Modeling of Fission Fragment Detection in 4H-SiC Schottky Diodes

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INTRODUCTION

Pyroprocessing is one of leading methods for the recovery of actinides from spent nuclear fuel, which is an integral part of a closed nuclear fuel cycle. Electorefining in molten salt, such as LiCl-KCl-UCl₃ at 500°C, is the key unit operation in the pyroprocessing. During the electorefining process, used nuclear fuel in metal form is loaded into an anodic basket and lowered into the molten salt electrolyte of the electorefiner. Once immersed in the salt, the active metal fission products and transuranic (TRU) elements are chemically oxidized via reaction with UCl₃ in the electrolyte. As electric current is passed across the electrodes, uranium is oxidized from the anode and instantaneously reduced on the cathode. The TRU elements accumulate in the salt because they are thermodynamically unstable in the presence of UCl₃. Once they have reached a suitable concentration in the salt, they can be cathodically recovered as U/TRU metal in a liquid cadmium cathode (LCC). Pyroprocessing utilizes the molten LiCl-KCl salt as it has a high solubility for actinide and fission product chlorides along with appropriate electrochemical potentials. An advantage that pyroprocessing has over the PUREX process is that pyroprocessing is unable to separate plutonium from uranium if both are present within the salt. This severely hinders the ability to divert special nuclear material (SNM), like plutonium, from the recycling process.

Even though the pyroprocessing process has a built in means of preventing the separation of SNM, it is still important to monitor for the diversion of SNM. The easiest way to perform this task is to have near online monitoring of the actinide concentrations within the electorefiner salt. More specifically, the specific isotope concentrations along with the elemental concentrations must be monitored over the electrorefiner 500°C. During the electorefining process, used nuclear fuel in metal form is loaded into an anodic basket and lowered into the molten salt electrolyte of the electorefiner. Once immersed in the salt, the active metal fission products and transuranic (TRU) elements are chemically oxidized via reaction with UCl₃ in the electrolyte. As electric current is passed across the electrodes, uranium is oxidized from the anode and instantaneously reduced on the cathode. The TRU elements accumulate in the salt because they are thermodynamically unstable in the presence of UCl₃. Once they have reached a suitable concentration in the salt, they can be cathodically recovered as U/TRU metal in a liquid cadmium cathode (LCC). Pyroprocessing utilizes the molten LiCl-KCl salt as it has a high solubility for actinide and fission product chlorides along with appropriate electrochemical potentials. An advantage that pyroprocessing has over the PUREX process is that pyroprocessing is unable to separate plutonium from uranium if both are present within the salt. This severely hinders the ability to divert special nuclear material (SNM), like plutonium, from the recycling process.

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testing of the SiC detectors ability to detect fission fragments. The experiment will then be conducted at the OSURR and compared to the MCNP results. An experimental setup at the OSURR that utilizes SiC detectors on either side of the U-238 layer in a sandwich configuration will also be tested for coincidence measurements of detection of fission fragments for the purposes of U-238 fast fission efficiency measurements.

SiC Detector

SiC has many properties that enable it to be used as a wide band gap semiconductor alpha particle detector. SiC can be fabricated into small detectors that have good energy resolution and a high stopping power while being gamma blind. Additionally, 4H-SiC has good corrosion resistance to molten salts while also maintaining good stability at elevated temperatures. The alpha particle detector was fabricated using a 300 μm bulk layer of 4H-SiC with a 21 μm thick epitaxial layer. On top of the epitaxial layer, a 100 nm layer of nickel and a 10 nm platinum capping layer were deposited to form the dead layer. On the bulk layer, a 100 nm layer of nickel was deposited to form the ohmic contact. The detector was then annealed at 650 °C to form a Schottky contact between the nickel and epitaxial 4H-SiC layer. After creating the detector, a protective packaging was placed around the detector for immersion in the molten salt. The detector can then be immersed in the salt and actinides electrodeposited onto the platinum capping layer for alpha particle spectroscopy. Figure 1 shows the detector structure and SNM deposition process.

The Ohio State University Research Reactor

The Ohio State University Research Reactor (OSURR) is an open pool, light water moderated research reactor that contains many dry irradiation facilities for a range of different sized experiments. Within the core there are three irradiation facilities: the central irradiation facility (CIF), auxiliary irradiation facility (AIF) and the peripheral irradiation facility (PIF). These dry tubes run vertically from the pool top into the reactor core. The CIF has a radius of 1.778 cm and the AIF and PIF have a radius of 3.175 cm. Any of these facilities could be used to house a SiC Schottky diode detector with U-238 deposited on the surface. These facilities would allow for a containment to be designed and placed around the detector with appropriate feedthroughs for electrical connections. The CIF experiences the largest total and fast neutron flux of any facility, 2.3e13 and 1.0e13 n/cm²/s respectively. Figure 2 shows an MCNP model of the OSURR with labels for the CIF, AIF and PIF.

MCNP Modeling of Detector and Fission Fragment Response

Modeling of the SiC detector began with creating the device in MCNP. Both the bulk and epitaxial layers of 4H-SiC were first generated using planes in the X, Y and Z directions. The bulk layer was modeled as a 5 mm by 5 mm square in the X-Y plane and extended 300 microns in the Z plane centered at the origin. Following this, the epitaxial layer was modeled as a 21 micron thick layer on top of the
bulk layer extending in the positive Z-direction and the same 5 mm by 5 mm square in the X-Y plane. A 100 nm layer was then added to both the top of the epitaxial layer and the bottom of the bulk layer for the Schottky and ohmic contacts. On top of the Schottky contact, the 10 nm platinum cap was added. Last, a 3 mm diameter, 1 micron think layer of depleted uranium was placed on top of the platinum cap, simulating electrodeposited SNM. Depleted uranium was chosen for the modeling purposes as the majority of the actinides deposited will be uranium and this setup will most closely match the experiment to be conducted at the OSURR. Depleted uranium will also serve as a conservative estimate for the energy loss within the electrodeposited SNM, as it has a larger density than the actual source material. Figure 3 below shows the detector modeled in MCNP and Figure 4 shows a zoomed in view showing the nickel, platinum and SNM layers.

Fig. 3. Top down and side view of SiC Schottky diode detector in MCNP VISED

Fig. 4. Side view of top layers of SiC Schottky diode

The SiC detector with SNM deposited was then placed within the CIF tube of the OSURR model. A k-code simulation was used to simulate an irradiation of the device within the reactor. Tallies were used to determine the neutron flux spectrum incident on the detector and determine the theoretical fission rate within the SNM layer deposited on the detector.

Pu-240 exhibits a double humped distribution of fission fragments similar to that of U-235. Two of the most common fission fragments emitted by Pu-240 are Ru-103 and Xe-134. These two isotopes were chosen as the representative light and heavy fission fragments respectively. Choosing only one heavy and one light isotope to model greatly simplifies the source term and the difference between similar light and heavy fission fragments should be negligible. The average energy for the light and heavy fission fragments are 68 MeV and 93 MeV respectively.

Results

To first model the fission fragments, SRIM and TRIM calculations were completed to determine the range of both heavy and light fragments.

Two different source configurations were first modeled in MCNP. The first was a planar source located along the top edge of the layer of SNM. The fission fragments were distributed evenly across the radius of the layer, but were all orientated parallel to the Z-axis facing the SiC detector. Next the source was distributed evenly throughout the entire volume of the SNM layer. These particles were started isotropically throughout the volume in both starting location and initial direction. These two configurations were modeled for both the light and heavy fission fragments. Figures 5 and 6 show the surface source distribution of the simulated pulse height distribution and the energy.
deposition of the fission fragments. Figures 7 and 8 show the volume source distribution of the simulated pulse height distribution and energy deposition of the fission fragments. All figures are normalized to a single source particle, as is standard for MCNP.

These plots show the energy deposition of the light and heavy fission fragments and the simulated detector pulse height observed by each. The surface sources have a nice gaussian peak for both energy deposition and pulse height, as the particles are orientated directly at the detector and thus only lose energy passing through the layer of the SNM. These particles have an average energy deposition within the detector of 62.8 MeV for the light fragment and 40.6 MeV for the heavy fragment. SRIM/TRIM calculations using this same geometry, starting energy and source orientation resulted in an average energy deposition of 65.94 MeV for the light fragment and 41.88 MeV for the heavy fragment.

The volume source had a much more scattered pulse height and energy deposition distributions as the particles can be orientated in any direction and start anywhere within the SNM layer. Both of these results show the fission fragments deposit significantly more energy than the Pu-240 alpha energy of 5.225 MeV.
Conclusion and Future Work

The results show that fission fragments within the SNM layer will reach the active layer of the SiC Schottky diode detector. These fission fragments will also deposit significantly more energy than the highest alpha energy emitted by the SNM layer. The SiC detector should be able to monitor the spontaneous fission of Pu-240 to determine the mass of the Pu-240, and thus determine the alpha activity of both Pu-240 and Pu-239. This will allow for the determination of the isotopic concentrations of both Pu-240 and Pu-239 during the pyroprocessing process.

Future work will include modeling of the SiC detector within the MCNP OSURR model. Experimentally testing the SiC detectors with a layer of SNM within the OSURR will be conducted to show that it is possible to measure the fission fragments of SNM using the SiC Schottky diode detectors.

REFERENCES