Sorption and diffusion of Np(V) in Opalinus clay

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Sorption and diffusion are the most important processes governing the migration behavior of ^{237}Np ($t_{1/2} = 2.1 \times 10^6$ a) and other long-lived radionuclides in the near and far fields of a high-level radioactive waste repository in an argillaceous host rock. Opalinus clay (OPA) from the Underground Research Laboratory in Mont Terri, Switzerland, was selected for studying the sorption and diffusion behavior of Np(V) in a natural clay.

The sorption of Np(V) on OPA was studied as a function of pH under ambient air ($p_{CO2} = 10^{-3.5}$ atm) and anaerobic ($p_{CO2} = 10^{-2.3}$ atm) conditions. The Np concentration was in the range of 9×10^{-12} to 8×10^{-6} M. The solid-to-liquid ratio was varied between 2-20 g/L. The sorption maximum for Np was at pH 8.5, i.e., 60% was sorbed under aerobic conditions and 90% under anaerobic conditions, respectively. The reduction of Np(V) to Np(IV) was found to be the reason for the stronger sorption of Np under anaerobic conditions. At pH > 8.5, the sorption of Np decreased with increasing pH due to complexation with carbonate in aqueous solution.

The diffusion of 8×10^{-6} M Np(V) in an OPA bore core with synthetic pore water (pH = 7.6) as mobile phase was studied under ambient air conditions. The direction of diffusion was perpendicular to bedding. The diffusion properties of the intact OPA were characterized by through- and out-diffusion experiments using HTO and ^{22}Na$^+$ as tracers [1]. The diffusion of Np(V) in OPA was studied by the in-diffusion method [2] due to the strong sorption of Np(V) on OPA. The diffusion parameters for Np(V) in OPA are $D_e = (6.9 \pm 1.1) \times 10^{-12}$ m^2/s and $\alpha = 243 \pm 4$. Using the diffusion accessible porosity $\varepsilon = 0.15 \pm 0.01$ of HTO, the distribution ratio of Np(V) in intact OPA was calculated as $K_d = (0.10 \pm 0.01) \times 10^{-2}$ m^3/kg. The K_d measured under identical conditions with OPA powder suspended in pore water was equal to $(1.5 \pm 0.9) \times 10^{-2}$ m^3/kg.

[1] Van Loon, Soler & Bradbury (2003) *J. Contam. Hydrol.* **61**, 73-83. [2] Van Loon & Eikenberg (2005) *Appl. Radiat. Isotopes* **63,** 11-21.

Hf and Th isotope considerations for the origin of continental basalts

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Enriched mantle (EM1 and EM2) sources are responsible for many Cenozoic intraplate basalts associated with the intermountain region of the southwestern U.S. Melt generation may be largely or entirely within the spinel stability field [1] and tap sources enriched by admixing with sediment [2]. New Hf isotope data for young, mostly magnesian basalts from enriched mantle domains in the western Great Basin and southwestern Utah range in ^{176}Hf/^{177}Hf from 0.282505 to 0.282956 (ε_{Hf} = -9.5 to +6.5) and scatter to >10 epsilon units above the OIB array at a given ε_{Nd}. ^{230}Th excesses in these basalts (updated from [3]) are variable in size, ranging from 1.5% to 33%. Apparent Lu/Hf and Sm/Nd fractionations reveal that (1) a garnet signature is present, reflecting melting in either the garnet stability field or of a source recently infiltrated with melts generated in the presence of garnet and (2) spinel peridotite contributes more to higher-degree partial melts. The large ^{230}Th excesses show that residual garnet is present during melting and this can be reconciled with inferred depths of melting only if eclogite and/or garnet pyroxenite are present at shallow depths in the mantle lithosphere. Lithologically heterogeneous source regions may also account for the complex relationship between inferred degrees of partial melting and the magnitudes of ^{230}Th excesses.

[1] Wang *et al.* (2002) *J. Geophys. Res.* **107**, (10.1029/2001JB000209). [2] Beard & Johnson (1997) *J. Geophys. Res.* **102**, 20149-20178. [3] Reid & Ramos (1996) *Earth. Planet. Sci. Letts.* **138**, 67-81.