Environmental Molecular Science Institute
University of Notre Dame

Actinides and Heavy Metals in the Environment
The Formation, Stability, and Impact of Nano- and Micro-Particles

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Civil Engineering and Geological Sciences
**Background:**

In order to clean up contaminated groundwater, and to plan for effective geologic disposal of nuclear waste, we must obtain a thorough understanding of the molecular-scale processes that control movement of contaminants in the subsurface.

**Scientific Objective:**

To determine the effects of bacteria, natural organic matter and other nano- to micro-scale particles on heavy metal (e.g., Cd, Cu, Pb) and actinide (e.g., U, Np) mobilities in groundwater.
Science/Engineering Projects

Mission: Determine the effects of nano- and micro-particles on heavy metal and radionuclide transport in geologic systems.

- Bacteria
- Natural Organic Matter
- Nanoscale Mineral Aggregates

National Lab/Industry Partnerships

- Argonne (APS; Actinide Facility)
- Sandia (molecular dynamics modeling)
- Oak Ridge (geomicrobiology)
- DuPont Engineering Technologies

Education/Outreach Projects

- REU Summer Program
- High School Student Internships
- Active Recruitment of Under-represented Groups with G.E.M.
- National Lab/Industry Internships
Fulvic/Humic Acid Adsorption and Metal Complexation Reactions

Maurice et al. (2003)
Nano-scale Mineral Aggregates

‘Old’ View
Standard X-ray Diffraction

Pu$^{4+}$ aggregates
Cu-Kα

‘New’ View
Advanced Photon Source
X-ray Scattering Data

Pair distribution function
G(r)

Net Intensity (arb. units)
Q(Å$^{-1}$)

Pu$^{4+}$ aggregates

Experiment
Calculation
Bacteria-Contaminant Interactions
One pK Model
Two pK Model
Three pK Model

Net Molality
Protons Added

0.1 M NaNO\textsubscript{3} only

pH
Bacterial Cell Wall Reactions

\[ R\text{-COOH}^o = R\text{-COO}^- + H^+ \quad \text{pKa} = 4.8 \]

\[ R\text{-POH}^o = R\text{-PO}^- + H^+ \quad \text{pKa} = 6.9 \]

\[ R\text{-OH}^o = R\text{-O}^- + H^+ \quad \text{pKa} = 9.4 \]
% Cd Adsorbed Onto *Bacillus subtilis*
Advanced Photon Source
Argonne National Laboratory
X-ray Absorption Fine Structure

- Attenuation of x-rays
  \[ I_t = I_0 e^{-\mu(E) \cdot x} \]

- Absorption coefficient
  \[ \mu(E) \propto \frac{I_f}{I_0} \]
X-ray-Absorption Fine Structure

Scattered Photoelectron

Outgoing Photoelectron

$r_i$
Normalized Cd adsorption vs pH

- carboxyl
- $H_2$-phosphoryl
- phosphoryl
- Total Cd

Bacterial surface
5 µm x 1 µm, 140 m²/g cell
Molecular Modeling of Metal Binding to Cell Wall Components
Cd Adsorption onto Natural Bacterial Consortia

The graph shows the percentage of Cd adsorbed against pH. The data points and trend line indicate an increase in Cd adsorption as the pH increases from 2 to 11.
Conclusions:

- Nano- and micro-particles can control heavy metal and radionuclide mobilities in the environment.

- A range of experimental, analytical, and modeling approaches are required in order to understand the molecular-scale processes that involve these particles.