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Neptunium substitution in synthetic uranophane and soddyite

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Abstract

Alteration of spent nuclear fuel in a geological repository under oxidizing conditions may result in uranyl compounds and incorporation of Np-237 into uranyl alteration phases could impact repository performance. Powders of synthetic soddyite, $(UO_2)_2(SiO_4)(H_2O)_2$, and uranophane, $Ca[(UO_2)(SiO_3OH)]_2(H_2O)_5$, were synthesized under mild hydrothermal conditions in the presence of Np^{5+}. Synthesis experiments were conducted at various temperatures and pH of the initial solutions. Powders of soddyite exhibit increasing Np concentration with the synthesis temperature at a pH of 4, consistent with substitution of Np^{5+} for U^{6+} in the structure. In contrast, the general decrease of the Np concentration in powders of uranophane with increasing synthesis temperature is inconsistent with incorporation of Np^{5+} into the structure of uranophane. These results further support the possibility that uranyl phases that form in a geological repository may impact Np mobility, but also demonstrate that additional studies of the impact of the crystal structure on incorporation are needed.

Keywords: Soddyite, uranophane, neptunium, nuclear waste, Yucca Mountain