**Introduction**

The complexation of radionuclides (e.g., plutonium (Pu) and 60Co) by co-disposed ethylenediaminetetraacetate (EDTA) has enhanced their transport in sediments at DOE sites. Pu(IV)-EDTA is not stable in the presence of relatively soluble Fe(III) degrading bacterium using the periplasmic binding protein, which has to bind to the periplasmic binding protein before transport can occur. EDTA can also enhance the bioreduction of insoluble metals. There is poor solubility Pu was between 4.8 and 3.2 micromolar at day 1 and 6, respectively, indicating rapid reduction. The micromolar Pu concentration after 6 days was 0.1 micromolar. EDTA enhancement of Pu(IV) reduction after 2 days incubation. EDTA significantly increased the concentration of Pu in solution, biosorbed, biocumulated.

**Summary**

**Task 1: Pu-EDTA Aqueous Chemistry**

**Research Objectives:** Under anaerobic conditions Pu(III) is most likely the mobile species. Therefore, fundamental data for Pu(III) reactions, expected to be important in geologic environments are needed. We will determine the solubility of a sparingly soluble Pu(III) compound as a function of pH and (EDTA).

**Hypothesis:** EDTA will enhance the solubilization of Pu(III) in abiotic systems.

**Approach:** Solubility of PuPO₄ will be measured as a function of pH, phosphate, and time to develop/validate thermodynamic data for this system (Figs. 1, 11, and 12).

**Concentration of EDTA and phosphate were fixed vs. pH and time to verify/determine the equilibrium constants for aqueous Pu(III)-EDTA complexes (Fig. 3).** Pu was maintained as Pu(III) in all experiments using reducing agents which included 0.004 M reduced AQDS solutions (pH < 6.0) and Na₂HPO₄ (pH > 6.0). Solvent extraction and UV-Vis-NIR were used for oxidation state analyses. Solids were separated from solutions using membrane filters.

**Conclusion:** A preliminary model was used to interpret solubility data. The results show that EDTA forms very strong complexes with Pu(III) and that Pu(III) is reduced by the EDTA enhancement of Pu(IV) reduction.

**Task 2: Reduction of Pu by Microorganisms**

**Research Objectives:** To elucidate the mechanism and rates of Pu(IV) and Pu-EDTA reduction by metal-reducing bacteria and determine where the Pu is located (in solution, biosorbed, biocumulated).

**Hypothesis:** PuO₂(am) with and without AQDS and EDTA by dissimilatory metal reducing bacteria. Third, completion of past work on periplasmic binding protein for transport.

**Approach:** Clone, express, and purify the periplasmic binding protein for transport of PuO₂(am) as an EDTA degrading bacterium.

**Results:** Calorimetric titration of EppA revealed the formation of free EDTA to bind to EppA for transport into the cell.

**Conclusions**

1. Pu(III) solubility data was collected and used to develop a model that demonstrated that PuEDTA is the dominant species occurring within the range of environmental conditions modeled.
2. S. oneidensis MR-1 rapidly produced Pu(IV) in dilute aqueous systems. This reduction was enhanced by the addition of electron donor (H₂) and electron shuttle (AQDS).
3. Degradability of metal-EDTA is related to the formation of free EDTA to bind to EppA for transport into the cell.

**References**
