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Fission Gas Measurement from Annular, LEU-foil Based Target

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ABSTRACT

The quantity of fission-product gas released from opening a high-density, LEU-foil target has not been previously characterized. We developed the hardware and a technique that has allowed us to open up an irradiated target and to collect and quantify such fission-product gas. The data collection metrology, which uses a helium sweep gas as a carrier and an activated charcoal filled condenser cooled to liquid nitrogen temperatures, has been calibrated using tracer runs of known quantities of Xe-133. A series of experiments is currently in progress to evaluate the fission-product gas release during disassembly of four different fission recoil barriers. Data collected to date has shown evidence of Xe-133 release of a magnitude that is a small fraction of 1%.

1. Introduction

The radioactive tracer Technetium-99m is widely used in medical imaging and is derived from its parent isotope Molybedenum-99 (Mo-99) by radioactive decay. The majority of Molybdenum-99 (Mo-99) produced internationally is extracted from high enriched uranium (HEU) dispersion targets that have been irradiated. The high concentration of U-235 in HEU-based targets makes them potential items of interest for rogue individuals. To alleviate proliferation concerns, the use of low-enriched uranium (LEU) targets is being mandated. However, the conversion of HEU to LEU based dispersion targets affects the Mo-99 yield. An increase in uranium density is required in LEU-based targets in order to recover the loss in Mo-99 production-per-target [1]. A possible approach to increase the uranium density is to use an LEU metal foil placed within an aluminum cladding to form a composite structure [2].

High density targets can help improve the economics of transitioning from high-enriched uranium (HEU) to low-enriched uranium (LEU). One high density target strategy is to use an LEU foil and to process the target post-irradiation using the modified Cintichem process [3]. An advantage of the modified Cintichem process is that the target may be cut open after irradiation and the foil removed to allow only processing of the LEU. Just dissolving the LEU foil, and not the cladding, reduces the amount of liquid waste that needs to be treated. However cutting open the target in the hot cell provides an opportunity for fission gases to be released into the hot cell and the facility stack. This paper describes an experiment that is currently ongoing at the University of Missouri Research Reactor (MURR) that evaluates the amount of fission gas released when a target is cut open, for a range of fission recoil barriers.

2. Background

The amount of fission gas produced will be distributed throughout the foil and not necessarily released into the cladding. Only a fraction of the gas generated will be at a location in the target where it may diffuse out of, or be produced on the surface of, the foil such that it can be released when the target cladding is cut open. The objective of this work is to determine the amount of fission gas that is released when an irradiated LEU fission target is cut open.

Neutron irradiation of U-235 creates fission products that include various gaseous isotopes of Xe, Kr, and I, in addition to Mo-99. The relative amounts of each isotope generated depend on the neutron energy distribution and duration of irradiation. An analysis of an LEU-foil (19.75% U-235) target placed in the N-1 position at MURR (8.0×10^{13} n/cm^2-s thermal neutron flux) for one irradiation cycle (150 hours) has been conducted using ORIGEN 2.0 to estimate the isotopic distribution of gaseous fission products [4]. A summary of the gas inventory from that analysis is shown in Table 1.

Table 1. Summary of Elemental Fission Gas Production from Irradiation of LEU Foil at MURR

Gas	Total Moles/Gram LEU
I	9.48×10^{-7}
Kr	8.34×10^{-7}
Xe	5.44×10^{-6}

Of particular interest are the gaseous isotopes with a half-life on the order of days as those isotopes can be used as a means to estimate the total gas production through spectroscopic counting. The most promising isotope that is expected to be present for counting multiple days after irradiation is Xe-133 (about 2.7 Ci/g LEU total remaining 3 days after irradiation). I-133 is also potentially promising if it can be kept from condensing on the target cladding and/or other structures in the hot cell before it can be counted (about 3.8 Ci/g LEU total remaining 3 days after irradiation).

The fission gas inventories described above describe the total amount generated. But the amount released when the target is cut open is expected to be a small fraction of the total amount generated. Something on the order of 1%-2% of the total inventory at most. Therefore an isotope counting process will need to be conducted in a very controlled manner to maximize the signal-to-noise ratio. And specifically, the gas will need to be removed from the proximity of the target for counting in order to make sure that the released gas counting does not get overwhelmed by and confounded with the gas trapped in the LEU foil matrix. So a mechanical gas collection and holding structure needs to be developed.

3. Experiment Design

There are at least two methods for collecting released fission gases. One approach is to use a target where fission gases are collected in a volume specifically created for gas capture [5]. In that strategy, after the target has been irradiated, a vacuum bottle attached to a piercing needle breaches the designated gas collection volume. The captured gas is then driven into the vacuum bottle that can be taken to a remote location for gas counting.

A second approach, and the one followed here, is based upon the strategy used by many investigators where an inert sweep gas like He or Ar is flowed over the test structure in an effort to entrain the fission gas [6-7]. The gas mixture is then routed to a cold finger condenser cooled by liquid nitrogen or some other cryogenic fluid to condense the fission gas products. Another strategy is to route the gas mixture through a condenser containing a high surface area adsorbent like activated charcoal to condense the fission gas products. After a period of time the sweep gas is turned off and valves to the condenser are closed to trap any condensed fission products. The condenser is then removed from the apparatus and taken to a location where the radiation emission from the trapped gas isotopes is counted. Or the condenser is sufficiently isolated such that counting can be done in-situ. Our experiment employs a sweep gas of He and a cryogenically cooled condenser that contains activated charcoal positioned in a 'counting' well beneath the hot cell. The positioning of the condenser allows for in-situ counting of gamma-ray emission.

In our experiment design, we need to be able to cut open the target in an enclosed environment within a hot cell. The annular target has conventionally been opened in previous experiments by cutting off each of the two welded ends and then slitting the outer tube of the target along the longitudinal direction using a manipulator actuated tool. That tool was modified to be actuated using stepper motors that are feedback controlled using an Arduino micro-controller. Chain transmissions are used between the stepper motors and the cutting wheel drive mechanisms. A photograph of the tube cutting device is shown placed in the fission box in Fig. 1.

A sealed, aluminum box was built to hold the cutter and to provide inlet and exit ports for the helium sweep gas. The box has doors on either side to allow samples to be set on the cutter. The doors are held closed with latches and are sealed with gaskets and vacuum grease. The sweep gas is introduced at the top of the box through a threaded hole and is drained at the bottom of the box through another threaded hole. Electric wires for controlling the cutting motors are passed into the box through sealed ports. The box with the cutter inside is shown in Fig. 1.

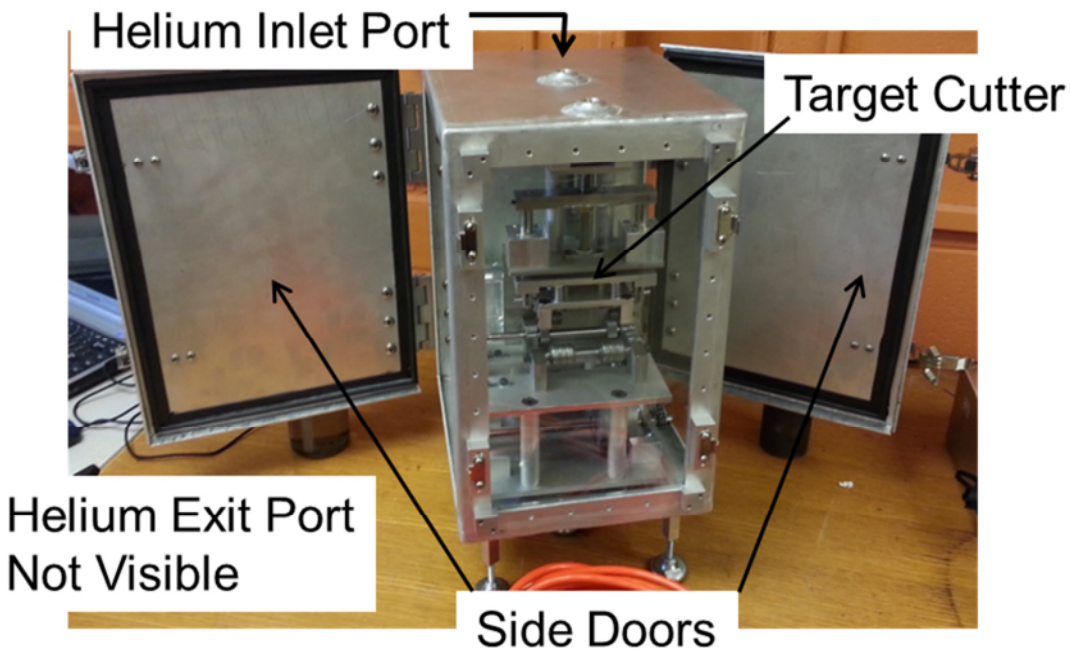

Figure 1. Photograph of Fission Collection Box with Target Cutter Placed Inside.

The condenser is designed to trap the fission gas entrained in the sweep gas. From the theoretical analysis presented above, it is expected that fission processes will generate a total of about 15 Ci of Xe-133 in the 5 gram LEU foil, 3 days after the end-of-irradiation at MURR stopped. Assuming a gas release of about 1% when the target cladding is cut open yields a potential signal of about 150 mCi.

The amount of fission gas release is relatively small, and of course the sweep gas will dilute the concentration of the fission gases further. If one only considers the open volume of the collection box, about 25 L, the mole fractions of both the Xe and Kr are on the order of 10^{-6}. Those very small concentrations suggest that a cold, large surface area is needed in order to capture countable amounts of active fission gas.

Activated charcoal is a unique material that has a large surface area-to-volume ratio. It is a material that has been proposed to adsorb fission gases as a means to scrub emissions from nuclear processing [8-12]. We added 28.7 grams of activated charcoal (NUSORB KITEG from NUCON International, Inc.) roughly centered vertically in a 6 inch length of copper tubing that is 1 inch in diameter. To further encourage the fission gases to precipitate from the helium sweep gas, the condenser was submerged in a Dewar containing liquid nitrogen at a temperature of about 77K. As illustrated in Fig. 2, heater tape was wrapped around the condenser inlet in order to keep the temperature of the quick disconnect greater than -40 ^{o}C. Thermocouples were attached and monitored during the experiment in order to make sure that the liquid nitrogen level was sufficiently high and the quick disconnect was sufficiently warm.

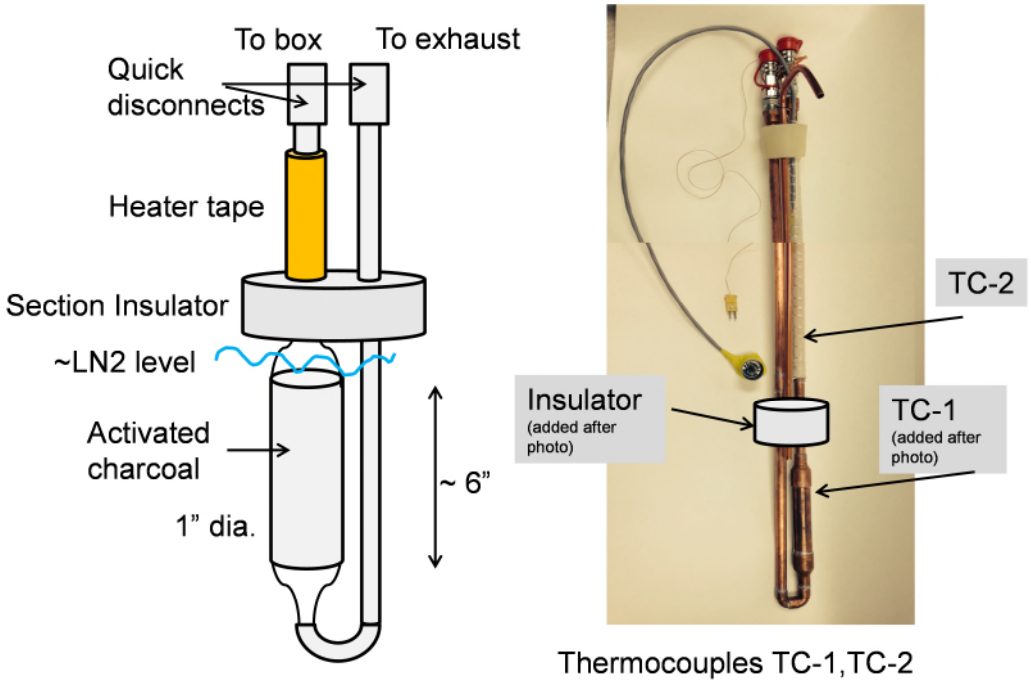

Figure 2. Photograph and Sketch of Condenser Design.

The 133Xe activity collected on the liquid nitrogen cooled activated carbon was measured using a HpGe detector (Canberra Model GC2018) and Inspector 2000 digital signal processor. The detector has a relative efficiency of 20% and a full-width-half-maximum resolution of 1.8 keV at 1.33 MeV. The detector was approximately thirty-six inches from the activated carbon trap. The detector was collimated with two inches of lead shielding with a 0.25 inch tall slit across the front face of the detector. A sketch of the overall fission gas collection system is shown in Fig. 3.

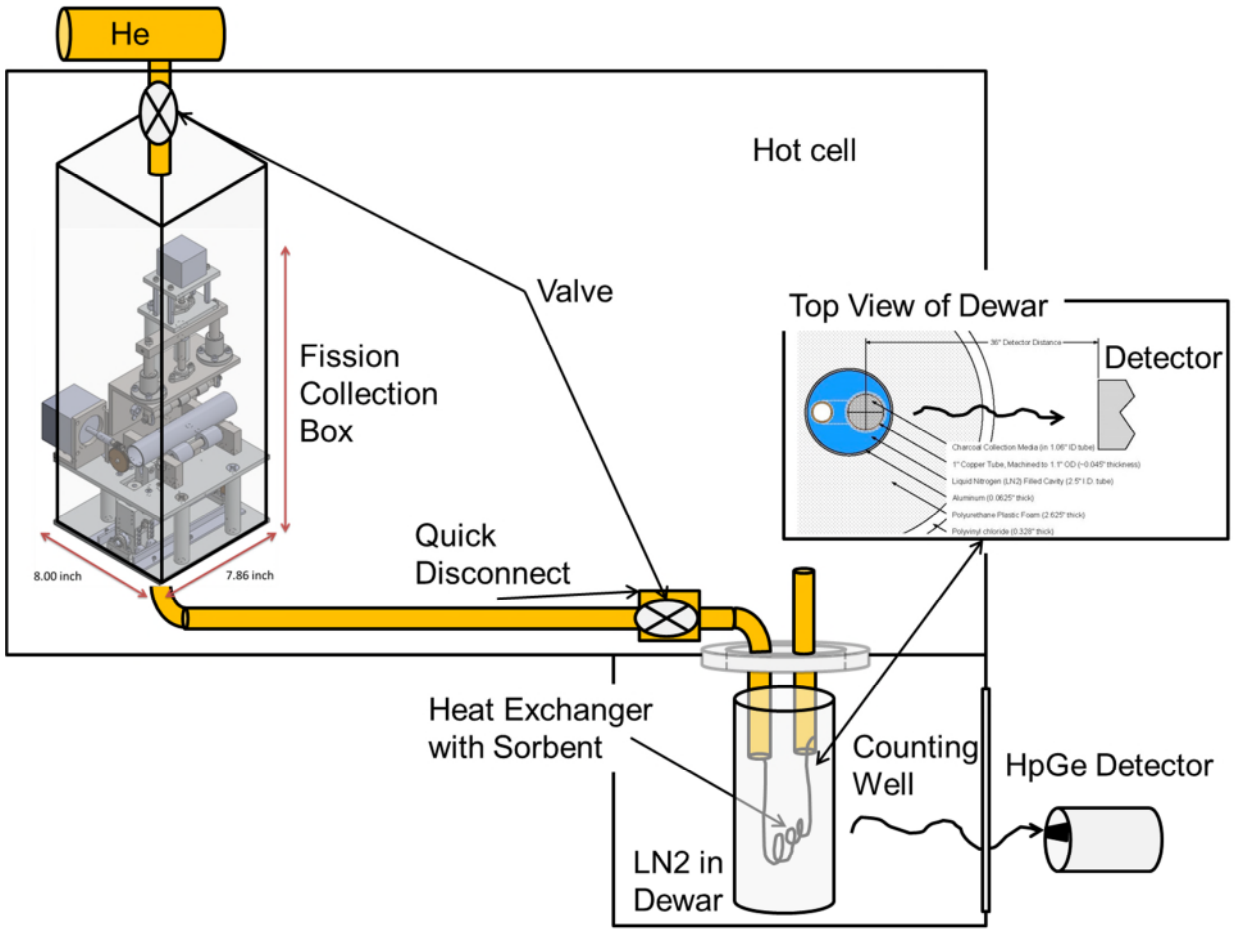

Figure 3. Layout for Fission Gas Collection and Counting System

The overall efficiency of the system for collecting and measuring Xe-133 was determined using one 10 and two 20 mCi Xe-133 sources. The calibrated sources were obtained from Lantheus Medical Imaging. The glass vial calibration sources were placed in the target cutting device and the radioactive gas was released by crushing the glass vial with the target cutter. Prior to crushing a calibration source, liquid nitrogen was introduced into the Dewar holding the activated carbon trap and the flow of liquid nitrogen was adjusted to maintain a constant temperature of -160 ^{o}C on the copper housing of the carbon trap during the entire measurement period (approximately 5 hours). Helium was introduced to the cutter box at a flow rate of 0.5 L/min to sweep the radioactive gas from the cutter box through the carbon trap. The glass vial calibration source was then crushed and a 300 second (live time) gamma spectrum of the carbon trap was recorded every five minutes. The dead time of the detector was less than 2% for all calibration and fission target measurements.

4. Results

The temporal variation in the count rate of the 81 keV gamma ray from Xe-133 over time for the three tracer calibration runs was similar to results observed for the LEU fission foil (see Fig. 4). In every case, it took approximately two hours for the measured count rate to reach a steady value and the count rate remained constant as long as the temperature of the activated carbon trap was held at -160 ^{o}C. As can be seen from Fig. 6, when the flow of liquid nitrogen was turned off

and the temperature of the trap rose above approximately -50 ^{o}C, the measured Xe-133 activity dropped. The average system response factor at steady state from the three calibration runs performed on three different days was 57.8 ± 1.4 counts per second/mCi of Xe-133 released in the fission collection box, at a 95% confidence interval.

The count rate of the 81 keV gamma ray from the LEU-foil wrapped with nickel recoil barrier target is shown in Fig. 4 as a function of time after the target was cut open. It can be seen that there is a significant uptake of Xe-133 at the condenser location where the counter was positioned. After about 2 hours the count rate leveled off at 127 ± 1.8 counts per second at a 95% confidence interval. This equates to 2.2 mCi of Xe-133 released by the target in the fission collection box, at the time of the target was cut open. Decay correcting this value 80 hours to the end of irradiation for the LEU foil and using the 20 Ci of Xe-133 activity predicted by ORIGEN at the end of irradiation yields a Xe-133 fission gas release of less than 0.017%.

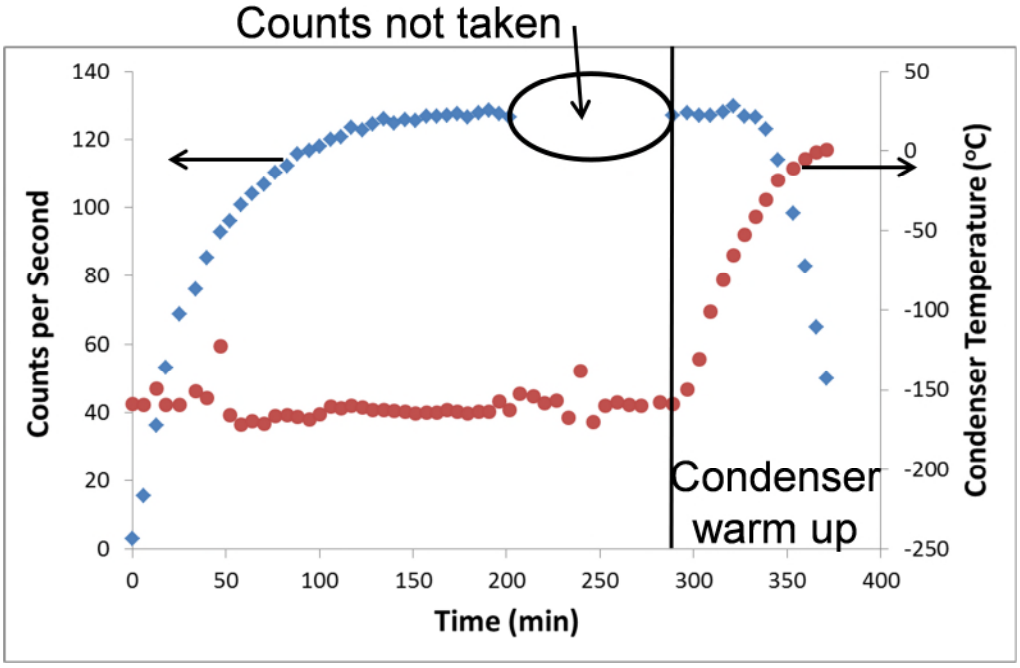

Figure 4. Xe-133 Counted in Condenser for LEU-foil Target with Ni Recoil Barrier.

After steady behavior was observed, the liquid nitrogen was allowed to fully evaporate from the Dewar and the condenser allowed to warm up to ambient temperature as can be seen on the right hand portion of Fig. 4. As the condenser warms up it can be noted that the count rate decreases, suggesting that the Xe-133 evaporates/desorbs from the activated charcoal. This behavior clearly demonstrates the need to keep the activated charcoal below the saturation temperature of the Xe. It also suggests that the activated charcoal trap can be regenerated to be used for repeated experiments.

5. Conclusions

A fission gas collection and counting system was constructed and demonstrated to be effective for trapping and counting Xe-133. Experiments using calibrated tracer amounts of Xe-133 in glass vials were carried out and used to characterize the apparatus, relating the number of counts observed by the gamma-detector to the amount of radioactive gas spatially released where

targets are cut open. The system was then used to collect and count the fission gas release from a 5g LEU-foil based target with a Ni foil recoil barrier. The results suggest that approximately 0.017% of the predicted Xe-133 within the LEU-foil was released by the target at the point in time when a Mo-99 producer might process the target. This would be if the producer were to cut open the target and remove the LEU-foil for dissolution.

6. Acknowledgements

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7. References

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