HAXPES At NRR CAT

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Introduction

Advanced nuclear energy systems are envisioned to operate at high temperatures, high burn-up, and harsh neutron irradiation and corrosive environments. Advanced materials and nuclear fuels with significantly improved performance, safety and reliability are needed for the increasingly demanding operating conditions. The fundamental challenge in developing new materials with improved irradiation, temperature, and corrosion resistance is to understand and control chemical and physical phenomena in complex systems over multiple time and length scales. The sustainability of current light water reactor (LWR) systems also largely relies on the development of the scientific basis for understanding and predicting nuclear fuel and cladding performance and long-term environmental degradation behavior of reactor materials. The Nuclear and Radiological Research Collaborative Access Team (NRR CAT) is designed to make synchrotron radiation techniques available to the nuclear material scientific user community. High–Resolution Hard X–ray Photoelectron Spectroscopy will be provided at the spectroscopy line at NRR CAT.

Nuclear Materials and Photoemission

It is difficult to study radioactive materials with photoelectron spectroscopy at synchrotron radiation facilities. Because the escape depth of electrons is small, the surface of the radioactive material must be cleaned before it can be studied. Figure 1 shows a plutonium

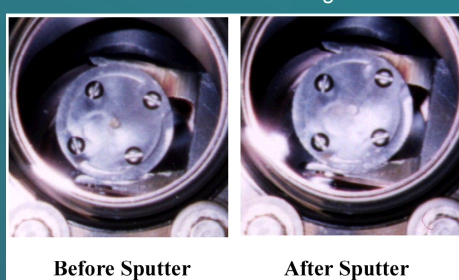

Figure 1: Sputter annealing plutonium metal removes the oxide layer that formed during shipping of the sample to the Advanced Light Source.

sample before and after cleaning by sputter annealing. The sample had been cleaned and shipped under vacuum (10^{-8} torr with battery powered ion pump) to the synchrotron. Shipping took 48 hours by dedicated truck. Cleaning the sample required the construction of a

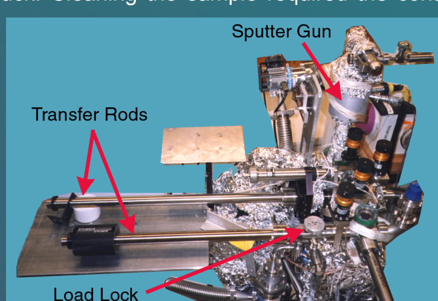

Figure 2: Sample Handling Integral Transfer system for Plutonium Intense Light Experiments housed a sputter annealing station for cleaning plutonium samples before introduction into the analysis chamber.

dedicated sputter cleaning chamber shown in Figure 2. This system was directly connected to a general user chamber. The small penetration depth of electrons prevents the sample from being encapsulated. There is always a risk of contamination whenever a photoemission measurement is conducted on a radioactive sample. The beamline will house a dedicated hard x–ray photoemission

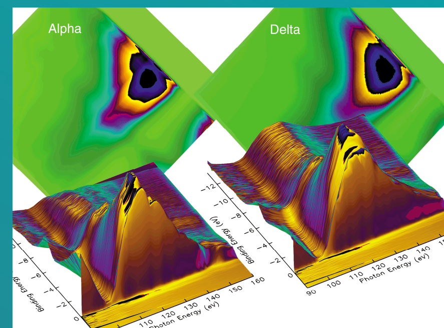

Figure 3: Resonant photoemisson through the Pu $O_{4,5}$ edge is shown. The lineshape has a Fano profile.

chamber. Contamination of the chamber will be expected but all efforts will be made to keep contamination at a minimum. Photoelectron spectroscopy can provide important information about the electronic structure of radioactive compounds. Figure 3 shows the resonant photoemission spectra from α– and δ–plutonium. The nature of the 5f electrons in plutonium is not well understood. Currently, it is

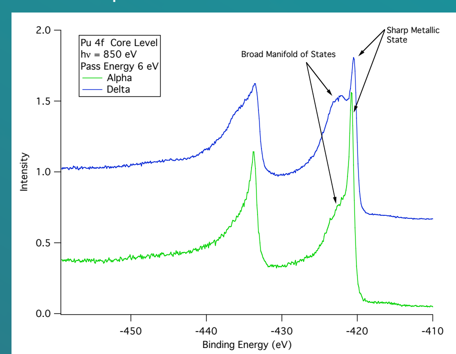

Figure 4: The Pu 4f core level spectra suggest that electrons are more localized in δ–plutonium.

believed that there is partial localization of the 5f electrons in δ–plutonium. Figure 4 shows the Pu 4f core level photoemission spectra the from α– and δ–plutonium. There is currently a debate about the mecahnism responsible for producing the broad manifold of states that is observed in the Pu 4f core level spectra. It could be a surface shift or poor screening from localized valence electrons. Hard x–ray photoemission (HAXPES) would allow measurements that minimized contribution from the surface due to the greater escape depths of higher kinetic energy electrons. HAXPES could help to determine the origin of this shifted component.

NRR CAT And HAXPES

The primary objective of the NRR CAT is to allow users to conduct a range of synchrotron x-ray experiments on highly-activated nuclear fuels and materials. This requires that the users be protected against radiation exposure and contamination. The proposed beamline will be designed to be a radiological facility per the DOE Standard. A specimen preparation hutch and glove boxes will provide the capabilities for specimen handling, preparation and processing, and short-term storage of specimens as needed. Inside the HEPA filtered hutch will be the HAXPES chamber. All routine handling will be performed remotely from outside of the controlled area.

References

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