Interaction between metal fission products and TRISO coating materials: A study of chemical bonding and interdiffusion: 1st quarter report, 2005

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Task 17: Interaction between metal fission products and TRISO coating materials: A study of chemical bonding and interdiffusion
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In this project we utilize a combination of state-of-the-art soft X-ray spectroscopies to understand the chemical bonding between metal fission products (Pd and Ag) with coating layers in TRISO fuel particles (SiC and pyrocarbon). We are primarily focusing 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. In the current first project year, we are beginning these investigations with the Pd/SiC interface, as discussed in the previous two quarterly reports. Our first experiments (both using our lab instrument as well as using high-brilliance synchrotron radiation at the Advanced Light Source (ALS) in Berkeley) indicate that the interface formation leads to three different carbon species, which are ascribed to a bulk species, an interface species, and “reacted” species due to intermixing at the interface. Furthermore, we find a significant charge transfer between Pd and Si, as evidenced by the synchrotron X-ray emission spectra presented in the previous report. These results are being presented by graduate student Goverdhan Gajjala at the 49th Annual Meeting of the Arizona-Nevada Academy of Sciences (UNLV, April 9) as well as at the American Nuclear Society Student Conference (Ohio State University, April 16).

In the last quarter, our efforts were primarily focused on completing the reassembly of our four-chamber ultra-high vacuum surface science instrument on the UNLV campus after its relocation from the University of Würzburg, Germany, at the end of 2004. Since the vacuum system as a whole and, in particular, most of its spectroscopy components are extremely sensitive instruments, the system was completely dismantled for the relocation, and hence significant work had to be done in the reassembly process. Fortunately, the damage incurred in transit was minimal – all electronic components work well and a ground-connection problem within the electron analyzer (previously encountered at the

Fig. 1: Graduate student Timo Hofmann operating the sample transfer in the four-chamber ultra-high vacuum system
University of Würzburg) could be resolved by our careful reassembly. Large effort had to be devoted to convert the gas handling systems (ultra-high purity (6N) Argon and Helium) to American standards in order to find reliable suppliers for the required high-purity gases. While Argon is necessary for sputter-cleaning surfaces in-situ, He is required for operating the UV-lamp necessary for UV photoelectron spectroscopy (UPS).

As shown in Fig. 1, the instrument is now completely reassembled and in use on the UNLV campus. Currently, we are working on a detailed and thorough optimization of the electron spectroscopy set-up (which includes the electron analyzer, the dual-anode Al/Mg X-ray source, a small-spot electron gun for local (200 nm) Auger electron spectroscopy, and the UV gas discharge source). These tasks comprise a careful alignment of all components as well as a detail electronic calibration. The latter task is currently being performed and entails the collection of reference spectra of an Ar-ion sputter-cleaned Au-foil reference sample for a variety of spectrometer settings (in order to optimize resolution vs. signal-to-noise ratio, several parameters can be tuned, including the opening angle of the spectrometer, the electron pass energy, and the slit shapes and widths within the spectrometer). As an example for the calibration spectra currently being collected, Fig. 2 shows UV- and X-ray excited photoelectron spectra of the Au foil reference sample.

![Hel UPS and Mg Kα XPS spectra](image)

**Fig. 2:** Uncalibrated UPS spectrum (left) and XPS survey spectrum (right) of a Au-foil reference sample currently being used to calibrate the electron spectrometer for all relevant experimental settings.

While these experiments are of enormous importance for the reliability and spectral quality obtained with our system in the future, they also present a unique opportunity for the graduate and undergraduate students to learn the operation (e.g., sample transfer) and scientific principles of our rather complicated instrument.

In parallel, all students are involved in the further analysis of our two previous experimental runs at the University of Würzburg and the Advanced Light Source. This analysis is of large importance, not only to fully understand the current data set in order to gain more insight into the Pd/SiC interface, but also to optimally plan the upcoming experimental runs, both at UNLV (after completion of the calibration experiments) as well as at the ALS (May 2005). While the November 2004 and May 2005 ALS campaigns were/are conducted in experimental time granted to the PI within other projects, we recently sub-
mitted a separate beamtime proposal for the fission product/TRISO coating project and were awarded eight experimental shifts on a highly-competitive undulator beamline (8.0) in the Fall of 2005.

Once the calibration experiments are complete, the next steps with our project will be to reproduce the previous findings discussed in the previous reports and the introduction above, to utilize realistic SiC-layers from TRISO coaters, and to investigate the possibilities of modifying the interface properties by varying the parameters and conditions under which the Pd/SiC interface is formed in our system.