

## Task 17

# Interaction between Metal Fission Products and TRISO Coating Materials

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## BACKGROUND

This project focuses on the chemical bonding and interface formation of metal fission products with the coating materials used in tri-isotropic (TRISO) fuel particles for gas-cooled reactors. By combining surface- and bulk-sensitive spectroscopic methods, intermediate chemical phases at the interface, intermixing/diffusion behavior, and the electronic interface structure as a function of material (metal and coating materials) and temperature are examined.

In detail, the project studies the interface formation of Pd, Cs, and Ag with SiC and pyrolytic carbon. Using SiC single crystals as substrates, interfaces are prepared under controlled conditions in an ultra-high vacuum environment and are studied with a combination of experimental methods, including Photoelectron Spectroscopy, Auger Electron Spectroscopy, Inverse Photoemission, X-Ray Emission Spectroscopy, and X-Ray Absorption Spectroscopy. Furthermore, microscopic techniques (Transmission Electron Microscopy, Scanning Tunneling Microscopy, Atomic Force Microscopy) and local scanning tunneling spectroscopy are being employed.

## RESEARCH OBJECTIVES AND METHODS

By combining results for the electronic and chemical structure of the fission product/TRISO layer interfaces, the research results give information about failure mechanisms of TRISO particles and fission product transport.

Furthermore, they can be used to derive strategies to tailor the interface properties for an optimization of TRISO particles in terms of, e.g., chemical and long-term stability.

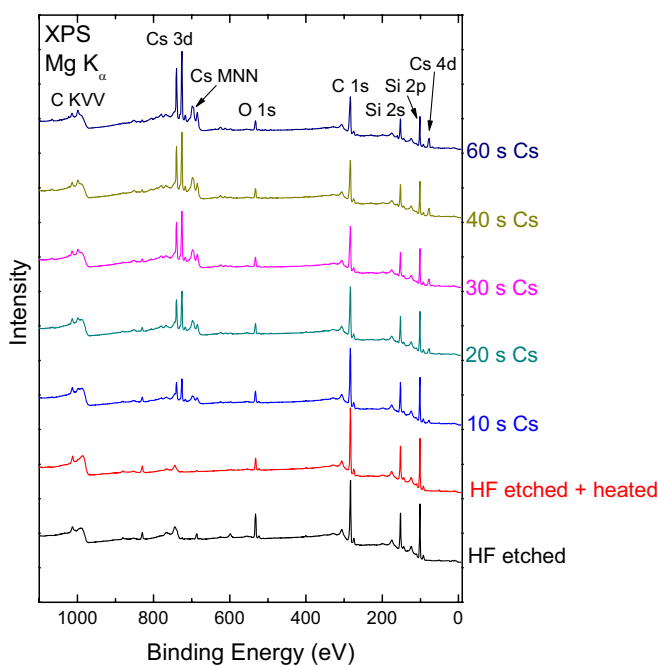
The experiments use two different experimental set-ups. First, a multi-chamber ultra-high vacuum system at UNLV (see photo in the 2005-2006 Annual Report) houses the majority of surface-sensitive methods. Second, the soft X-ray fluorescence (SXF) endstation at Beamline 8.0 at the Advanced Light Source, Lawrence Berkeley National Lab is used to perform X-ray absorption and emission spectroscopy. The apparatus at UNLV consists of a surface spectroscopy chamber, a preparation chamber, and an ultra-high vacuum scanning probe microscope. Samples can be introduced into the machine through a glove box with integrated load-lock and then transferred to each of the chambers via a distribution chamber. The analysis chamber is equipped with a state-of-the-art electron analyzer, an X-ray source, an ultraviolet (UV) source, and an inverse photoemission setup consisting of a low-energy high-flux electron gun and a UV detector. The preparation chamber is used for cleaning samples with an ion source and for the deposition of metal films with an evaporator.

## RESEARCH ACCOMPLISHMENTS

In the past year, emphasis was placed on a detailed analysis and description of the Cs/SiC interface formation process.

Great care was taken to prepare a clean SiC surface. It was found that the purchased SiC single crystals exhibit strong surface contaminations with oxygen. Based on an in-depth literature research, two different cleaning routes were investigated. The first route is an HF etch of the SiC surface, which is reported to result in a clean, C-terminated SiC surface. The second route is a sputter-cleaning of the surface with Ar<sup>+</sup> ions. It was found that it was not possible to entirely remove the O with either of the techniques, showing that the crystals were significantly oxidized into the bulk. As a consequence of this investigation, a new vendor was identified and new SiC crystals were ordered.

For the deposition of Cs films on the SiC substrate, a Cs evaporator was designed and constructed. Cs films of different thickness were deposited on SiC substrates (exhibiting some degree of surface oxidation, as discussed above). Exemplarily, the survey spectra of films with different Cs thicknesses are shown (left). These spectra illustrate the stepwise increase in Cs intensity after subse-



XPS survey spectra of an HF-etched SiC single crystal surface, after heat treatment at approximately 400°C, and after various deposition steps of Cs (given is the deposition time in seconds).

quent deposition steps, indicating an increase in the Cs film thickness. In parallel, the Si and C intensities decrease due to attenuation by the Cs film. A careful quantitative analysis shows that this attenuation is weaker than expected, suggesting either an inhomogeneous growth of the Cs film or an intermixing at the interface.

To gain further insight into this question, and to learn more about the detailed chemical environment at the Cs/SiC interface, detailed core level X-ray photoelectron spectroscopy (XPS) spectra were recorded. As an example, a series of C 1s core levels as a function of sample preparation and Cs deposition time (below right). As is evident in the figure, a second carbon species at higher binding energies emerges, which is directly indicative of the chemical interaction between Cs and SiC. Interestingly, no changes are observed in the Si core levels, suggesting that the chemical attack of SiC by Cs is initiated at the C sites.

The X-ray photoemission experiments were supplemented by UV photoemission data (not shown), which gives detailed information about the valence electrons. The students in the project have learned about the chemical and electronic information that can be exerted from such spectra, and a detailed analysis of the Cs/SiC dataset is currently ongoing. In the previously investigated case of Pd/SiC, similar data could be used to draw the picture of the formation of a Schottky barrier at the Pd/SiC interface.

Finally, selected Cs/SiC interfaces were also investigated at the Advanced Light Source using X-ray emission spectroscopy of the Si L<sub>2,3</sub> and the C K edge, giving additional insight into the intermixing behavior at the interface between Cs and SiC.

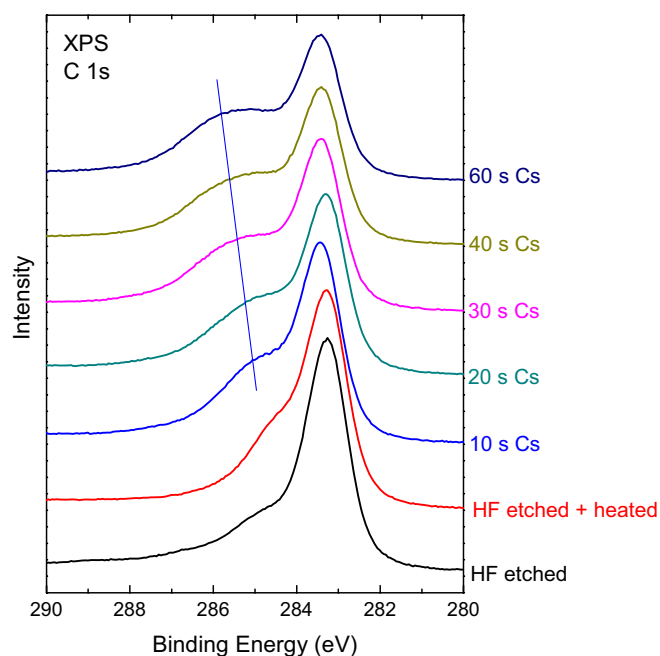
## FUTURE WORK

The experiments on Cs/SiC will be continued including the deposition on differently prepared substrates. In addition, the interaction between Cs and pyrolytic graphite surfaces will be studied. The focus will be on single crystalline surfaces and also on rough surfaces, which will be prepared by Ar<sup>+</sup> ion bombardment. This will allow an investigation of the influence of substrate roughness on the chemical Cs-SiC and Cs-C interactions.

Furthermore, the substrate temperature will be varied during deposition, up to approximately 1200 C. Both experiments will help to correlate the results to the real case of a TRISO particle with microscopically rough interfaces involving the SiC diffusion barrier and the pyrocarbon layers. In addition, the experiments will simulate the “natural” high temperature environment present in the nuclear reactor.

## ACADEMIC YEAR HIGHLIGHTS

- ◆ Participation of all graduate and undergraduate students in experimental campaigns at the Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA (October 2006 and March 2007).
- ◆ C. Heske, “How to reveal the chemical and electronic properties of interfaces, buried layers, and liquids with soft X-ray spectroscopy,” Physics Department, Boston University, December 8, 2006.
- ◆ C. Heske, “The surface and interface analysis ‘tool chest’ at UNLV,” Group seminar Prof. T. Moustakas, Boston University, November 17, 2006.
- ◆ C. Heske, “How to reveal the chemical and electronic properties of interfaces, buried layers, and liquids with soft X-ray spectroscopy,” Materials Science and Engineering Department, Stanford University, August 31, 2006.
- ◆ C. Heske, “Surface and Interface Analysis in Las Vegas,” Seminar Experimentelle Physik II, University of Würzburg, Germany, July 20, 2006.



XPS detail spectra of the C 1s core level, indicating the presence of a second, reacted C species after deposition of the Cs films (indicated by the blue slanted line).

### Research Staff

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