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Pu(VI) redox behavior in water solutions

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Plutonium aqueous solution chemistry is fascinating and highly complex. This element has the potential for simultaneous existence in solution in four oxidation states under certain conditions and the chemical behaviour of each of them is different. Previously, Kvashnina et al [1] showed that the formation of PuO_2 nanoparticles from Pu(VI) solutions by adding ammonia, occurs through the formation and subsequent dissolution of the $NH_4Pu^{V}O_2CO_3$ phase. Nitsche et al [2] observed the formation of $NaPu^{V}O_2CO_3$ phases from Pu(V) and Pu(VI) solutions in a brine simulant reveal to the Waste Isolation Pilot Plan. These studies demonstrate the potential stability of Pu(V) solid phases; however, the available information is currently insufficient.

In current work, formation of Pu(V) phases from Pu(VI) aqueous solutions has been studied between pH 2 and 12 in air condition. Several solutions were prepared with an initial concentration of Pu(VI) 10^{-4}M using NaOH or KOH for pH adjustment. During the experiment, the percentage of plutonium-containing phases precipitation and the change in pH were monitored. Redox reactions were also monitored by measuring Eh, determining the oxidation states of Pu in solution by liquid extraction (HDEHP) and spectrophotometry.

After 10 – 20 days, the formation of a solid phase assumed as $MePuO_2CO_3$ (Me = Na, K) was observed in solutions with pH > 6. This solid phase was determined by X-ray diffraction analysis, X-ray absorption spectroscopy (XANES, EXAFS) and high-resolution transmission electron microscopy (HRTEM). The structure of Pu(V) carbonates was described in detail in current work. After one year of aging, the solid phase recrystallized in solution into PuO_2 nanoparticles of various sizes.

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References

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[2] H. Nitsche et al., *Radiochimica Acta* **66-67**, 3 (1994).