Plutonium Distribution among Acidic Processing Waste at the Hanford Site, USA: Implications to Long-Term Subsurface Migration

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Between 1955 and 1962, 4 million liters of plutonium (Pu) reprocessing waste from the Plutonium Finishing Plant at the Hanford Site, containing an estimated 50-140 kg Pu, were pumped into the sediments of the 216-Z-9 (Z-9) trench. The waste consisted of high salt (~ 5M NO_3^-, ~ 0.6M Al^{3+}), acidic (pH ~ 2.5) solutions, that also contained the organic solvents: CCl_4, TBP, DBBP, and lard oil. While most of the Pu was observed to precipitate within the first several centimeters,[1] a small fraction of Pu has been detected in the vadose zone at depths of 37 m.[2] The mechanisms controlling Pu migration beneath the trench are unknown. In this study, a series of binary and ternary batch experiments were used to determine Pu partitioning behavior between aqueous, organic, and solid phases representative of the waste constituents and natural sediments of the Z-9 trench. The Pu partitioning behavior observed in the binary and ternary experiments was then used to develop a conceptual model for the transport of Pu in the subsurface.

Our results show that Pu can migrate as a Pu-TBP-nitrate complex in the presence of a TBP-containing organic phase under low pH and high nitrate conditions. Reducing the nitrate concentrations or increasing the pH will lead to Pu partitioning into the aqueous phase and subsequent sorption to native Hanford sediments. Our findings suggest that Pu migration in the subsurface is likely controlled by weak sorption of aqueous Pu under acidic conditions as well as with the formation of Pu-TBP-nitrate complexes in the organic phase. Dispersion of the nitrate plume and the transient nature of the low pH conditions will limit long-term Pu mobility beneath the trench.

1. Ames, L. L. Characterization of Actinide bearing soils: Top sixty centimeters of 216-Z-9 enclosed trench; Richland, WA, **1974**.
2. Cantrell, K. J.; Riley, R. G. A review of subsurface behavior of plutonium and americium at the 200-PW-1/3/6 operable units; **2008**.