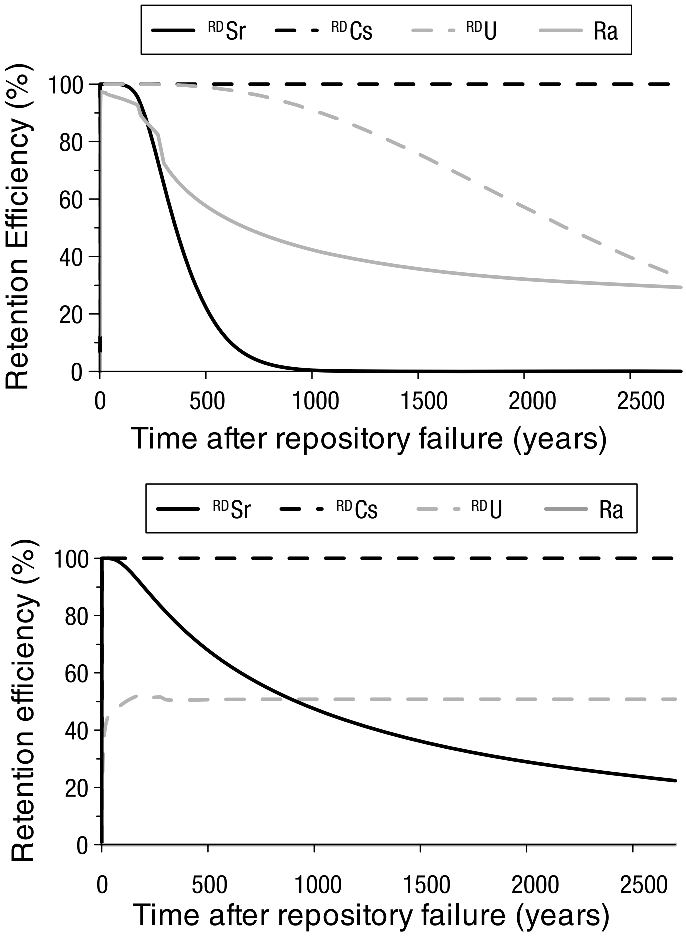

Figure 3. Predicted retention efficiency of the radionuclides derived from the deep repository through time.

Fe(III) oxyhydroxide surface although the efficiency decreases through time due to the dissolution of this mineral. On the other hand, the redox potential in the clay porewater is low enough to allow the precipitation of amorphous uraninite. Finally, E for radium is only considered in the till aquifer since the precipitation of (Ba,Ra)SO_4 solid solution does not occur in the clay environment.

Acknowledgments

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