

# Task 17

## Interaction between Metal Fission Products and TRISO Coating Materials

C. Heske

### BACKGROUND

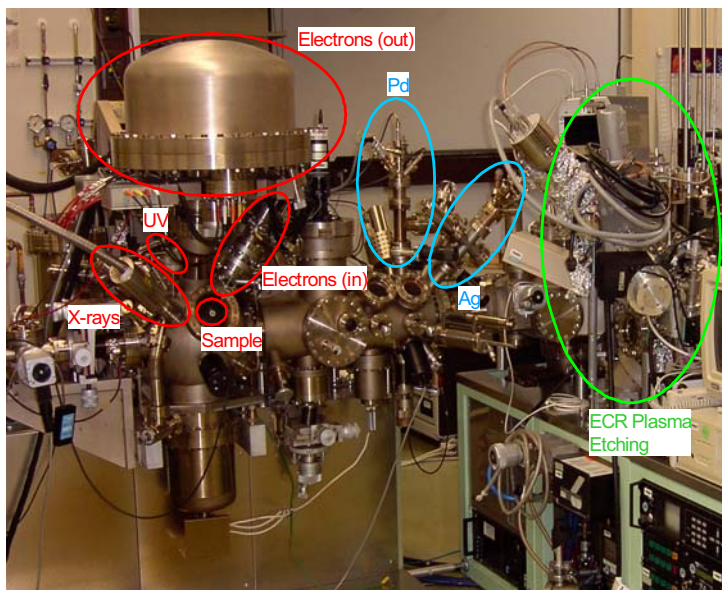
The goal of this project is to elucidate the chemical bonding and interface formation of metal fission products with the coating materials used in state-of-the-art TRISO fuel particles. Particular emphasis is placed on an analysis of intermediate chemical phases at the interface, the intermixing/diffusion behavior, and the electronic interface structure as a function of material choice (metal and coating materials), temperature, and external stress. Furthermore, the chemical state of some of the metal fission products will be assessed.

This project studies the interface formation of Pd, Ag, and Cs with SiC and pyrolytic carbon. Using the TRISO coating materials (and single crystal references) as substrates, interfaces will be prepared under controlled conditions in an ultra-high vacuum environment and will be studied with a variety of different spectroscopic and (when applicable) structural methods. In addition, realistic microstructures will be studied.

### RESEARCH OBJECTIVES AND METHODS

The research objectives of this project are as follows:

- To give valuable information about failure mechanisms of TRISO particles and fission product transport.
- To give, through simulating experiments, indications for optimized irradiation testing and post-irradiation examinations within the AFCI effort at ORNL.
- To derive strategies to tailor the interface properties for an optimization of TRISO particles in terms of, e.g., chemical and long-term stability.



Picture of the four-chamber ultra-high vacuum apparatus. Red: surface spectroscopy; blue: metal deposition, green: surface modification.

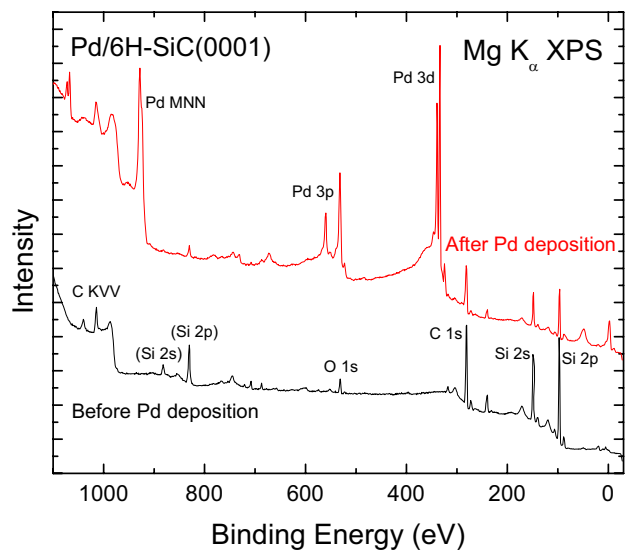
The combination of surface sensitive techniques (e.g., photoelectron spectroscopy) with bulk sensitive methods (e.g., X-ray emission spectroscopy) will probe the chemical properties as well as the diffusion behavior in several complementary ways. In addition, spatially resolving methods will be employed to characterize cross sectional profiles. A variety of surface modification methods will be applied ex-situ (e.g., for stressed coating layers) or in-situ (e.g., by ion bombardment) prior to or after the interface formation to study the dependence of the interface properties on the surface/interface morphology and quality of the coating material.

### RESEARCH ACCOMPLISHMENTS

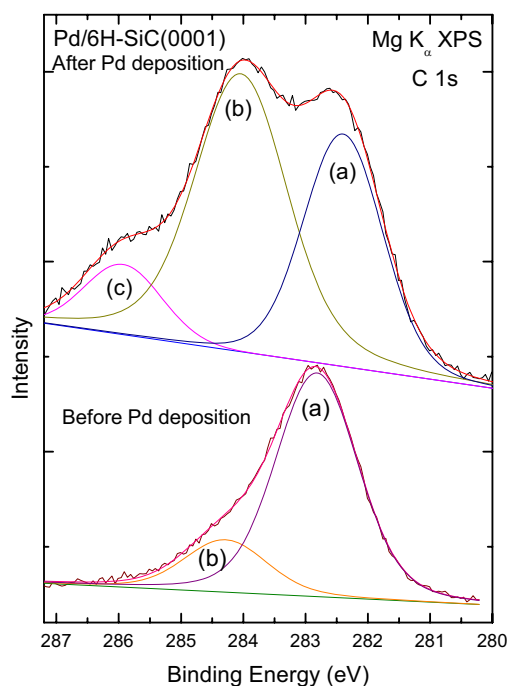
The project utilizes a four-chamber ultra-high vacuum surface science instrument (shown in the figure below, left), which combines several experimental techniques, including X-ray and UV photoelectron spectroscopy (XPS and UPS), X-ray and small-spot-electron-excited Auger spectroscopy (XAES and AES), low-energy electron diffraction (LEED), and in-situ ellipsometry. It allows the modification of surfaces by ion bombardment, plasma etching, annealing, and metal evaporation under strictly controlled conditions. The instrument was shipped from the University of Würzburg, Germany, in September of 2004 and arrived on campus in November 2004. After successful reassembly, all components are fully operational, and first calibration and surface science experiments are currently under way. Before dismantling the system for shipment at the University of Würzburg, first investigations of the Pd/SiC interface formation were conducted in Würzburg during the summer of 2004.

A second line of experiments is being conducted at the Advanced Light Source, Lawrence Berkeley National Laboratory (Beamline 8.0, SXF endstation). The first experimental campaign, in which the Pd/SiC interface was studied by soft X-ray emission spectroscopy and X-ray absorption spectroscopy, was conducted at the end of November 2004.

Several experimental runs were conducted to optimize the surface cleaning procedures for commercial SiC(0001) single crystal substrates. The figure on the next page (top, left) shows an XPS survey spectrum taken after a series of ion-sputter/annealing cycles to remove surface contaminants from the surface polishing process and the air exposure (in particular Ca, O, and water). As can be seen, only a small residual signal of oxygen is present, while all other prominent peaks are associated with the SiC bulk material. This surface was used to collect first data on the room temperature Pd/SiC interface formation by depositing a thin (less than 1 nm) layer of Pd (top spectrum in the figure). This leads to the observation of Pd-related lines and an attenuation of the SiC peaks. The Pd/SiC interface, a detailed analysis of the individual XPS peaks has been conducted. As an example, the figure (bottom, left) shows a close-up of the Carbon 1s peak.



*X-ray photoelectron survey spectrum of a SiC(0001) single crystal before (bottom, black) and after (top, red) Pd deposition.*



*X-ray photoelectron spectra of the C 1s peaks before (bottom) and after (top) Pd deposition.*

## ACADEMIC YEAR HIGHLIGHTS

- ◆ Research on interactions between metal fission products and TRISO coating materials,” by C. Heske, G. Gajjala, and V. Marepally, was presented at the semi-annual meeting of the AFCE, Salt Lake City, UT, September 2004.
- ◆ “Interaction between metal fission products and TRISO coating materials (SiC),” by G. Gajjala, V. Marepally, T. Hofmann, J. White, C. Heske, L. Weinhardt, S. Hansen, O. Fuchs, and E. Umbach, was presented at the 49<sup>th</sup> Annual Meeting of the Arizona-Nevada Academy of Sciences, UNLV, April 9, 2005.
- ◆ “Interaction between metal fission products and TRISO coating materials (SiC),” by G. Gajjala, V. Marepally, T. Hofmann, J. White, C. Heske, L. Weinhardt, S. Hansen, O. Fuchs, and E. Umbach, was presented at the ANS Student Conference, Ohio State University, April 16, 2005.

Again, the bottom spectrum pertains to the clean SiC surface, while the upper spectrum was derived from the carbon atoms at and near the Pd/SiC interface. As indicated by the detailed peak-fitting analysis, it is evident that a bulk (a) and a surface (b) carbon component exist at the SiC surface. Upon deposition of Pd, a third species (c) is observed, which is tentatively interpreted as an interdiffusion-related carbon species. Furthermore, a strong interface species (b), a weakened bulk species (a), and a general upward shift of all electronic levels is observed. In an initial-state picture, the fact that the new species appear at higher binding energies indicates a charge transfer away from the carbon atoms. This finding is corroborated by the Si L<sub>2,3</sub> X-ray emission spectra (not shown) in which increased spectral weight was observed above the valence band maximum, i.e., near the Fermi energy, and interpret this as a Pd-induced transfer of partial charge to the Si atoms.

## FUTURE WORK

Several parallel experimental paths will be followed in the upcoming project year. First, the study of the Pd/SiC interface will be continued to reproduce initial results (in particular the finding of two interface-related C species) and to investigate their dependence on external parameters, such as deposition temperature, Pd flux and thickness, and, in particular, substrate morphology. In the latter case, SiC films from the TRISO coating process will be investigated, as well as utilize suitable surface modification techniques (plasma etching, ion bombardment, etc.) to modify the interface properties.

### Research Staff

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### Students

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