First high temperature safety tests of AGR-1 TRISO fuel with the Fuel Accident Condition Simulator (FACS) furnace

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Abstract

Three TRISO fuel compacts from the AGR-1 irradiation experiment were subjected to safety tests at 1600 and 1800°C for approximately 300 h to evaluate the fission product retention characteristics. Silver behavior was dominated by rapid release of an appreciable fraction of the compact inventory (3 to 34%) at the beginning of the tests, believed to be from inventory residing in the compact matrix and outer pyrocarbon (OPyC) prior to the safety test. Measurable release of silver from intact particles appears to become apparent only after ~60 h at 1800°C. The release rate for europium and strontium was nearly constant for 300 h at 1600°C (reaching maximum values of approximately 2×10^{-3} and 8×10^{-4} respectively), and at this temperature the release may be mostly limited to inventory in the compact matrix and OPyC prior to the safety test. The release rate for both elements increased after approximately 120 h at 1800°C, possibly indicating additional measurable release through the intact particle coatings. Cesium fractional release from particles with intact coatings was $<10^{-6}$ after 300 h at 1600°C or 100 h at 1800°C, but release from the rare particles that experienced SiC failure during the test could be significant. However, Kr release was still very low for 300 h 1600°C ($<2 \times 10^{-6}$). At 1800°C, krypton release increased noticeably after SiC failure, reflecting transport through the intact outer pyrocarbon layer. Nonetheless, the krypton and cesium release fractions remained less than approximately 10^{-3} after 277 h at 1800°C.

Keywords: TRISO, FACS, irradiated fuel, coated particle fuel, accident testing, safety testing

1. Introduction

Tristructural isotropic (TRISO) coated particle fuel is a robust fuel form designed for use in high temperature gas-cooled reactors (HTGRs) [1, 2]. The fuel provides a high degree of fission product retention during reactor operation and during depressurized loss of forced cooling, allowing for high reactor core and coolant temperatures. US TRISO fuel development was re-initiated in the early 2000s as part of the US Department of Energy Advanced Gas Reactor (AGR) Fuel Development and Qualification Program [3]. The objective of the program is to perform the requisite research and development needed to qualify TRISO fuel for use in an HTGR. As part of this program, a series of fuel irradiation experiments is being conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). These experiments are intended to provide data on fuel performance under irradiation, support fuel process development, qualify the fuel for normal operating conditions, provide irradiated fuel for safety testing, and support the development of fuel performance and fission product transport models. The first of these irradiation tests, designated AGR-1, began in the ATR in December of 2006 and ended in November 2009.

Post-irradiation examination (PIE) of the AGR-1 fuel is currently in progress at INL and Oak Ridge National Laboratory (ORNL). The PIE focuses on assessing the fuel performance during the irradiation, primarily by quantifying the level of fission product release from particles and fuel compacts and characterizing irradiation-induced changes to the fuel kernel and coating microstructures in order to better understand potential causes of coating failures [4-9].

A key component of TRISO fuel performance evaluations is post-irradiation safety testing, in which fuel specimens (either intact fuel elements or loose particles) are heated to elevated temperatures representative of reactor accident scenarios and the release of fission products is measured [10-17]. The tests can provide information on fission product diffusion through intact coating layers, failure rates for various coating layers, and the total fraction of fission products released at high temperatures. These data comprise a vital part of reactor design and safety assessments.

Safety testing of the AGR-1 fuel is in progress at INL and ORNL, involving a number of AGR-1 compacts spanning a range of irradiation conditions (burnup, fast neutron fluence, and irradiation temperature) and safety test temperatures between 1600 and 1800°C [18]. While peak core temperatures are generally not expected to exceed approximately 1600°C during a loss of forced coolant flow incident, temperatures as high as 1800°C are used as margin tests and to accelerate thermally driven phenomena (e.g., fission product diffusion and coating failure) to aid in determining their temperature dependence. The results of the first three AGR-1 safety tests using the recently commissioned Fuel Accident Condition Simulator (FACS) furnace at INL are presented here.

2. Experimental

2.1 AGR-1 fuel and irradiation

The AGR-1 fuel particles consist of 350 µm diameter spherical kernels comprised of a heterogeneous mixture of uranium oxide and uranium carbide (denoted UCO), with 19.7% uranium enrichment [19]. The kernels were coated with porous carbon buffer, inner pyrolytic carbon (IPyC), silicon carbide (SiC), and outer pyrolytic carbon (OPyC) layers. The coated particles were pressed into right cylindrical compacts nominally 25 mm in length and 12.3 mm in diameter containing approximately 4,100 coated particles each. The final compact consists of a uniform distribution of coated particles in a carbonaceous matrix. A baseline fuel and three fuel variants were included in the AGR-1 irradiation, with each variant fabricated by varying one step of the coating process to produce slightly different IPyC or SiC coating properties [20]. One key goal of the experiment is to identify any fuel performance differences between the fuel types, either during the irradiation or during post-irradiation high temperature safety testing, in order to support optimization of the fuel fabrication process and eventual selection of a reference fuel for qualification.

A total of 72 compacts were irradiated in the AGR-1 experiment in six separate capsules. Each capsule contained 12 compacts of a specific fuel type [21]. Each compact has a unique identifier in the format X-Y-Z that denotes the original position in the irradiation experiment. X indicates the capsule, Y indicates the vertical level within the capsule, and Z indicates the stack as described in Reference 5. The experiment completed 620 effective full power days in the reactor and achieved a compact-average peak burnup of 19.6% fissions per initial heavy metal atom (FIMA) with zero TRISO coating failures observed based on the measured fission gas release-to-birth ratios during the irradiation [22, 23].

2.2 Safety testing fuel specimens

The three AGR-1 fuel compacts selected for these tests are listed in Table 1 along with selected fuel properties and irradiation conditions. Note that Compact 6-4-1 was AGR-1 Baseline fuel, while Compacts 4-3-3 and 4-3-2 were Variant 3 fuel, which was fabricated to have a smaller SiC grain size relative to the Baseline fuel.

Table 1. Properties [24] and irradiation conditions [25, 26] for the safety test fuel specimens.

Compact ID	AGR-1 6-4-1	AGR-1 4-3-3	AGR-1 4-3-2
AGR-1 fuel type	Baseline	Variant 3	
²³⁵ U enrichment (%)	19.74 (± 0.05)	19.74 (± 0.05)	
Kernel diameter (μm)	349.7 (± 9.0)	349.7 (± 9.0)	
Buffer (μm)	103.5 (± 8.2)	104.2 (± 7.8)	
IPyC (μm)	39.4 (± 2.3)	38.8 (± 2.1)	
SiC (μm)	35.3 (± 1.3)	35.9 (± 2.1)	
OPyC (μm)	41.0 (± 2.1)	39.3 (± 2.1)	
Particles/compact	4145	4126	
Particle packing fraction (%)	36.9	36.0	
Burnup (%FIMA)	13.4	18.6	16.4
Fast fluence ×10 ²⁵ (n/m ²)	2.43	4.16	3.68
Irradiation temperature ¹ (°C)	1041	1094	1057

¹ Time-average, volume-average irradiation temperature.

2.3 Safety testing

The fuel compacts were heated in the FACS furnace located at the Materials and Fuels Complex (MFC) at INL. The FACS furnace utilizes a graphite resistance-heating element and has a refractory metal hot zone (Fig. 1). Fuel specimens are supported on a tantalum holder inside a tantalum flow tube. High purity helium is injected into the flow tube from the bottom, sweeps past the fuel specimen and out the top of the flow tube, and then flows out of the furnace to the Fission Gas Monitoring System (FGMS). A water-cooled cold finger is used to position a steel condensation plate immediately above the hot zone and in the path of helium flow as it exits the flow tube. Condensation plates are used to collect condensable fission products during a test, and can be exchanged during furnace operation so that time-dependent data on fission product release from the fuel can be obtained. The furnace is capable of heating specimens as high as 2000°C. The main furnace chamber is installed in the shielded hot cells of the Hot Fuel Examination Facility (HFEF) at MFC, and is controlled from a user workstation located adjacent to the hot cell window. The atmosphere inside of the HFEF main hot cell is argon with O₂ and H₂O impurities controlled below approximately 100 ppm.

The FGMS consists of two lead-shielded, cryogenically-cooled charcoal traps used to retain fission gases (primarily ⁸⁵Kr) that may be released from the fuel during a test and carried to the traps along with the the helium sweep gas. Each cold trap is continuously monitored with a high purity germanium gamma detector during a test to provide real-time data on the buildup of radioactive fission gases. During initial testing and calibration of the FGMS system, it was observed that a radioactive gas source injected into the FACS furnace in a helium carrier gas flow of 1L/min was first detected in the FGMS cold traps in approximately 4 minutes, with 99% of the activity collected in the traps within approximately 30 minutes.

Both the FACS furnace and FGMS have been described in further detail previously [27]. The tests presented in this paper mark the first use of these systems with irradiated fuel. In addition to providing important data on AGR-1 fuel performance at elevated temperatures, the tests were also used as a performance evaluation of the equipment and methods used at INL. Although destructive analysis of the AGR-1 compacts is often performed following safety tests [28], this will not be discussed in this paper.

Fig. 1. Cutaway drawing of the FACS furnace main chamber.

The compacts were heated in ultra-pure helium flowing at 1 L/min. The peak temperatures for the tests were 1600°C (6-4-1 and 4-3-3) and 1800°C (4-3-2). The furnace temperature ramp profile was similar for all of the tests, and included the following steps:

1. Ramp to 400°C at 120°C/h and hold for 2 h
2. Ramp to 1250°C at 120°C/h and hold for 12 h
3. Ramp to target temperature (1600 or 1800°C) at 50°C/h and hold for a nominal duration of 300 h

The total inventory of ^{85}Kr measured in the traps was decay-corrected to the end of the AGR-1 irradiation and compared with the predicted inventory in the fuel compact to express the inventory as a compact fraction. During some of the tests, small quantities of ^{85}Kr were detected in the traps prior to heating of the specimen, apparently due to influx of very low levels of contamination present in the hot cell which were not completely purged from the furnace prior to the start of the test. It is therefore unclear how much of the calculated ^{85}Kr release fractions less than approximately 10^{-6} (corresponding to total measured activity of ~100 to 1500 Bq for the AGR-1 fuel specimens used in these tests) presented in the following sections represent release from the fuel specimen or the influence of impurities unrelated to the fuel specimen. Additional testing is planned to better understand the influence of impurities in the hot cell atmosphere.

FACS condensation plates were exchanged periodically during the tests and each plate was placed in an individual clean container for transport to the analytical radiochemistry laboratory, located in a separate facility. The inventory of gamma-emitting fission products (including $^{110\text{m}}\text{Ag}$, ^{134}Cs , ^{137}Cs , ^{154}Eu , and ^{155}Eu) on the plates was determined using a high purity germanium detector. The inventory of ^{90}Sr on the plates was determined by leaching the plates in nitric acid and analyzing the solutions with gas-flow proportional counting. The inventory of fission products released from the compact during the test was converted to a fraction of the total original inventory using Eq. 1, where F is the inventory fraction on the plate, I is the decay-corrected cumulative plate inventory (in moles), e is the condensation plate collection efficiency, and P is the predicted inventory in the entire compact based on physics simulations of the irradiation experiment [25]. The collection efficiency of the condensation plates for each specific element was experimentally determined during FACS furnace testing performed previously [27].

$$F = \frac{I}{e \cdot P}$$

Eq. 1

3. Results

The experimental results for each safety test are presented below in terms of the fraction of the compact inventory released during the test. Table 2 presents the total cumulative release fractions for each isotope for each safety test. With approximately 4,100 particles per compact (the average value varies slightly between Baseline and Variant 3 fuel, as indicated in Table 1), a release fraction of approximately 2.4×10^{-4} represents 100% release from a single average particle in the compact.

Note that ^{137}Cs and ^{155}Eu data were collected from the condensation plates but are not presented in this paper. The ^{137}Cs activity was in relatively good agreement with the ^{134}Cs data for all of the tests; however, when release levels from the fuel are very low (resulting in low total activity on the condensation plates), the ^{137}Cs release fraction tends to be biased slightly higher than ^{134}Cs due to higher levels of trace ^{137}Cs contamination in the hot cells compared to ^{134}Cs . Therefore ^{134}Cs is considered to be a slightly better indicator of cesium release from the fuel specimen. The ^{155}Eu data were always in good agreement with ^{154}Eu , with the calculated release fractions from the two isotopes within 20% or less in most cases, so only the ^{154}Eu data were used in this paper to represent europium behavior and simplify the figures.

As with the other fission products discussed here, $^{110\text{m}}\text{Ag}$ fractions are based on the total predicted inventory in the compact at the end of irradiation, and the calculation does not consider any release that occurred during the irradiation.

Table 2. Cumulative release fractions at the end of each safety test.

Compact	Temperature (°C)	Duration (h)	$^{110\text{m}}\text{Ag}$	^{134}Cs	^{154}Eu	^{90}Sr	^{85}Kr
6-4-1	1600	300	3.2×10^{-2}	1.2×10^{-4}	2.2×10^{-3}	1.9×10^{-4}	1.9×10^{-6}
4-3-3	1600	300	3.4×10^{-1}	6.8×10^{-7}	8.2×10^{-4}	7.6×10^{-4}	1.4×10^{-6}
4-3-2	1800	277 ^a	1.4×10^{-1}	1.2×10^{-3}	1.8×10^{-2}	9.2×10^{-3}	4.7×10^{-4}

^a Test 4-3-2 was terminated approximately 1 day before the nominal duration of 300 h due to the apparent onset of multiple TRISO failures.

3.1 Compact 6-4-1

Compact 6-4-1 was heated to 1600°C and held for a duration of 300 h. Fission product release fractions are plotted in Fig. 2 along with the sample temperature. Approximately 3% of the total predicted compact $^{110\text{m}}\text{Ag}$ inventory was released very early in the test (the majority of this was released by the end of the initial 12-h hold at 1250°C). After approximately 12 h at 1600°C, very little additional $^{110\text{m}}\text{Ag}$ release was observed, and the final release fraction was 3.2×10^{-2} . The fraction of ^{134}Cs released during the ramp up to target temperature (i.e., based on the inventory on the first two plates) was very low ($<10^{-5}$). However, after approximately 12 h at 1600°C a notable increase occurred. At an elapsed time of approximately 120 h the release fraction had increased to 1.13×10^{-4} , but further release was very low for the remainder of the test. The final ^{134}Cs release fraction was 1.2×10^{-4} . The ^{154}Eu release during the ramp steps was $<10^{-5}$. Once reaching 1600°C, the release rate was almost constant for the remainder of the test, with a final release fraction of 2.2×10^{-3} . Strontium behaved in a similar manner to europium, with only a slight decrease in release rate during the 1600°C hold and a final ^{90}Sr release fraction of 1.9×10^{-4} . The ^{85}Kr release rate was low and relatively constant for the duration of the test, with a final release fraction of 1.9×10^{-6} .

Fig. 2. Fractional release of fission products during the 1600°C test of AGR-1 Compact 6-4-1 (Baseline fuel). The dashed line represents the release fraction equivalent to the inventory in a single average particle.

3.2 Compact 4-3-3

Compact 4-3-3 was heated to 1600°C and held for 300 h. Fission product release fractions are plotted in Fig. 3 along with the sample temperature. A problem developed with several limit switches on the FACS condensation plate transfer arm during the test, which prevented plate exchanges starting roughly half-way through the test. The test was therefore completed without further condensate plate exchanges after an elapsed time of 190 h. Consequently, a single condensation plate was used to collect solid fission products released from the fuel during the last ~140 h of the test.

Approximately 34% of the total predicted compact $^{110\text{m}}\text{Ag}$ inventory was released during the ramp to 1600°C, with very little additional release for the remainder of the test. The ^{134}Cs release rate was extremely low and nearly constant for most of the hold at 1600°C, reaching a final release fraction of 6.8×10^{-7} . Strontium and europium release behavior were similar, with a nearly constant release rate once the temperature reached 1600°C. The final release fractions were 7.6×10^{-4} and 8.2×10^{-4} for ^{90}Sr and ^{154}Eu , respectively. The ^{85}Kr release was very similar to that from Compact 6-4-3, with a nearly constant release rate throughout the test and a final release fraction of 1.4×10^{-6} .

Fig. 3. Fractional release of fission products during the 1600°C test of AGR-1 Compact 4-3-3 (Variant 3 fuel). The dashed line represents the release fraction equivalent to the inventory in a single average particle.

3.3 Compact 4-3-2

Compact 4-3-2 was heated to 1800°C and held for 277 h. The fission product release fractions are plotted in Fig. 4 along with the sample temperature. The silver release behavior during the latter portion of the test differed from that observed in the 1600°C tests. The $^{110\text{m}}\text{Ag}$ release fraction immediately following the ramp to 1800°C was 6.1×10^{-2} , followed by a period of approximately 65 h with very little additional release. After a total elapsed time of approximately 100 h, the $^{110\text{m}}\text{Ag}$ release rate began to increase, continuing until the end of the test and resulting in a final release fraction of 1.4×10^{-1} . Once the temperature reached 1800°C, the ^{154}Eu and ^{90}Sr release rates were relatively constant until an elapsed time of approximately 155 h (~120 h at 1800°C), after which the release rate noticeably increased. The final release fractions were 1.8×10^{-2} and 9.2×10^{-3} for ^{154}Eu and ^{90}Sr , respectively. The ^{134}Cs release was very low during the initial portion of the test, reaching a release fraction of 8.8×10^{-7} at 142 h (~100 h at 1800°C). After this the release fraction increased significantly, initially to 3.5×10^{-4} at 214 h, followed by additional increases for the remaining duration of the test, achieving a final release fraction of 1.2×10^{-3} .

The ^{85}Kr release underwent several notable transitions, indicating loss of coating integrity in several particles. Fig. 5 highlights the different regions of interest in the ^{85}Kr release plot on a linear scale, and provides the release fraction corresponding to the different transitions. Release during the first half of the test was very low, reaching a value of 1.6×10^{-6} at 168 h (132 h at 1800°C). At 168 h an abrupt increase occurred, followed by a period of ~70 h with constant release at a rate of $1.7 \pm 0.03 \times 10^{-7} \text{ h}^{-1}$ (Fig. 5a). The initial rapid ^{85}Kr release fraction at 168 h was 7.6×10^{-6} (3% of a single particle inventory), while the total fraction of ^{85}Kr released from the start of this event until the next transition in ^{85}Kr release (at an elapsed time of 246 h) was 2.1×10^{-5} , equivalent to 8.5% of a single particle inventory (see Fig. 5a). A larger ^{85}Kr release was observed at 246 h (Fig. 5b). In this instance, the fraction released during the discrete event at 246 h itself was 2.0×10^{-4} (83% of a single particle inventory). As described in detail in the next section, it is believed that the ^{85}Kr release between 168 and approximately 250 h was from the same particle; the total ^{85}Kr release fraction during this period was 2.2×10^{-4} , equivalent to 92% of a single particle inventory (see Fig. 5b).

Starting at ~253 h a small release (release fraction 1.2×10^{-5} , equivalent to 5.0% of a single particle inventory) was observed, followed by ~38 h of constant ^{85}Kr release (with release rate of $1.7 \pm 0.2 \times 10^{-7} \text{ h}^{-1}$, which is the same as that observed between 175–245 h), as shown in Fig. 5c. The cumulative ^{85}Kr release fraction during this time period was 1.9×10^{-5} (7.8% of a single particle inventory). At ~293 h a large ^{85}Kr release was observed, constituting a release fraction of 2.2×10^{-4} . The total release fraction between 253 and ~300 h was 2.4×10^{-4} (100% of a single particle inventory, see Fig. 5d).

A final small ^{85}Kr release was detected at 313 h as the test was terminating (Fig. 5d). The incremental fractional release was measured to be 9.2×10^{-6} (3.8% of a single particle inventory), similar in magnitude to the releases observed at 168 h and 253 h. No additional ^{85}Kr releases were observed after 313 h, since the test was terminated at this stage and the temperature rapidly decreased.

Fig. 4. Fractional release of fission products during the 1800°C test of AGR-1 Compact 4-3-2 (Variant 3 fuel). The dashed line represents the release fraction equivalent to the inventory in a single average particle.

Fig. 5. Selected regions of the ^{85}Kr release data for Compact 4-3-2 (1800°C). Labels indicate the release fraction associated with various transitions and the slope of selected portions of the data.

4. Discussion

4.1 Krypton and cesium

Krypton is effectively retained by any intact dense coating layer (either pyrocarbon or SiC), even at temperatures between 1600–1800°C [10, 13]. However, complete breach of all three dense TRISO coating layers (referred to in this discussion as “TRISO failure”) would result in rapid release of krypton from the particle, with the degree of krypton retention in the kernel dependent on temperature. Cesium is retained primarily by intact SiC, while intact pyrocarbon provides very little retention, making cesium the main indicator of SiC layer degradation or failure [13, 29]. Therefore, the release behavior of these two elements is the key metric of coating integrity during the safety tests. A particle that experiences a failure of the SiC layer while still maintaining at least one pyrocarbon layer intact would be expected to release a significant inventory of cesium while still experiencing very good retention of krypton at 1600°C.

Fig. 6 shows the ^{134}Cs release fraction and release rate for each of the three tests (data points for the release rate have been placed at the middle of each condensation plate exchange interval to represent the average rate for the interval). The ^{134}Cs release during the 4-3-3 test was extremely low, indicating no failed coating layers and excellent retention by intact SiC even after 300 h at 1600°C. The early release of ^{134}Cs during the 6-4-1 test, along with the very low ^{85}Kr release, suggests at least one particle experienced a failed SiC layer early in the test but maintained an intact pyrocarbon layer throughout the test. The total fraction released by an elapsed time of approximately 120 h (1.1×10^{-4}) is equivalent to approximately 45% of a single particle inventory. While it is not possible to determine the exact number of particles with SiC failure based solely on the observed release, the level (significantly less than the inventory of one particle) suggests that this was a single particle, and also suggests appreciable retention of cesium either within the particle or in the compact matrix. Multiple safety tests performed at ORNL followed by post-

safety test examination of particles with breached SiC have shown similar partial releases of cesium, with most of the balance still retained inside the particle [7]. From an elapsed time of 140 h until the end of the 6-4-1 test, the additional released fraction amounted to only 2.7×10^{-6} , indicating that cesium release from the particle was essentially complete within ~ 100 h at 1600°C and that no further appreciable releases of cesium from the compact occurred. It is possible that this particle had coating damage prior to the test that lead to failure of the SiC layer immediately upon heating to 1600°C . Damage in coating layers due to irradiation has been previously observed in as-irradiated AGR-1 particles for which no evidence of complete in-pile failure was observed [4].

Fig. 6. ^{134}Cs release fraction and fractional release rate (h^{-1}) for each safety test.

The ^{85}Kr release during both 1600°C tests was very low, indicating excellent retention by the TRISO coating layers. In addition, the ^{85}Kr release fraction for the 6-4-1 test was only marginally higher than that from 4-3-3, indicating only negligible krypton release through the intact pyrocarbon layer(s) of the particle with failed SiC at this temperature.

Analysis of the krypton and cesium data from the 4-3-2 test provides information on the types of coating failures that occurred during this test. Note that the timing of the ^{85}Kr release at 168 h roughly corresponds to the rapid release of ^{134}Cs that occurred at an elapsed time of ~ 150 h (Fig. 4). This suggests that the SiC layer of a particle failed at this point, causing a significant portion of the ^{134}Cs inventory to be rapidly released because both the IPyC and OPyC provide little impedance to Cs transport. With the loss of the SiC layer, the ^{85}Kr was diffusing only through pyrocarbon. The data also demonstrate that cesium

release from this particle with a SiC failure preceded the krypton release as long as one of the pyrocarbon layers remained intact. The krypton undergoes an induction time (diffusive breakthrough) before diffusion through the pyrocarbon is detected. This behavior has been observed in previous TRISO heating experiments [13].

It is noteworthy that the total fraction of ^{134}Cs released from Compact 4-3-2 at an elapsed time of 214 h (3.5×10^{-4}) corresponds to approximately 150% of the inventory of a single particle. There is some uncertainty in the reported release fractions of metallic fission products due to uncertainty in the collection efficiencies used for the FACS condensation plates, which were determined previously with surrogate samples using a test temperature of 1600°C [27]. It is possible that the cesium efficiency is higher at a test temperature of 1800°C than the value of 0.66 determined at 1600°C . For example, using a cesium collection efficiency of 1.0, the ^{134}Cs release fraction at 214 h would be 2.3×10^{-4} , or 94% of an average particle inventory. Ultimately, additional testing with the FACS furnace over a range of temperatures is needed to determine if there are variations in collection efficiency with test temperature.

The proposed chronological sequence of coating failures during the 4-3-2 safety test, involving three different particles, is summarized in Table 3, which describes the type of coating failure (SiC or full TRISO coating), the time of the first observed krypton release, and the approximate timing of the first cesium release. Note that there is some uncertainty in the timing of the first cesium release because of the condensation plate exchange interval, which was approximately 24 h.

The large ^{85}Kr releases at 246 h and 293 h were clearly the result of TRISO failure, as the amount of ^{85}Kr released was nearly equivalent to a single particle inventory in both cases. It is also likely that the first TRISO failure (^{85}Kr release at 246 h) was the same particle that first experienced a SiC failure at approximately 150 h (resulting in increased ^{85}Kr release starting at 168 h, as discussed above), indicating that the particle first experienced a breach in the SiC layer and then experienced a breach in the remaining intact pyrocarbon, since the total released ^{85}Kr fraction amounted to 92% of a particle inventory and the release rate immediately following the TRISO failure at 246 h was roughly zero (see Fig. 5).

The small ^{85}Kr release observed between ~253–293 h (Fig. 5c) is very similar to the release observed at 168 h (both in the amount of ^{85}Kr released and the release rate), and therefore is believed to be from another SiC failure. As before, it appears that the same particle then experienced failure of the remaining intact pyrocarbon at 293 h, and the total released ^{85}Kr fraction during this interval was equivalent to 100% of a particle inventory (Fig. 5d). Note that both SiC failures resulted in a small instantaneous release of ^{85}Kr followed by a steady release rate, and the TRISO failures resulted in rapid release of the expected remaining ^{85}Kr inventory in the particle.

After a slight decrease in the ^{134}Cs release rate at ~200 h (see Fig. 6), the rate increased again at ~230 h. Because it is expected that cesium is not impeded by intact pyrocarbon alone, it is unlikely that the failure of the pyrocarbon at 214 h would result in a significant increase in cesium release rate, and the likely conclusion is that this ^{134}Cs release was due to the second SiC failure (^{85}Kr release observed at 253 h). Thus, as with the first SiC failure, this is consistent with cesium release preceding the release of krypton when the SiC layer failed with the pyrocarbon still intact. Finally, after the ^{134}Cs release rate decreased at ~275 h, it began to increase once again at ~300 h (Fig. 6), corresponding to the small increase in ^{85}Kr at the end of the test. The conclusion, therefore, is that there was a third particle that experienced a SiC failure at the end of the test.

Table 3. Chronological sequence of coating failures during the 4-3-2 safety test. The times provided correspond to the first observed ^{85}Kr and ^{134}Cs release associated with each event.

Particle	Description	First observed ^{85}Kr release	First observed ^{134}Cs release ^a
Particle 1	SiC failure	168 h	143–167 h
Particle 1	TRISO failure (failure of remaining intact pyrocarbon)	246 h	N/A
Particle 2	SiC failure	253 h	214–238 h
Particle 2	TRISO failure (failure of remaining intact pyrocarbon)	293 h	N/A
Particle 3	SiC failure	313 h	287–311 h

^a Note that the range provided for the timing of the first ^{134}Cs release is governed by the condensation plate exchange interval, which was approximately 24 h after an elapsed time of 55 h.

Note that the ^{134}Cs release rate at the beginning of the 4-3-2 test was very low (Fig. 6), similar to that from Compact 4-3-3. After the first SiC failure in Compact 4-3-2 (~150 h), the release rate increased to a level more consistent with the rate at the beginning of the 6-4-1 test, also believed to be related to SiC failure as described above. Thus a release rate of $\sim 10^{-6}$ to 10^{-5} h^{-1} corresponds to release from particles with failed SiC. The release rate for Compact 6-4-1 fell to a level $< 10^{-8} \text{ h}^{-1}$ later in the test, which is more consistent with Compact 4-3-3 and representative of release levels when there are no particles with failed SiC. However, for the remainder of the 4-3-2 test the ^{134}Cs release rate remained relatively high ($> 10^{-6}$), consistent with the occurrence of several SiC failures during the remainder of this test.

The final ^{134}Cs release fraction during the 4-3-2 test was 1.2×10^{-3} . This is equivalent to the inventory in approximately 5 particles. If a higher cesium collection efficiency of 1.0 is used (discussed above), the corresponding end-of-test release fraction would be 8.0×10^{-4} (equivalent to the inventory in 3.3 particles). This corresponds better with the ^{85}Kr data, which suggests that a total of three particles experienced SiC failure by the end of the test (Table 3). The implication is that these particles released effectively their entire cesium inventory.

There is also the possibility that additional SiC failures could have occurred during the later stages of the test but were not detected, either because (a) it becomes increasingly difficult to detect the slight increase in ^{85}Kr associated with a SiC failure as the total activity in the cold trap increases (note that the ^{85}Kr increase from the third SiC failure at 313 h, for example, amounted to less than a 2% increase in the total activity in the cold trap), or because (b) the SiC failures were masked by other events occurring simultaneously (such as the second TRISO failure, for example). Post-irradiation destructive examination of Compact 4-3-2 and other AGR-1 compacts heated at ORNL is expected to shed further light on this issue; however, this is beyond the scope of this paper.

4.1.1 Krypton diffusion in pyrocarbon

The PARFUME (PARTicle FUEL MODEL) code [30] was used to determine the fractional release of krypton during the Compact 4-3-2 test for comparison with the empirical data. PARFUME is an advanced gas-cooled reactor fuel performance modeling and analysis code developed at INL. It is an integrated mechanistic code that evaluates the thermal, mechanical, and physico-chemical behavior of TRISO fuel particles during irradiation and safety testing to determine particle failure probability and fission product release. A specific methodology was developed with PARFUME to conduct comparisons between model prediction and PIE measurement of fission product release [31]. In the case of Compact 4-3-2, the methodology was applied to the prediction of the release of krypton by the first particle in the compact

assumed to have experienced a full failure, i.e., consecutive failures of its IPyC, SiC, and OPyC layers (Particle 1 in Table 3).

As explained in [31], PARFUME models fission product release from a single TRISO particle by classic Fickian diffusion using effective diffusion coefficients recommended by the IAEA [32]. Theoretically, to obtain the release from Compact 4-3-2 it is necessary to combine the release from the intact particles in the compact and the release from any particles with failed coatings. However, the contribution of intact particles to the release fraction of krypton from Compact 4-3-2 is predicted by PARFUME calculations to be 1.6×10^{-3} after 155 hours, and about 3×10^{-2} at the end of the safety test, whereas safety test data indicate a fractional release of about 1.5×10^{-6} after 155 hours (see Fig. 4). This over-prediction can be attributed to an over-estimation of the diffusivities in the kernel, pyrocarbon, and/or SiC layers used in modeling. The behavior of Particle 1 in Compact 4-3-2 provides a unique opportunity to assess the diffusivity of krypton in pyrocarbon during the period when the OPyC layer was the only intact layer remaining in that particle and, therefore, allows a comparison to the diffusivities provided in Reference 32. It foremost serves as an indicator that experimentally-observed krypton release corresponding to transport of krypton through intact OPyC can be modeled with classic Fickian diffusion.

It is assumed for the purposes of this analysis that SiC failure is preceded by failure of the IPyC layer, with the OPyC layer remaining intact for some time until its eventual failure. This assumption is based on extensive analysis of AGR-1 particles that experienced SiC failure during safety testing [7]. It is also assumed that the timing of the SiC failure can be approximated by the first observed cesium release, which occurs between 143-167 h (Table 3). The OPyC failure is estimated by the timing of krypton release, which occurs at 246 h. The modeling therefore assumed simultaneous IPyC and SiC failures at 155 h (in the middle of the range implied by the ^{134}Cs release data) and OPyC failure at 246 h. The diffusivity of krypton in the kernel provided in Reference 32 is two to three orders of magnitude higher than in PyC, so it is assumed that the release of krypton between 155 h and 246 h is driven primarily by diffusion in the OPyC. With these assumptions, the over-prediction of the release from intact particles is assumed, a priori, to be caused by an over-estimation of the diffusivity in the SiC layer, rather than in the kernel or PyC layers. Considering the low release from these intact particles observed experimentally, the calculation of krypton release from Compact 4-3-2 that is considered here does not include their contribution, and is solely due to the first particle to experience SiC failure (Particle 1). As krypton release through intact SiC is not considered in this calculation, there is no predicted release from Particle 1 during the first 155 h of the test (prior to the failure of the SiC layer).

Figure 7 shows the comparison of the fractional release of krypton in Compact 4-3-2 measured during the 1800°C test and calculated by PARFUME. The slowly increasing release between times ~175 h and ~245 h corresponds in PARFUME to diffusion through the intact OPyC, as failed layers are assumed to release krypton instantaneously and the diffusivity of krypton in the kernel is more than two orders of magnitude higher than the diffusivity in the OPyC at 1800°C. The calculated release rate of $2.4 \times 10^{-7} \text{ h}^{-1}$ between 200 and 245 h is in reasonable agreement with the rate of $1.7 \times 10^{-7} \text{ h}^{-1}$ for the experimental data over the same time interval. This result supports the interpretation that the observed release indeed corresponds to release through an intact OPyC layer following failure of the SiC layer. PARFUME calculations show that a rate of $1.7 \times 10^{-7} \text{ h}^{-1}$ could be achieved by reducing the diffusivity of krypton in PyC by about 20% or, alternatively, by reducing the diffusivity of krypton in the kernel by a factor of 200. However, such a reduction in kernel diffusivity would result in <100% krypton release from a particle with completely failed TRISO layers by the end of the safety test, while the experimental data clearly indicate rapid release of krypton from the particle once the final coating layer fails. Therefore, on first approximation, it is more likely that the slight discrepancy between PARFUME and experiment is caused by the over-estimation of the diffusivity in the OPyC layer. The relatively small correction of ~20% confirms the reasonable agreement between PIE and modeling predictions as it pertains to explaining the release of krypton through the intact OPyC layer between ~175 h and ~245 h. It also justifies a posteriori that the

over-predicted krypton release from intact particles could be attributed to an over-estimation of the diffusivity of krypton in the SiC layer, and not in the PyC layers.

The stepwise increase in released ^{85}Kr that occurs at ~ 168 h in the PIE data is not fully understood at this time, and is not duplicated by the PARFUME prediction. It is possible that additional mechanisms impact the behavior when krypton is first released from the compact that are not included in the PARFUME model.

Fig. 7. Comparison of experimental data and PARFUME prediction of krypton release from Compact 4-3-2 during the 1800°C test. Inset shows a portion of the two curves on a linear scale and the corresponding slopes in the region 200 – 245 h.

4.2 Silver

The $^{110\text{m}}\text{Ag}$ release fractions and release rates for the three safety tests are plotted together in Fig. 8. Both of the 1600°C tests were marked by similar silver behavior: rapid release of a significant fraction of the compact inventory during the ramp to 1600°C (in both tests $>96\%$ of the total $^{110\text{m}}\text{Ag}$ released during the test was released by the time the test temperature reached 1600°C), with very little additional release during the 300 h hold at 1600°C. Table 4 provides the cumulative releases during the ramp to 1600°C, during the hold at 1600°C, and the total release fraction at the end of the tests. It is believed that this behavior is indicative of silver released from particles during the irradiation but retained within the compacts outside of the particle SiC layer (i.e., within the OPyC or carbonaceous compact matrix) that is rapidly released during initial heating of the compact in the furnace. Similar behavior has been observed previously (for example, the heating test results of German UO_2 TRISO fuel element HFR-K3/1 [12]) and post-safety test leaching of AGR-1 fuel has shown that silver typically found outside of intact SiC is absent after safety testing, while gamma analysis of the particles has shown that many still contain appreciable silver [28].

It has long been known that silver can diffuse readily through intact SiC at reactor operating temperatures [34, 35]. Analysis of several AGR-1 compacts has indicated that the fraction of the $^{110\text{m}}\text{Ag}$ inventory retained in the compacts outside of the SiC layer ranged from 2.0×10^{-4} to 1.2×10^{-1} at the end of the irradiation [5,6], so values of the order of magnitude seen in these safety tests are plausible. Due to this substantial initial release and the often negligible activity of $^{110\text{m}}\text{Ag}$ on subsequent condensation plates, it is difficult to determine if any lower-level release through the intact coatings occurs during the test,

although the data indicate that such releases in these two tests would be below the level of 5×10^{-3} (Table 4).

Table 4. ^{110m}Ag release fractions during the ramp to maximum temperature, during the hold at maximum temperature, and at the end of each test.

Compact	Release fraction		
	During ramp to maximum temperature	During hold at maximum temperature	Total release fraction
6-4-1	3.1×10^{-2}	$< 1.1 \times 10^{-3}^a$	3.2×10^{-2}
4-3-3	3.4×10^{-1}	4.6×10^{-3}	3.4×10^{-1}
4-3-2	6.1×10^{-2}	7.9×10^{-2}	1.4×10^{-1}

^a Most of the plates collected during the hold at 1600°C had no measurable ^{110m}Ag activity. A minimum detectable activity was determined and used to calculate an upper bound on the fraction released during each interval.

The silver behavior during the initial portion of the 1800°C test was similar to that seen at 1600°C , with 6.1% of the compact inventory released during the ramp to 1800°C . However, the increasing release rate observed at an elapsed time of approximately 100 h (~ 60 h at 1800°C) deviates from the behavior at the lower temperature (Fig. 8), and may be an indication of diffusive release of silver from the intact particles. Table 4 provides the fractional releases during the ramp to 1800°C and the 277 h hold at 1800°C . By the end of the test, 56% of the total ^{110m}Ag release occurred during the 1800°C hold, and the release rate was approximately two orders of magnitude higher than the rate for the 1600°C tests (Fig. 8).

Fig. 8. ^{110m}Ag release fraction and release rate for three safety tests

4.3 Europium and strontium

Europium and strontium generally behaved in a similar manner in these tests. The ^{154}Eu release fractions and the instantaneous release rates during each test are plotted together in Fig. 9 for direct comparison. At 1600°C the release rate was relatively constant for both europium and strontium. As with silver, it is possible that the release at this temperature is primarily from inventory that resided in the compact outside of the SiC layer prior to the safety test. Analysis of the AGR-1 irradiation capsules and several as-irradiated AGR-1 compacts has indicated that europium and strontium were released to a certain extent through intact SiC during the irradiation [5, 6, 8]. The fractions of these isotopes found in several as-irradiated AGR-1 compacts outside of the SiC layer ranged from 2.4×10^{-4} to 1.3×10^{-2} for ^{154}Eu and 1.5×10^{-6} to 3.1×10^{-3} for ^{90}Sr [6]. For both isotopes, these values bound the cumulative release fractions found in this study at the end of 1600°C tests (Table 2). While the total release fractions for these two tests are consistent with europium and strontium release through intact SiC, it cannot be determined from the results with certainty what portion was released through SiC during the irradiation (and subsequently released from the compact during the safety tests) vs. the inventory released through intact SiC during the safety test.

Fig. 9. ^{154}Eu release fraction and release rate for three safety tests.

Strontium has commonly been found to have significant retention in the carbonaceous matrix of UO_2 TRISO spherical fuel elements during post-irradiation heating tests [12, 13]. While less discussion has

been devoted to the behavior of europium in the literature (perhaps due to the shorter half-life and the lower fission yield of ^{154}Eu relative to ^{90}Sr), ^{154}Eu has also been found in the matrix of spherical fuel elements at levels that indicate release from intact coating layers [12]. In addition, ^{154}Eu release from a spherical TRISO fuel element (HFR-EU1/3) during heating at 1600 and 1800°C has recently been reported [35]. The chemistry of both strontium and europium should be similar in UCO fuel kernels, and is expected to be significantly different compared to oxide fuel. In AGR-1 UCO, the carbide phase of these elements should persist to burnups of ~20% FIMA or more, whereas in oxide fuel both elements would be present as oxides [36]. The vapor pressure of the elements Eu and Sr is higher above their respective carbides compared to the oxides [37-41], and this may contribute to higher levels of release from UCO fuel compared to oxide fuel.

The europium and strontium behavior at 1800°C suggests that additional release through intact coatings may become significant after approximately 120 h at 1800°C (elapsed time of ~155 h), at which point the release rate began to increase during the Compact 4-3-2 test (Figs. 4 and 10). This behavior bears some similarity to $^{110\text{m}}\text{Ag}$ (discussed above). The release fractions of ^{90}Sr and ^{154}Eu in the 1800°C 4-3-2 test are plotted together on a linear scale in Fig. 10 for comparison. Note that the increase in release occurred at approximately the same time for both elements.

Fig. 10. Comparison of ^{154}Eu and ^{90}Sr release fractions from Compact 4-3-2 (1800°C).

4.4 FACS system performance

The FACS system performed well during these first safety tests with irradiated fuel. The power supply and heating element functioned as planned with no interruptions to the programmed heating profiles. Power output during the tests was very stable, indicating negligible drift in the heating element performance or thermocouple readings. The issue during the 4-3-3 test that prevented condensation plate exchanges was traced to electrical wiring in the control cabinet and has been repaired.

The FGMS also functioned extremely well during these tests. The cold traps and data acquisition systems performed as designed, and the system exhibited good sensitivity to ^{85}Kr activity in the traps. A minimum activity of $7.4 \times 10^1 \text{ Bq}$ (2 nCi) of Kr-85 is detectable during an 8-hour counting period with the FGMS. The FGMS has the feature to save spectra at frequent intervals, typically 1 hour. A longer counting time is useful during early phases of a test when the activity in the traps is very low. Once the trap activity increases above approximately 2×10^2 (5 nCi), the 1-hour interval spectra can be extracted while still maintaining relatively low uncertainty on each data point. This provides for better time resolution of krypton releases, which would be difficult to resolve with longer time steps.

5. Conclusions

The test results presented here highlight the generally excellent performance of the AGR-1 fuel at extreme temperatures. In particular, the krypton and cesium releases were approximately 10^{-3} or less after almost 300 h at 1800°C. Some of the key observations from these three safety tests are summarized below.

- Early release of silver during the tests appears to be from inventory retained in the compact matrix at the end of irradiation. Additional release during 300 h at 1600°C was minimal, and falls below the level of 5×10^{-3} . After approximately 60 h at 1800°C, the results indicate that additional release from intact particles may have been significant (an additional 7.9% of the compact inventory was released during this time).
- The release behavior of europium and strontium were very similar. The release from the compacts at 1600°C may be dominated by the inventory present in the matrix at the end of irradiation. After approximately 120 h at 1800°C, the release rate of both elements increased, suggesting that diffusion through intact coatings may be important.
- Cesium releases appear to be related to abrupt failure of the SiC layer that results in relatively rapid release of a significant portion of the particle inventory. Release levels in the absence of such failures were very low ($<10^{-6}$ for 300 h at 1600°C or 100 h at 1800°C). However, if a particle's SiC layer fails, as much as ~50% or more of its cesium inventory can be released over the course of several days. It is estimated that one such SiC failure occurred during 1600°C testing, and as many as three at 1800°C. Thus with the AGR-1 fuel it is expected that cesium release will be driven predominantly by the SiC defect and failure fractions.
- The cesium and krypton data during the 1800°C test have enabled some detailed observations about the progression of coating failures. Two TRISO failures were observed during testing at 1800°C, with approximately 100% of the expected particle ^{85}Kr inventory released during each failure. The evidence suggests that the SiC layer in these particles failed first, followed by the intact pyrocarbon. Increases in the krypton release rate associated with the loss of the SiC layer containment were observed, and cesium release preceded the krypton release, consistent with holdup of krypton by the pyrocarbon. Model predictions of krypton release through the intact OPyC layer from the first particle with failed SiC indicate a reasonable agreement in the total release rate using diffusion coefficients from Reference 32.

It is not practical to determine the dependence of post-irradiation fuel safety test performance on irradiation conditions (including burnup, fast neutron fluence, and irradiation temperature history) or fuel type (e.g., AGR-1 Baseline, Variant 3) based on the number of tests reported here. The ongoing AGR-1 safety testing campaign will include numerous compacts tested at ORNL and INL, as well as extensive post-safety-test characterization of fuel compacts and particles which provides a great deal of information regarding fuel performance during the tests and also aids in interpreting safety test fission product release data [7]. Comprehensive conclusions regarding AGR-1 fuel performance at 1600-1800°C (including broad trends in average fission product release fractions, coating failure fractions, and the effect of irradiation conditions and fuel type) will require integration of data from all of these experiments and will be reported in subsequent publications. In addition, comparison of fuel performance and fission product transport models to the AGR-1 safety testing data is in progress.

Acknowledgments

This work was supported by funding from the US Department of Energy, Office of Nuclear Energy. The support of staff at the Hot Fuel Examination Facility (FACS furnace operation) and Analytical Laboratory (condensation plate analysis) are gratefully acknowledged. Dr. Jason Harp also provided valuable support with condensation plate analysis.

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