Neptunium(V) diffusion in Opalinus Clay

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Argillaceous rocks are considered in several countries, including Germany, as potential host rock for the construction of repositories for high-level radioactive waste. Opalinus Clay (OPA) is currently investigated as a candidate host rock formation for the Swiss nuclear waste repository because of the very low hydraulic conductivity (10^{-14} m/s < K_h <10^{-13} m/s) and high sorption ability of OPA. Diffusion is considered to be the dominating processes governing the transport of radionuclides in clay. To better understand the diffusion behavior of Np(V) in OPA, in-diffusion experiments with an abrasive peeling technique [1] were performed under ambient conditions using synthetic pore water (pH 7.6) as mobile phase.

The in-diffusion set-up is described in [2]. OPA (BAE-25/10, 25.4 F x 11 mm) used was from Mont Terri without special pre-treatment. Pore water was spiked with ^{237}Np(V) and $^{22}Na^+$. $^{22}Na^+$ was used to check the reliability of the in-diffusion technique. The abrasive peeling technique [1] was used to determine the concentration profile of ^{237}Np in OPA. The activities were measured directly by γ-ray spectroscopy using the γ-lines of ^{237}Np at 29.37 and 86.48 keV and of ^{22}Na at 1274.5 keV, respectively.

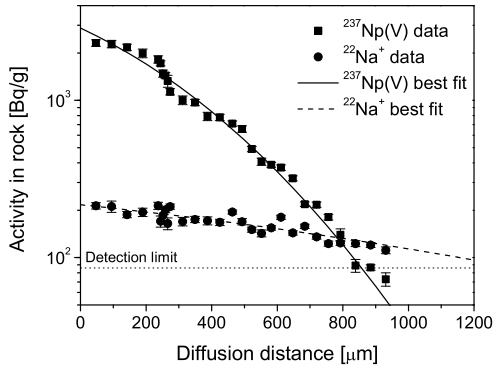

Figure 1. Concentration profile of ^{237}Np and ^{22}Na in OPA (Mont Terri) under ambient conditions. Synthetic pore water (pH 7.6) was the mobile phase.

The analysis of the experimental data (Fig. 1) assumed a single reservoir with decreasing concentrations for ^{237}Np and ^{22}Na due to in-diffusion into a semi-infinite sample [3]. An in-house computer code was used to obtain the effective diffusion coefficient D_e and the rock capacity factor α from the experimental data. The distribution ratio K_d was deduced using the relationship $\alpha = \varepsilon + \rho \cdot K_d$, where the porosity ε was measured by a through-diffusion experiment with HTO.

The quality of the diffusion parameters D_e and α was tested by using them as input parameters for the calculation of the radionuclide concentrations in the source reservoir (Fig. 2).

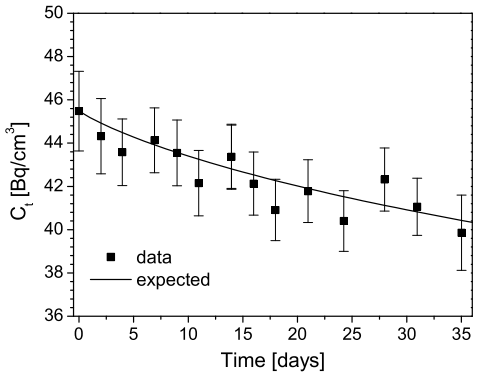

Figure 2. Concentration of ^{237}Np in the source reservoir as a function of time.

The results of the in-diffusion-experiment and the parameters used to reproduce the experimental data for ^{237}Np(V) and $^{22}Na^+$ are summarized in Table 1.

Table 1. Diffusion parameters for ^{237}Np and ^{22}Na

	^{237}Np(V)	$^{22}Na^+$
C_0 (Bq/m^3)	$(45.5 \pm 1.8) \times 10^6$	$(1.00 \pm 0.05) \times 10^9$
V (ml)	210	20
t (s)	3.1×10^6	6.1×10^4
ρ (kg/m^3)	2420 ± 8	2420 ± 8
d (cm)	2.54 ± 0.01	2.54 ± 0.01
l (cm)	1.10 ± 0.01	1.10 ± 0.01
P_b (m/s)	$(3.8 \pm 0.7) \times 10^{-8}$	-
l_b (mm)	1.57± 0.01	-
ε	0.15 ± 0.01	0.15 ± 0.01
D_e (m^2/s)	$(6.9 \pm 1.1) \times 10^{-12}$	$(1.8 \pm 0.2) \times 10^{-11}$
α	243 ± 4	0.44 ± 0.03
K_d (m^3/kg)	$(10.0 \pm 1.0) \times 10^{-2}$	$(1.4 \pm 0.1) \times 10^{-4}$

In order to study the diffusion behavior of ^{237}Np(V) in OPA under reduction conditions, future diffusion experiments will be carried out under an argon atmosphere.

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