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Biodegradation of PuEDTA and Impacts on Pu Mobility

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RESULTS TO DATE: Ethylenediaminetetraacetate (EDTA) and nitrilotriacetate (NTA) are synthetic chelating agents, which can form strong water-soluble complexes with radionuclides and metals and has been used to decontaminate and process nuclear materials. Synthetic chelating agents were co-disposed with radionuclides (e.g., 60Co, Pu) and heavy metals enhancing their transport in the subsurface. An understanding of EDTA biodegradation is essential to help mitigate enhanced radionuclide transport by EDTA. The objective of this research is to develop fundamental data on factors that govern the biodegradation of radionuclide-EDTA. These factors include the dominant EDTA aqueous species, the biodegradation of various metal-EDTA complexes, the uptake of various metal-EDTA complexes into the cell, the distribution and mobility of the radionuclide during and after EDTA biodegradation, and the enzymology and genetics of EDTA biodegradation.

Research has focused on completing collaborations on modeling metal-NTA complex biodegradation, elucidating cellular transport of EDTA, and determining the kinetics and thermodynamics of Pu-EDTA complexes at environmentally relevant pHs.

A model (CCBATCH) was used to simulate the biodegradation of NTA by Chelatobacter heintzii. NTA biodegradation using C-14 was used to develop stoichiometric representations of the degradative pathway and kinetic constants necessary for model development. Modeling also showed the formation of intermediates in solution (e.g., iminodiacetate and glyoxylate), which may also impact metal mobility. Finally, modeling and experimental data determined that the rate-limiting substrate form for the biodegradation of NTA was CaNTA-. Thus the formation of CaNTA- would enhance the degradation of NTA by C. heintzii.

A putative ABC type transporter lying upstream from the EDTA monooxygenase gene has been cloned and sequenced. Research is now focused on expressing this gene in E. coli to determine if it codes for an EDTA periplasmic binding protein and a transport protein. Expression of the binding and transport proteins in E. coli would greatly simplify studies to understand the form of the EDTA transported into the EDTA degrading bacterium BNC1.

Pu(IV) rapidly formed strong complexes with EDTA, which enhanced Pu solubility by eight orders of magnitude when EDTA was present at 0.1 mM and PuO2 was present at 0.4 mM. The speciation of PuEDTA was complex with three species present as a function of pH including Pu(OH)EDTA from pH 2-5.5, Pu(OH)2EDTA from pH 6-9, and Pu(OH)3EDTA from pH 9-11. The implications are that Pu(OH)2EDTA2- will be the dominant PuEDTA species in biodegradation experiments at pH 7 with CaEDTA2-, MgEDTA2-, and HEDTA3- also present.

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