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Fission Product Inventory and Burnup Evaluation of the AGR-2 Irradiation by Gamma Spectrometry

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Gamma spectrometry has been used to evaluate the burnup and fission product inventory of different components from the US Advanced Gas Reactor Fuel Development and Qualification Program's second TRISO-coated particle fuel irradiation test (AGR-2). TRISO fuel in this irradiation included both uranium carbide / uranium oxide (UCO) kernels and uranium oxide (UO₂) kernels. Four of the 6 capsules contained fuel from the US Advanced Gas Reactor program, and only those capsules will be discussed in this work. The inventories of gamma-emitting fission products from the fuel compacts, graphite compact holders, graphite spacers and test capsule shell were evaluated. These data were used to measure the fractional release of fission products such as Cs-137, Cs-134, Eu-154, Ce-144, and Ag-110m from the compacts. The fraction of Ag-110m retained in the compacts ranged from 1.8% to full retention. Additionally, the activities of the radioactive cesium isotopes (Cs-134 and Cs-137) have been used to evaluate the burnup of all US TRISO fuel compacts in the irradiation. The experimental burnup evaluations compare favorably with burnups predicted from physics simulations. Predicted burnups for UCO compacts range from 7.26 to 13.15 % fission per initial metal atom (FIMA) and 9.01 to 10.69 % FIMA for UO₂ compacts. Measured burnup ranged from 7.3 to 13.1 % FIMA for UCO compacts and 8.5 to 10.6 % FIMA for UO₂ compacts. Results from gamma emission computed tomography performed on compacts and graphite holders that reveal the distribution of different fission products in a component will also be discussed. Gamma tomography of graphite holders was also used to locate the position of TRISO fuel particles suspected of having silicon carbide layer failures that lead to in-pile cesium release.

I. INTRODUCTION

The AGR-2 TRISO Fuel irradiation is the second test in a series of irradiations designed to qualify tristructural isotropic (TRISO) fuel for use in next generation reactors. The first irradiation, AGR-1, and its associated post-

irradiation examination (PIE) and safety testing demonstrated the fuel performance of lab-scale-produced TRISO fuel.^{1, 2} The AGR-2 irradiation and its associated PIE is designed to demonstrate the fuel performance of production-scale TRISO fuel.^{3, 4} Additionally, while AGR-1 contained exclusively uranium oxide / uranium carbide (UCO) TRISO kernels, AGR-2 contained a capsule with uranium oxide (UO₂) TRISO fuel.⁵

The irradiation contained six separate capsules. Capsules 2, 3, 5, and 6 contained fuel fabricated in the US and are the subject of this paper. Capsules 2, 5, and 6 contained fuel compacts with UCO TRISO particles, while Capsule 3 contained compacts with UO₂ particles. The AGR-2 UCO fuel consisted of nominally 425 μm diameter kernels with 14.0% ²³⁵U enrichment. UO₂ fuel consisted of nominally 500 μm diameter fuel kernels with 9.6% ²³⁵U enrichment. The TRISO coatings on both types of kernels had nominal thickness of 100 μm (buffer), 40 μm , (inner and outer pyrolytic carbon), and 35 μm (SiC). The coated particles were formed into right cylindrical compacts approximately 12.3 mm in diameter and 25.1 mm in length. The particle packing fractions were 37% and 23% for the UCO and UO₂ compacts, respectively.⁵

The test train contained 6 independently controlled and monitored capsules.^{6, 7} The major components of the AGR-2 capsules are shown in Fig. 1 and include the fuel compacts, the graphite holder that supports the compacts, the graphite spacers, the capsule shell, and the gas exit lines. In each of the four AGR-2 capsules discussed in this work there were 12 compacts with 4 in each of the three stacks. The compact numbering scheme for AGR-2 is shown in Fig. 2 where compacts are numbered by capsule number, axial level, and stack number.

The AGR-2 irradiation was performed in the Idaho National Laboratory (INL) Advanced Test Reactor (ATR) for 12 cycles or 559.2 effective full power days and reached calculated burnups ranging from 7.3 to 13.2 % FIMA (fission per initial heavy-metal atom) for UCO fuel and 9.0 to 10.7 % FIMA for UO₂ fuel. The time-averaged-volume average temperature for UCO fuel ranged from 987°C to

1296°C for the irradiation and for UO₂ fuel from 996°C to 1062°C for the irradiation.⁷

Gamma spectrometry was used previously to successfully evaluate the burnup of the AGR-1 TRISO fuel compacts⁸ as well as the inventory of Ag-110m remaining in the compacts.⁹ Additionally, gamma spectrometry of the graphite support structures in AGR-1 was able to identify compacts containing defective SiC TRISO particles.¹⁰ Gamma spectrometry was also utilized to create an inventory of gamma-emitting fission products of each of the capsules in AGR-1.⁹ This combined experience from AGR-1 has been applied to the similar compacts and capsule components in AGR-2. This work will focus on gamma spectrometry results from the four US fuel capsules.

II. EXPERIMENT

Gamma scanning was accomplished using the INL Hot Fuel Examination Facility (HFEF) Precision Gamma Scanner (PGS). The PGS has three major components: the detector system, collimator, and stage. The detector system is a high purity germanium (HPGe) detector surrounded by a Compton suppression detector, and a digital multi-channel analyzer. The collimator is approximately 2.13 m long with an aperture that has a fixed width of 2.22 cm and a variable height of 0.254 to 0.00254 cm. The collimator can also be rotated 90° so that the fixed width of the aperture is vertical instead of horizontal. The stage suspends items before the collimator and moves them in a plane parallel to the face of the collimator. The stage can also rotate items about a central axis.

Compacts were suspended before the PGS collimator in a thin-walled aluminum tube and were individually encapsulated in an additional aluminum container. Calibration sources (Eu-152) packaged in equivalent containers and placed inside the same aluminum tube were scanned before the first compact and after the last compact to confirm the performance of the system. Each compact was typically scanned in 0.254 cm steps for a live time of

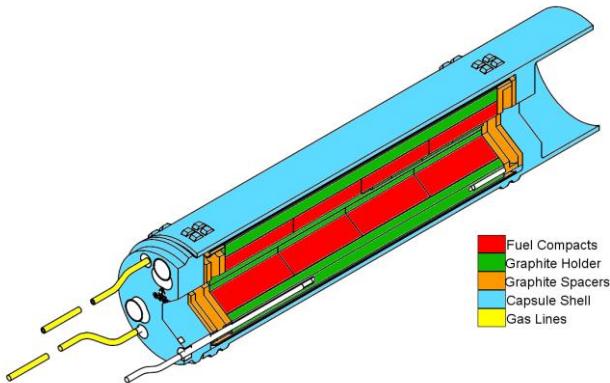


Fig. 1. Major components of a representative AGR-2 capsule.

30 minutes. The collected spectra were analyzed to evaluate both the fission product content of each compact and the burnup of each compact. Fission product inventories were compared to the predicted fission product inventories from Reference 11. Burnup was calculated from both the measured Cs-137 activity and the measured Cs-134 to Cs-137 activity ratio and compared to the predicted burnup from Reference 11. Burnup determination follows the technique discussed in Reference 8. Burnup is derived from both the absolute Cs-137 decay-corrected activity measured in each compact and by the relative decay-corrected activity ratio of Cs-134 to Cs-137. Burnup derived from Cs-137 only can only be calculated as an average for the entire compact, since the starting inventory of fissile material is not known for the thin section of the compact characterized in each scan. The total activity for a compact is determined by summing the local activity from all the different measurements of a single compact. The total is then converted to burnup. The burnup determined by the ratio of Cs-134 to Cs-137 can either be determined over the entire compact based on total activities or locally for every spectrum collected from each compact. This local measurement of burnup reveals some significant burnup gradients in some compacts. The activity of several different gamma-emitting fission products present in the compacts was also quantified, including Zr-95, Ru-106, Ag-110m, Cs-134, Cs-137, Ce-144, and Eu-154. The quantification of Ag-110m was given special attention and some additional longer scans

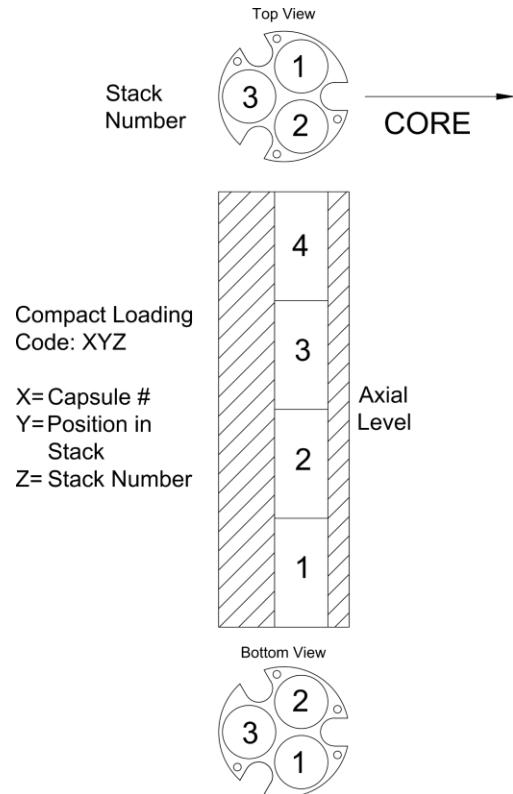


Fig. 2. Compact numbering scheme for AGR-2 compacts

were performed on specific compacts to better quantify the Ag-110m content of the compacts.

In addition to the compacts, the graphite holders were also gamma scanned with the PGS. The goal of these scans is to estimate the inventory and distribution of fission products in the graphite. The distribution of different fission products can be used to infer the fuel performance of compacts during irradiation, and information about fission product release from compacts can be gathered. Graphite holders are scanned in two ways. Initially, they are scanned in two off-axis sweeps to identify axial levels of interest and to estimate the total activity of different isotopes in the holders. In the off-axis scans, the half of the holder that held Stacks 2 and 3 is typically scanned first, and the half of the holder that held compacts from Stacks 1 and 3 is scanned second. See Fig. 3 for an illustration of the off-axis scanning. If an axial level of interest is identified, a tomographic scan of that level is performed, during which the PGS collimator is rotated to a vertical orientation and the holder is turned about its axial centerline after the PGS scans across the holder at a particular level and angle. The scans from several angles are used to reconstruct an activity intensity map of a particular isotope in the graphite holder. Image reconstruction utilizes Gamma Emission Computed Tomography and produces isotope-specific tomograms.¹² The axial levels of interest are typically levels that contain Cs-134 and Cs-137 signals, which indicate that the compact from that location during the irradiation possibly had a TRISO particle with a failed SiC layer similar to what was seen in Reference 10.

III. RESULTS

III.A. Compact Gamma Spectrometry Results

Data from the gamma spectrometry of compacts have been processed into burnup estimates and compared to the predicted burnup from physics calculations.¹¹ This is shown in Fig. 4 for all U.S. capsules (2, 3, 5, and 6). The numerical values for the predicted and measured by the Cs-134 to Cs-137 ratio are shown in Table 1. The ratio based burnup value is shown in favor of the Cs-137 only activity because it is less susceptible to biases introduced by determining the absolute efficiency of the detector system. Burnup values are plotted in relation to the vertical displacement from ATR core centerline. This places 3 data points at the same horizontal position on the graph, since there are three compacts at each level, each in a different stack (Fig. 2). Because of the geometry of the capsule in the ATR core, the compacts in Stack 1 and 2 were irradiated with roughly equivalent neutron fluence, and the data from Stack 1 and Stack 2 largely overlap. The compacts in Stack 3 were shielded from the core by the other stacks and saw a lower neutron fluence and thus burnup. The burnup based on the total Cs-137 activity in

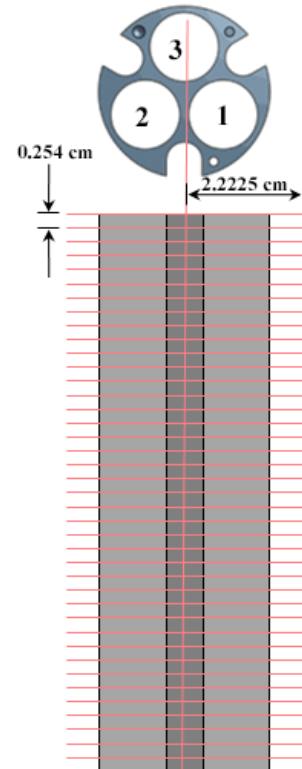


Fig. 3. Illustration of the off-axis scanning of AGR-2 graphite holders.

each compact is shown by the orange squares, and the local burnup determined from the ratio of Cs-134 to Cs-137 is shown by the blue diamonds (data from individual compacts cannot be distinguish readily in Fig. 3 because there was no separation between the compacts in each stack). Local burnup can vary by 1 to 2% FIMA across a single compact, which is a statistically significant variation. Measured burnup ranged from 7.3 to 13.1 % FIMA for UCO compacts and 8.5 to 10.6 % FIMA for UO₂ compacts. See Table 1 for more detail. Overall, the burnup values based on the ratio of Cs-134 to Cs-137 and the direct Cs-137 activity agree reasonably well with the predicted burnup.¹¹ This level of agreement is in line with AGR-1.⁸

In the Capsule 5 data, there is significantly more scatter in Stack 1 and 2 Cs ratio data than was typically seen in PGS scans of AGR-1 and in scans of the compacts from the other AGR-2 capsules. This is due to a statistically significant variability in the activity of Cs-137 and Cs-134 across the compacts from scan to scan, although the cause is unknown. The difference between measured and predicted burnup values in Stack 1 and Stack 2 (the higher-burnup stacks) in Capsule 6 is similar to what was seen in AGR-1 comparisons for Capsule 6, which was in a similar axial position relative to the core.⁸

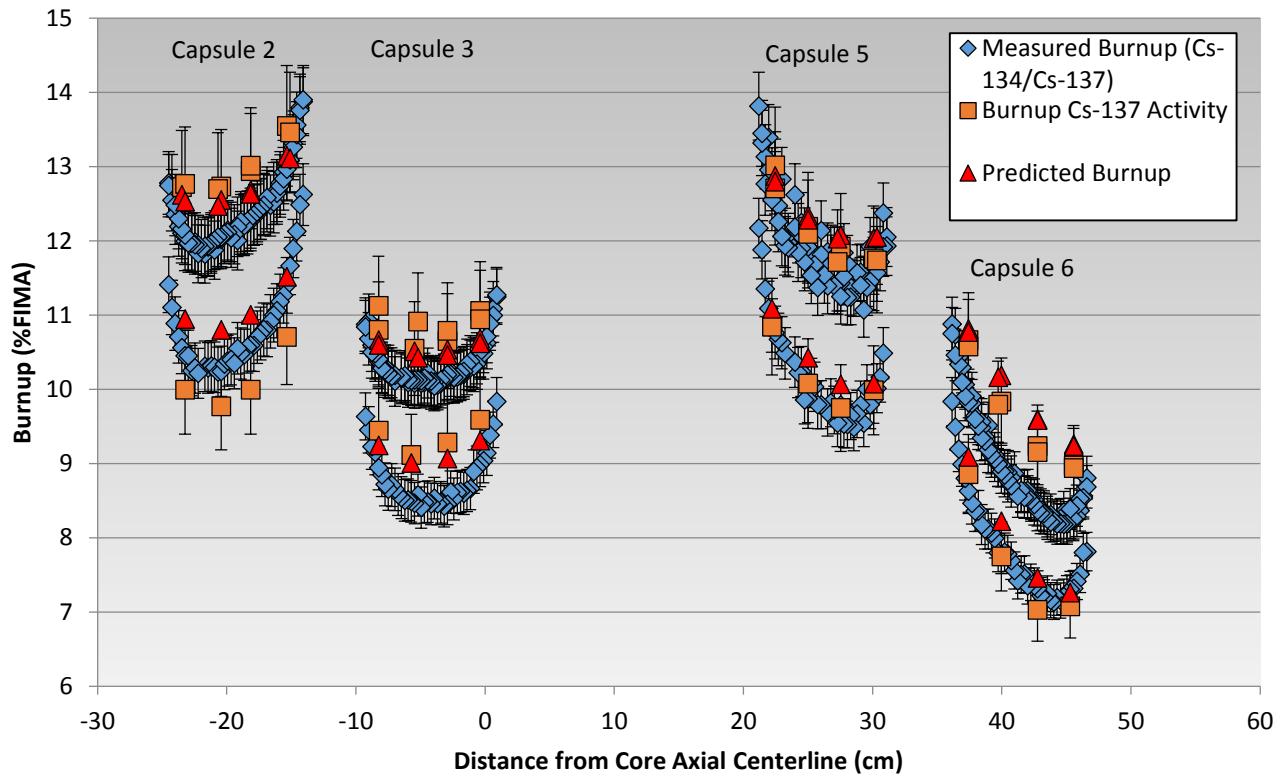


Fig. 4. Burnup evaluation of AGR-2 based on gamma spectrometry of AGR-2 compacts.

Table 1. Predicted burnup compared to measured burnup calculated by the ratio of Cs-134 to Cs-137 for the AGR-2 compacts. The asterisk (*) in each compact ID number corresponds to the capsule number.

Compact	Capsule 2		Capsule 3		Capsule 5		Capsule 6	
	Predicted	Measured	Predicted	Measured	Predicted	Measured	Predicted	Measured
*43	11.52	11.6	9.31	9.1	10.08	10.0	7.26	7.9
*33	11.00	10.6	9.07	8.5	10.07	9.6	7.46	8.0
*23	10.80	10.3	9.01	8.5	10.42	10.0	8.22	8.6
*13	10.95	10.6	9.25	9.0	11.09	11.1	9.09	9.3
*42	13.15	13.1	10.69	10.6	12.03	11.8	9.26	8.8
*32	12.68	12.3	10.54	10.1	12.08	11.6	9.60	9.1
*22	12.55	12.0	10.51	10.1	12.34	12.0	10.19	9.7
*12	12.62	12.2	10.66	10.4	12.88	12.9	10.81	10.6
*41	13.11	13.1	10.62	10.6	12.05	11.6	9.24	8.8
*31	12.63	12.3	10.46	10.2	12.03	11.5	9.59	9.1
*21	12.47	12.0	10.43	10.1	12.28	11.8	10.16	9.7
*11	12.53	12.2	10.60	10.4	12.80	12.5	10.77	10.5

Table 2. Percent of Ag-110m retained in irradiated compacts from AGR-2 Capsules 2, 3, 5, and 6. The asterisk (*) in each compact ID number corresponds to the capsule number.

Compact	% Retained			
	Capsule 2	Capsule 3	Capsule 5	Capsule 6
*43	5.7% (-0.7%, +2.9%)	114.1% (-1.4%, +1.4%)	63.0% (-2.3%, +2.4%)	58.5% (-4.5%, +5.2%)
*33	25.4% (-1.2%, +1.2%)	86.6% (-1.3%, +1.3%)	23.3% (-1.0%, +1.9%)	33.5% (-3.3%, +6.3%)
*23	30.1% (-1.1%, +1.2%)	84.7% (-1.4%, +1.4%)	17.0% (-1.3%, +2.3%)	20.6% (-2.5%, +7.2%)
*13	35.2% (-1.3%, +1.4%)	109.6% (-1.6%, +1.6%)	80.7% (-2.4%, +2.4%)	63.7% (-2.9%, +4.6%)
*42	0.9% (-0.4%, +4.8%)	115.6% (-1.1%, +1.1%)	50.6% (-1.6%, +4.2%)	66.5% (-3.0%, +3.1%)
*32	6.8% (-0.8%, +4.0%)	94.2% (-1.1%, +1.1%)	1.8% (-0.5%, +4.0%)	15.0% (-2.0%, +6.8%)
*22	12.7% (-1.0%, +3.0%)	93.7% (-1.1%, +1.1%)	21.8% (-1.1%, +3.0%)	18.3% (-2.1%, +6.3%)
*12	19.8% (-1.1%, +3.4%)	98.2% (-1.0%, +1.7%)	62.8% (-2.0%, +3.7%)	48.0% (-2.3%, +5.9%)
*41	0.8% (-0.3%, +4.7%)	117.4% (-1.1%, +1.1%)	85.7% (-2.1%, +2.1%)	69.1% (-3.7%, +3.8%)
*31	16.0% (-1.1%, +1.7%)	94.1% (-1.0%, +1.0%)	21.0% (-0.9%, +1.0%)	14.1% (-1.6%, +8.5%)
*21	12.2% (-1.1%, +3.1%)	94.4% (-1.1%, +1.1%)	33.7% (-0.9%, +0.9%)	4.7% (-1.1%, +8.9%)
*11	20.1% (-0.9%, +4.1%)	114.6% (-1.1%, +1.1%)	93.7% (-1.8%, +1.8%)	46.0% (-2.2%, +5.9%)

In addition to burnup, another key metric for fuel performance and subsequent PIE is the percentage of Ag-110m retained in each compact. This percentage is shown in Table 2 for the four U.S. capsules from AGR-2. The values were calculated by dividing the measured Ag-110m inventory to the predicted inventory. The retention percentage is a ratio of experimentally measured activity divided by the calculated activity for a compact and the calculated activity may be under-predicted in some compacts resulting in a retention percentage greater than 100%. The time-average-volume-average temperatures for AGR-2 have been calculated for each compact in AGR-2¹³, and it is possible to relate Ag-110m release to these temperatures to some degree. However, experience from AGR-1 and AGR-2 has shown that the release of Ag-110m is related to temperature, the time a compact stays at elevated temperatures, and the amount of Ag-110m present in the compact while it is at temperature. The retention is then a multi-physics problem that is dependent on several time varying phenomena such as temperature, neutron flux, neutron fluence, and radionuclide inventory. Due to a large number of spectra that had no detectable Ag-110m in the initial scans, the Level 2 and 3 compacts from Capsule 5 were rescanned with longer scan times to better evaluate the percentage retained.

The uncertainty on the percentage retained is not symmetric. The lower uncertainty band is equal to the

square root of the sum of the squares of the activity uncertainty for each scan with a detectable amount of Ag-110m present for all scans that correspond to a particular compact. The upper uncertainty band is also equal to the square root of the sum of the squares of the activity uncertainty for each scan with a detectable amount of Ag-110m present for all scans that correspond to a particular compact; however, in the case of a scan with an undetectable amount of Ag-110m, the activity uncertainty is replaced with the minimum detectable activity. Thus, the upper uncertainty band is always larger than the lower uncertainty band. Typically, the Stack 1 and Stack 2 compacts that were irradiated at the same level (e.g., Compacts 6-4-1 and 6-4-2) are similar in percentage of Ag-110m retained, but this trend does not always hold true (e.g., Compacts 5-3-2 and 5-3-1). More investigation into why compacts that had very similar irradiation conditions, but very different Ag retention, is warranted. Destructive examination¹⁴ and safety testing¹⁵ are in progress on the AGR-2 compacts, and will provide additional information on silver retention. In addition, the silver retention of the fuel compacts has been compared to fuel performance models.¹⁶

III.B. Holder Gamma Spectrometry Results

The off-axis scans for Capsules 2 and 3 for a selection of different fission products are shown in Fig. 5 as examples. The scans are plotted so that the left side scan that contains Stack 2 and half of Stack 3 is shown on the left of the plot and the right side scan containing Stack 1 and half of Stack 3 is shown on the right of the plot. Holder 2 axial scans detected the presence of Ag-110m, Eu-154, Cs-134, and Cs-137 in the graphite. Most notably, the scans contained a consistently strong Cs-134 and Cs-137 signal isolated about Level 2 (location of the second level of compacts from the bottom, see Fig. 2). Tomographic scans were able to identify that the cesium activity was predominantly located adjacent to the original location of Compact 2-2-3 in the graphite holder. The tomogram for Cs-134 distribution at Level 2 of Holder 2 is shown in Fig. 6. This suggests that this compact may have contained one or more particles that experienced SiC layer failure, releasing relatively high fractions of cesium isotopes. Subsequent PIE showed that there was a TRISO particle with a defective SiC layer in AGR-2 Compact 2-2-3.¹⁴ Capsule 2 was run at relatively high temperatures (time-average, volume-average temperature for all compacts in the capsule was 1252°C),¹³ which likely contributed to the release of Eu-154 into the holder. This was not observed in any of the AGR-1 capsules, none of which were operated at such high temperatures for long durations. Fig. 5 and Fig. 7 indicate that Eu-154 released locally to the graphite but did not migrate significantly.

The only significant fission product present in Holder 3 was Ag-110m, and it was only present at Levels 2 and 3 of the holder (see Fig. 5). This is consistent with Capsule 3

compact measurements, which indicated elevated release of Ag-110m from the Level 2 and 3 compacts for all three stacks. Tomograms of Holder 3 Level 2 indicate Ag-110m is distributed locally near the compacts away from the hot center of the graphite holders in the cool regions of the graphite near each stack¹³ as seen in Fig. 8. Holder 3 contained no detectable Cs-134 or Cs-137, indicating TRISO particles with a failed SiC layer were unlikely to be present in this capsule.

Off-axis scans of Capsule 5 indicated large amounts of Ag-110m at Levels 2 and 3 in the holder (Fig. 9). Small amounts of Cs-137 and Cs-134 were also located at Levels 2 and 3 of the holder. No other fission products were detected in the graphite holder. The Cs spikes centered around Levels 2 and 3 indicate that there may be one or more TRISO particles with a failed SiC layer in at least one of the compacts at these levels. Gamma tomography was performed on both of these levels. The Cs-134 tomograms from this analysis are shown in Fig. 10. These images indicate that the Cs activity is adjacent to the compact in Stack 3 at both levels, suggesting the possibility that Compact 5-2-3 and/or Compact 5-3-3 may contain TRISO particles with failed SiC layers.

Off-axis scans of AGR-2 Holder 6 were also performed. In the case of Holder 6, elevated levels of Ag-110m were detected at the axial ends of the holder (results not shown). This is similar to what was seen in Holder 1 and Holder 6 in AGR-1. Radioactive Cs isotopes were found at very low levels at Levels 2 and 3 of the holder. Tomographic scans that split Levels 2 and 3 of Holder 6 again indicate Cs release from Stack 3 (see Fig. 11). The data suggest that Compact 6-2-3 and/or Compact 6-3-3 may contain particles with failed SiC layers.

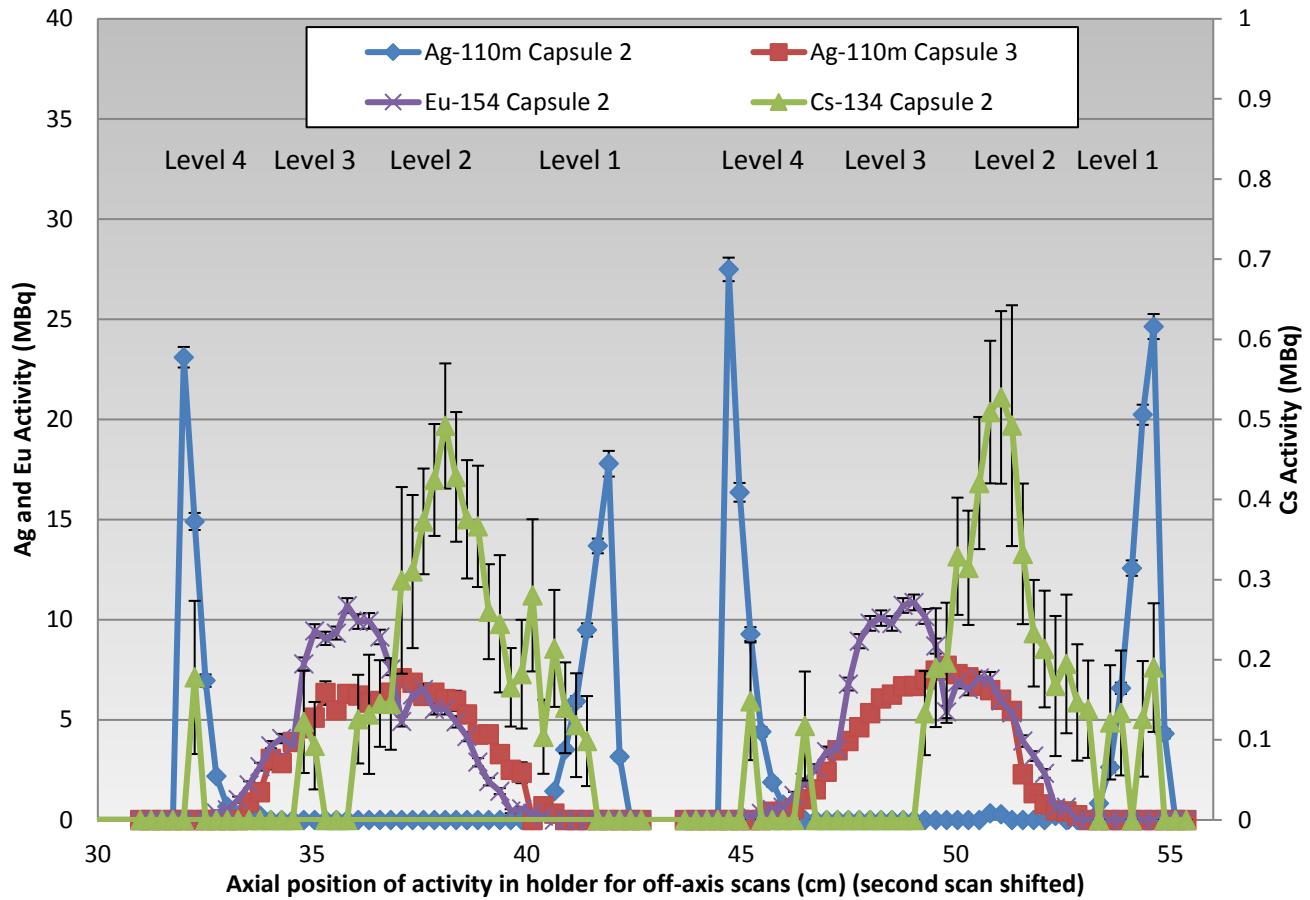


Fig. 5. Off-axis distribution of different fission products detected in left and right side axial scans of Holders 2 and 3.

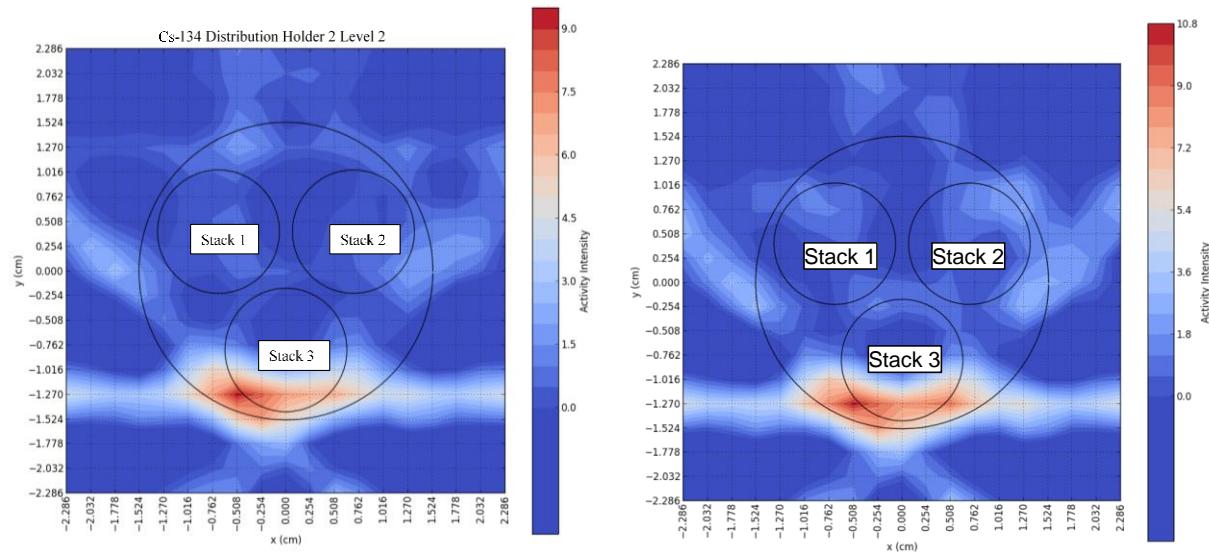


Fig. 6. Cs-134 distribution in Holder 2 Level 2 (left) and Cs-137 distribution in Holder 2 Level 2 (right).

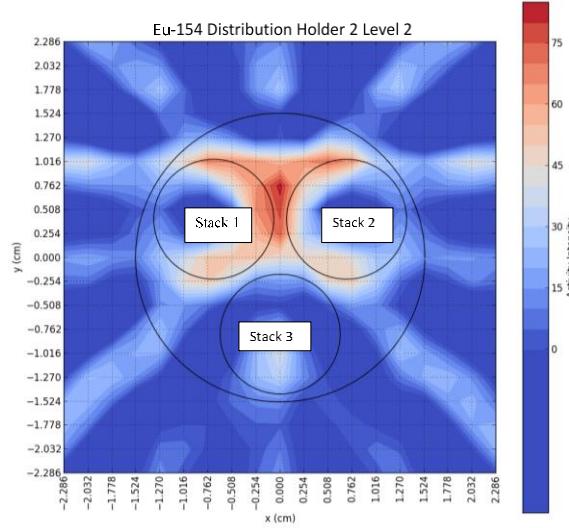


Fig. 7. Eu-154 distribution in Holder 2 Level 2.

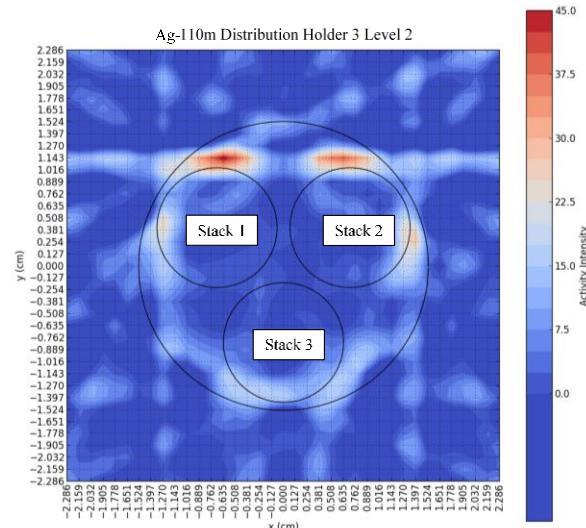


Fig. 8. Ag-110m distribution in Holder 3 Level 2.

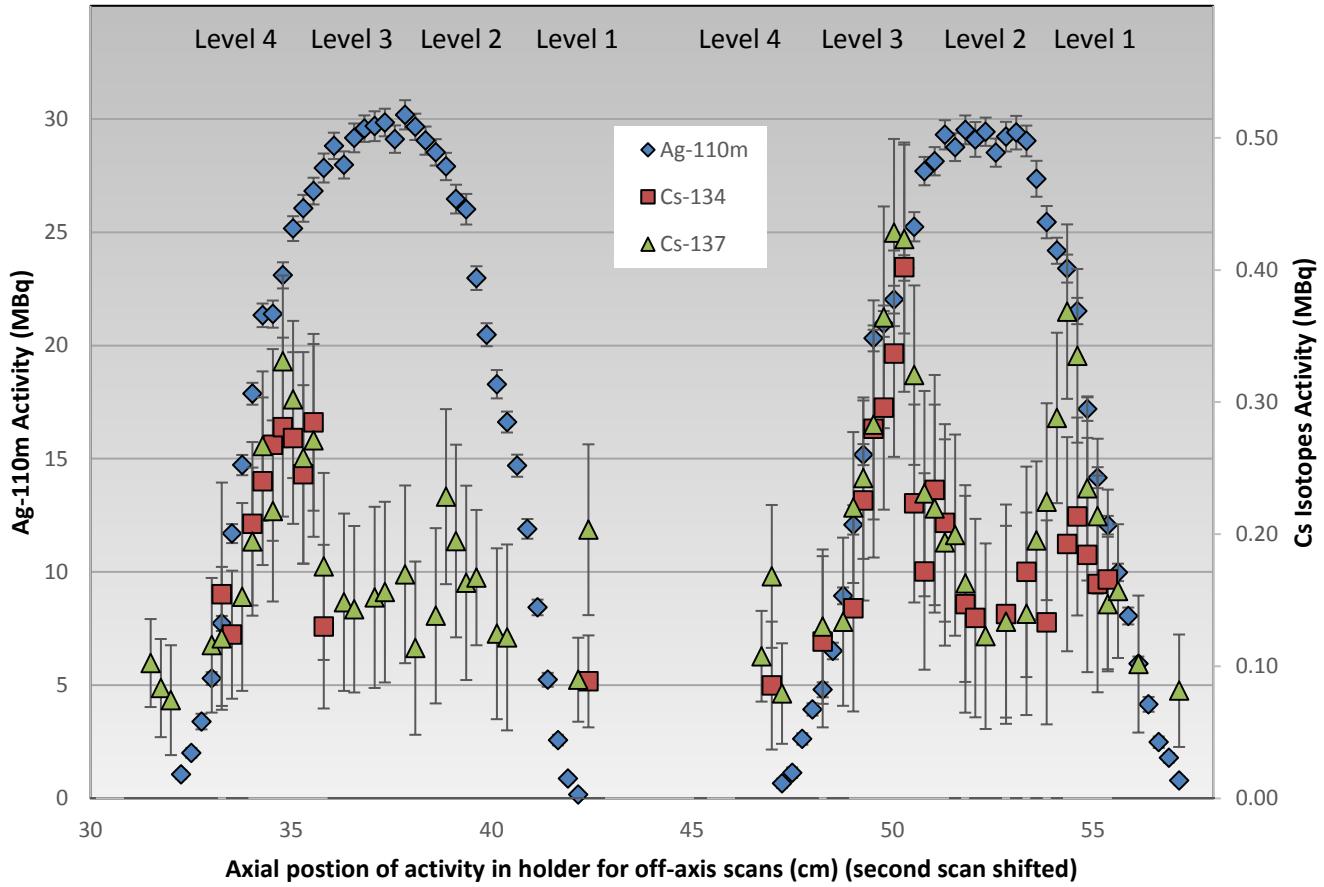


Fig. 9. Off-axis distribution of different fission products detected in left and right side axial scans of Holders 5

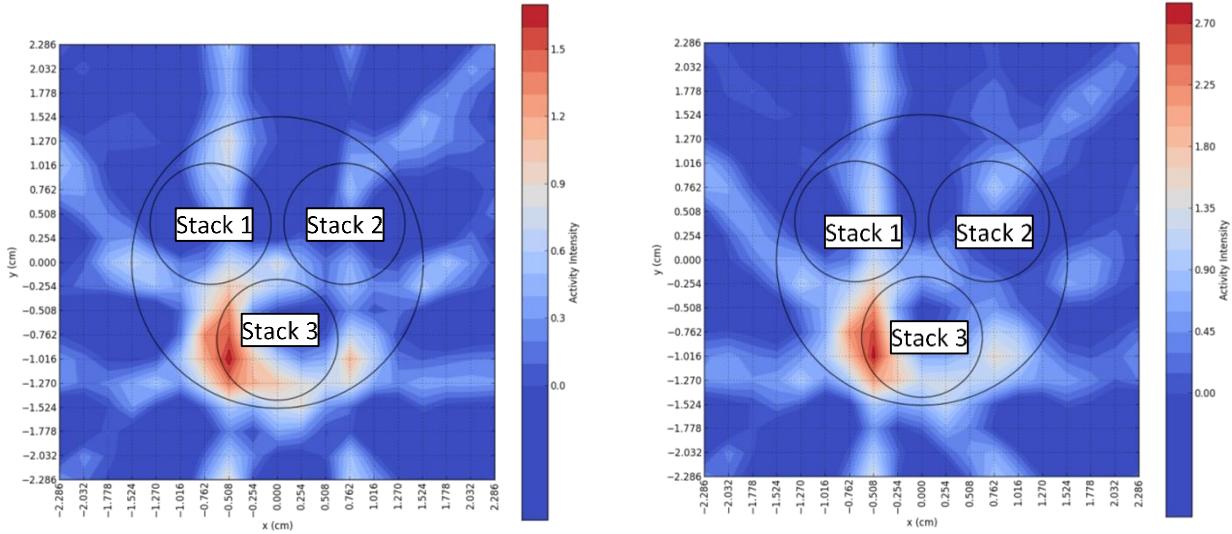


Fig. 10. Tomographic Cs-134 gamma scans from Levels 2 (left) and 3 (right) of AGR-2 Holder 5.

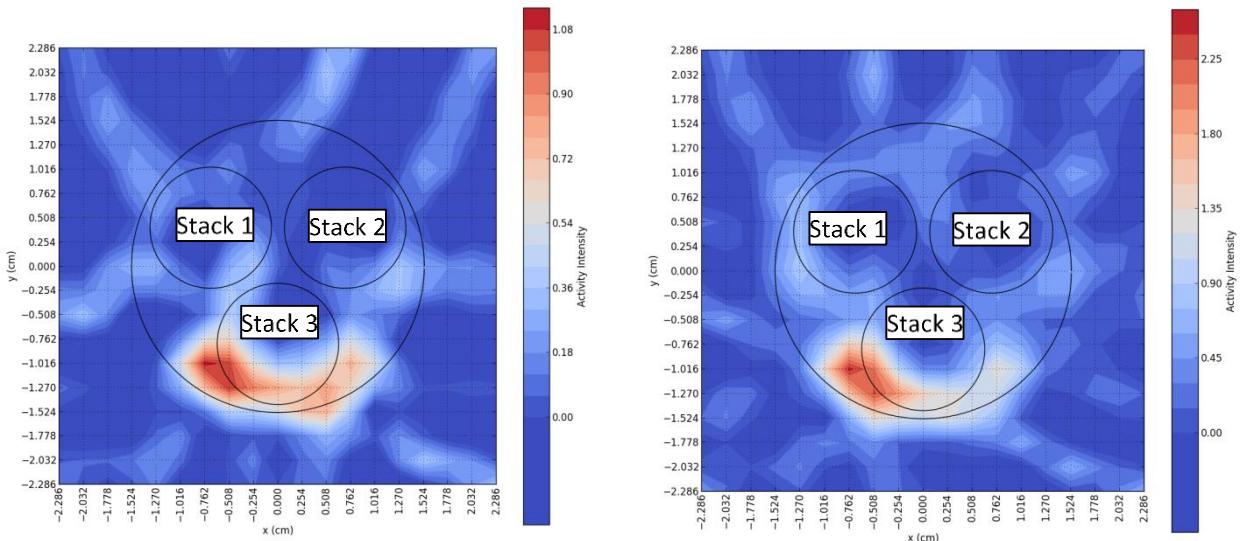


Fig. 11. Tomographic Cs-134 gamma scans from Levels 2 (left) and 3 (right) of AGR-2 Holder 6.

III.C. Silver in Additional Capsule Components

In addition to scanning with PGS, capsule components were sent to the hot cells at the Analytical Laboratory at the INL Materials and Fuels Complex for quantitative gamma spectrometry analysis of fission product inventories. The graphite spacers were gamma counted on an out-of-cell HPGe detector. The capsule shells were leached, and the leachate for each capsule shell was analyzed for gamma emitting fission products and Sr-90. Because of experience in AGR-1, negligible fission product activity was found in gas exit lines, the exit lines were not evaluated in the AGR-2 analysis. The amount of decay-corrected Ag-110m detected in the compacts and in each capsule component was combined and compared to the expected value from

simulations to create a total Ag-110m mass balance for each capsule, shown in Fig. 12. The apparent over-recovery of Ag-110m in Capsule 5 and Capsule 3 are likely due to under-prediction of the amount of Ag-110m in those capsules. A portion of the under-recovery of Ag-110m in Capsule 6 and Capsule 2 may be due to an over-prediction of the burnup in these capsules (especially Capsule 6, as shown in Fig. 4) in addition to potential losses during chemical processing of the capsule components. This is especially true when large portions of the Ag-110m are found on the capsule hardware or capsule shells, as this measurement has high experimental uncertainties. Overall this agreement is acceptable and similar to what was seen in AGR-1. In AGR-1, the Ag-110m inventory balance for

capsules with large amounts of Ag-110m on the capsule hardware was lower than when more was retained in the holders and the compacts.⁹

IV. CONCLUSIONS

Gamma spectrometry scans of the US capsules from AGR-2 have been completed. The gamma-emitting fission product inventory of each compact has been determined and compared to simulations of the irradiation test. The burnup of each compact from AGR-2 has been estimated using gamma spectrometry on both a whole-compact level by the direct Cs-137 inventory and axially along the compact length by the Cs-134 to Cs-137 ratio. The experimentally-measured burnup agrees well with the predicted burnup.

The retention of Ag-110m in the compacts was also measured. The release of Ag-110m is a complex multi-physics phenomenon and this measurement will provide additional information to help evaluate the modeling of Ag-110m release. The retained fraction of Ag-110m from these measurements also helps to guide which compacts are chosen for further PIE such as destructive exams and safety testing that can provide additional data on the mechanisms of Ag-110m release.

Fission product distribution and inventory in the graphite holders was also examined. The axial distribution

of fission products guides additional scanning that can be used to identify the location of TRISO particles that are potentially defective or failed. The inventory of Ag-110m in other capsule components was also evaluated and compared against the predicted inventory. These results show that a majority of the Ag-110m generated in the test has been captured.

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V. REFERENCES

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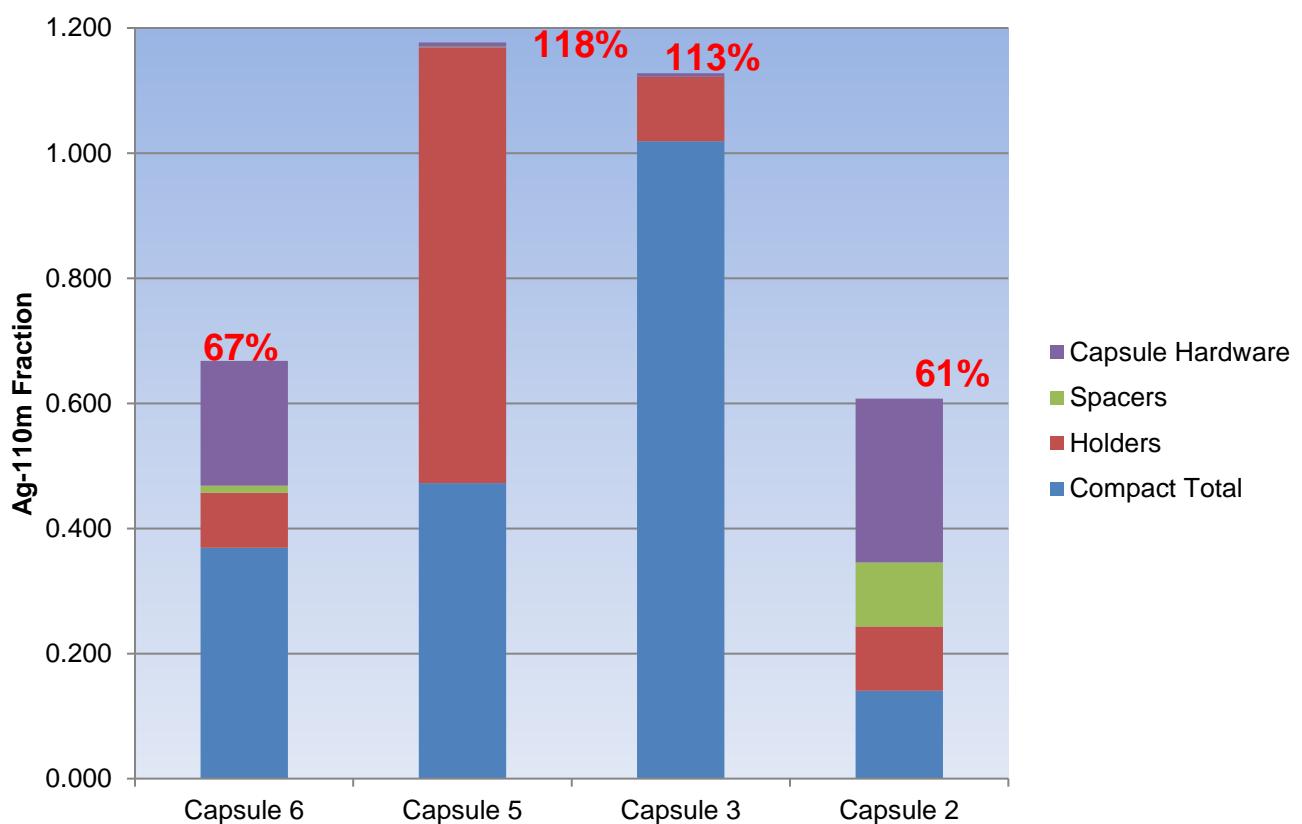


Fig. 12. Inventory of Ag-110m in different capsule components compared to the predicted inventory for each capsule

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