Identification of Pd-Si Compounds in Diffusion Couple Studies to Complement AGR PIE reports

Jordan Stone, Steven Cavazos, Katherine Montoya, Thomas Kirtley, Gisele George, Elizabeth Sooby

University of Texas at San Antonio, San Antonio, TX 78249 USA

ABSTRACT

Palladium (Pd) and silver (Ag) are known fission products of TRISO particle nuclear fuels [1]. Pd is known to corrode the silicon carbide (SiC) layer of a TRISO particle, which can compromise the overall fuel performance [2,3]. This study sets out to identify Pd-Si formations in SiC in order to better understand the corrosion of the SiC layer, and therefore the diffusion of Ag out of the TRISO layers found in AGR PIE reports. As of present, Pd has been observed to degrade SiC at 1000 °C; the extent of Pd diffusion and Pd-SiC compounds have yet to be determined.

BACKGROUND

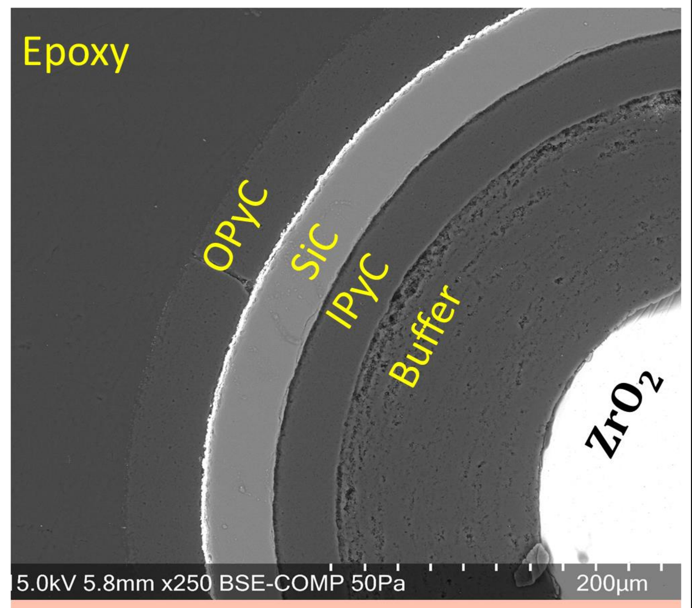

Figure 1: An Electron image of Pd coated TRISO particle post 1150 °C anneal with layers labeled taken with SEM.

A TRISO particle is a nuclear fuel form that utilizes alternating layers of Pyloric Carbon (PyC) and SiC in order to provide structural support and regulate diffusion of materials out of the nuclear fuel kernel (UCO, UO_2). In this experiment, the nuclear fuel kernel has been replaced with a surrogate ZrO_2.

TRISO fuel is used in High Temperature Gas Reactors (HTGRs), and significant fuel performance studies of TRISO fuels have been conducted through the Advanced Gas Reactor (AGR) program. Post irradiation exams (PIEs) have reported high fractional release rates of the ^{110m}Ag isotope between the temperatures of 850 °C – 1150 °C, as seen in Figure 2 [1].

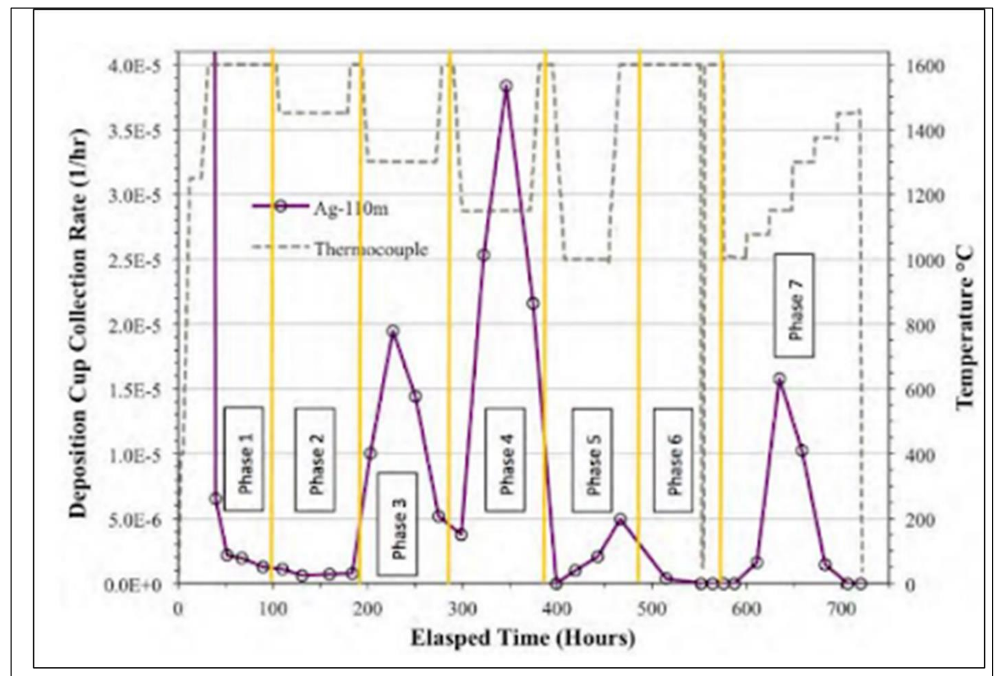

Figure 2: The Deposition Cup Collection rate of 110 Ag as a function of temperature and time [1].

The ^{110m}Ag isotope is a high gamma emitter. This can cause hazardous conditions for reactor workers as they may come in to contact with this fission product. This study focuses on the transport methods that allow for Ag to diffuse through the SiC layer of TRISO. Pd is also a fission product. Previous studies at UTSA conducted by graduate student Steven Cavazos - showcased Palladium's ability to corrode the SiC layer of TRISO particle at 1150C, as seen in Figure 3. Pd corrosion could aid Ag diffusion. By evaluating the formation of Pd-Si that form in the SiC layer during annealing, we can better understand Pd's corrosive interactions. Therefore, furthering our understanding of Ag thermal transport.

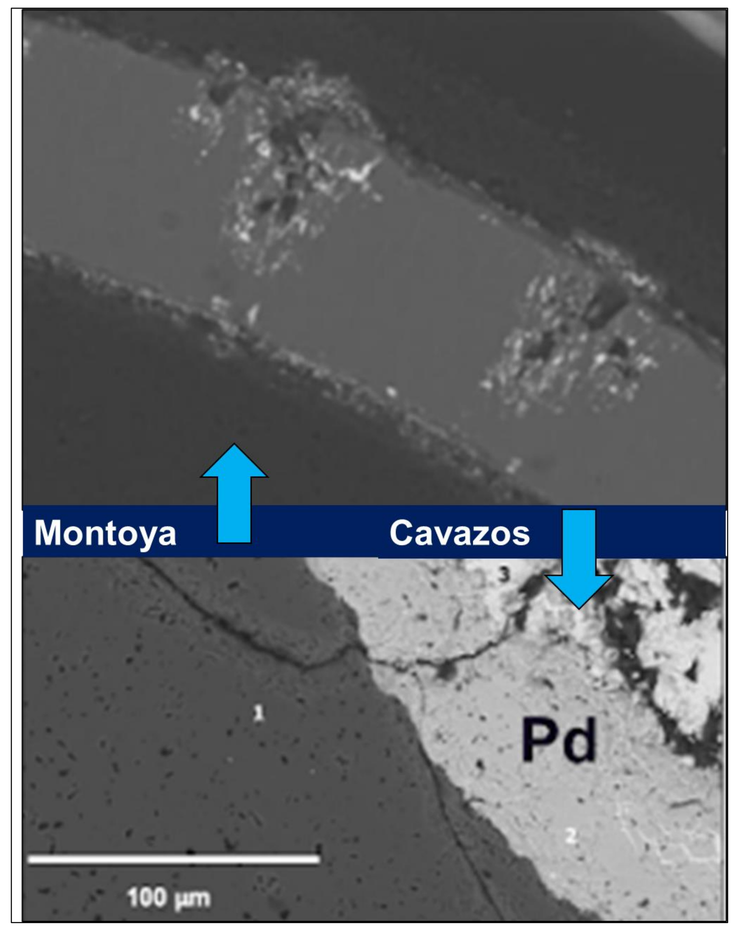

Figure 3: Electron image taken of corrosion of SiC in TRISO caused by accidental Pd exposure during 1500 °C for 72Hrs with 100ppm O_2 (Top). Pd degradation of SiC after annealing at 1150 °C for 4 Hrs (Bottom)

EXPERIMENTAL METHODS

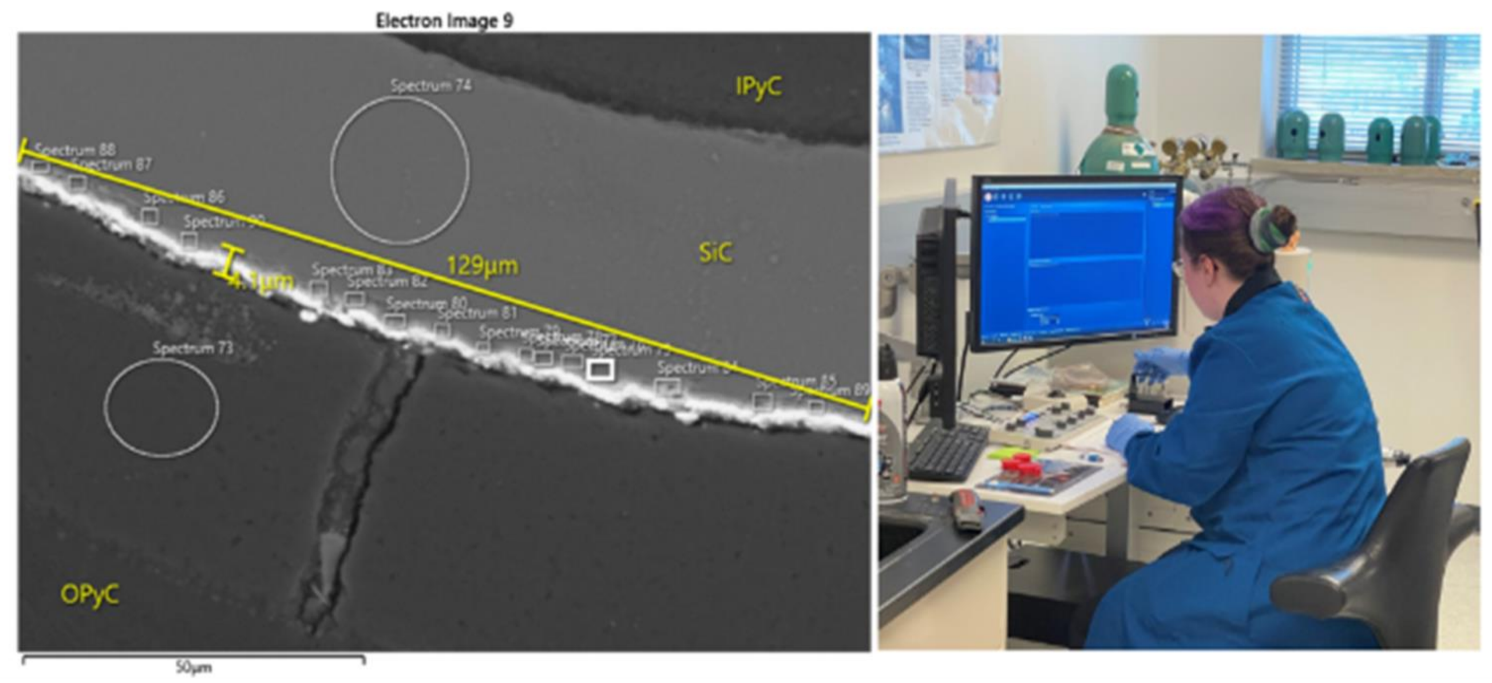

Figure 4: An electron image of 1150°C annealed Pd coated TRISO with EDS spot scans along the Pd/SiC interface (Left). Using a SEM to obtain EDS spot scans of TRISO (Right).

TRISO particles annealed at 850 °C and 1150 °C were imaged in SEM, where Pd corrosion was observed as seen in Fig. 4. The size of Pd-Si formations identified where not suitable for accurate EDS analysis.

In order to recreate the interactions between the SiC layer of a TRISO particle and Pd, SiC blocks of roughly 8.17mm in length by 6.01mm in width and height were leveled and polished. Pd foil of 1.0mm thickness was secured between 2 SiC blockers via alumina, Al_2O_3 jig to form the diffusion couple sample. The bulk diffusion couple assembly is showcased in Fig. 5. These couples were introduced to 1000°C for a 4-hour anneal in a tube furnace with a 5°C per minute ramp up.

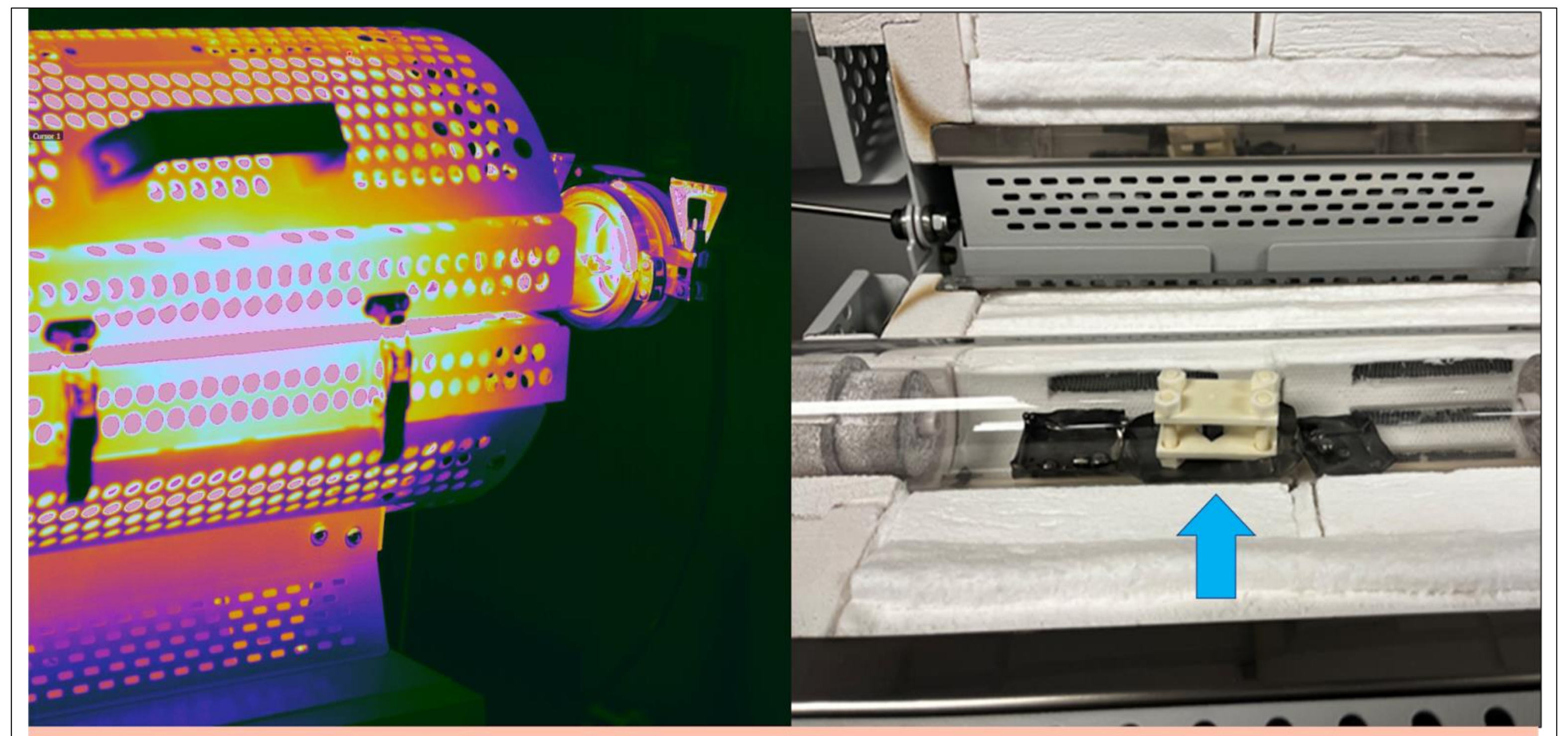

Figure 5: The alumina jig assembly inside of the tube furnace (Right). An IR photo taken of the tube furnace at dwell temp, 1000C. (Left).

After the couple cooled to room temperature, their level of diffusion was evaluated. A block with an area of diffusion of 6.15mm by 3.10mm was placed in epoxy resin. 2.5mm of material was ground off of this sample after the epoxy resin was hardened in order to expose the diffusion interface. The interface was then polished by hand in order to create a better surface for imaging.

Figure 6: Comparing temperatures with Tube furnace via IR camera.

RESULTS

- Pd corrosion was observed in post anneal SiC blocks.
- Possible Pd-Si formation likely due to Pd diffusion through SiC.

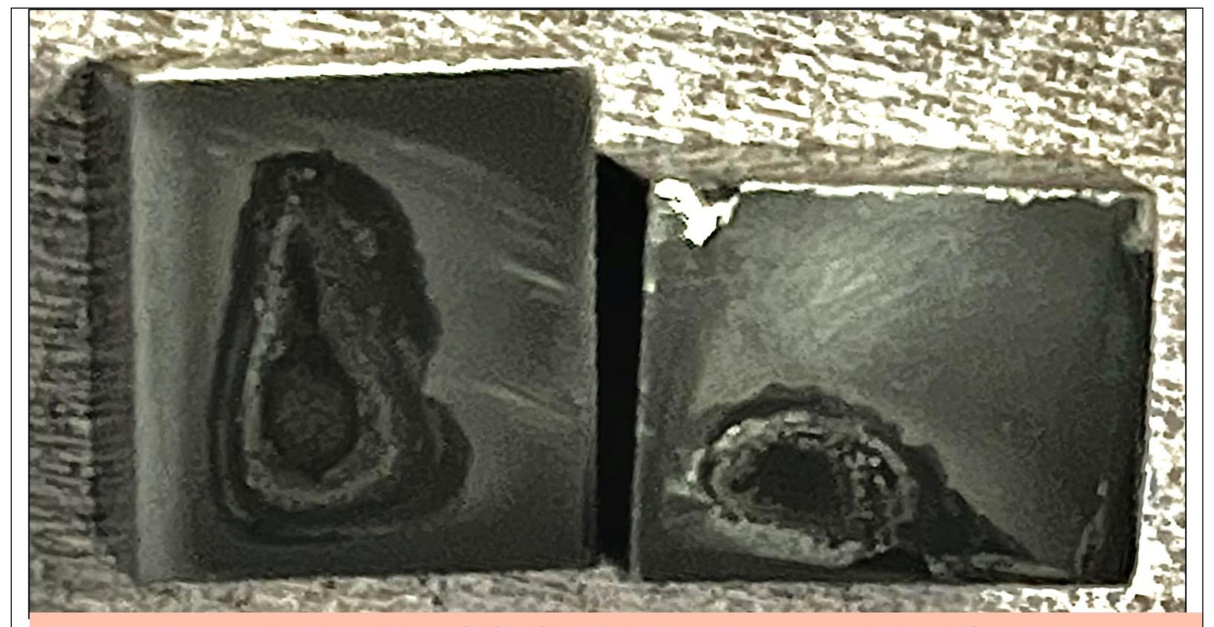

Figure 7: A photo of SiC blocks post 1000 °C 4Hr diffusion couple. Notable corrosion on surfaces that were in contact with Palladium foil.

CONCLUSIONS

- SiC degradation can occur when coupled with Pd at 1000 °C.
- Level of Pd diffusion and Pd-Si formations have yet to be determined as of present.

FUTURE WORK

The interface will have Au deposited on the surface and imaged using a scanning electron microscope. The sample will then Backscatter imaging will allow for easier viewing of different compounds that had formed from Pd diffusion. Possible Pd-Si areas will be scanned using EDS analysis to determine their weight precents. The weight precents of Pd and Si where then used to identify the Pd-Si compounds as seen in Figure 8.

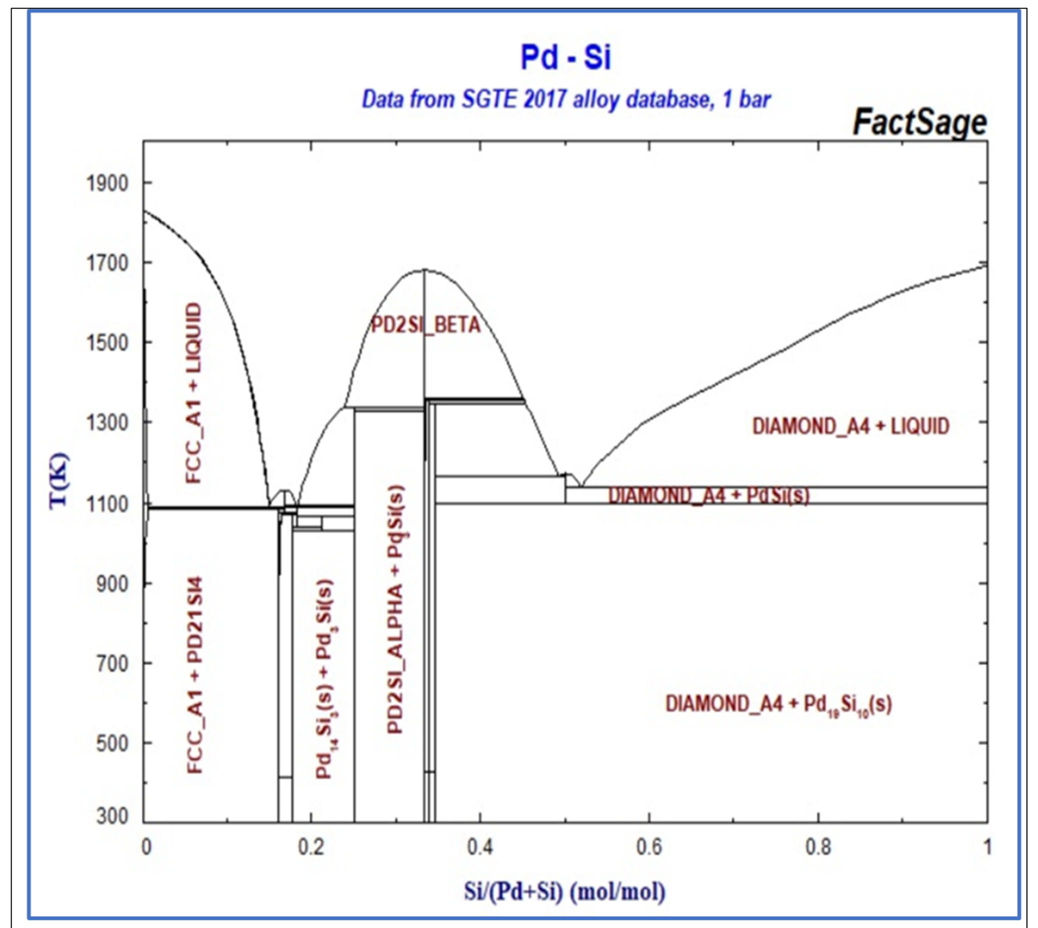

Figure 8: Phase diagram of Pd-Si compounds, used to identify compounds with mole fraction at specific temperatures [4].

REFERENCE

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