Los Alamos SFA:
Pu/Actinides in the Environment

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Pu/Actinide SFA Motives


- Pu Inventories at Several DOE Sites
  - Los Alamos
  - Hanford
  - Idaho
  - ORNL
  - Savannah River
  - Rocky Flats
  - Nevada Test Site

- What Controls Pu/Actinide Fate in the Subsurface?
  - Intrinsic colloid formation, Pu association with natural colloids
  - Biogeochemical and hydrological processes that affect Pu subsurface fate and transport (e.g., Pu-organic forms; Pu redox; colloid formation)

- How Do We Predict Pu/Actinide Subsurface Transport?
Integrated LANL Research Program

Program Scope & Direction

Colloid-Facilitated Transport

Pu/Actinide Fate & Transport in the Environment

Pu/Actinide Biogeochemistry

Site-Specific Processes and Samples

Modeling Fate and Transport

Field Experiments

Incorporate Coupled Processes for Long-Term Stewardship Decisions

Biogeochemical & Hydrological Environment

Hypothesis Testing and Modeling
Fundamental Understanding Leads to Application at Multiple Scales

- Colloid Transport
- Program Scoping
- Biogeochem.
- Site-Specific Processes and Samples
- Modeling
- Field Testing
- Applications at Multiple Scales
  - Field
  - Column
  - Pore
  - Molecular

Fundamental Process Understanding

Los Alamos National Laboratory
Earth and Environmental Sciences
SFA Drivers: Public and Programmatic

- Significant Environmental Inventory
  - LANL, INL, Hanford, NTS, and maybe 11 other sites in US
  - Larger international inventory?
  - TRU and Nuclear Repositories (YMP, WIPP)

- High Public Visibility
  - Pu contamination, perceived or real, is scrutinized, publicized, and criticized by government agencies and public interest groups (300 hits for Pu on Concerned Citizens website alone).

- Complex Behavior, not Understood Well
  - Colloid-facilitated transport: a significant factor.
  - Biological processes affect Pu redox & speciation.
  - In the Lab: multiple redox states and distinct species.
  - In environment: sorption, move as or with colloids, or soluble species.
Technical and Scientific Research Questions

- What Controls Pu/Actinide Fate and Transport?
  - Source terms and source forms.
  - Range of redox and chemical conditions in actual subsurface environments.
  - Relevant biogeochemical & hydrological mechanisms in subsurface environments?
  - Colloid and colloid-facilitated transport; soluble Pu-complexes?
  - Data needs for models?

- How are Fate and Transport Predicted in Subsurface Environments?
  - Subsurface characterization needs?
  - Modeling approaches and appropriate scales?
  - Data needs for models?
  - Calibration of model predictions/simulations?
Guiding Ideas and Hypotheses

- We can bound the range of biogeochemical hydrological conditions from site characterization data.
- Colloids and complexed Pu are key forms for transport.
- Initial waste form and subsurface biogeochemistry determine Pu species formation and stability.
- Site-specific conditions and transients are key to understanding transport.
- Redox cycling is an important process in Pu fate and transport.

*Coupling of colloid behavior, biogeochemical and hydrological processes will be integrated via modeling.*
Bound Biogeochemical & Hydrological Environments using Site Characterization

- Riley & Zachara review (1992) and new site characterization since (e.g., EM Programs at LANL; work at Rocky Flats; RIBRA at INL)

- Applications of new technology to existing subsurface data; Information from IFCs

- Source form could be Pu oxides, aqueous Pu, associated/complexed Pu or ?

- Source terms could vary from less than pCi/kg levels to $>10^6$ pCi/kg depending on site, processes, medium.

- Interaction between waste form and site-specific hydrology and biogeochemistry will be key in fate and transport.
Pu Transports as Colloids

- Significant fraction of source term that transports is in colloidal or Pu-colloidal form.
- Colloids/Pu-colloids behave as “fast-lane vehicles” for transport in subsurface environments.
- Pore-scale colloid attachment and detachment kinetics are important.
- Pu partitioning between solution, immobile matrix, and mobile colloids.
- Sufficient quantity, limited colloid filtration, colloids stable
Pu Transport as Soluble Complexes

- Biogeochemical and hydrological conditions (e.g., redox, pH) favorable for formation of soluble Pu complexes (e.g., Pu-siderophores; Pu-EDTA; Pu-carbonates)
  - Conditions that stabilize oxidized Pu(V) and Pu(VI) (both more soluble than reduced Pu(III) and Pu(IV))
  - Under reducing conditions redox cycling between Pu(III) and Pu(IV) that leads to increased solubility.

- Pu partitioning between solution, immobile matrix, and mobile colloids.
LANL SFA Proposed Timeline

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How Will We Accomplish This?

**Fundamental Understanding**
- Lab experiments
  - Experiments conducted with site-specific materials
  - Model experimental results
  - Molecular to column scale
  - Collaborations (SSRL, INL)

**Field-Derived Samples**
- Lab results & methods as guides
  - Move into larger scales (column, larger)
  - Bound experiments with biogeochemical-hydrological conditions.
  - Continue collaborations

**Field Testing**
- LANL as Collaborator with IFC(s)
  - Models of processes at different scales

Coupled Processes used for Long-Term Stewardship

FY 08

Review and Redirect

FY 12

Review and Redirect