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Characterization of pentavalent and hexavalent americium in nitric acid using X-ray absorption fine structure spectroscopy and first-principles modeling

The minor Actinides (MAs), Neptunium (Np), Americium (Am), and Curium (Cm) accumulate in irradiated nuclear fuels. To reduce the potential long-term hazard of radioactive wastes, transmutation of MAs is considered to be an important option for the future nuclear fuel cycle. Mixed oxide containing MAs (MA-MOX) are promising candidate fuels for transmutation in fast reactors. The local and electronic structures around the Am atom in oxide fuels would yield an immense amount of information since the valence state of Am strongly affects the oxygen potentials and thermal properties of MA-MOX fuels. X-ray absorption fine structure (XAFS) spectroscopy, is an excellent method for examining the local and electronic structures surrounding an Actinide atom in oxide fuels. The redox behavior of Am in the presence of Sodium bismuthate ($NaBiO_3$) oxidizer has been previously established, however, its use as an oxidizing agent for Am in acidic solution has not been studied using XAFS. In this work, the local and electronic structures around the AmO_2^+ and AmO_2^{2+} in nitric acid solution are characterized by transmission XAFS measurement. Density functional theory (DFT) was combined with XAFS to predict the equilibrium geometries and properties of the $AmO_2^{+/2+}$ aquo complexes and their relative kinetic stability and chemical hardness.

Biography

Dr. Catherine L Riddle completed her PhD in radiochemistry from the University of Nevada, Las Vegas in 2014 and she has been a research scientist at INL for 18 years'. Catherine's research spans multiple areas and disciplines including; Actinide separations and speciation in the investigation of Actinides and Lanthanides for the expansion of new technologies for used nuclear fuel recycling and nuclear national security. Catherine holds patents in oil/heavy metal remediation, advanced neutron/antineutrino detection, and used nuclear fuel recycling processes. She is a principle investigator for multiple projects at INL and has forewarded many technologies into the commercial market.

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