DIFFUSION OF STRONGLY SORBING CATIONS (^{60}Co AND ^{152}Eu) IN COMPACTED FEBEX BENTONITE

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Compacted bentonite is used as an engineered barrier in high-level radioactive waste (HLRW) repositories because is a swelling clay of very low permeability and high sorption capability for many solutes. The transport of radionuclides through compacted bentonite is a diffusion-controlled process retarded by sorption. Performance assessment calculations of a repository need diffusion coefficients data of relevant radionuclides.

Several studies on diffusion behaviour of neutral, anionic and weakly sorbing elements on clay exist (Kozaki *et al.*, 2008; Descostes *et al.*, 2008) while very few studies are available for moderately sorbing elements (Van Loon *et al.*, 2005), and almost no studies for Eu, a highly sorbing element are reported.

In this study, diffusion experiments with strongly sorbing radionuclides, as ^{60}Co and ^{152}Eu, have been performed through compacted FEBEX bentonite. Diffusion essays with these strongly sorbing radionuclides are not straightforward to carry out because they are very time consuming essays, but also because sorption on the diffusion cells, tubing, filters and reservoirs, typically used in the classical through-diffusion or in-diffusion methods make hard the interpretation of the experimental results and the calculation of the diffusion coefficients.

FEBEX bentonite was selected as Spanish reference buffer materials, and used in many national and international projects. The clay comes from the Cortijo de Archidona deposit (Almería, Spain), and has a smectite content greater than 90% (93 ± 2%), with quartz (2 ± 1%), plagioclase (3 ± 1%), cristobalite (2 ± 1%), potassic feldspar, calcite, and trydimite as accessory minerals. The specific weight of the FEBEX bentonite is 2.7 g/cm^3.

Diffusion experiments were performed using the instantaneous plane source method. In this setup, a paper filter tagged with a tracer is introduced between two compacted tablets, avoiding contact between the tracer and the experimental vessels. The tracer can diffuse into both tablets. The apparent diffusion coefficient (D_a) can be obtained analysing the tracer concentration profile in the samples at the end of the experiment.

FEBEX bentonite was compacted, at 1.65 g/cm^3 of dry density (the density considered in the Spanish reference concept for radioactive waste repositories in granite and clay), in cylindrical stainless-steel rings of 38 mm in diameter and 25 mm of length. A filter paper with the tracer was placed between two rings with the compacted clay and the system was closed with two end-pieces and two sintered filters. The diffusion cells were introduced in a vessel to avoid humidity loss. Synthetic water simulating the pore-water composition at 1.65 g/cm^3 was used for bentonite saturation.

Three experiments with each tracer (^{60}Co $1.0 \cdot 10^{-8}$ M and ^{152}Eu $3.9 \cdot 10^{-8}$ M) were performed. The experiments lasted 843 and 852 days, for ^{60}Co and ^{152}Eu respectively, time enough to obtain an evolved profile. At the end of the experiments, the cell is disassembled, the bentonite is sliced and the tracer activity is measured directly in each slice to obtain the tracer concentration profile within the compacted sample. As both tracers used are gamma emitters, a Packard Cobra II auto-gamma counter was used to measure the samples activity.

The experimental results were modelled using the analytical solution for the instantaneous injection of a solute in a one-dimensional (1D) semi-infinite medium, equation (1):

$$C(x,t) = \frac{M}{2.A\sqrt{\pi \cdot D_a t}} \exp - \left(\frac{x^2}{4 D_a t} \right) \tag{1}$$

where C is the tracer concentration in the clay, *M* is the mass of tracer injected uniformly across the cross-sectional area *A* at point x = 0 at time t = 0, and the initial width of the tracer source is infinitesimally small, and D_a is the apparent diffusion coefficient of the tracer diffusing in the *x* direction during the time *t*.

A stochastic 1D model with GoldSim 9.60 computer code was additionally used to analyze the role of the paper filter in which the tracer was spiked in the calculations.

Figure 1 shows examples of the normalized concentration profile obtained with each tracer.

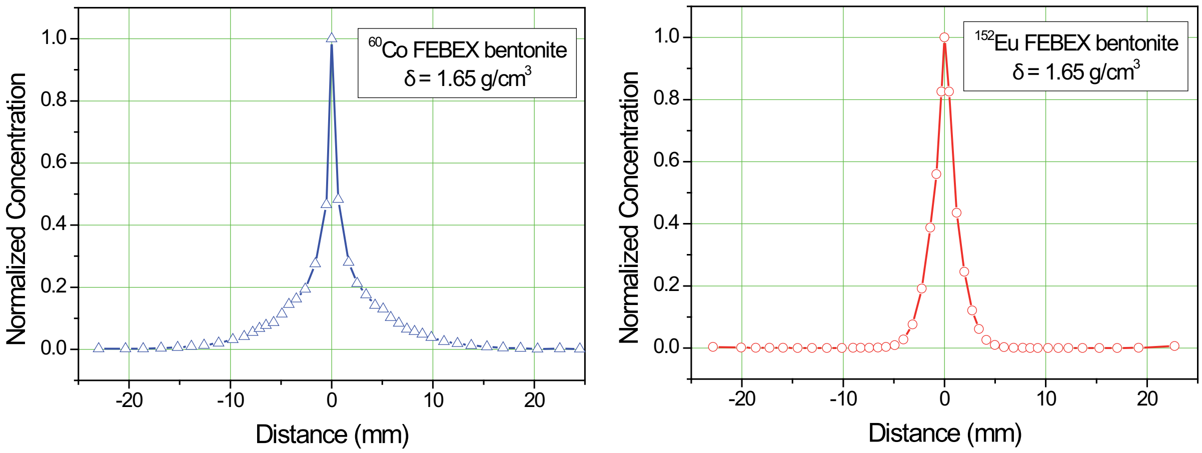

Figure 1: Examples of the experimental concentration profiles. Left: ^{60}Co concentration profile after 843 days. Right: ^{152}Eu concentration profile after 852 days.

The range of values for the apparent diffusion coefficients, Da, obtained with the analytical and numerical solution are $D_a(Co) = (1–3) \cdot 10^{-13}$ m^2/s and $D_a(Eu) = (1–2) \cdot 10^{-14}$ m^2/s. Always the first slice fit better with the lower value.

The filter paper or instantaneous plane source is therefore, a simple and useful method to determine apparent diffusion coefficients for sorbing species, such as Co, and even for strongly sorbing elements such as Eu.

References:

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