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Abstract – In the High Temperature Gas-Cooled Reactor (HTGR) the TRISO particle fuel serves as the primary fission product containment. However the large number of TRISO particles present in proposed HTGRs dictates that there will be a small fraction ($\sim 10^{-4}$ to $10^{-5}$) of as-manufactured defects and in-pile particle failures that will lead to some fission product release. The matrix material surrounding the TRISO particles in fuel compacts and the structural graphite holding the TRISO particles in place can also serve as sinks for containing any released fission products. However data on the migration of solid fission products through these materials is lacking. One of the primary goals of the AGR-3/4 experiment is to study fission product migration from intentionally failed TRISO particles in prototypic HTGR components such as structural graphite and compact matrix material. In this work, the potential for a Gamma Emission Computed Tomography (GECT) technique to non-destructively examine the fission product distribution in AGR-3/4 components and other irradiation experiments is explored. Specifically, the feasibility of using the Idaho National Laboratory (INL) Hot Fuels Examination Facility (HFEF) Precision Gamma Scanner (PGS) system for this GECT application was considered. Previous experience utilizing similar techniques, the expected activities in AGR-3/4 rings, and analysis of this work indicate using GECT to evaluate AGR-3/4 will be feasible. The GECT technique was also applied to other irradiated nuclear fuel systems currently available in the HFEF hot cell, including oxide fuel pins, metallic fuel pins, and monolithic plate fuel. Results indicate GECT with the HFEF PGS is effective.

I. INTRODUCTION

In the High Temperature Gas-Cooled Reactor (HTGR) the TRISO particle fuel serves as the primary fission product containment. HTGR reactor design specifications generally allow for a small fraction ($\sim 10^{-4}$ to $10^{-5}$) of as-manufactured defects and in-pile particle failures that will lead to some fission product release. The matrix material surrounding the TRISO particles in fuel compacts and the structural graphite holding the TRISO particles in place can also serve as sinks for any released fission products. However data on the migration of solid fission products through these materials is lacking. One of the primary goals of the AGR-3/4 experiment in the US is to study fission product migration from failed TRISO particles in prototypic HTGR components such as structural graphite and compact matrix material. This data is critical for determining the radiological source term released from the reactor core during accidents.

In this work, the potential for a Gamma Emission Computed Tomography (GECT) technique to non-destructively examine the fission product distribution in AGR-3/4 components and other irradiation experiments is explored. Specifically the feasibility of using the Idaho National Laboratory (INL) Hot Fuels Examination Facility (HFEF) Precision Gamma Scanner (PGS) system for this GECT application is considered. To test the feasibility, the response of the PGS system to idealized fission product distributions has been
modeled using Monte Carlo radiation transport simulations. The GECT technique was also applied to other irradiated nuclear fuel systems that were currently available in the HFEF hot cell to help develop the technique.

The PGS system consists of a collimator that penetrates the HFEF hot cell wall, a high purity germanium (HPGe) gamma spectrometry system with Compton suppression, and an in-cell stage used to position samples for examination. The hot cell side of the collimator consists of tungsten alloy blocks that can be adjusted to create a slit height varying from 0.254 to 0.00254 cm (0.1 inches to 0.001 inches). The width of the collimator opening is fixed at 2.22 cm (0.875 inches) creating a rectangular viewing area for the PGS system. An illustration of the PGS collimator is shown in Fig. 1. Gamma-rays entering the collimator travel through a thin aluminum window (right side of Fig. 1), pass through the initial tungsten collimator blocks (Aperture Insert and Upper Cam in Fig. 1), through a second tungsten alloy beam scraper, and then to the HPGe detector. The PGS collimator can be rotated from a horizontal slit orientation 90 degrees to a vertical slit orientation when needed for specific scanning procedures.

The use of a gamma tomography technique for the evaluation of the fission product distributions in AGR-3/4 was first considered after successfully using a similar but simpler technique in the examination of the AGR-1 TRISO fuel experiment. The distribution of Ag-110m and Cs-134 in AGR-1 components was examined, and specific fuel compacts containing particles with a failed SiC layer were identified with this method [1]. This was done by scanning the structural graphite fuel holders horizontally with the PGS collimator rotated into the vertical slit orientation followed by rotating the graphite holder 90 degrees and then scanning horizontally a second time. The Cs-134 activity in the graphite holders in the vicinity of compacts containing particles with failed SiC was small relative to the total activity present in an irradiated compact. For example, about 1.1 MBq (30 μCi) of Cs-134 was observed in an isolated area of the AGR-1 Capsule 5 graphite holder, which is equivalent to a ~5x10^-3 fraction of the entire inventory of an irradiated compact. The relatively simple implementation of the technique during the AGR-1 PIE was somewhat limited; while the resulting activity maps allowed specific compacts of interest to be identified, the resolution of the activity intensity was limited. Subsequent post irradiation analysis of the identified compacts revealed the presence of 3 particles (of about 8300 in the two compacts) with failed SiC layers [2].

The GECT technique utilizes the inverse Radon transformation to convert a set of collected gamma ray data into a tomograph. The gamma ray data set is collected in a regular geometric pattern and can be gamma-emitting isotope specific. This technique has been applied several times to examine the distribution of Cs-137, Eu-154, Ce-144 and other fission products in irradiated Light Water Reactor (LWR) fuel [3], [4], [5].

An illustration of an AGR-3/4 irradiation experiment capsule cross section is shown in Fig. 2. There were 12 capsules in AGR-3/4 each with 4 TRISO fuel compacts that each contained 20 designed-to-fail (DTF) TRISO particles in a line down the central axis of the fuel compacts [6]. These particles consist of a fuel kernel coated with a

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**Fig. 1.** Assembly drawing of PGS collimator
thin layer of pyrocarbon with relatively high anisotropy which is designed to fail during the irradiation [7]. Once they fail, these particles provide a known source of fission products that will diffuse into the capsule components.

The fuel compacts are surrounded by the Inner Ring fabricated of matrix material or graphite. The Inner Ring has an inner diameter of 1.247 cm and a nominal outer diameter of 2.377 cm. The Inner Ring is nested inside the graphite Outer Ring which is nested inside the Graphite Sink. The Outer Ring has a nominal inner diameter of 2.454 cm and an outer diameter that varies between 3.350 and 3.962 cm. This variation in diameter is used to achieve different temperatures in the experiment capsules during irradiation. The Graphite Sink is placed inside the capsule shell. The primary objective of the AGR-3/4 post-irradiation examination is to measure the distribution of fission products in the Inner and Outer Rings, and use this data to characterize the diffusion behavior of fission products in these materials. A more detailed description of the AGR-3/4 irradiation experiment can be found in Reference [6].

In this work GECT has been performed by collecting data sets that can be run through an inverse Radon transformation with filtered back projection. The input of the inverse Radon transformation consists of a $n \times m$ matrix of line integrals, which for this work are either simulation response or counts, created by the $m$ evenly spaced, ordered measurements for each $n$ angles of collected information. This data, along with the angles and spacing information, is used in the inverse Radon transformation to create a $m \times n$ Radon matrix that is representative of the intensity of emitted particles (for this case, gamma-ray production) over the scanned region. During the transformation the data is filtered to help reduce edge blurring in the features of the resulting tomographs. The $n \times m$ matrix collected in this work is the count rate of a specific isotope from each collected gamma-ray spectrum. The shape of the PGS aperture results in tomographs that are representative of the response from a 2.22 cm axial section of the item being scanned. The inverse radon transformation was carried out using the scikit-image package for SciPy [8], and the data input and output is handled using a simple Python script.

II. COMPUTATIONAL SIMULATION OF GECT APPLIED TO AGR-3/4 COMPONENTS

The response of the HFEF PGS system was modeled using a Monte Carlo photon transport code (MCNP) for several different source distributions that are expected in the inner and outer rings of the AGR-3/4 experiment. The results of these simulations and how they can be used to evaluate the effectiveness of the PGS as a GECT tool are discussed. A method of interpretation of the resulting tomographs to produce diffusion coefficient parameters is explored. The limits of detection for the PGS system and the implications for using PGS to scan the different AGR-3/4 components are evaluated.

II.A. MCNP Simulation of GECT on PGS

In previous work involving the PGS, a representative MCNP model was created of the PGS collimator and detector system that was shown to reasonably produce the response of the PGS system [9], [10]. This model was modified to represent gamma rays emitted from both inner and outer rings from AGR-3/4. To simulate the analysis of the AGR-3/4 rings, the rings and gamma sources (i.e. the distribution of initial starting positions for particles in the Monte Carlo simulation) were sequentially moved a set distance each step past the collimator opening as illustrated in the sketch in Fig. 3. The simulated system response for each step was recorded and compiled as input for the inverse Radon transformation. The simulated source distributions were azimuthally symmetric so it was not necessary to simulate the different angles in cases where the entire graphite ring was considered for scanning. Some simulations were performed where the ring was represented as sectioned in half or quartered axially. In these cases it was necessary to simulate all desired angles and record the different responses. Two different source distributions were used in the simulations: a source with a constant radial distribution and a source that decayed logarithmically approximately 2 orders of magnitude radially from the inner radius of the ring to the outer radius of the ring being simulated. The same source distributions were applied to the inner and the outer ring. These distributions are based on the predictions

![Fig. 2. Cross-section diagram of an AGR-3/4 irradiation capsule. Labels indicate the key experiment components.](image-url)
in the AGR-3/4 Irradiation Test Plan [11]. Depending on the temperature conditions of the capsule, the test plan predicts a fission product distribution varying from constant to dropping two orders of magnitude across the inner ring. The radial distributions that were used in the MCNP simulations are shown in Fig. 4.

The scanning for the simulations moved the source in a progressive pattern at 0.635 mm steps. The source was then rotated around the azimuthal axis of symmetry and again moved past the collimator. The simulated source was 661.62 keV Cs-137 gamma rays. The images in Figures 5 and 6 are intensity maps from the two different simulated sources of the inner ring of an AGR-3/4 capsule. Only 8 angles were used to create these tomographs. In the images, the outline of the inner ring is shown by the two black circles. There is significant presence of false intensity signals outside of the region where the original source was placed in the simulation, especially in Fig. 6, caused in part by the back projection filter used in the inverse Radon transformation and by the relatively low number of angles (compared to typical tomography applications) used to create the image. This false signal is often seen in image processing and is called “ghosting”. Limitations with collimator size and the Fourier nature of the analysis technique also make abrupt edge definition difficult. This causes the constant radial distribution in Fig. 5 to exhibit appreciable reduction near the edges. Unitless activity intensity data have been extracted by integrating the data around the axis to create an average radial distribution. This is shown in Fig. 7 for the constant distribution and in Fig. 8 for the decaying distribution. It should be noted that in Fig. 8 there is some activity outside of the bounds of the ring. This is caused by the application of the back projection filter and the resulting ghosting that accompanies the application of filtering when only a few angles are used. Additional scanning angles would eliminate this artifact in both the tomography (Fig. 6) and the integrated distribution (Fig. 8).

These distributions, once derived, must also be interpreted. In the case of the constant fission product distribution, there is little diffusion data that can be extracted. A constant distribution indicates the fission products were mobile enough and the temperature was high enough that an equilibrium distribution was achieved. In the case of the logarithmically decaying radial distribution, a portion of the data can be fit using linear least squares fitting assuming a simple model given in Equation (1)

\[ A(r) = A_0 e^{-Cr} \]  

where \( A \) is the activity intensity, \( r \) is the radius, and \( C \) is some constant for radial diffusion. The \( C \) value can be determined from the radial distribution data using least squares fitting. This is done in Fig. 9 and compared to the original slope of the radially decaying source distribution. Ideally, the two slopes would be identical. The current technique shows promise in terms of reproducing the slope of the original activity distribution, with the slope from simulation deviating from the original slope by 4.4%. Further development of the GECT technique with the PGS may lead to improvements in reproducibility; however, the PGS system will always have some inherent limitations.

The above example is illustrative of the ability of GECT data to be interpreted to recreate the data from a specific model. The physics and models that govern fission product distributions in AGR-3/4 are slightly more complex. The diffusion equation governs the concentration or activity profiles in the rings. As an approximation the diffusion in this experiment can be considered one dimensional in the radial direction, and the diffusion coefficients can be assumed to be governed by a simple Arrhenius relationship as shown in the following equations.
Fig. 5. GECT recreation of a constant radial source with azimuthal symmetry in an AGR-3/4 inner ring.

Fig. 6. GECT recreation of an azimuthally symmetric source that decreases in intensity logarithmically by two orders of magnitude across the thickness of an AGR-3/4 inner ring.
Where $C_i$ is the concentration of the fission product, $r$ is the radius of the ring, $D$ is the diffusion coefficient, $D_0$ is the pre-exponential factor, $E$ is the migration energy of the fission product, $k$ is Boltzmann's constant and $T$ is the temperature of the system. The conditions (e.g., temperature) of each capsule should be kept roughly constant over the course of the experiment. The distributions of each ring will be roughly a Bessel function of the first kind of the 0th order. The derived fission product distributions will need to be fit to the proper Bessel function and related back to a diffusion coefficient value for that temperature. The diffusion coefficient values for several rings of different temperatures will be needed to derive a functional relationship for the diffusion coefficient like the one shown in Equation (3).

II.B. Expected Activity in Rings and the Impact on GECT Feasibility

One of the inherent limitations in the PGS system is the limits of detection. These are driven in part by the system geometry (significant separation between specimen and detector due to the installation outside of a shielded hot cell) as well as the background radiation from within the hot cell. Based on experience from AGR-1, the PGS system can detect Cs-137 and Cs-134 down to about 37 kBq (1 μCi) in activity in the area viewed by the PGS collimator. Anything below this tends to be obscured by the inherent background. The minimum detectable activity of Ag-110m is also around 37 kBq (1 μCi). These minimum activities are based on examining the AGR-1 graphite holders which contained relatively small activities of gamma emitters and the live time for measurements was limited to 2 hours. Similar conditions are expected when examining AGR-3/4 diffusion rings, so it is reasonable to expect similar minimum detectable activities.

In evaluating the suitability of GECT using the PGS for the AGR-3/4 rings, it is advantageous to start with an estimate of the range of fission product activities that may be present in the specimens. Predicted inventories of fission products in the AGR-3/4 rings have been generated based on modeling, as described in Reference [11]. The activity concentration for Cs-137 and Ag-110m is

\[
\frac{\partial C}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( rD \frac{\partial C}{\partial r} \right) \tag{2}
\]

\[
D = D_0 \exp\left(-\frac{E}{kT}\right) \tag{3}
\]

Fig. 7. Integrated activity intensity across the ring thickness for the constant source distribution.

Fig. 8. Integrated activity intensity across the ring thickness for the decaying source distribution.

Fig. 9. Least squares fit to original source distribution and the radial distribution from simulation.
expected to vary several orders of magnitude with temperature and several orders of magnitude radially over the inner and outer rings. In the inner ring, modeling predicts the Cs-137 concentration to be at least 1 g/m² and Ag-110m concentration to be at least 0.001 g/m². By assuming some geometric simplifications, this minimum concentration can be converted to an approximate activity viewed by the PGS for each scan of an inner ring. The PGS will view approximately 2.4MBq (66 μCi) of Cs-137 and 133kBq (3.6 μCi) of Ag-110m. This Cs-137 concentration is promising for GECT applications since there would be adequate Cs-137 to scan the inner ring in a reasonable period of time even if the final inner ring concentration were much lower than predicted by the data in Reference [11]. The Ag-110m concentration as predicted is adequate for applying GECT to the inner ring, but it may be problematic to collect adequate counts at the edges of the ring, where there is less material and where actual concentrations may be lower than predicted by simulations. However, based on experience from AGR-1 graphite holder scanning [1], there will probably be large amounts of Ag-110m released from intact TRISO particles for detection and mapping that is not accounted for in the predictions from Reference [11], which only considers release from the designed-to-fail particles, which comprise about 1% of the particles in each compact.

The outer ring will be more challenging. At higher capsule temperatures, the concentration of Cs-137 should still be high enough to allow application of the GECT technique. The same is true for Ag-110m. However, in the lower temperature capsules (in which outer ring temperatures are expected to be in the range of 750 to 825°C), the concentration of both Cs and Ag will probably be too low to measure with GECT. The higher temperature capsules (in which outer ring temperatures are expected to be in the range of 900 to 1100°C) should have adequate activity concentrations for mapping.

The time required to complete a scan must also be considered. Scanning the outer diameter of the inner ring at 0.0635 cm (0.025-inch) increments requires 42 scans per angle (extra scans to insure edge capture are included) and at least 8 angles should be scanned to obtain good GECT images. At the expected activities, 30 minutes is predicted to be sufficient time to quantify the Cs and Ag fission product activities in each scan. This results in approximately 7 days of scanning to produce a single tomographic intensity map, assuming the detector dead time is negligible.

Axial scans of the rings similar to the two axial scans of the graphite fuel holders in AGR-1 [1] will be performed on all the inner and outer rings from AGR-3/4 prior to performing the more time-intensive GECT analysis. These scans will be used to roughly quantify the axial distribution of fission product activity in each ring and to identify specific levels of interest in the rings (levels of interest may be determined based on the relative activity at that level, or the amount of axial variation observed in the activity). The GECT technique will then be applied to specific levels of interest in selected rings. By focusing the analysis effort in this manner, the entire axial length of all 24 AGR-3/4 rings will not be characterized with GECT, reducing the overall time required for this experimental campaign. However, it may be desirable to investigate creating a more sophisticated timing software for the PGS system that would complete a scan either after a set time or after a certain number of counts were collected in a set of key peaks. Scanning the outer rings using the GECT approach will be more time intensive. With half hour scans, an eight angle scan would require approximately 11 days. However, the expected activity in the outer ring may be too low to use half hour scans necessitating longer scanning times.

III. EXPERIMENTAL DEMONSTRATION OF THE GECT APPROACH

To demonstrate the GECT technique using experimental data, multiple angular scans (9 from 0 to 180º) of a metallic fuel plate were performed using the PGS. The selected fuel plate was from the RERTR-12 irradiation experiment [12]. A sketch of the plate and the scanning area is shown in Fig. 10. The dimensions of the fuel plate are 10.147 cm long by 2.540 cm wide by 0.140 cm thick (3.995 x 1.000 x 0.055 inches). The uranium molybdenum alloy (U-Mo) fueled zone is nominally 8.255 cm long by 1.905 cm wide and 0.0254 cm thick. The plates are formed by using a hot isostatic press to bond the U-Mo foil between two aluminum plates. The plate was fitted into a fixture for gamma scanning that allowed the plate to hang in front of the detector so that the length of the plate was aligned with the vertical axis of the PGS collimator. The plate was scanned by rotating the PGS collimator to vertical collimator orientation, and the plate was initially aligned so that the face of the plate and the face of the collimator were orthogonal. The plate was initially scanned horizontally across its width with the collimator open to 0.254 cm in 0.0635 cm steps. Additional steps beyond the expected width of the plate were included to insure full coverage in the scans for a total of 53 measurements per angle. The plate was then rotated through 9 different angles between 0 and 180º and scanned over the same set of steps. This resulted in 477 spectra that were analyzed and processed through the inverse Radon transformation to produce tomographic images that represent the relative activity of different fission products in the fuel plate.
The resulting map of Cs-137 intensity is shown.

Fig. 10. Sketch of RERTR plate scanned by PGS

Fig. 11. GECT intensity map of Cs-137 in an RERTR plate coordinates are in cm.

The resulting map of Cs-137 intensity is shown
in Fig. 11. In this scan the plate was hanging vertically out of the plane of the page so that the map is a cross section of the plate. The foil in the plate is 1.905 cm wide (Fig. 10). This corresponds well with the length of high Cs-137 activity along the y-axis of Fig. 11. The thickness of the foil is only 0.0254 cm. However because the step size (0.0635 cm) was smaller than the collimator opening (0.254 cm) some smearing of the foil has occurred spreading the thickness of the foil over 4 different scans in the GECT image. A smaller collimator opening would help alleviate this artifact and is feasible in this case where the count rate is very high. The peaks in Cs-137 activity points A and B in Fig. 11 are real and caused by additional material that increased neutron reflection on either side of the plate. The difference in Cs-137 peaking intensity may be due to power tilting in the ATR core. RERTR 12 was irradiated in the south large B position in ATR (position B11). The power in this position is an average between the southeast and southwest lobes of ATR which are typically run with a power difference of 3±1 MW. This difference could lead to the higher Cs-137 activity from increased fission at point A in Fig. 11.

In addition to the plate-type fuel, fuel with cylindrical geometry was also scanned with the HFEF PGS for GECT analysis. A fuel pin from the AFC-2E irradiation experiment [13] was examined. A representative cross section of an AFC-2E
irradiation experiment is shown in Fig. 12. The scanned AFC-2E fuel pin consisted of 70% U, 20% Pu, and 10% Zr metal alloy sodium bonded to HT-9 cladding with an inner diameter of 0.493 cm and an outer diameter of 0.584 cm. The fuel and cladding were encapsulated in a helium bonded 316 stainless steel outer capsule (outer diameter 0.899 cm) [13]. The GECT scans demonstrated here were performed while the fuel pin was still in the outer capsule. The capsule was scanned in 0.0635 cm (0.025 inch) increments, the collimator slit width was set to 0.0635 cm (0.025 inch), the live time for each spectrum was 60 s and nine angles were captured in 22.5° increments from 0° to 180°. Tomographs revealing the distribution of Cs-137 and Ru-106 in AFC-2E rodlet 2 are shown in Fig. 13 and Fig. 14. The black circles in the figures from innermost out represent the cladding inner diameter, the cladding outer diameter/inner capsule diameter interface, and the outer diameter of the capsule. The results show remarkably different behavior between the two fission products. Cesium is known to be mobile in metallic fuel and appears to have concentrated to one side of the fuel pin (to the right in Fig. 13). This may indicate that the fuel pin was cooler on that side during irradiation (azimuthal orientation of the pin within the reactor was not maintained during removal and transport to the PIE facility). Ruthenium is known to be relatively immobile in metallic fuel and that appears to be the case in AFC-2E rodlet 2 as shown by much more constant distribution shown in Fig. 14. Data extracted from the same set of gamma-ray spectra can be used to create tomographs that show steel activation products in the capsule region and the rodlet cladding region as expected. Tomographs of other fission products (Ce-144, Zr-95, Nb-95, etc.) have also been extracted from this data but are not shown in this paper.

The results from the two fuel types demonstrate the feasibility of the GECT approach using the PGS system on irradiated fuel, and offer promise that the GECT can be applied to the AGR-3/4 diffusion rings to quantify fission product distributions. At this time the intensity values in the tomographs are only relative quantities. The magnitude of the “Activity Intensity” in the tomographs is related to the magnitude of the activities used as input to the

![Fig. 14. Ru-106 activity distribution in an AFC-2E rodlet coordinates are in cm.](image-url)
inverse Radon transformation. Additional development is necessary to fully understand if the values from the tomographs can be scaled to quantitative activity concentrations (e.g. Bq/cm$^2$) or if the data from tomographs can only be used to identify relative trends.

IV. CONCLUSIONS

Preliminary simulations and experiments of the HFEF PGS system demonstrate that it will be feasible to analyze the distribution of fission products in the AGR-3/4 diffusion rings using the GECT technique. Simulated system response has demonstrated that GECT tomographs can be analyzed to produce radial fission product distributions that can be used to help determine the diffusion coefficient of different fission products in graphite and compact matrix material. The expected activity in the AGR-3/4 rings in certain capsules is sufficient to allow the PGS to collect the necessary number of gamma-ray spectra in a reasonable time frame. Lessons learned from applying GECT with PGS to other fuel systems will help to further optimize the technique for AGR-3/4 application. Further investigation is needed to determine if it is possible to convert the GECT output into quantitative values versus solely trending data. Plans also exist to physically sample AGR-3/4 rings in order to determine the distribution of Sr-90 and other non-gamma-emitting fission products in the rings. Results from these tests will also help to benchmark GECT results.

REFERENCES


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