MODELING OF Cs⁺ DIFFUSION AND RETENTION IN THE DI-A2 EXPERIMENT (MONT TERRI). UNCERTAINTIES IN SORPTION AND DIFFUSION PARAMETERS

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Indurated argillaceous formations are investigated in a number of countries (e.g. France, Belgium, Switzerland) as host rocks for radioactive waste disposal. Such formations are characterized by very low permeability and thus may retard the migration of radionuclides for very long time periods. The Mont Terri underground rock laboratory in the northwest of Switzerland offers the unique opportunity to study radionuclide migration in the Opalinus Clay.

In the DI-A2 experiment (Wersin *et al.*, 2007, 2008), several non-reactive and reactive tracers were injected as a pulse in a packed-off borehole. Unlike the previous DI-A1 test, the design of the Teflon filter in the injection borehole forced the water to flow through the filter and the open space between the filter and the borehole wall (the filter itself did not act as a diffusion barrier between the circulating solution and the rock). The decrease in tracer concentration in the liquid phase was monitored during a period of one year. Afterwards, the borehole section was overcored and the tracer profiles in the rock were analyzed. A main interest of this experiment was to understand the chemical behavior of sorbing tracers: Cs^+ (stable), $^{85}Sr^{2+}$, $^{60}Co^{2+}$ and Eu^{3+} (stable). The complete dataset (except for Eu^{3+} because of strong sorption to experimental equipment) was analyzed with a 2D diffusion-reaction model and the derived diffusion and sorption parameters were compared with laboratory data (Soler, 2007; Wersin *et al.*, 2008). As in DI-A1, a difference by a factor of about 2 was obtained for the sorption capacity of Cs^+ between in-situ and laboratory batch sorption experiments.

Recent experimental and modeling studies (Appelo *et al.*, 2009; Jakob *et al.*, 2009; Van Loon *et al.*, 2009) have shown equivalent Cs^+ sorption on intact and disaggregated Opalinus Clay samples. Cation exchange instead of a simpler Freundlich isotherm was used in the modeling. Additionally, Jakob *et al.* (2009) obtained a larger ($\times$ 4) effective diffusion coefficient for Cs^+ normal to bedding (Dell = 1.8e-10 m^2/s), compared to previous studies. In view of these developments, new modeling of Cs+ diffusion and retention in the DI-A2 experiment has been performed, including a cation exchange model to account for the retention of Cs^+.

A difference in sorption capacity by a factor of about 2 still remains depending on the use of the original model by Bradbury and Baeyens (2000) or the modified model by Van Loon *et al.* (2009). Van Loon *et al.* (2009) decreased Cs^+ sorption on the type-II sites to refine the fit of the exchange model to the experimental data in both compacted and disaggregated clay samples. A value of Dell (parallel to bedding) equal to 2e-10 m^2/s was obtained when using the modified model by Van Loon *et al.* (2009; Figure 1); Dell = 4e-10 m^2/s was obtained when using the original model by Bradbury and Baeyens (2000), which was also used by Jakob *et al.* (2009). Clearly, the values of De obtained are correlated with the strength of sorption in the model, with higher sorption leading to larger De values. This correlation may be the reason, at least in part, behind the larger value for Dell obtained by Jakob *et al.* (2009). Discrimination between the two versions of the exchange model is not possible when using only the results of the in-situ test.

Additionally, at early times (t < 10 d) the drop in Cs^+ concentration in the circulation system is slower than expected. Due to the experimental setup, this slow decrease in concentration cannot be caused by the filter in the contact between borehole and rock. Poor mixing in the circulation system could explain this effect (Figure 1).
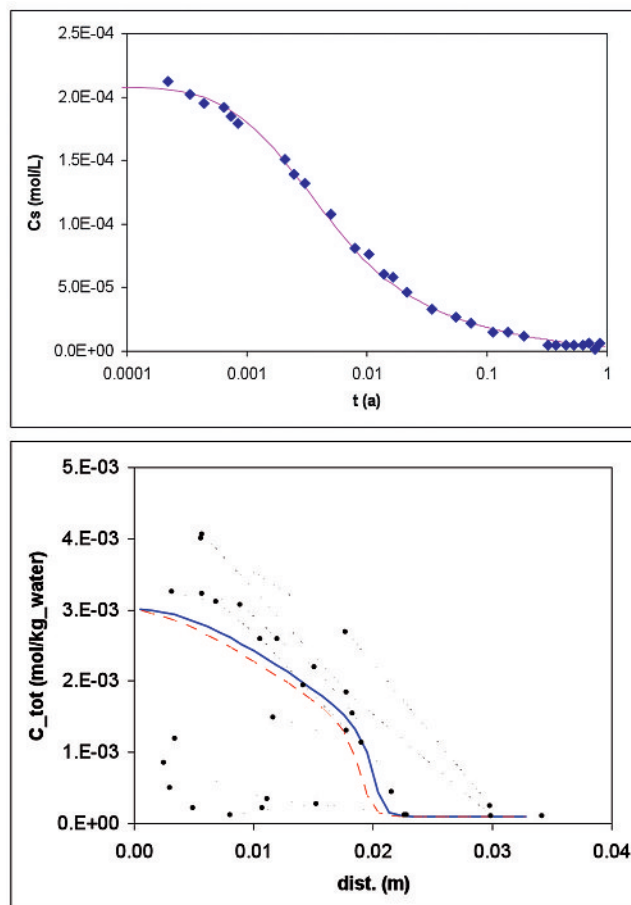
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Figure 1: (a) Cs$^+$ concentration in the circulation system vs. time for the case with $D_e = 2 \times 10^{-10}$ m^2/s and poor mixing in the circulation system. (b) Tracer distribution profiles in the rock (total [sorbed + solution] concentration vs. distance to borehole wall). Dots correspond to experimental data and thick lines correspond to modeling results. Measured profiles with small concentrations correspond to the upper and lower ends of the injection interval. The solid and dashed thick lines correspond to model profiles along the strike (solid) and dip (dashed) of the bedding.

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