Growth and Interaction of Pd on TRISO fuel layer ZrC at elevated temperture.

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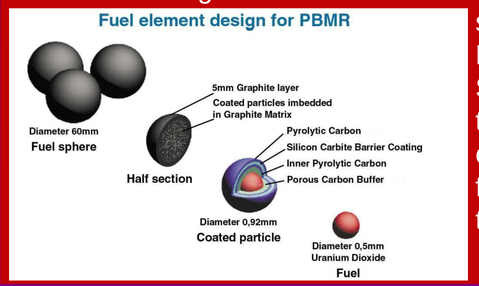
Objective

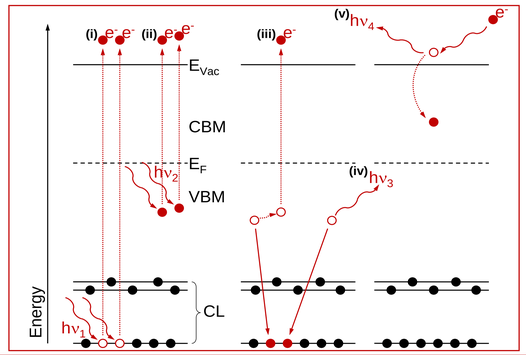
Next generation nuclear fuels will consist of a layered sphere that contains a fissiable material at its core surrounded by coatings serving various purposes. During the fission process nearly every element is produced to some degree. The diffusion barrier is designed to contain these products. In pervious designs SiC was used as a diffusion barrier. During reactor test Cs was found to escape the fuel. ZrC was purposed to replace SiC due to its strength and stability at high temperatures. Palladium is of great interest due to its catalytic properties. This study will use X-ray Photoelectron Spectroscopy to monitor the chemical environment of Pd and ZrC as a function of Pd growth and temperature (32-1200˚C).



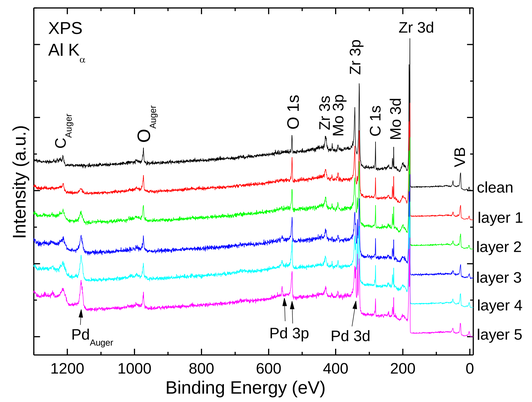
Spectroscopic Techniques

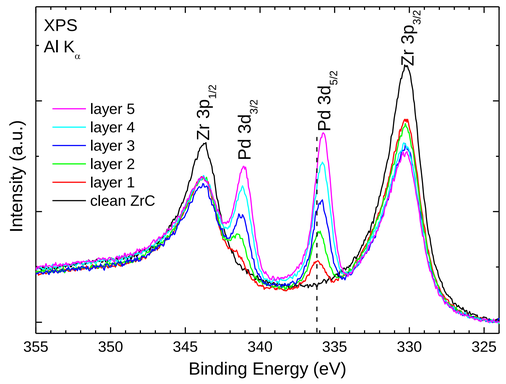
(i) XPS (X-ray Photoelectron Spectroscopy)
(ii) UPS (UV Photoelectron Spectroscopy)
(iii) XAES (X-ray Excited Auger Electron Spectroscopy)
(iv) XES* (X-ray Emission Spectroscopy)
(v) IPES (Inverse Photoemission Spectroscopy)
(vi) XAS* (X-ray Absorption Spectroscopy)

* performed at the Advanced Light Source, Lawrence Berkeley National Laboratory



The growth pattern of Pd on ZrC

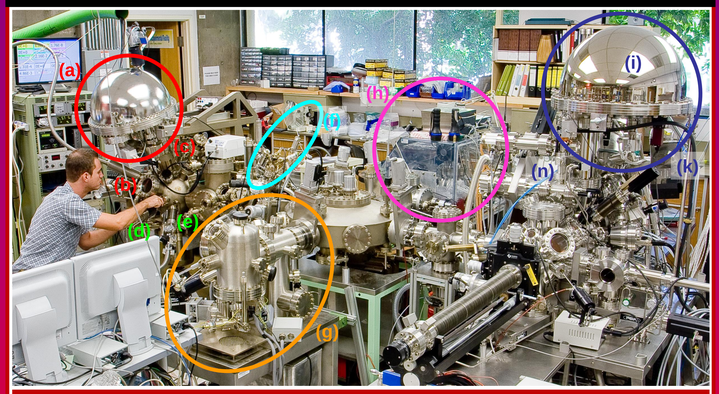




- Mo peaks are contributions from the sample holder.
- O is present on the surface even after cleaning.
- As layer thickness increases there is an increase in intensity of the Pd peaks.

- Pd intensity increases with thinker layers.
- Zr intensity decreases with thickness of Pd layers.
- Pd peaks shift to higher binding energies with increased thickness.

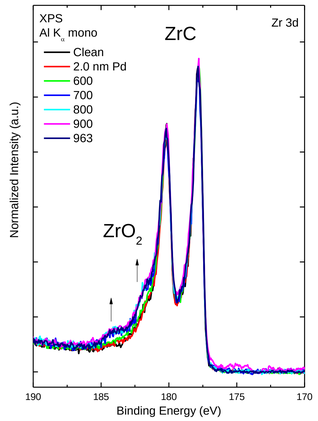
Equipment

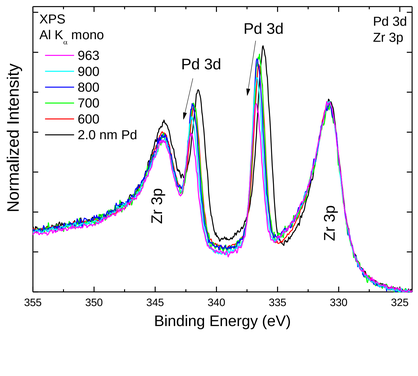


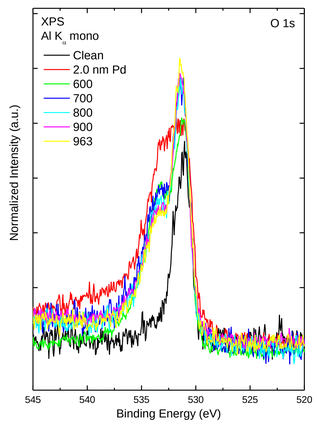
(a) High dynamic range Electron Analyzer
⇒ **XPS, XAES, UPS**
(b) X-ray Source ⇒ **XPS, XAES**
(c) UV-Source ⇒ **UPS**
(d) e^--Gun ⇒ **IPES**
(e) IPES detector ⇒ **IPES**
(f) Ar^+-Ion Gun ⇒ **sample cleaning**
(g) Scanning Probe Microscope ⇒ **AFM, STM, STS, KPFM**
(h) Glove Box ⇒ clean sample entry
(i) High resolution Electron Analyzer ⇒ **XPS, XAES, UPS**
(k) Monochromatized X-ray Source ⇒ **XPS, XAES**
(n) Monochromatized UV-Source ⇒ **UPS**

Chemical environment of Pd/ZrC at elevated temperature

After the deposition of Pd thin film on ZrC the sample was heated in 100˚C steps from 600˚C to 963˚C. Between heating steps the sample was allowed to cool to ≈200˚C for measurement.







- Growth of ZrO2 peaks with an increase in temperature
- Intensity of the ZrC peaks remains constraint with increasing temperature

- Pd peaks decrease in intensity with respect to the Zr 3d peaks
- Pd shifts to higher binding energies with increases in temperatures

- At least two components of oxygen on the surface
- The higher binding energies species appears after the growth of Pd film and decreases with temperature
- The lower binding energy species grows with every measurement

Results/Future Work

The Pd peak shifts to higher binding energies during growth on ZrC. A shift to higher binding energies is then present when heated. There is also a decrease in intensity of the Pd peaks when heated. The shift and decrease in intensity can be explained by diffusion into the bulk which could destabilized the ZrC layer of the TRISO fuel particle. The changes could also be explained by the evaporation of Pd off the ZrC surface.

Further investigation of the Pd chemical environment is necessary. Imaging of the ZrC surface is planned to further explain the growth pattern of Pd. Imaging will also be used to aid in the understanding of the Pd chemical shift during growth and heating. If Pd is defusing into the bulk of ZrC holes will be seen in the microscopy images.

Acknowledgement

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OAK RIDGE National Laboratory