AAPG Annual Meeting
March 10-13, 2002
Houston, Texas

Robert J Finch[1] (1) Argonne National Laboratory, Argonne, IL

Precipitation of Neptunium Solids During the Aqueous Corrosion of Np-Doped Uranium Oxides

Because of its long half-life and potential mobility in oxidizing groundwaters, Neptunium-237 is a radioisotope that is of special concern for assessing the viability of a high-level nuclear-waste repository, such as the potential repository at Yucca Mountain, Nevada. Although crystalline Np(IV) dioxide NpO2 is believed to be the thermodynamically stable Np solid in groundwaters similar to those found in and around Yucca Mountain, NpO2 may be kinetically inhibited from precipitating from homogeneous Np(V)-containing solutions below approximately 200 C [1]. In addition, low concentrations of dissolved Np in oxidizing, uranium-saturated waters may favor the co-precipitation of Np in U(VI) (uranyl) solids, as reported recently in corrosion experiments on spent nuclear fuel [2]. Here we report recent corrosion experiments with Np-doped U oxides, in which the starting solids were exposed to water-saturated air for several weeks in sealed stainless-steel vessels. Our results indicate that uranyl oxyhydroxides incorporate small amounts of Np, and that crystalline NpO2 will precipitate at 150 C and 90 C. The coexistence of Np-bearing uranyl compounds with pure Np oxides helps to define the limit of Np solubility in the structures of uranyl compounds under the experimental conditions. Applying our results to a recently developed model for predicting dissolved Np concentrations in U-saturated groundwaters shows reasonable agreement with experimentally measured Np concentration from studies on the aqueous corrosion of spent nuclear fuels.

[1] Roberts et al. (1999). ‘Migration “99’ Abstracts volume, program #A1-02. [2] Buck et al. (1998) Materials Research Society Symposium Proceedings Vol. 506, 87-94.