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## Probing Forensic Signatures of Nuclear Materials

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Processes conducted to extract uranium out of natural ore, recover uranium from leach solutions, and purify feed material for specific end uses are chemical in nature (1). These activities provide the opportunity for chemical reagents or reaction intermediates to carry over into uranium end-products. Measurements of chemical signatures provide information that is important to forensic analyses of process materials such as uranium oxides. However, chemical speciation may transform over time as a result of exposure to environmental conditions. To better understand detectable process signatures, we have initiated an effort to examine temporal changes in morphology and chemical speciation of high-purity uranium oxide powders subjected to environmental temperatures and relative humidities.

The range of chemical compositions of uranium oxides is considerable and complicated, providing challenges to measurement and interpretation, particularly in non-crystalline materials. We have explored the use of X-ray diffraction analysis and synchrotron-based X-ray Absorption Spectroscopy to probe temporal changes in the valence states and chemical speciation of uranium oxide powders. These tools provide complementary means for characterizing subtle changes in amorphous samples. Results are compared with images collected by Scanning Electron Microscopy employed to evaluate morphology.

We have prepared a series of high-purity, well characterized uranium oxide powders (UO<sub>2</sub>, U<sub>3</sub>O<sub>8</sub>, and UO<sub>3</sub>), which have been exposed to carefully controlled simulated environmental conditions for several years. Using the analytical tools described above, we have collected an extensive set of physicochemical data that can be used as a benchmark suite for both the development of scientific insight and for comparison with “real-world” process materials.

Our ultimate goal is to exploit the rich, albeit complex, information from these complimentary diagnostic probes to support the reconstruction of a samples' process history. Success in this effort will require expertise in analytical methods, chemistry, materials science, process engineering and theory. LA-UR 13-29048

### References:

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**Primary author:** Dr WILKERSON, M. (United States of America)

**Co-authors:** Dr PUGMIRE, A. (Los Alamos National Laboratory, USA); Ms TAMASI, A. (Los Alamos National Laboratory, USA); Dr SCOTT, B. (Los Alamos National Laboratory, USA); Mr WAGNER, G. (Los Alamos National Laboratory, USA); Dr ELLIS, J. (Los Alamos National Laboratory, USA); Dr WALENSKY, J. (University of Missouri-Columbia, USA); Dr ZIMMER, M. (Los Alamos National Laboratory, USA); Dr MARTIN, R. (Los Alamos National Laboratory, USA); Dr CONRADSON, S. (Los Alamos National Laboratory, USA); Dr KOZIMOR, S. (Los Alamos National Laboratory, USA)

**Presenter:** Dr WILKERSON, M. (United States of America)

