Comparison of fission product release predictions using PARFUME with results from the AGR-1 irradiation experiment

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Abstract – The PARFUME (PARticle FUel ModEl) code was used to predict fission product release from tristructural isotropic (TRISO) coated fuel particles and compacts during the first irradiation experiment (AGR-1) of the Advanced Gas Reactor Fuel Development and Qualification program. The PARFUME model for the AGR-1 experiment used the fuel compact volume average temperature for each of the 620 days of irradiation to calculate the release of fission products silver, cesium, and strontium from a representative particle for a select number of AGR-1 compacts. Post-irradiation examination (PIE) measurements provided data on release of fission products from fuel compacts and fuel particles, and retention of fission products in the compacts outside of the silicon carbide (SiC) layer. PARFUME-predicted fractional release of these fission products was determined and compared to the PIE measurements. Results show an overall over-prediction of the fractional release of cesium by PARFUME. For particles with failed SiC layers, the over-prediction is by a factor of about two, corresponding to an over-estimation of the diffusivity in uranium oxycarbide (UCO) by a factor of about 100. For intact particles, whose release is much lower, the over-prediction is by an average of about an order of magnitude, which could additionally be attributed to an over-estimated diffusivity in SiC by about 30%. The release of strontium from intact particles is also over-estimated by PARFUME, which also points towards an over-estimated diffusivity of strontium in either SiC or UCO, or possibly both. The measured strontium fractional release from intact particles varied considerably from compact to compact, making it difficult to assess the effective over-estimation of the diffusivities. Furthermore, the release of strontium from particles with failed SiC is difficult to observe experimentally due to the release from intact particles, preventing any conclusions to be made on the accuracy or validity of the PARFUME predictions and the modeled diffusivity of strontium in UCO. In the case of silver, the comparisons between PARFUME and PIE are better than for cesium and strontium. They show a trend of over-prediction at low burnup and under-prediction at high burnup. PARFUME has limitations in the modeling of the temporal and spatial distributions of the temperature and burnup across the compacts, which affects the accuracy of its predictions. Nevertheless, the comparisons lie in the same order of magnitude.

I. INTRODUCTION

Post-irradiation examination measurements were compared to model predictions of silver (Ag), cesium (Cs), and strontium (Sr) release from selected tristructural isotropic (TRISO) fuel particles and compacts during the first irradiation test of the Advanced Gas Reactor (AGR) program (AGR-1). The modeling was performed using the particle fuel model computer code PARFUME developed at
Idaho National Laboratory (INL). PARFUME is an advanced gas-cooled reactor fuel performance modeling and analysis code ([1]). It has been developed as an integrated mechanistic code that evaluates the thermal, mechanical, and physico-chemical behavior of fuel particles during irradiation to determine the failure probability of a population of fuel particles given the particle-to-particle statistical variations in physical dimensions and material properties that arise from the fuel fabrication process, accounting for all viable mechanisms that can lead to particle failure.

The code also determines the diffusion of fission products from the fuel through the particle coating layers, and through the fuel matrix to the coolant boundary. The subsequent release of fission products is calculated at the compact level (release of fission products from the compact) but it can also be assessed at the particle level by adjusting the diffusivity in the fuel matrix to very high values. Furthermore, the diffusivity of each layer can be individually set to a high value (typically $10^{-6}$ m$^2$/s) to simulate a failed layer with no capability of fission product retention. In this study, in order to more accurately reproduce the number of particles with non-retentive coating layers potentially observed in PIE, or lack thereof, it was decided to model intact particles and particles with failed layers separately. The resulting respective release fractions were then combined accordingly in each compact to compare to the measured release data.

During the AGR-1 irradiation campaign, the fuel kernel produced and released some fission products, which migrated through the successive layers of the TRISO-coated particle and through the compact matrix. The release of these fission products was measured in PIE and modeled with PARFUME. PARFUME calculates the release fraction as the ratio of the number of atoms released from the compact to the amount produced in the compact fuel kernels and through uranium contamination. The silver, cesium, and strontium release fractions determined by both PIE and PARFUME for selected compacts or TRISO-coated particles were then compared.

II. AGR-1 IRRADIATION AND POST-IRRADIATION EXAMINATION

The Department of Energy (DOE) Advanced Gas Reactor Fuel Development and Qualification Program was established to qualify TRISO fuel for use in High Temperature Gas Reactors (HTGRs). The primary goal of the program is to provide a baseline fuel qualification data set in support of the licensing and operation of an HTGR ([2]).

AGR-1 is the first of these irradiation tests. Irradiation began in the Advanced Test Reactor (ATR) at INL in December of 2006 and ended in November 2009. AGR-1 was intended to serve as a shakedown test of a multi-capsule design to be used in subsequent irradiations, and to test early variants of the fuel produced under the DOE AGR program ([3]). A total of 72 compacts were irradiated in the AGR-1 experiment in six different capsules. The experiment completed 620 effective full power days (EFPD) in the reactor and achieved calculated peak burnup of 19.6% fissions per initial heavy metal atom (FIMA) and fast neutron fluence of $4.30 \times 10^{25}$ n/m$^2$ (E > 0.18 MeV) ([4]).

At completion of the irradiation, PIE and high temperature safety testing were conducted. The objectives of the AGR-1 PIE and associated safety testing are to: assess the overall performance of the test train and components and provide data to verify the test train thermal analyses, verify the particle coating integrity and evaluate the fission product retention of the fuel during normal irradiation and during high temperature post-irradiation safety tests, and characterize the fuel compacts and individual particles to assess the condition of the matrix material, kernels, and coatings ([5]). Comparison between PIE measurements and model predictions focused on silver, cesium, and strontium.

The results of the AGR-1 PIE are discussed in detail in a separate paper in these proceedings [6]. As described in Ref. [6], information on fission product release was obtained through a number of different measurements. The primary means of assessing the level of fission product release was through direct analysis of the released inventory, which included the inventory released from particles but retained in the compacts outside of the SiC layer and the inventory released from the compacts and measured on the irradiation capsule components. Silver represents a special case, in which the level of release can be sufficiently high (i.e., in excess of several percent) that the fraction retained can be estimated by measuring the remaining inventory in the fuel. In all cases, the measured inventory was first decay-corrected to the end of the AGR-1 irradiation and then compared with the predicted inventory from as-run neutronics calculations of the AGR-1 experiment [7] to calculate corresponding release or retention fractions. The PIE data used for comparison with PARFUME predictions originate from four different measurement techniques ([6]):

- Gamma scanning of 56 irradiated AGR-1 compacts was used to determine their inventory of Ag-110m. The irradiation characteristics of these compacts are shown in Figure 1 (black dots) and detailed in Ref. [8].
- Deconsolidation-leach-burn-leach (DLBL) was used on nine compacts to determine the amount of Cs-137 and Sr-90 that was retained within the compact outside of the SiC layer following

...
irradiation. Table 1 lists the nine compacts that underwent DLBL.

- Individual particles from these nine deconsolidated compacts were gamma counted in order to quantify their Ag-110m inventory.
- The inventory of fission products on the AGR-1 capsule components was measured to determine the level of release from the fuel compacts during irradiation.

Fig. 1: Irradiation conditions of the AGR-1 gamma scanned compacts (TAVA stands for time-average volume average).

Table 1: Compacts used in DLBL and particle gamma counting measurements.

<table>
<thead>
<tr>
<th>Compact</th>
<th>Burnup (%FIMA)</th>
<th>Fast Fluence ($\times 10^{25}$ n/m$^2$, $E &gt; 0.18$ MeV)</th>
<th>TAVA (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6-3-2</td>
<td>11.43</td>
<td>2.38</td>
<td>1070</td>
</tr>
<tr>
<td>6-1-1</td>
<td>15.25</td>
<td>3.00</td>
<td>1111</td>
</tr>
<tr>
<td>5-3-1</td>
<td>16.93</td>
<td>3.60</td>
<td>1040</td>
</tr>
<tr>
<td>5-2-3</td>
<td>17.42</td>
<td>3.77</td>
<td>1059</td>
</tr>
<tr>
<td>5-2-1</td>
<td>17.37</td>
<td>3.71</td>
<td>1057</td>
</tr>
<tr>
<td>4-4-2</td>
<td>16.74</td>
<td>3.59</td>
<td>1024</td>
</tr>
<tr>
<td>4-1-1</td>
<td>19.38</td>
<td>4.13</td>
<td>1072</td>
</tr>
<tr>
<td>3-2-1</td>
<td>19.07</td>
<td>4.21</td>
<td>1051</td>
</tr>
<tr>
<td>1-3-1</td>
<td>16.14</td>
<td>3.22</td>
<td>1092</td>
</tr>
</tbody>
</table>

For cesium and strontium, the inventory of fission products released from the particles was taken to be the sum of the inventory found in the compact outside of the SiC layer (i.e., in the outer pyrolytic carbon (OPyC) and matrix, based on DLBL measurements) and the inventory found on outside of the compacts on the capsule components. The DLBL data necessarily includes the contribution of fission products generated by fission of uranium contamination in the coating layers and in the matrix, but since the uranium contamination for AGR-1 compacts was on average lower than $4 \times 10^{-7}$ for each fuel variant, any release significantly above this level was attributed to release from the particles. For silver, the retained inventory in particles and compacts was determined directly based on measurement of the inventory remaining in the fuel (compact gamma scanning and individual particle gamma counting). The release fraction was then determined by subtracting the retained fraction from 1.0.

III. PARFUME MODELING

III.A. AGR-1 Geometry and Fuel Characteristics

The AGR-1 test train contains six capsules arranged vertically. Each AGR-1 capsule contains 12 fuel compacts arranged in three vertical stacks with each stack containing four compacts, as seen in Figure 2. Each compact is uniquely identified in the format X-Y-Z, where X indicates the capsule, Y the axial level within the capsule, and Z the stack.

Fig. 2: Layout of an AGR-1 capsule. The cross-section view at the top of the figure shows the orientation of the three fuel stacks within the graphite fuel holder and the axial cutaway shows the four axial levels.

AGR-1 compacts are right cylinders nominally 25.1 mm in length and 12.4 mm in diameter. Each compact contains ~4,150 fuel particles uniformly dispersed in a matrix composed of a thermosetting carbonaceous material. Each particle consists of a UCO kernel (diameter ~350 μm) coated with layers of porous carbon buffer (~100 μm), inner pyrolytic carbon (IPyC, ~40 μm), silicon carbide (~35 μm), and outer pyrolytic carbon (~40 μm) layers. AGR-1 fuel comprises a baseline fuel and three fuel variants, with each variant fabricated by varying the deposition parameters for either the IPyC or SiC layer, resulting in minor differences in properties for that layer. Detailed fuel characteristics of the AGR-1 fuel can be found in Ref. [3].
III.B. Boundary and Initial Conditions

PARFUME is designed to evaluate fuel performance based on user inputs for fast neutron fluence and burnup with a corresponding set of thermal conditions. The neutronics and thermal conditions for all the compacts used for comparison to PIE measurements are based on results obtained from as-run neutronics calculations and as-run thermal analysis ([7, 9]). The calculations were computed prior to completion of PIE, and they are based on an earlier version of the as-run neutronics calculations ([10]). The difference between the two sets of calculations is on burnup values, and it does not exceed 1.2%. Therefore, there is no impact on the calculated results. All subsequent plots show the burnup values used in PIE analysis.

PARFUME assumes that all particles in a compact experience similar irradiation and thermal histories over the course of irradiation. Practically, PARFUME models one particle using the average burnup and fast neutron fluence and the volume-averaged temperature of the whole compact. The thermal history evolves on a daily basis. For each compact, the daily temperatures of all the calculation nodes are averaged, and PARFUME uses the resulting volume-averaged compact daily temperatures to set the thermal history of the modeled TRISO particle. The daily temperatures are set as boundary conditions at the outer edge of the OPyC. From the OPyC boundary temperature, PARFUME calculates the temperature profile between the OPyC and the kernel center. The daily temperatures were used in the modeling because the time-average volume-average (TAVA) temperatures were determined to not be a suitable metric to correlate fission product release to temperature ([8]).

III.C. Input Parameters

The input parameters needed to model the AGR-1 irradiation with PARFUME originate from the AGR-1 Irradiation Experiment Test Plan ([3]) for the fuel characteristics, particle geometry, compact characteristics, and material non-mechanical properties, and from a CEGA Corporation report ([11]) for the material mechanical properties. In addition, as-run neutronics calculations and as-run thermal analysis are used for the boundary conditions (see Section III.B). The diffusion coefficients used for fission product transport are derived from the International Atomic Energy Agency (IAEA) Technical Document 978 ([12]) and displayed in Table 2. The corresponding diffusivities can be calculated using these diffusion coefficients in the following Arrhenius-type equation:

\[
D = D_{0,1}e^{-\frac{Q_{0,1}}{RT}} + D_{0,2}e^{-\frac{Q_{0,2}}{RT}}
\]  
(Eq. 1)

where

\[
D_{0,i} = \text{pre-exponential factor (m}^2\text{/s)}
\]

\[
Q_{0,i} = \text{activation energy (kJ/mol)}
\]

\[
R = \text{gas constant (8.3142} \times 10^{-3} \text{kJ/mol/K)}
\]

\[
T = \text{temperature (K)}
\]

PARFUME calculates fission product release at the compact level ([1]). The release from the particles can be obtained by setting the diffusivity in the matrix to a high value (typically \(10^{-6} \text{ m}^2\text{/s}\)) to force fission product release from the matrix. In addition, release from particles with failed layers can also be obtained by setting the diffusivities in these layers to the same value of \(10^{-6} \text{ m}^2\text{/s}\). In these calculations, PARFUME models the fission product release from both intact particles and particles with both failed IPyC and SiC layers. The results are then compared to the measured release data.

Table 2: Diffusion coefficients used in PARFUME.

| Species | \(D_{i,1}\) (m
\[
\] \text{\textnormal{^2}}\text{/s)} | \(Q_{i,1}\) (kJ/mol) | PyC | SiC | Matrix graphite |
<table>
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<tbody>
<tr>
<td>Ag</td>
<td>(6.7 \times 10^{-4})</td>
<td>65*</td>
<td>5.3 \times 10^{-4})</td>
<td>154</td>
<td>3.6 \times 10^{-4})</td>
</tr>
<tr>
<td>Cs</td>
<td>(5.6 \times 10^{-4})</td>
<td>209*</td>
<td>6.3 \times 10^{-4})</td>
<td>222</td>
<td>5.5 \times 10^{-4})</td>
</tr>
<tr>
<td>Sr</td>
<td>(5.2 \times 10^{-4})</td>
<td>362*</td>
<td>5.2 \times 10^{-4})</td>
<td>362</td>
<td>1.6 \times 10^{-4})</td>
</tr>
</tbody>
</table>

Note: UO
\[
\] _2 values are used for the kernel; the buffer has a set diffusivity of \(10^{-4} \text{ m}^2\text{/s}\).

IV. SILVER RELEASE

Gamma scanning of 56 compacts was used to measure the retained Ag-110m at the compact level. In addition, gamma counting of particles from the deconsolidated compacts was used to estimate the average retention in the particles from each compact, as described in Ref. [6].

IV.A. Compact Gamma Scanning

The silver release fractions from PIE measurements ([6, 13]) and PARFUME predictions for the 56 selected compacts are displayed in Figure 3. The data points are plotted according to the AGR-1 capsule in which they were irradiated with the top of the experiment (Capsule 6) on the left and bottom (Capsule 1) on the right. For each capsule, the compacts are plotted from the left for Stack 3 to
Stack 1 and a decreasing axial level within each stack. Negative release indicate apparent bias in the measured PIE values.

From Figure 3, the following observations can be made:

- PARFUME under-predicts the PIE data in Capsules 1 and 6.
- PARFUME under-predicts the PIE data in Capsule 5 on average.
- PARFUME over-predicts the PIE data in Capsules 2 and 3.
- PARFUME mostly over-predicts the PIE data in Capsule 4.

Since the burnup of the AGR-1 fuel was higher in the middle capsules than it was at the ends, these observations indicate that PARFUME under-predicts PIE data in the lower burnup capsules (end capsules) and over-predicts PIE data in the higher burnup capsules (middle capsules).

**IV.B. Particle gamma counting**

Typically 40 to 120 particles were selected per compact. The silver retention fraction was determined for each particle by calculating the ratio of the measured Ag-110m inventory to the predicted inventory. The average particle retention fraction for each compact was then calculated by averaging the individual particle values, as described in Ref. [6]. The average silver release fraction per particle was then compared to the values calculated by PARFUME.

PIE results ([6]) and PARFUME results are displayed in Figure 4. Similar to the compact gamma scanning comparisons, PARFUME over-predicts release in Capsules 3 and 4, and it under-predicts release in Capsules 1, 5 and 6. PIE data for Compacts 1-3-1, 5-2-1, 5-2-3, 5-3-1, and 6-3-2 show a range that account for uncertainty in the average particle retention fraction due to several gamma-counted particles from each compact exhibiting Ag-110m inventory below the detection limit (see Ref. [6] for more details).

**IV.C. Silver Matrix Content**

Sections IV.A and IV.B showed reasonable agreement between PIE and PARFUME for silver release from compacts or particles. In the case of PARFUME, the calculated release from particles or from their host compact is essentially identical because of the high diffusivity assumed for silver in the matrix material (see Table 2 of Section III.C). Direct PIE measurements performed by DLBL on the compacts in Table 1 showed that the amount of silver outside the SiC layer (referred to here as “matrix content”, even though the experimental technique cannot distinguish between inventory in the OPyC or in the matrix) is generally limited to a few percents of the silver inventory or less, with the exception of Compact 6-1-1, whose silver matrix content reaches 12% ([6]). On the other hand, results from the AGR-1 safety testing experiments ([14]) showed on a few occasions a high release of silver, up to ~30%, in the first hours of the tests. Based on the nature of this prompt release, it is believed that it comes primarily from silver that was released from particles during the irradiation but retained within the compacts outside of the SiC layer, and subsequently rapidly released when the compacts were heated to 1600°C [14]. To a first approximation then, these rapid silver releases are taken as a measure of silver outside of the SiC layer in the compacts, comparable to the DLBL measurements.

The silver matrix content was assessed for 17 compacts (18). These are the compacts for which data on both Ag-110m in the compact matrix (either from DLBL measurement or inferred from safety test data) and total compact Ag-110m inventory (from compact gamma scanning) were available. Hypothetically, subtracting the measured inventory in the matrix from the total compact inventory gives an assessment of the average fractional release from the particles, which is compared to PARFUME in Figure 5. In Figure 5, the data labeled “PIE” is based
solely on the total compact inventory data (from gamma scanning) and is identical to that presented in Figure 3 for these specific compacts. The data labeled “Corrected PIE” has been determined as described in this paragraph (i.e., correcting the total compact inventory by the amount estimated to be present in the compact outside the SiC layer at the end of irradiation). PARFUME does not predict any silver retention outside the SiC layer, hence it only has one set of results. The numbering of the compacts follows the same rule as in Figure 3 for the 17 available compacts.

![Figure 5: Compact (PIE) and particle (Corrected PIE) release compared to predicted PARFUME release.](image)

Figure 5 shows that the previously observed trends of under/over-predictions are mostly not modified by the correction of the matrix content. In the case of under-prediction, the discrepancy between PIE and PARFUME is increased, while in the case of over-prediction, the corrected PIE results are closer to the PARFUME results. The exception is Compact 4-3-3 (#10 in Figure 5) that goes from being over-predicted to being under-predicted.

**IV.D. Discussion and Analysis on Silver Release**

As explained in Section III.B, TAVA temperatures are not a suitable metric to correlate silver release to temperature because they do not adequately reflect the thermal state of the compacts throughout irradiation. Daily temperatures were therefore used to calculate the predicted silver release. In addition to their distribution around the compact TAVA temperature, the daily temperatures also widely vary spatially throughout a compact with typically a spread of several hundreds of degrees from the cold side to the hot side of a compact ([9]). This can lead to a significant range in silver release from one particle to the other. Figure 6 illustrates the calculated range in silver release between the coldest and hottest particles in Compact 6-3-2 during the AGR-1 irradiation, assuming a particle is associated with the coldest and hottest nodes in the thermal calculations. The nominal value is obtained using the volume-average daily temperatures of the compact.

![Figure 6: Silver retained fraction of 60 randomly selected particles from Compact 6-3-2 compared to the range of values predicted by PARFUME.](image)

The calculations show that particles from Compact 6-3-2 are predicted to release between 9% (cold particle) and 67% (hot particle) of their silver, with a nominal value of 41%. The predicted release from Compact 6-3-2 is compared to PIE in Figure 6, where the retained fraction (1 – release fraction) is displayed, as the inventory of Ag-110m remaining in the particle is measured by gamma counting. Because of the gamma counting detection threshold, Figure 6 shows the distribution of particles with detectable silver and adds the contribution of particles for which the Ag-110m activity was below the detection limit (where the detection limit was used as a conservative upper bound for the Ag-110m activity). For Compact 6-3-2, the distribution of retained silver spans from 8 to 88%, which almost encompasses all of the predicted range of 33 to 91%. The agreement between PARFUME and PIE is not as close on Compact 1-3-1 for which the measured and predicted ranges are displayed in Figure 7. The measured Ag-110m retention fraction ranges from 6 to 53%, while the PARFUME prediction ranges from 64 to 98%, showing a significant offset although the widths of both ranges are somewhat comparable.
The discrepancy observed between PARFUME and PIE when comparing the average fractional release (or retained fraction) can be largely attributed to the wide distribution of temperatures experienced by a compact during irradiation. PARFUME uses the volume-average daily temperatures to compute the average fractional release for each compact, while gamma measurements are performed on a subset of particles whose exact irradiation history is not known because information regarding the original location of the particles in the compact was lost during the deconsolidation. Furthermore, in the case of PARFUME, the release fraction of a particle bearing the volume-average daily temperatures is not equivalent to the average of the release fractions of all individual particles in the compact because the diffusivity does not vary linearly with temperature (see Eq. 1 in Section III.C). Therefore, using the volume-average daily temperatures under-estimates the release by neglecting the relatively larger release of hotter particles.

The silver release is also impacted by burnup as can be shown in Figure 8. Figure 8 shows that the silver fractional release decreases with burnup, which can be correlated with the temperature state of the compacts throughout irradiation. Figure 9 displays the correlation between burnup and the time-average volume-average temperatures calculated over the full 620 days of irradiation and calculated over the last 310 days of irradiation. It demonstrates that the temperature in the compacts globally was higher in the second half of the irradiation compared to the first half and that the lower burnup compacts tended to have higher temperatures during the second half of irradiation. This is easily explained by considering that the lower burnup compacts had more fissile material inventory towards the end of irradiation than higher burnup compacts, and they therefore had higher fission rates which ultimately led to higher heat generation rates and, consequently, higher temperatures.

The larger release of silver is obtained when high temperatures force the diffusion of a large amount of silver through the TRISO particle. This is typically the case towards the end of irradiation, when the silver source has accumulated significantly with time, and only if the temperature of the compact is high enough. Because compacts with lower burnup became hotter towards the end of irradiation, they yielded higher silver release on average.

### V. CESIUM AND STRONTIUM RELEASE

**V.A. Deconsolidation-Leach-Burn-Leach**
DLBL was performed on the nine compacts listed in Table 1 (see Section II). For compacts containing all intact particles, the DLBL measurement includes the contribution from the amount of fission products released through intact coatings but retained in the compact outside of the SiC layer, as well as fission products from uranium contamination in the OPyC and the compact matrix. As mentioned in Section II, the uranium contamination in AGR-1 compacts was on average lower than $4 \times 10^{-7}$ for each fuel variant so any release significantly above this level can be assumed to come from other sources. The DLBL data therefore represents a partial inventory of fission products released from intact particles, as it does not account for fission products released from the compacts and found on capsule components.

If any particles with failed SiC are present – and the particles are not removed from the population before the burn-leach is performed – then the inventory of the kernels from these particles will also be included in the post-burn leach solutions, and may greatly exceed the level of fission products attributed to diffusive release through coatings.

Based on analysis of fission products in the capsule components ([15]), it was determined that compacts 6-3-2, 5-2-3, and 5-2-1 may have one or more particles with a non-retentive SiC layer. DLBL results from Compact 6-3-2 indicated the presence of one such particle, the kernel of which was dissolved during post-burn leaching (and therefore the DLBL totals include the contribution from this kernel) ([16]).

Gamma analysis of particles from Compact 5-2-3 revealed that this compact contained two such particles. Compact 5-2-1 was determined to have one such particle. In both cases, these particles were removed from the population before DLBL analysis, and so the remaining inventory in the kernels was not dissolved during the DLBL process ([16]).

Two sets of calculations were performed with PARFUME. The first set calculates the diffusive release from compacts containing intact particles only. The second set calculates the diffusive release from compacts containing one or two particles with failed IPyC and SiC layers, in order to model the compacts that PIE determined to have leaking particles. Because PIE measurements indicate very little cesium release from intact particles, the second set of calculations assumes no diffusion of cesium from intact particles in order to compare PARFUME predictions of release from particles with failed SiC to PIE measurements. However, PIE data indicate a non-negligible strontium release from intact particles. Thus all strontium calculations account for strontium released by intact particles because their contribution is not negligible when compared to the amount of strontium released by the particles with failed SiC.

For each compact, the PIE measurements were corrected to take into account the amount of cesium and strontium released from the compacts and collected on the capsule components. In each capsule, this amount cannot be easily attributed to the specific compacts it originated from, so bounding values were determined. For most cases, a lower bound assumes that none of the cesium and strontium found on the capsule components originates from the analyzed compact (label “DLBL Min” in Figures 10 and 11). On the other hand, an upper bound assumes that all of the cesium or strontium found on the capsule components originates from the analyzed compact (“DLBL Max”).

In some cases, the contribution from fission product inventory on the capsule components could be assessed more accurately and attributed to one or more specific compacts ([8]). This is specifically true for cesium release, where it was found experimentally that only particles with failed SiC contributed significantly to the inventory released from the compacts in each capsule. For example, in Capsule 5, Compacts 5-2-3 and 5-2-1 were found to contain particles with failed SiC, and therefore the overwhelming majority of cesium found on the capsule components was from these two compacts; thus it can be assumed that Compact 5-3-1 (also from Capsule 5) contributed a negligible amount of cesium to the capsule component inventory. In these cases only one “DLBL” value is displayed in the histograms. The data are displayed in Figures 10 and 11. In these figures, “PARF Intact” represents the calculated release fractions of compacts containing intact particles only, and “PARF N Fail” represents the calculated release fractions of compacts containing N particles (N = 1 or 2) with failed IPyC and SiC layers. In the case of cesium, “PARF N Fail” excludes the contribution from intact particles, and only represents the calculated release from the particles with failed SiC. In the case of strontium, “PARF N Fail” represents the sum of the calculated release from intact particles and from particles with failed SiC. “PARF Intact” is also displayed in the figures corresponding to compacts containing particles with failed SiC to separately show the calculated contribution from the intact particles. It shows that the calculated release from intact particles is not negligible compared to the calculated release from particles with failed SiC, which is in disagreement with what PIE data show.
V.B. Discussion and Analysis on Cesium and Strontium Release

Figure 10a shows that, on average, PARFUME over-predicts the measured cesium release from intact particles by up to two orders of magnitude, with the exception of Compact 4-4-2 for which the calculated release is slightly lower than the measured release. For strontium, the PARFUME over-prediction for particles with intact SiC is up to 2.5 orders of magnitude, except for Compact 1-3-1 where the calculated release is slightly lower than the measured release (see Figure 11a).

While comparisons between PARFUME and PIE for cesium and strontium suffer from the same issues encountered with silver (e.g., the temperature and burnup distributions that are not taken into account in the modeling make it difficult to make detailed predictions), the magnitude of the discrepancy in the measured versus calculated release from intact particles is striking. PARFUME’s over-prediction of the release of cesium and strontium indicates that the IAEA diffusivities of these species in SiC are too high at the AGR-1 irradiation temperatures.

Figure 10b shows that the contribution to the release of cesium from one or two particles with failed SiC is slightly over-estimated by PARFUME. The PARFUME predictions “PARF N Fail” in Figure 10b exclude the contribution of intact particles, as PIE showed that the release is dominated by the contribution of particles with failed SiC. A failed SiC layer gives direct access to the diffusivity in the kernel, assuming the PyC layers are lowly retentive and that there is no holdup of cesium due to the formation of immobile species elsewhere within the particle. Figure 10b seems then to indicate that the IAEA diffusivity of cesium in the kernel is too high. This could also explain why the predicted cesium release is too high in intact particles, as shown in Figure 10a. It could therefore be expected that the diffusivity of cesium in UCO is lower than its diffusivity in UO2, the latter being used in the PARFUME modeling due to a lack of empirical data for UCO. Based on Figure 10b, the predicted fractional release is about a factor of two higher than the PIE measurements, which at this relatively high level of cesium release corresponds to an over-estimation of the diffusivity of cesium in the kernel by a factor of about 100. In addition, although the cesium release from intact particles is much lower than from particles with a failed SiC layer, the larger relative discrepancy between PARFUME calculations and PIE data on the release from intact particles would indicate that the diffusivity of cesium in SiC given by the IAEA is also too high at the AGR-1 irradiation temperatures. Taking into account the correction on the diffusivity of cesium in the kernel, the over-prediction of its diffusivity in SiC would be around 30%.

In the case of strontium, the available PIE data do not allow a conclusive assessment of the over-prediction of the diffusivities in the kernel or SiC layer since the respective contributions from intact particles and particles with failed SiC are not clearly predicted release is slightly lower than the measured release.
separable. The PARFUME predictions in Figure 11b include the contribution of intact particles, as PIE showed it was not negligible compared to the release from particles with failed SiC. Figure 11a shows a large over-prediction of the release from intact particles in most cases (with the exception of Compact 1-3-1). This over-prediction is also evident in Figure 11b for Compact 6-3-2, though it is much more limited for Compacts 5-2-1 and 5-2-3. Figure 11b shows that the release fraction of strontium from Compact 5-2-3 is about 0.3%. The contribution to the release fraction from particles with failed SiC cannot exceed 4.8×10^{-4}, corresponding to two failed particles out of ~4150 in the compact. Therefore, most of the measured release from Compact 5-2-3 comes from the intact particles. Similarly, the release from the intact particles of Compact 5-2-1 is approximately 0.16%, making up most of the compact release despite having one particle with failed SiC. Such a high release from intact particles would be consistent with the release observed from Compact 1-3-1 (see Figure 11a) but it does not offer any indication about the strontium release from particles with failed SiC. In this case, the over-prediction observed on the release from intact particles can be attributed to an over-estimation of the effective diffusivity of strontium in SiC, or in UCO, or both. It would also be possible that one diffusivity is over-estimated while the other one is under-estimated, the larger magnitude of the over-estimation leading to the overall over-prediction on the release.

VI. CONCLUSION

The PARFUME modeling code was used to predict fission product release from TRISO-coated fuel particles and compacts during the AGR-1 irradiation, and compare the predictions to PIE measurements.

Comparisons between PARFUME and PIE were conducted on the release of fission products silver, cesium, and strontium, both at the particle and compact levels. Post-irradiation examination measurements were made by gamma scanning of 56 compacts, gamma counting individual particles from nine deconsolidated compacts for silver, by deconsolidation-leach-burn-leach of nine compacts for cesium and strontium, and measurement of silver, cesium, and strontium on the irradiation capsule components.

The results of these comparisons are summarized below:

- Silver
  (a) PARFUME predictions and PIE measurements are in reasonably good agreement.
  (b) The large distribution in silver release from particle gamma counting can be partially explained by the spatial distribution of temperatures across a compact, as demonstrated by PARFUME calculations.
  (c) Low burnup capsules tend to have a higher silver release because they became hotter towards the end of irradiation when silver was readily available for release.
  (d) Both PARFUME calculations and PIE data show the trend of higher release at lower burnup, but it is more strongly marked in PIE measurements. PARFUME tends to under-predict PIE in the lower burnup capsules and to over-predict PIE in the higher burnup capsules.
  (e) PIE data showed non-negligible retention of silver by the matrix, though retention is not predicted by PARFUME due to the historical high diffusivity of silver through graphite.

- Cesium
  (a) The predicted cesium release from particles with failed SiC is too high by a factor of about two, which could mean that the IAEA diffusivity of cesium in the kernel is also too high, by a factor of about 100.
  (b) There is an overall over-prediction by PARFUME of cesium release from intact particles by an order of magnitude. Taking into account the over-prediction in kernel diffusivity, then the IAEA diffusivity of cesium in SiC is too high by about 30%.

- Strontium
  (a) The release from intact particles is large enough so that the contribution from a few particles with failed SiC cannot be assessed. Furthermore, the PIE data are too scarce to draw conclusions about the validity of the diffusivity of strontium in UCO.
  (b) There is an overall over-prediction by PARFUME of strontium release, suggesting that the IAEA diffusivities of strontium in SiC, UCO or both are too high. The data do not allow a straightforward derivation of more suitable diffusion coefficients for strontium in UCO or SiC.

It should be duly noted that these conclusions rest upon results obtained on a relatively small number of specimens (especially for cesium and strontium) and with a modeling issue with temperature and burnup spatial distributions across the compacts. Some conclusions have a reasonable level of confidence, but the analysis in terms of correct values of the diffusivities is only tentative. There are major factors affecting both the data and calculations used in this fractional release assessment, namely:

- Accuracy of the diffusivities (PARFUME)
- Accuracy of the predicted compact temperatures (PARFUME)
• Spatial distribution of the compact daily temperatures (PARFUME)
• Spatial distribution of the sources (PARFUME)
• Uncertainty from gamma scanning measurements (PIE)

Refining the modeling with spatial burnup and temperature distributions would allow better predictions of the fractional release of the fission products and more accurate comparisons between model predictions and compact gamma scanning or leach-burn-leach results. Comparison with particle gamma counting results poses an additional problem of unknown specific irradiation conditions experienced by the subset of particles, which is related to their original location within the compacts, which does not seem to be easily solved.

REFERENCES