Uranium Analysis with X-ray Microscopy

Research Team: Andrew Duffin, Jesse Ward, Gregory Eiden, Steven Smith, Bruce McNamara, Edgar Buck
Key Collaborators: David Prendergast, David Shuh (LBNL)

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965
ST3, 1.10; October 2013-September 2015

Purpose

- Chemical fingerprinting of anthropogenic and mineral uranium leading to chemical age dating of reactive uranium samples
- Develop x-ray and/or electron microscopy protocol for non-destructive uranium sample analysis

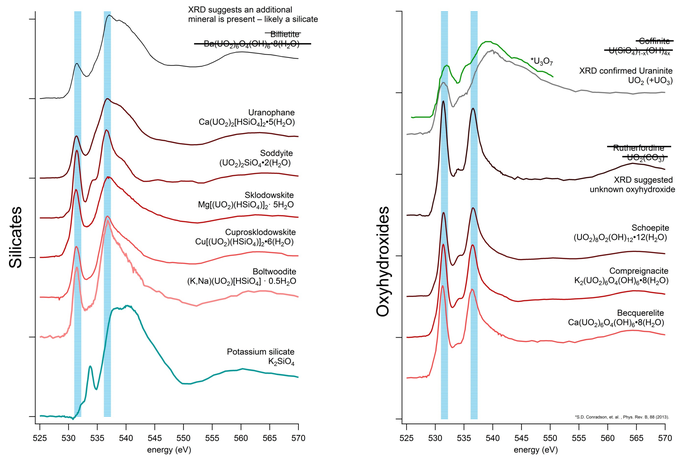
Key Successes

- Positive classification of uranyl compounds via oxygen K-edge NEXAFS
- Excellent fit of calculated spectra to experiment will lead to identification of states responsible for the spectral features
- Developed method for uranyl fluoride particle production

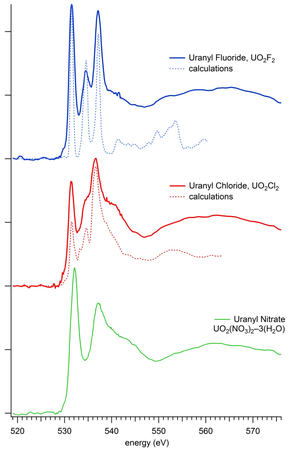
Research Accomplishments

Building Spectral Library

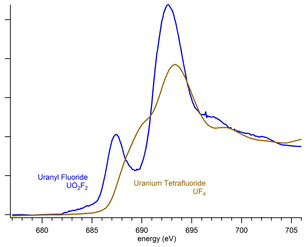
Uranyl Minerals



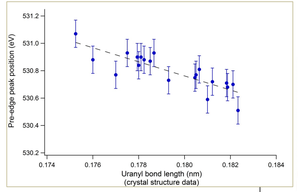
Anthropogenic Uranium



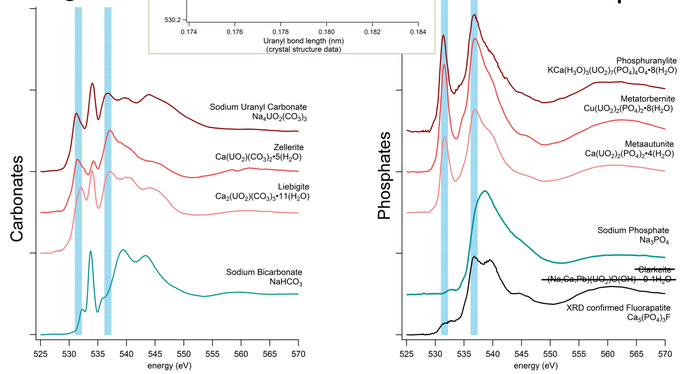
Calculated X-ray absorption spectra show excellent agreement with the experimental spectra



Pre-edge peak position correlates with bond length



Uranium minerals distinguished by oxygen K-edge spectra



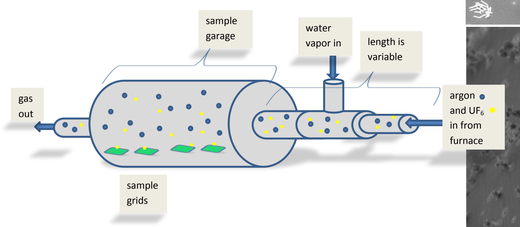
Reactive Sample Production

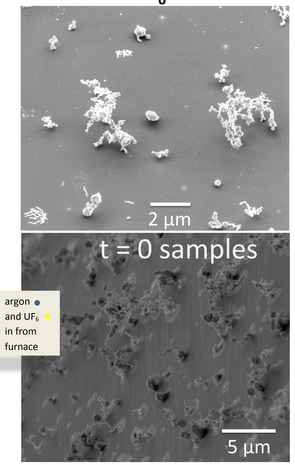
Uranyl fluoride particles are produced by the reaction of UF_6 with moist air and deposited on sample windows.

$$UO_{2(s)} + \frac{2}{3}NF_{3(g)} = UO_2F_{2(s)} + \frac{1}{3}N_{2(g)}$$

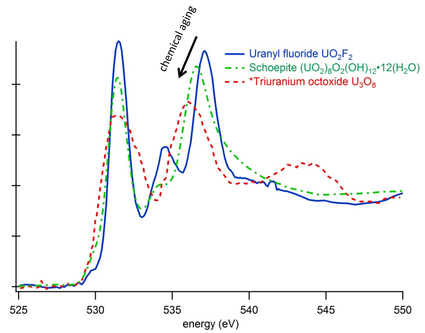
$$UO_2F_{2(s)} + \frac{4}{3}NF_{3(g)} = UF_{6(g)} + \frac{2}{3}N_{2(g)} + O_{2(g)}$$

$$UF_{6(s,g)} + 2H_2O_{(s,g)} = UO_2F_2 \cdot 2H_2O_{(s)} + 4HF_{(s,g)}$$





Next Steps



- Age-prepared samples under various humidity and temperature conditions
- Chemically imaging aged particles to reveal changes with exposure (July 2014 beam run at SSRL)
- Use calculations to model the expected spectral changes with chemical aging
- Expand on the spectral library
- Request for follow-on funding submitted to National Nuclear Security Administration

PNNL-SA-103584