# Investigation of the Chemical State of Ag and Pd in SiC Shell of Irradiated TRISO Particles via XAFS

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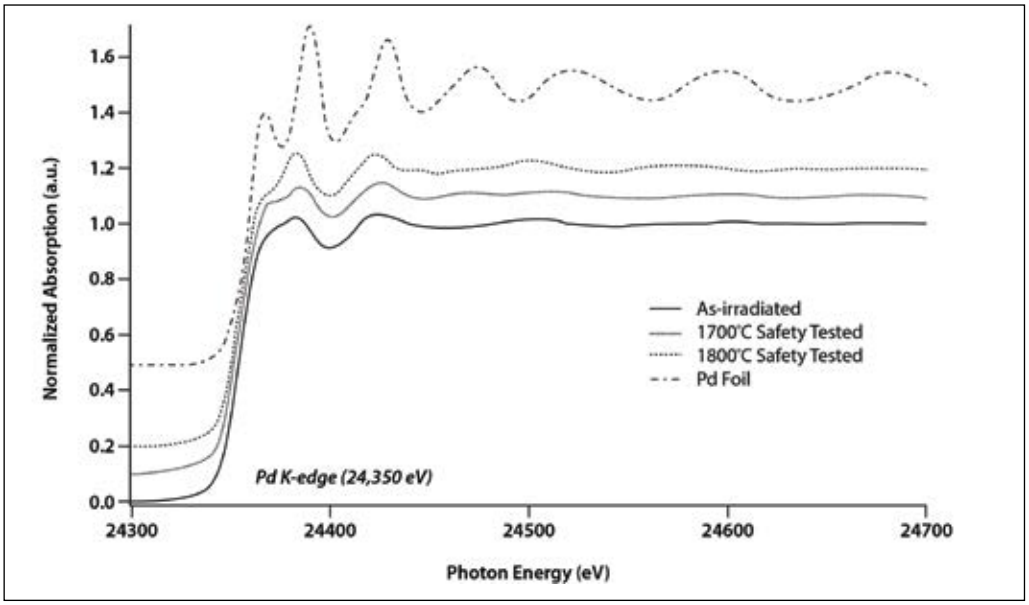


*Figure. 1. Palladium XAFS in SiC shells from the various particles compared with metallic Pd foil that clearly shows a different character.*

***This experiment for the first time provides insights into specific mechanisms of radionuclide transport in irradiated TRISO particles, providing the basis for fundamental understating that in turn allows informed enhancements to the fuel design.***

Tristructural isotropic (TRISO) fuel particles consist of a fuel kernel contained within a multiple ceramic spherical coating shells. The purpose of these ceramic coating layers, namely graphite and silicon carbide (SiC), is to limit the release of radionuclide fission products from the fuel kernel. However, a number of these fission products, particularly precious metals such as Ag, do transport within these layers under reactor normal operating and off-normal conditions. This study examines the mechanism of transport of these species in the SiC coating layer of TRISO particles.

## Project Description

TRISO fuel form has enjoyed decades of development and testing and is currently considered a mature technology. This fuel form, originally developed for high temperature gas-cooled reactor (HTGR) applications, is now being considered for a number of other reactor platforms. To further enhance the safety of these reactors, the mechanism of transport and release of a few select problematic radionuclides, namely Ag and Pd, from these particles needs to be understood. A multitude of computational studies exist that have examined the various possibilities for the transport of these species

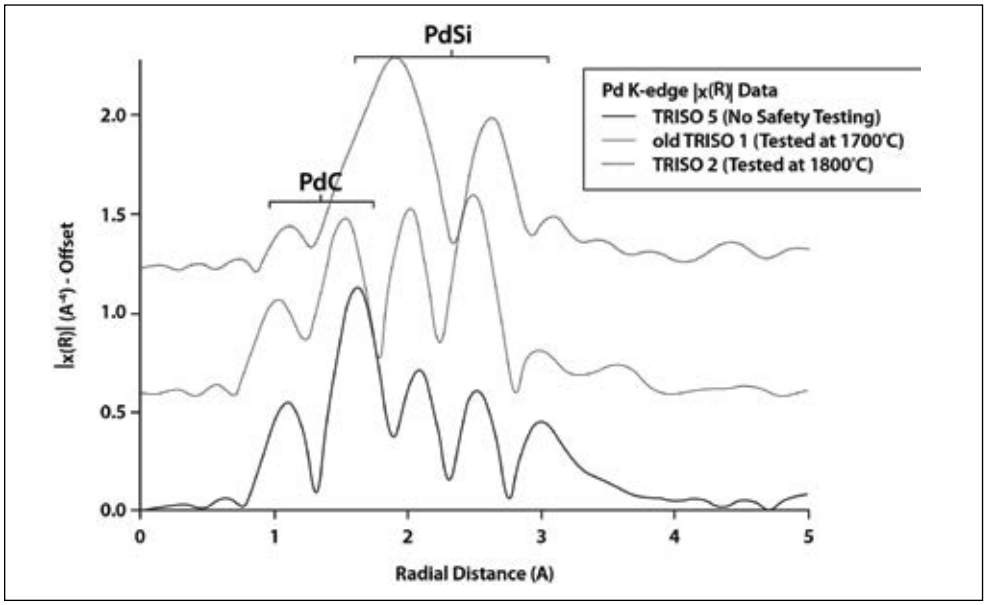


*Figure 2. Radial Distribution Function (RDF) around Pd in the various SiC shells showing different chemical forms of Pd.*

inside the SiC coating layer of TRISO particles. These computational studies are complemented by only a handful of experimental studies, most of which have poorly simulated the prototypical fuel operating environment. Therefore, the computational studies merely serve to contemplate various scenarios without any constraints and experimental data to narrow down any particular mechanism. In this study, care was taken to extract the SiC coating layer from actual TRISO fuel particles irradiated inside the reactor to high burnups. These SiC fragments were then examined via X-ray absorption fine structure (XAFS) technique, using a synchrotron, to specifically and accurately probe particular chemical species of interest. The results of these studies can pinpoint the chemical state of the species of interest after various fuel operating and testing scenarios. In this manner, given the experimental data, specific mechanisms of transport and release may be reinforced or dismissed. These studies will provide the mechanistic understating of radionuclide transport and release in the coated particle fuel forms. Accordingly, this will allow fuel scientists to enhance safety in the reactors employing this fuel form.

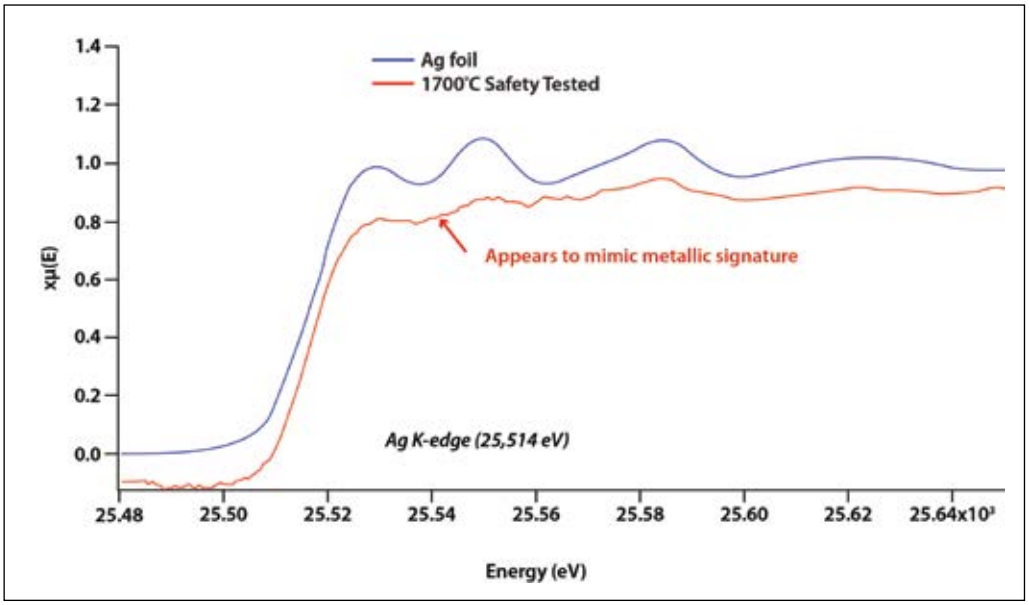


*Figure. 3. Silver XAFS in 1700°C safety-tested SiC shell compared to a metallic Ag foil. The data show a metallic character for the Ag in the SiC shell.*

## Accomplishments

SiC fragments from three specific TRISO fuel particles were selected for synchrotron XAFS examination. One of the particles was in the as-irradiated state, while the other two had been safety tested (after irradiation, held at elevated temperature isothermal conditions to simulated off-normal conditions) at 1700 and 1800°C. The particles had undergone gamma assay to determine the inventory of Ag still retained in the particles, the amount of which varied widely between these three particles. The particles were crushed, burned to remove all the graphite layers, and leached with acid to remove all the fuel. This process was repeated twice to then end up with pristine SiC coating shells. These shells were then extracted from Oak Ridge National Laboratory's (ORNL's) fuel hot-cell and shipped to Materials Research Collaboratory Access Team (MRCAT) for synchrotron characterization. The basic fluorescence spectra showed the presence of Pd, U, and Pu in these shells for all the particles. However, Ag, in reasonable detectable amounts, was only present in the SiC shell from the 1700°C safety-tested particle. XAFS studies were carried out on

all of these shells and the dominant chemical state of Pd and U for all the shells from the various particles was determined. Long collection times were dedicated to collection of XAFS data for Ag in the 1700°C safety-tested SiC shell. The data closely resemble that of metallic Ag, though a metallic alloy cannot be ruled out at this time. Longer collection times are needed to probe the XAFS regime to determine the bond length of the Ag near neighbors to answer this question. However, this is a major finding that implies Ag does not transport inside the SiC as a single atomic solute. Instead, it is in metallic clusters. The graduate student collaborator in this project, Rachel Seibert (IIT), meticulously and patiently collected and analyzed the XAFS data.

### Future Activities

Future activities involve electron microscope characterization of these SiC shells. The shells have already been returned to ORNL and the microscopy characterization is currently underway in FY 2016.

*The ability, facilitated via NSUF, for preparation of samples at hot cell facilities and subsequent examination at synchrotron resources provides unprecedented experimental data.*

**— Kurt Terrani, Staff Scientist - ORNL**

**Distributed Partnership at a Glance**

| NSUF and Partners | Facilities and Capabilities |
|---|---|
| Illinois Institute of Technology | Materials Research Collaborative Access Team (MRCAT) facility at Argonne National Laboratory's Advanced Photon Source |
| **Collaborators** | |
| Illinois Institute of Technology | Jeff Terry (collaborator), Rachel Seibert (collaborator) |
| Oak Ridge National Laboratory | Kurt Terrani (principal investigator) |