Actinides and Rare Earths Focus Topic
Room Hall D - Session AC-TuP

Chemistry and Physics of the Actinides and Rare Earths Poster Session

AC-TuP1 Spatially Resolved Uranium Speciation in Nuclear Materials by Scanning Transmission X-ray Microscopy, ***Joseph Pacold***, Lawrence Berkeley National Laboratory; *M.J. Kristo, K.B. Knight, K.S. Holliday,* Lawrence Livermore National Laboratory; *W.W. Lukens, C.H. Booth, S.G. Minasian, T. Tyliszczak, D.K. Shuh,* Lawrence Berkeley National Laboratory

The production and manipulation of nuclear material can leave distinct physical and chemical signatures, which can later be characterized to provide evidence of the origin and process history of an unidentified specimen, a field known as 'nuclear forensics'. A broad variety of analytical chemistry techniques can provide information about interdicted and post-detonation materials. Here, we present the results of several research studies of uranium-bearing forensic specimens by soft X-ray scanning transmission X-ray microscopy (STXM). STXM yields X-ray absorption spectroscopy data with 25-nm or better spatial resolution, making it possible to quantitatively evaluate variations in oxidation state and other chemical properties across a heterogeneous specimen. Operating in the soft X-ray regime provides access to the $N_{IV,V}$ edges of the actinides and the oxygen *K* edge, which is highly sensitive to variations in U-O bonding, and consequently carries unique fingerprints of uranium oxides and their hydrates. Thus, this approach makes it possible to evaluate the oxidation state and the heterogeneity of nuclear forensic samples, yielding information on formation or process history, and/or past storage conditions. In studies of U-bearing glassy materials, the *L* edges of some transition metals (particularly iron, which influences the redox behavior of uranium) can provide additional insights. Technical developments in STXM operations relevant to forensics are also summarized. In particular, we report on improvements in sample preparation and rapid data analysis methods implemented in STXM experiments at Beamline 11.0.2 of the Advanced Light Source.

AC-TuP2 XPS Investigation on the Reduction of Aged UO_2 Powders during Exposure to Vacuum, ***Scott Donald***, *M. Davisson, Z. Dai, A.J. Nelson,* Lawrence Livermore National Laboratory

The evolution in vacuum of UO_2 powder samples, previously aged under a controlled environment composed of 20% O_2 and relative humidity varying from 34% to 98%, was studied using core level x-ray photoelectron spectroscopy. Upon exposure to ultra-high vacuum, the U 4f peak position was found to shift to a lower binding energy with time, consistent with a decrease in the overall oxidation state of the uranium. No samples were found to reduce fully to stoichiometric UO_2 under vacuum; those exhibiting the greatest decay with time asymptotically approached a mean uranium valence of +4.5, analogous to U_4O_9, within 14 days. The reaction rate was found to be variable with the initial mean uranium valence, with more oxidized samples reducing more quickly. Scanning electron microscopy images of the aged powders showed corresponding structural variation and enhanced surface cracking, both of which may serve to facilitate the reduction process.

The work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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