Task 17

Interaction between Metal Fission Products and TRISO Coating Materials

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BACKGROUND

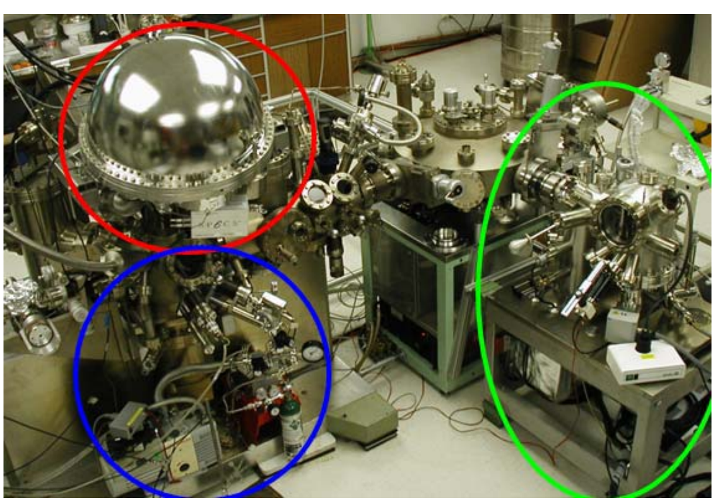
In this project the chemical bonding and interface formation of metal fission products with the coating materials used in TRISO fuel particles is investigated. A combination of surface- and bulk-sensitive spectroscopic methods are used to analyze intermediate chemical phases at the interface, intermixing/diffusion behavior, and the electronic interface structure as a function of material (metal and coating materials) and temperature.

The interface formation of Pd, Cs, and Ag with SiC and pyrolytic carbon is studied in detail. Using the SiC single crystals and TRISO coating materials as substrates, interfaces are prepared under controlled conditions in an ultra-high vacuum environment and are studied with a photoelectron spectroscopy, Auger electron spectroscopy, Inverse Photoemission, X-ray emission spectroscopy, and X-ray absorption spectroscopy. Recent additions to the experimental approach include microscopic techniques (Transmission Electron Microscopy, Scanning Tunneling Microscopy, Atomic Force Microscopy) and local scanning tunneling spectroscopy.

RESEARCH OBJECTIVES AND METHODS

The research results of this project are expected to:

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



Bird's eye view of the modified multi-chamber ultra-high vacuum apparatus. Red: replacement electron analyzer; blue: inverse photoemission setup, green: scanning probe microscope.

The experiments use two different experimental apparatuses, namely a multi-chamber ultra-high vacuum system at UNLV, and the SXF endstation at Beamline 8.0 at the Advanced Light Source Lawrence Berkeley National Lab. In the past project year, significant improvements of the UNLV system could be implemented. Several upgrades could be installed, in particular a new state-of-the-art electron analyzer, a partially custom-built inverse photoemission (IPES) set-up, and, very recently, a scanning probe microscope (all instruments are funded through other projects). In particular the replacement of the electron analyzer now allows us to take reliable electron spectra at a significantly increased count rate, while avoiding the saturation effects, software communication problems, and background issues of the old analyzer. The achievable signal-to-noise ratio is now improved by several orders of magnitude, while also improving the spectral resolution greatly. The IPES set-up now allows investigation into the conduction band, and hence gives valuable information about the interface formation from the viewpoint of the unoccupied electronic states. The new scanning probe microscope will, once fully commissioned, give insights into the sample morphology and chemical and electronic structure on a *local* (i.e., for single crystal systems, *atomic*) scale.

RESEARCH ACCOMPLISHMENTS

In the second year of the project, focus was placed on a detailed analysis and description of the Pd/SiC interface formation process. This description is documented in the Masters thesis of Goverdhan Gajjala, which was successfully defended at the UNLV Department of Electrical Engineering. As an example, a series of X-ray photoelectron spectroscopy (XPS) survey spectra, that were taken for a SiC single crystal substrate after introduction into the system, after sputter-cleaning with Ar^{+} ions (2 keV), and after several Pd deposition steps are illustrated. In this particular case, the Pd deposition was performed at approx. 800 degrees C to simulate elevated temperatures in TRISO fuel. Furthermore, the SiC surface was very strongly sputtered, which induces a significant number of structural defects at the surface and is hence intended to simulate real SiC surfaces. An example of a detailed UV photoelectron spectroscopy (UPS) study of the corresponding samples is also provided, indicating how the electronic surface structure is converted from an adsorbate-induced character to a semiconductor (SiC) to a metallic thin film (for increasing Pd overlayer thickness). A detailed analysis of the valence band maxima and Fermi edges, together with a study of the work function (which can be derived from the secondary electron cutoff in the UPS spectra) gives detailed insight into the electronic structure of the Pd/SiC interface, which can be interpreted in view of pronounced intermixing effects, as well as the formation of an electronic Schottky barrier.

In the thesis of Goverdhan Gajjala, the results obtained from the sample series are compared with a similar series that was obtained at room temperature. Furthermore, the first experimental series, which was taken at the University of Wuerzburg (before relocation of the instrument to UNLV), can be well compared. In all cases, the presence of two additional carbon species at the interface was found, one associated with a short-range charge transfer (the actual chemical bonding) and a longer-range interdiffusion species (in addition to the expected SiC bulk species). Combining all spectral regions (in XPS and UPS), a detailed picture of the Pd/SiC interface formation can be painted, in particular when taking additional soft X-ray emission and absorption data into account, which was taken in the experimental campaigns at the Advanced Light Source.

FUTURE WORK

The Pd/SiC results are currently being prepared for publication in a peer-reviewed journal. For this purpose, some additional data is currently collected in order to allow a reliable quantitative analysis of the bonding and interdiffusion processes. This is necessary since the old electron analyzer showed several age-related electronic problems.

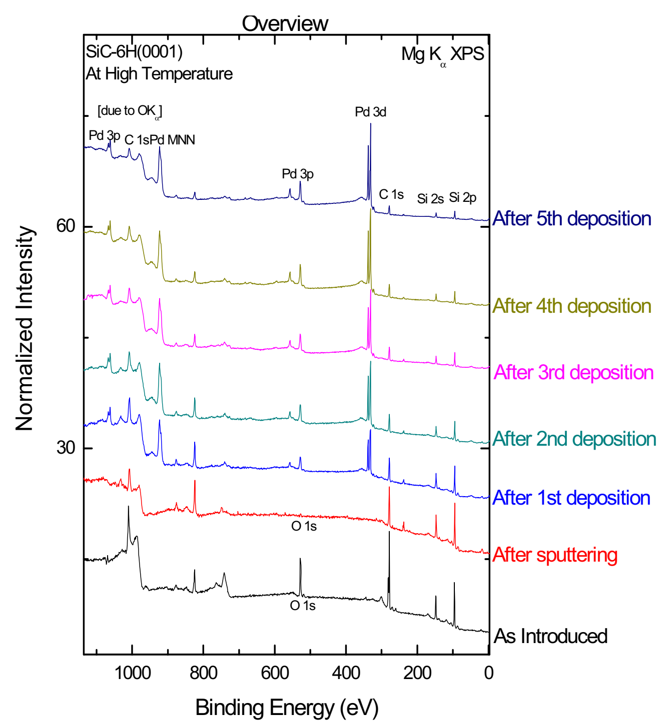
As mentioned above, the electron analyzer has been replaced by a modern state-of-the-art instrument (SPECS PHOIBOS 150MCD) that not only allows a much more reliable quantitative analysis, but also a significantly improved energy resolution, shorter accumulation time, and drastically enhanced signal-to-noise ratio. These experiments will be very important to reproduce and calibrate some of the experiments presented in G. Gajjala's thesis before they are published in a peer-reviewed journal.

In addition to completing the existing Pd/SiC data set, the investigation of the Cs/SiC interface is being prepared – a Cs evaporator has been designed and is currently being constructed by Sharath Sudarshanam. Again, these studies will be performed as a function of external parameters such as deposition/diffusion temperature, surface/interface modification, SiC preparation method, etc.

Further studies will focus on other material combinations, in particular Ag and SiC, as well as various metals (Pd, Ag, Cs, …) and their interaction with the pyrocarbon layers in TRISO fuel. By combining these studies, a deeper knowledge and a more comprehensive picture of the chemical properties of interfaces in TRISO fuels and the interaction of metallic fission products with TRISO coating layers are expected.

ACADEMIC YEAR HIGHLIGHTS

- G. Gajjala, "Interaction Between Pd and SiC: A Study for TRISO Nuclear Fuel," Masters Thesis, Department of Electrical & Computer Engineering, UNLV, April 2006 (submission), May 2006 (defense).
- C. Heske, "Soft X-ray spectroscopy of compound semiconductors: how to reveal the chemical and electronic properties of buried interfaces," Tutorial at the Florida American Vacuum Society Meeting, Orlando, FL, March 2006.
- C. Heske, "X-ray spectroscopy of buried things: interfaces, liquids, and dirty powders", Stanford Synchrotron Radiation Lab Workshop on Soft X-ray Science, Stanford, CA, February 2006.
- C. Heske, "Soft X-ray spectroscopy of compound semiconductors: how to reveal the chemical and electronic properties of interfaces and buried layers", Pacifichem 2005, Honolulu, HI, December 2005 (featured in an article in Chemical & Engineering News 84, 35 (2006)).



XPS survey spectra of an as-introduced SiC-6H(0001) single crystal surface, after 2 keV Ar^+ ion sputtering, and after several Pd deposition steps at approx. 800°C.

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