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Comparison of fission product release predictions using PARFUME with results from the AGR-1 safety tests

Blaise P. Collin, David A. Petti, Paul A. Demkowicz, John T. Maki
Idaho National Laboratory
2525 Fremont Avenue, Idaho Falls, ID 83415, USA
Phone: +1-208-526-2217, blaise.collin@inl.gov

Abstract – *Safety tests were conducted on fourteen fuel compacts from AGR-1, the first irradiation experiment of the Advanced Gas Reactor (AGR) Fuel Development and Qualification program, at temperatures ranging from 1600 to 1800°C to determine fission product release at temperatures that bound reactor accident conditions. The PARFUME (PARticle FUel ModEl) code was used to predict the release of fission products silver, cesium, strontium, and krypton from fuel compacts containing tristructural isotropic (TRISO) coated particles during the safety tests, and the predicted values were compared with experimental results. Preliminary comparisons between PARFUME predictions and post-irradiation examination (PIE) results of the safety tests show an overall over-prediction of the fractional release of these fission products, which is largely attributed to an over-estimation of the diffusivities used in the modeling of fission product transport in TRISO-coated particles. Correction factors to these diffusivities were assessed for silver and cesium in order to enable a better match between the modeling predictions and the safety testing results. In the case of strontium, correction factors could not be assessed because potential release during the safety tests could not be distinguished from matrix content released during irradiation. In the case of krypton, all the coating layers are partly retentive and the available data did not allow to determine their respective retention powers, hence preventing to derive any correction factors.*

	Correction Factor Kernel			*Correction Factor SiC*		
	1600°C	*1700°C*	*1800°C*	*1600°C*	*1700°C*	*1800°C*
Silver	-	-	-	10^2-10^3	-	10-10^2
Cesium	10^2-10^3	-	10^3	10	-	-
Strontium	-	-	-	-	-	-
Krypton	-	-	-	-	-	-

The estimated correction factors to the diffusivities are based on limited statistics and on safety testing data that show differences from one compact to another at a given temperature, which leads to uncertainties in the assessment of these correction factors. At this point, these correction factors, or lack thereof, are preliminary, while a more definitive assessment must await additional data from future AGR safety testing campaigns.

I. INTRODUCTION

Model predictions were compared to measurements of silver (Ag), cesium (Cs), strontium (Sr), and krypton (Kr) release from fourteen tristructural isotropic (TRISO) fuel compacts that underwent safety testing following the first irradiation test of the Advanced Gas Reactor program (AGR-1). The safety tests on these compacts were conducted at both Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory (INL) at isothermal temperatures of 1600, 1700, and 1800°C. The results of these tests are presented elsewhere in these proceedings ([1]). A companion paper presents the results of fuel performance modeling for the AGR-1 fuel during irradiation ([2]).

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 ([3]). 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). 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.

PARFUME calculates the release fraction from a compact 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 release fraction obtained from safety tests was calculated by measuring the amount of fission products released from the compact and by normalizing that amount to a predicted compact source term obtained by as-run neutronics calculations of the AGR-1 experiment ([4]).

II. AGR-1 SAFETY TESTING

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 ([5]).

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. 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 4.30×10^{25} n/m^2 ($E > 0.18$ MeV) ([6]).

At completion of the irradiation, safety testing of selected irradiated compacts was conducted at the ORNL Core Conduction Cooldown Test Facility (CCCTF) and at the INL Fuel Accident Condition Simulator (FACS). A description of the two experimental systems has been provided previously ([7, 8]). Safety tests involved heating compacts for at least 300 hours to maximum temperatures of 1600 to 1800°C, where 1600°C is the expected maximum temperature during a high-temperature gas-cooled reactor depressurization conduction cooldown event, and 1800°C is the expected temperature at which the performance of the silicon carbide (SiC) layer will begin to degrade. The times used in these tests are much greater than calculated to occur in an actual modular HTGR, but they are typical of historic testing conducted during German TRISO fuel development.

Table 1 lists the fourteen safety-tested compacts, while Figure 1 shows the safety test heating plans ([7]).

Table 1: Compacts used for safety testing (compacts tested at 1600°C are highlighted in blue, at 1700°C in green, and at 1800°C in red).

Compact	Burnup (%FIMA)	Fast Fluence ($\times10^{25}$ n/m^2, $E > 0.18$ MeV)	Safety Test Temperature (°C)
6-4-3	13.35	2.46	1600
6-4-1	13.35	2.43	1600
6-2-1	14.20	2.87	1600
5-3-3	17.01	3.65	1600
5-1-3	18.19	3.82	1800
4-4-3	18.99	4.06	1700
4-4-1	18.96	3.99	1800
4-3-3	18.63	4.16	1600
4-3-2	16.38	3.68	1800
4-1-2	17.39	3.72	1600
3-3-2	17.02	3.8	1600
3-3-1	19.07	4.23	1700
3-2-3	19.12	4.28	1800
3-2-2	17.02	3.79	1600

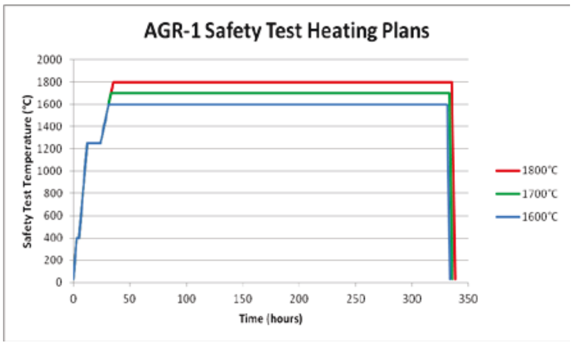

Fig. 1: AGR-1 safety test heating plans.

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.

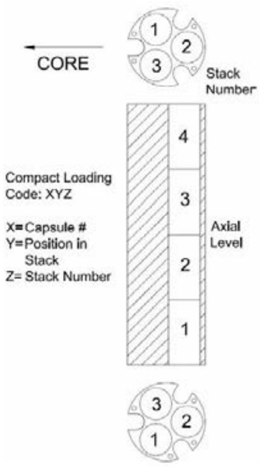

Fig. 2: Layout of an AGR-1 capsule. The cross-section view at the top of the figure shows the orientation of the three fuel stacks within the graphite fuel holder and the axial cutaway shows the four axial levels.

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 (OPyC, ~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. [9].

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 ([4, 10]). The calculations were computed prior to completion of PIE, and they are based on an earlier version of the as-run neutronics calculations ([11]). 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 of the irradiation phase.

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.

Following the modeling of the irradiation phase, PARFUME models the heating phase using the designed heating plans (see Figure 1 in Section II). The heating plans for isothermally-tested compacts consists of two temperature ramps of 120°C/hr followed by temperature plateaus of 2 hours and 12 hours, respectively, and then a temperature ramp of 50°C/hr until the maximum temperature (1600, 1700, or 1800°C) is reached. A heating phase of at least 300 hours follows at maximum temperature and then the compacts are brought back to room temperature (30°C) with a ramp of -600°C/hr ([7]). During safety tests, the burnup and fast neutron fluence do not evolve and they keep their end-of-irradiation values.

The objective of the PARFUME calculations is to provide the estimated fractional release of fission product species Ag, Cs, Sr, and Kr during the heating phase. The corresponding release fraction is defined as the amount of fission products released during the heating phase relative to the total inventory produced during irradiation.

III.C. Input Parameters

The input parameters needed to model the AGR-1 irradiation with PARFUME originate from he AGR-1 Irradiation Experiment Test Plan ([9]) for the fuel characteristics, particle geometry, compact characteristics, and material non-mechanical properties, and from a CEGA Corporation report ([12]) 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 ([13]) and displayed in Table 2. The corresponding diffusivities can be calculated using these diffusion coefficients in the following Arrhenius-type equation:

$$D = D_{0,1} e^{-\frac{Q_{0,1}}{RT}} + D_{0,2} e^{-\frac{Q_{0,2}}{RT}} \qquad \text{(eq. 1)}$$

where

$D_{0,i}$ = pre-exponential factor (m^2/s)
$Q_{0,i}$ = activation energy (kJ/mol)
R = gas constant (8.3142×10^{-3} kJ/mol/K)
T = temperature (K)

Table 2: Diffusion coefficients used in PARFUME.

Species	$D_{0,i}$ (m^2/s) $Q_{0,i}$ (kJ/mol)	Kernel (UO_2)	PyC	SiC	Matrix graphite
Ag	$D_{0,1}$	6.7×10^{-9}	5.3×10^{-9}	3.6×10^{-9}	1.6
	$Q_{0,1}$	165	154	215	258
	$D_{0,2}$	-	-	-	-
	$Q_{0,2}$				
Cs	$D_{0,1}$	5.6×10^{-8}	6.3×10^{-8}	5.5×10^{-14}	3.6×10^{-4}
	$Q_{0,1}$	209	222	125	189
	$D_{0,2}$	5.2×10^{-4}	-	1.6×10^{-2}	-
	$Q_{0,2}$	362		514	
Sr	$D_{0,1}$	2.2×10^{-3}	2.3×10^{-6}	1.2×10^{-9}	10^{-2}
	$Q_{0,1}$	488	197	205	303
	$D_{0,2}$	-	-	1.8×10^{6}	-
	$Q_{0,2}$			791	
Kr	$D_{0,1}$	8.8×10^{-15}	2.9×10^{-8}	3.7×10^{1}	6.0×10^{-6}
	$Q_{0,1}$	54	291	657	0
	$D_{0,2}$	6.0×10^{-1}	2.0×10^{5}	-	-
	$Q_{0,2}$	480	923		

Notes: UO_2 values are used for the kernel; the buffer has a set diffusivity of 10^{-8} m^2/s; PARFUME does not use the fluence-dependent diffusivity of Cs in SiC; Kr coefficients for the kernel and SiC layer are specific to accident conditions.

PARFUME calculates fission product release at the compact level ([2]). The release from particles with failed layers can be obtained by setting the diffusivities in these layers to a value of 10^{-6} m^2/s. In these calculations, PARFUME models the fission product release from both intact particles and particles with both failed IPyC and SiC layers. The failures of the coating layers are assumed to occur at the beginning of the heating phase. The results are then compared to the measured release data.

IV. FISSION PRODUCT RELEASE

As mentioned in Section III.C, fission product release is modeled from both intact particles and particles with both failed IPyC and SiC layers. Based on PIE measurements after the safety tests, the number of particles whose SiC layers failed during safety testing was assessed and reported in Table 3 [14]. The numbers reported in Table 3 were then used to model the compacts containing particles with failed SiC layers to enable comparisons with the release data.

Table 3: Compacts containing particles with failed SiC (compacts tested at 1600°C are highlighted in blue, at 1700°C in green, and at 1800°C in red).

Compact	Number of particles with failed IPyC & SiC	Safety Test Temperature (°C)
6-4-3	0	1600
6-4-1	1	1600
6-2-1	0	1600
5-3-3	0	1600
5-1-3	7	1800
4-4-3	0	1700
4-4-1	2	1800
4-3-3	0	1600
4-3-2	5*	1800
4-1-2	1	1600
3-3-2	1	1600
3-3-1	4	1700
3-2-3	11	1800
3-2-2	0	1600

Note: Compact 4-3-2 is modeled with 2 exposed kernels and 3 particles with failed IPyC and SiC layers.

IV.A. Silver Release

The safety test data for silver release include silver promptly released during the first ~24 hours of the tests spent at temperature, which is believed to be silver retained in the matrix, or possibly in the OPyC layer, at the end of the irradiation phase, and which was then quickly driven out of the compact at temperatures above 1600°C ([1]). Consequently, the fractional release during the heating phase is assumed to be obtained by considering the amount of silver released after the first 24 hours spent at the safety test temperature.

Figures 3 and 4 show the fractional release of silver from compacts containing only intact particles and from compacts containing particles with failed SiC layers, respectively. They exclude the release measured during the first ~24 hours of the safety tests spent at temperature. Compacts 3-2-2 and 3-3-2 are also excluded from these plots because of unplanned temperature ramps at the beginning of their respective safety tests that caused some silver release which is believed to be from intact particles, and not from silver outside the SiC at the end of irradiation.

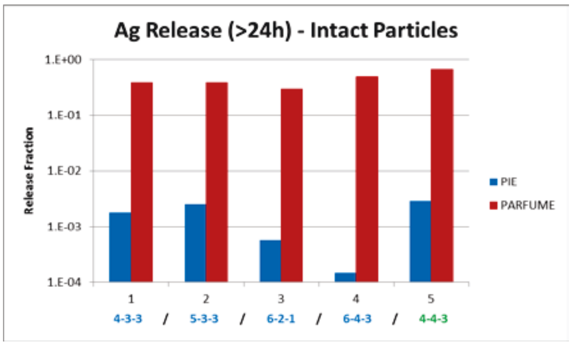

Fig. 3: Silver fractional release from compacts containing only intact particles corrected to exclude release during the first 24 hours of the safety tests spent at temperature.

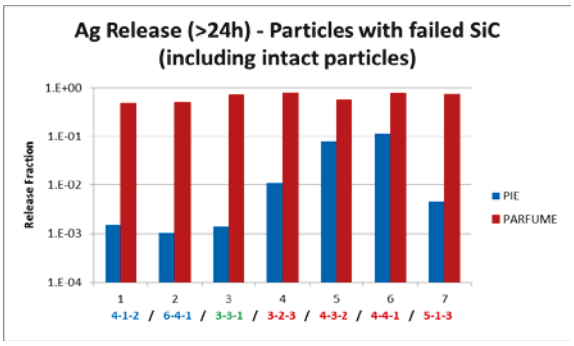

Fig. 4: Silver fractional release from compacts containing particles with failed SiC layers corrected to exclude release during the first 24 hours of the safety tests spent at temperature. PARFUME predictions include the release from intact particles.

Comparison of safety testing data between Figures 3 and 4 shows that the release is associated with intact particles: the measured release from compacts containing particles with failed SiC (Figure 4) is similar to the contribution from compacts containing only intact particles (Figure 3), when data at similar temperatures (1600 and 1700°C) are compared. Because of the high release from intact particles, silver release from failed particles cannot be discerned.

Figures 3 and 4 show a large over-prediction of the release from compacts containing only intact particles and from compacts containing particles with failed SiC layers, respectively. The over-predicted release is either from (Figure 3) or largely dominated by (Figure 4) intact particles and, therefore, the over-predictions are associated with an over-estimation of the diffusivity of silver in the SiC. The potential over-estimation of the diffusivity in the kernel cannot be assessed.

Figure 5 shows the impact of a reduction in the diffusivity of silver in SiC on the calculated release fractions from intact particles at 1600 and 1800°C. The IAEA diffusivity was divided by successive powers of ten and the subsequent release fractions were calculated at these two safety testing temperatures. Also shown on the plot are the maximum measured release fractions that are obtained from all twelve heating tests at 1600 and 1800°C since silver release is dominated by intact particles even in compacts containing particles with failed SiC. The maximum release fractions at 1600 and 1800°C are used to obtain upper limits of the measured release, which subsequently leads to upper limits for the diffusivity of silver in SiC. The PIE data at 1700°C consist of one release fraction result for each category of compact – containing only intact particles or containing particles with failed SiC. Therefore, compacts tested at 1700°C (Compacts 4-4-3 and 3-3-1) were excluded from the analysis because of the measurement uncertainty on their respective release fractions and the lack of statistical significance associated with the scarcity of available measured data.

The intersection of the curves of the diffusivity-dependent release fractions with the maximum measured release fractions provide a minimum estimate of how much the diffusivities in SiC need to be reduced for the calculated release fractions to match PIE measurements. At 1600°C, the correction factor is close to 10^3, and it is between 10 and 10^2 at 1800°C.

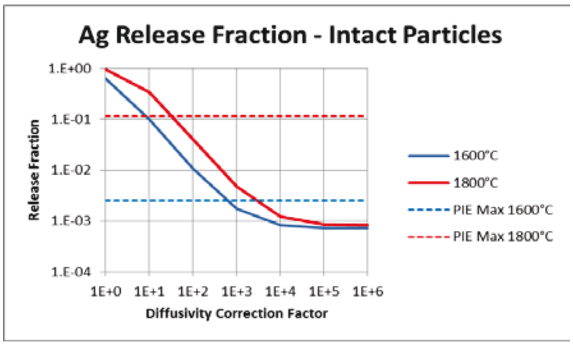

Fig. 5: Calculated silver release fraction versus diffusivity in SiC.

IV.B. Cesium Release

Figures 6 and 7 compare the calculated and measured fractional releases of cesium for compacts containing only intact particles and compacts containing particles with failed SiC layers, respectively. Figure 6 shows that the measured release from intact particles is limited. Moreover, it is largely over-predicted by PARFUME. It is suspected that a significant portion of this release is from inventory outside the SiC at the end of irradiation ([1]), which would mean that the release from intact particles would be even lower than shown in Figure 6, and the over-prediction by PARFUME even larger.

For compacts containing particles with failed SiC, Figure 7 shows a fairly good agreement between safety testing data and PARFUME, when considering no release from the intact particles in the PARFUME predictions, with an over-prediction of a factor of 3 at most. This would indicate that the diffusivity in the kernel is somewhat over-estimated, because the release from a particle with a failed SiC layer is controlled by the diffusivity in the kernel since the PyC layers are non-retentive to cesium. Part of the over-prediction may be attributed to the irradiation phase and part to the heating phase ([15]). Indeed, it was shown that the diffusivity of cesium in the kernel under irradiation might be over-estimated by a factor of about 100 ([2]). Because of the respective magnitude of the measured release fractions in Figure 6 (<10^{-5}) and in Figure 7 (>10^{-4}), the release from compacts containing particles with failed SiC can be mostly attributed to the particles rather than the matrix.

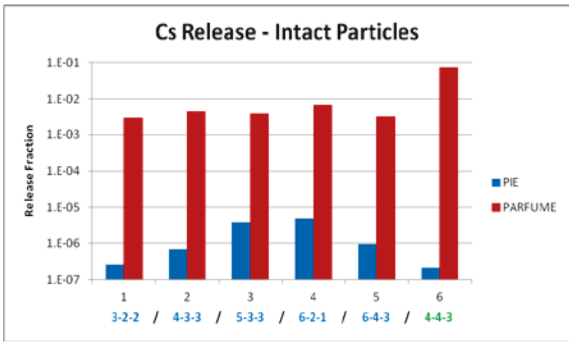

Fig. 6: Cesium fractional release from compacts containing only intact particles.

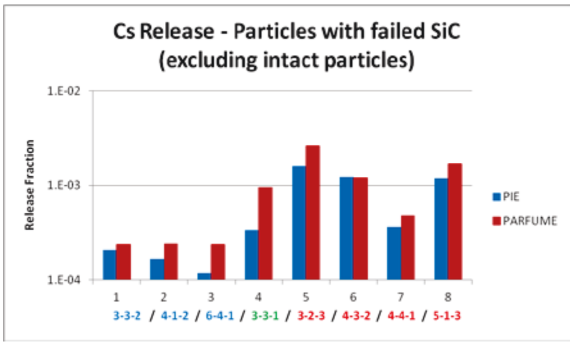

Fig. 7: Cesium fractional release from compacts containing particles with failed SiC layers. PARFUME predictions exclude the release from intact particles.

Figures 8 and 9 show the fractional release of cesium as a function of time for two compacts, one containing only intact particles (Compact 5-3-3 tested at 1600°C) and one containing particles with failed SiC (Compact 3-2-3 tested at 1800°C). Figure 8 demonstrates the over-prediction of cesium release by PARFUME. It is also noticeable in the constantly increasing calculated release, while the measured release is somewhat flat. This means that the predicted diffusion of cesium through the SiC layer is too large, as PIE shows a very small release rate of cesium. The prompt release of cesium from Compact 3-2-3 predicted by PARFUME is the result of the modeling of SiC failures from the start of the test (see III.C) as cesium is not held up in the particle and is being released right away. In the case of Compact 3-2-3, PIE shows a slow diffusive release which might be indicative of the successive failures of SiC layers in the eleven particles that experienced SiC failure during this test (see Table 3 in Section IV). Note that the sharpness of the PARFUME curves is due to time-step management in the modeling and it does not reflect any changes in the physical conditions of the safety tests or fission product diffusion behavior.

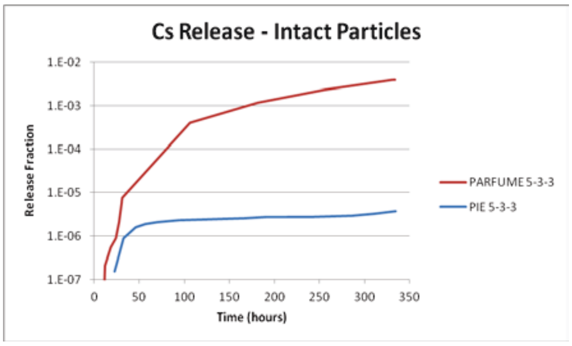

Figure 8: Cesium fractional release as a function of time from compacts containing only intact particles.

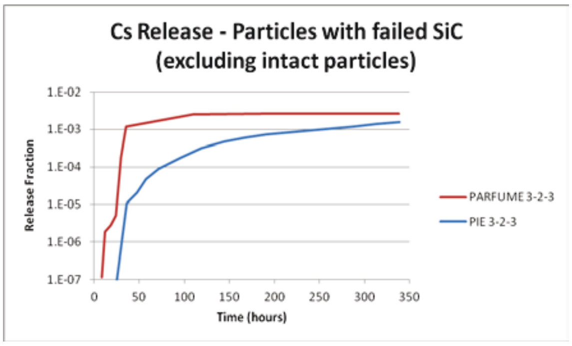

Fig. 9: Cesium fractional release as a function of time from compacts containing particles with failed SiC layers. PARFUME predictions exclude the release from intact particles.

Figure 10 shows the impact of a reduction in the diffusivity of cesium in the kernel on the calculated release fractions from the kernel at 1600 and 1800°C, when the diffusivity in the kernel is reduced by a factor of 100 during irradiation, as suggested by Ref. [2]. Figure 10 shows that the resulting fraction of cesium outside the kernel at the end of irradiation is about 35%. Because PARFUME over-predicts the fractional release from the kernel, Figure 10 also includes the minimum release fractions from the kernel at 1600 and 1800°C that PARFUME should predict to match PIE measurements of the release from particles with failed SiC. They are used as targets to reduce the diffusivity of cesium in the kernel so that PARFUME results would match these PIE measurements. Compacts tested at 1700°C were again excluded from the analysis because of the measurement uncertainty on their respective release fractions and the lack of statistical significance. The subsequent correction factor on the diffusivity is at least between 10^2 and 10^3 at 1600°C and approximately 10^3 at 1800°C.

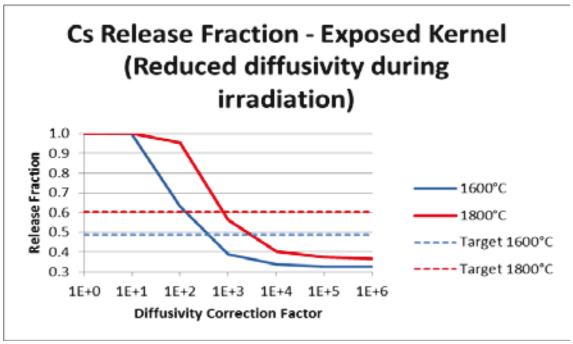

Fig. 10: Calculated cesium release fraction versus diffusivity in the kernel. The diffusivity of cesium in the kernel during irradiation was reduced by a factor of 100.

In addition to the over-estimation of the diffusivity of cesium in the kernel during irradiation by a factor of 100, comparisons between PIE and PARFUME also showed that the diffusivity of cesium in SiC during irradiation could be over-estimated by about 30% ([2]). These two corrections were taken into account to determine the over-estimation of the diffusivity of cesium in the SiC layer during safety testing. Additionally, the correction factor for the diffusivity of cesium in the kernel during the heating phase was set to 500 at 1600°C. There was no analysis made at 1700°C because of the lack of statistical significance at this temperature. There was also no analysis made at 1800°C since all the compacts tested at this temperature contain particles with failed SiC layers whose release largely dominates the release from the compact and obscures any release from intact particles. Combining all three corrections on the respective diffusivities (kernel diffusivity under irradiation, SiC under irradiation and kernel diffusivity under heating test conditions), Figure 11 shows the impact of a reduction in the diffusivity of cesium in SiC during heating tests on the calculated release fractions from intact particles at 1600°C. The maximum measured release fraction at 1600°C is also plotted in Figure 11. It might partly or completely correspond to cesium present outside the SiC at the end of irradiation, and, therefore, it provides an over-estimated upper bound of the cesium release from compacts containing only intact particles. In order to match safety testing results, the diffusivity in SiC has to be reduced by at least a factor of 10 at 1600°C, but this factor could be higher depending on if any of the cesium was present outside the SiC at the end of irradiation rather than directly released from the particles during the safety tests.

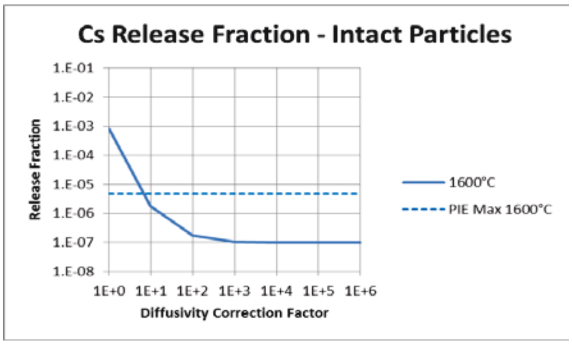

Figure 11. Calculated cesium release fraction versus diffusivity in SiC. The diffusivities of cesium in the kernel and in SiC during irradiation were reduced by a factor of 100 and by 30%, respectively, and the diffusivity of cesium in the kernel during safety testing was reduced by a factor of 500.

IV.C. Strontium Release

Figures 12 and 13 show the fractional release of strontium for compacts containing only intact particles and compacts containing particles with failed SiC layers, respectively. Comparison of safety testing data between Figures 12 and 13 shows that, overall, the release is dominated by intact particles. Releases from compacts containing particles with failed SiC (Figure 13) are similar to releases from compacts containing only intact particles (Figure 12), when data at similar temperatures (1600 and 1700°C) are compared. Because of the dominant release from intact particles, strontium release from failed particles cannot be discerned.

It is not possible to determine directly if the strontium release measured during safety testing is due to release through intact SiC or simply from residual inventory outside the SiC at the end of irradiation. The total inventory of strontium release from the compacts during safety tests at 1600°C is similar to the levels found in as-irradiated compacts outside of the SiC layer [16]. At the end of the safety tests, the amount of strontium outside the SiC is not negligible compared to the amount collected on the deposition cups in the furnace [1]. Therefore, it is not clear whether any of the strontium released during safety testing is from intact particles, if it simply is due to slow release from strontium already outside of the SiC at the end of irradiation, or a combination of both. However, the bulk of PIE data suggests that the majority of the strontium release from safety testing is due to inventory residing outside of SiC at the end of irradiation ([1]).

Figures 12 and 13 show a large over-prediction of the release from compacts containing only intact particles and from compacts containing particles with failed SiC layers by 2 to 3.5 and by 1.5 to 2.5 orders of magnitude, respectively. The over-prediction might be even larger if the measured release does not originate from the particles.

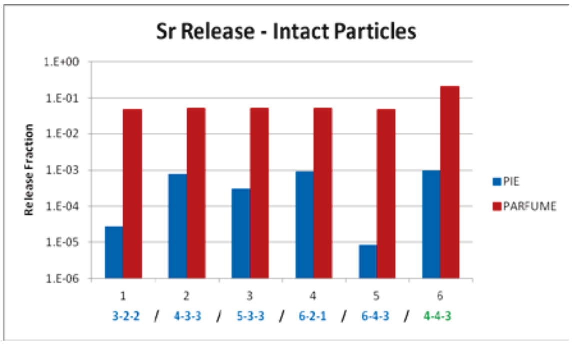

Fig. 12: Strontium fractional release from compacts containing only intact particles.

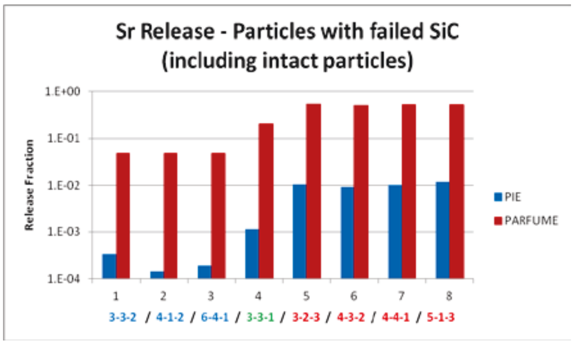

Fig. 13: Strontium fractional release from compacts containing particles with failed SiC layers. PARFUME predictions include the release from intact particles.

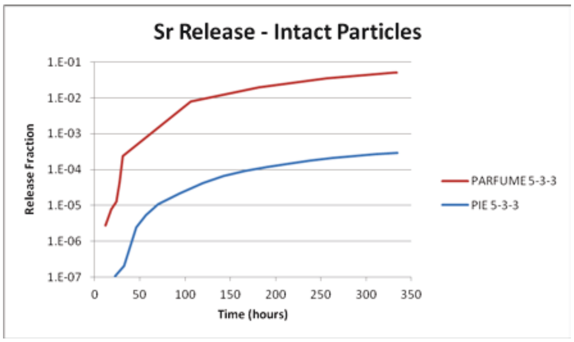

Fig. 14: Strontium fractional release as a function of time from compacts containing only intact particles.

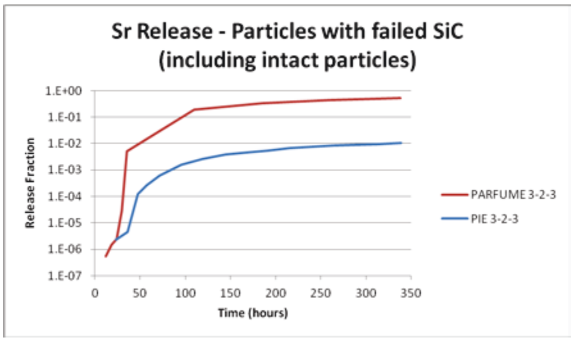

Fig. 15: Strontium fractional release as a function of time from compacts containing particles with failed SiC layers. PARFUME predictions include the release from intact particles.

Figures 14 and 15 show the fractional release of strontium as a function of time for Compact 5-3-3 and Compact 3-2-3. The figures demonstrate the over-prediction of strontium release by PARFUME. The prompt release of strontium from Compact 3-2-3 predicted by PARFUME is again the result of the modeling of SiC failures from the start of the test, but it is less prompt than for cesium because the kernel acts as a retentive barrier to strontium. In both cases, the similar slopes in the measured and predicted releases seem to indicate that strontium is released from the particles rather than from the matrix, since PARFUME does not predict any matrix retention.

As with cesium, part of the over-prediction may be attributed to the irradiation phase. Indeed, comparisons between PIE and PARFUME on strontium release at the end of irradiation showed that the diffusivity of strontium is over-estimated in either the kernel or in the SiC layer, or possibly in both, but the available data do not allow a precise assessment of the effective over-estimation of these diffusivities ([2]). Therefore, the magnitude of the over-prediction of strontium release during safety testing might be over-estimated because the discrepancies in strontium retention during irradiation are not captured by the safety testing calculations.

In the end, PARFUME shows a large over-prediction in strontium release during safety testing. However, this over-prediction could be increased or decreased depending on whether the release is from the matrix rather than from the particles, or if strontium is better retained during irradiation than predicted, respectively. In any case, correction factors cannot be derived from the measured data because all three diffusivities in the kernel, SiC, and matrix might be over-estimated, which is too many unknowns to apply the method used for silver and cesium.

IV.D. Krypton Release

Krypton is unique because it has both the SiC and PyC layers as diffusion barriers. Krypton release is therefore indicative of the failure of all three coating layers, and is the primary indicator for particle failure during the heating test.

For krypton, there is no reliable release data for compacts heated up at 1600°C, partly due to the potential for background contamination from the hot cell contributing to the measured krypton activity, and partly because the release fractions were sufficiently low that released activity was below detection limits. The measured release, including background contamination, was $\leq 10^{-5}$ for all 1600°C tests, which indicates a low release from intact particles.

Compacts tested for Krypton release at 1700 and 1800°C all contained particles with failed SiC. Figure 16 shows the fractional release of krypton for these compacts, and an over-prediction by 1 order of magnitude at 1700°C, and by up to 1.5 orders of magnitude at 1800°C.

To be consistent with PIE observations, the particles with failed SiC layers are also modeled with failed IPyC layers, but intact OPyC layers. The over-prediction of the krypton release from compacts containing particles with failed SiC is therefore attributed to the over-estimation of the diffusivity of krypton in the kernel or in the OPyC, or a combination of both. Historically, the diffusion of noble gases in PyC, including krypton, has been a major focus of study in bistructural isotropic (BISO) coated particles, and the diffusivity of krypton in PyC is better known than its diffusivity in the kernel. On the other hand, the best comparison arises from Compact 4-3-2 which is modeled with two exposed kernels and three particles with failed SiC (and consequently failed IPyC). This would indicate that the discrepancies between PARFUME and PIE are larger when modeling the release from particles with intact OPyC.

In the case of krypton, the paucity of safety testing data and the complexity of its release due to multiple retention barriers do not allow the determination of correction factors for the diffusivities in the kernel and the successive coating layers.

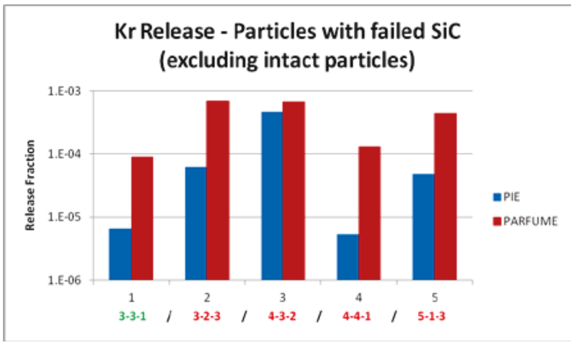

Fig. 16: Krypton fractional release from compacts containing particles with failed SiC layers. PARFUME predictions exclude the release from intact particles.

V. CONCLUSION

The PARFUME modeling code was used to predict fission product release from fourteen TRISO-coated fuel compacts during AGR-1 safety tests at 1600, 1700, and 1800°C. Predictions were then compared to PIE measurements.

Comparisons were made for both compacts containing only intact particles and for compacts containing particles with both failed IPyC and SiC layers. The latter comparisons theoretically allow for corrections to be made for the over-estimation of the diffusivities of the fission products in the kernel, provided that the OPyC layer is non-retentive. In the case of significant release from intact particles, the former comparisons allow the determination of the over-estimation of the diffusivity of fission products in the SiC layer.

The predictions showed different trends in the comparison to PIE measurements depending on the fission product species, which leads to different conclusions regarding the diffusivities used in the modeling of fission product transport:

- *Silver*

(a) The diffusivity of silver in SiC might be over-estimated by a factor of at least 10^2 to 10^3 at 1600°C and at least 10 to 10^2 at 1800°C.

(b) The diffusivity of silver in UCO might also be over-estimated, but the available data do not allow definite conclusions.

- *Cesium*

(a) The diffusivity of cesium in UCO might be over-estimated by a factor of at least 10^2 to 10^3 at 1600°C and 10^3 at 1800°C. The analysis of these over-estimates takes into account corrections for the diffusivities of cesium in the kernel and in SiC during irradiation, as it was shown that these diffusivities are over-estimated by a factor of 100 and by 30%, respectively.

(b) The diffusivity of cesium in SiC might also be over-estimated by a factor of 10 at 1600°C, as estimated by the comparison between calculated and measured release fractions from intact particles. There is no available estimate at 1800°C since all the compacts heated up at 1800°C contain particles with failed SiC layers whose release largely dominates the release from intact particles.

- *Strontium*

(a) There is evidence suggesting that the measured release is predominantly of strontium present outside the SiC at the end of irradiation rather than strontium released by particles during the safety tests.
(b) Comparisons between PARFUME and PIE show an over-prediction of the release from compacts containing only intact particles by 2 to 3.5 orders of magnitude.
(c) Comparisons show an over-prediction of the release from compacts containing particles with failed SiC by 2 orders of magnitude.
(d) The over-prediction might be even larger if the measured strontium is from the matrix and not the particles.
(e) Correction factors for the diffusivities cannot be derived from the measured data because there are too many diffusivities (kernel, SiC, matrix) to correct.

- *Krypton*

(a) There is no reliable release data for compacts heated up at 1600°C, which includes all the compacts containing only intact particles.
(b) At 1700 and 1800°C, comparisons show an over-prediction of the release from compacts containing particles with failed SiC by 1 to 1.5 orders of magnitude.
(c) There are not sufficient data from these heating tests to determine which of the TRISO-coating's layers diffusivities are under or over-estimated.

Table 4 summarizes the correction factors to be applied to the IAEA diffusivities in order for the PARFUME predictions to match measured release fractions from the AGR-1 PIE. For cesium, the diffusivity during irradiation was reduced by a factor of 100 in the kernel and by 30% in SiC.

Table 4. Diffusivity correction factors.

	Correction Factor Kernel			**Correction Factor SiC**		
	1600°C	**1700°C**	**1800°C**	**1600°C**	**1700°C**	**1800°C**
Silver	-	-	-	10^2-10^3	-	10-10^2
Cesium	10^2-10^3	-	10^3	10	-	-
Strontium	-	-	-	-	-	-
Krypton	-	-	-	-	-	-

The estimated correction factors to the diffusivities enable a better match between the modeling predictions and the safety testing results. They are based on limited statistics and on safety testing data that show differences from one compact to another at a given temperature, which leads to uncertainties in the assessment of these correction factors. At this point, these correction factors, or lack thereof, are preliminary, while a more definitive assessment must await additional data from future AGR safety testing campaigns.

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