

Neptunium incorporation in sodium-substituted metaschoepite

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ABSTRACT

Uranyl oxide hydrate minerals are common in the altered zones of U deposits and are expected to form where spent nuclear fuel is altered in an oxidizing environment. Consistent with all known uranyl oxide hydrate minerals, metaschoepite $[(\text{UO}_2)_4\text{O}(\text{OH})_6](\text{H}_2\text{O})_5$, contains sheets of uranyl polyhedra with H_2O groups located in the interlayer. Several crystals along the series from metaschoepite to Na-substituted metaschoepite (Na-MS), approximate formula $\text{Na}[(\text{UO}_2)_4\text{O}_2(\text{OH})_5](\text{H}_2\text{O})_5$, have been synthesized and their structures have been determined. Each contains sheets of uranyl pentagonal bipyramids that are topologically identical to those in metaschoepite, schoepite and fourmarierite. The sheets are electroneutral in metaschoepite, but substitution of O for OH located at the equatorial vertices of the uranyl polyhedra gives a charged sheet that is balanced by incorporation of Na in the interlayer. Synthesis of crystals of Na-MS from a solution containing ~ 500 ppm Np^{5+} , followed by analysis of the crystals using laser-ablation inductively coupled-plasma mass-spectroscopy, demonstrated that the crystals incorporate Np. This is in contrast to earlier studies that showed no incorporation of Np^{5+} in synthetic metaschoepite (which has electroneutral sheets), and supports the hypothesis that Np^{5+} incorporation is more likely in uranyl oxide hydrates with charged species in the interlayer.

Keywords: Metaschoepite, uranyl, uranium, nuclear waste, geologic repository, crystal structure, neptunium