Performance of AGR-1 High-Temperature Reactor Fuel During Post-Irradiation Heating Tests

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1. INTRODUCTION

The U.S. Department of Energy's Office of Nuclear Energy is supporting the development and qualification of tri-structural isotropic (TRISO) coated particle fuel needed to accelerate the commercialization of high-temperature gas-cooled reactors. TRISO-coated particles consist of four concentric coatings on a spherical kernel of fuel: a low-density carbon buffer, a dense inner pyrolytic carbon (IPyC), a polycrystalline SiC, and a dense outer pyrocarbon (OPyC). These layers are designed to contain fission products throughout the life of the fuel particle. The AGR-1 experiment was the first of a recent series of irradiation tests being performed on cylindrical, graphite-matrix compacts containing TRISO-coated uranium oxide plus uranium carbide (UCO) fuel kernels (Petti et al., 2010). The AGR-1 irradiation test train consisted of six independently monitored capsules, each holding twelve compacts made from one of four TRISO particle composites, one with a baseline TRISO coating design and three with minor coating process variations (Hunn et al., 2007). Post-irradiation, elevated-temperature performance tests are being conducted at both Idaho National Laboratory (INL) and Oak Ridge National Laboratory (ORNL) to explore the fission product release characteristics of this fuel at temperatures that may be reached during a depressurized conduction cooldown event, which is the accident scenario in which the fuel is exposed to the highest temperatures; the first test was conducted at 1600 °C in a pure helium atmosphere (Baldwin et al., 2014) and subsequent tests have been conducted at 1600, 1700, and 1800 °C.

The objectives of these furnace tests are as follows: (1) heat the compact in a pure helium atmosphere, and measure the high-temperature time-dependent release of gaseous and condensable fission products from the compact with the Core Conduction Cooldown Test Facility (CCCTF) at ORNL (Baldwin et al., 2014) and with the Fuel Accident Condition Simulator (FACS) furnace system at INL (Demkowicz et al., 2012), (2) measure the inventory of fission products released from the particles but retained in the compact matrix by deconsolidation-leach-burn-leach analysis (DLBL) following the furnace test, (3) examine TRISO fuel particles recovered after the deconsolidation to quantify the individual particle retention of specific fission products (including ^{110m}Ag , ^{134}Cs , ^{137}Cs , ^{144}Ce , and ^{154}Eu) with the ORNL Irradiated Microsphere Gamma Analyzer (IMGA), and (4) perform microanalysis on selected particles to better understand the correlation between particle microstructure and fission product retention. Specific details of the CCCTF (Baldwin et al., 2014) and FACS (Demkowicz et al., 2012) design and experimental methods have been previously reported and will not be repeated in this paper.

Briefly, the ORNL CCCTF and INL FACS are high-temperature furnaces designed to test the behavior of coated-particle fuel under simulated accident conditions. The CCCTF furnace (Fig. 1) operates within a dedicated hot cell and can be heated up to a maximum temperature of 2000 °C. The test compact is placed inside a graphite holder within a tantalum can that separates it from the surrounding furnace components. High-purity helium (99.9998%) is used as a sweep gas to purge the interior of the can through a series of liquid-nitrogen-cooled charcoal traps outside the CCCTF cell. The traps adsorb any condensable gases and are continuously monitored with NaI detectors to provide real-time measurement of radioactive fission gas (Kr and Xe) indicative of diffusive-release or TRISO coating failure. An airlock at the top of the furnace allows for the exchange of a water-cooled cold finger assembly that resides in the upper section of the furnace. The cold finger has a replaceable deposition cup on the end that collects volatile fission products (e.g., Cs, Ag, Pd, I, Sr, and Eu) while it is inside the furnace, thus allowing for periodic measurement of metallic fission product release.

The INL FACS furnace system has a different design (Demkowicz et al., 2012) but performs the same basic functions as the CCCTF. Fission gases released from the fuel to the sweep gas are monitored in real time using dual cryogenic traps fitted with high purity germanium detectors. Condensable fission products are collected on a plate attached to a water-cooled cold finger that can be exchanged periodically and automatically without interrupting the test. A schematic of the furnace system is shown in Fig. 2. While both furnace systems have mostly tantalum parts exposed to the test atmosphere, the CCCTF differs in that it has a graphite fuel holder surrounding the fuel compact that introduces a significant amount of graphite into the test environment. This fuel holder simulates the graphite surrounding the fuel in a prismatic block reactor and can act as a sink for the less volatile fission products such as Eu and Sr.

The fourteen irradiated AGR-1 compacts used in this furnace test series are described in Table 1. Some of the AGR-1 Baseline fuel properties are shown in Table 2, and additional data on this fuel and the three other AGR-1 fuel types are reported in references (Hunn et al., 2007) and (Phillips et al., 2010). AGR-1 Variant 1 had an increased IPyC deposition temperature resulting in decreased density and anisotropy; while Variant 3 involved an argon diluent in the hydrogen and methyltrichlorosilane used for SiC deposition to produce a finer grain structure at lower deposition temperature. The TRISO particles were overcoated prior to compacting with a resinated powder containing 64 wt% natural graphite, 16 wt% synthetic graphite, and 20 wt% thermosetting resin (Pappano et al., 2008) that was similar to the German A3-3 matrix recipe (Kania et al., 2015).

Fig. 1. Schematic of the CCCTF furnace (Baldwin et al., 2014).

Fig. 2. Schematic of the INL FACS furnace (Demkowicz et al., 2012).

Table 1. Furnace Test Series Compacts

Compact	Burnup (% FIMA ⁽¹⁾)	Fast Fluence (10 ²⁴ n/cm ²)	TAVA ⁽²⁾ (°C)	Test Temp (°C)	Fuel Type ⁽³⁾	Test Lab
6-2-1	14.2	2.87	1135	1600	B	ORNL
6-4-1	13.4	2.43	1041	1600	B	INL
6-4-3	13.4	2.46	1041	1600	B	ORNL
3-2-2	17.0	3.79	1019	1600	B	ORNL
3-3-2	17.0	3.80	1020	1600	B	ORNL
4-1-2	17.4	3.72	1042	1600	3	ORNL
4-3-3	18.6	4.16	1094	1600	3	INL
5-3-3	17.0	3.65	1042	1600	1	ORNL
3-3-1	19.1	4.23	1051	1700	B	ORNL
4-4-3	19.0	4.06	1059	1700	3	ORNL
3-2-3	19.1	4.28	1053	1800	B	ORNL
4-3-2	16.4	3.68	1057	1800	3	INL
4-4-1	19.0	4.00	1057	1800	3	ORNL
5-1-3	18.2	3.82	1042	1800	1	ORNL

⁽¹⁾ FIMA: fissions per initial metal atom

⁽²⁾ TAVA: Time-averaged, volume-averaged temperature

⁽³⁾ Fuel Type: Baseline (B) or Variants 1–3

Table 2. Average Baseline AGR-1 Fuel Properties

Property	Dimension
Kernel diameter	350 µm
²³⁵ U enrichment	19.74%
Buffer thickness	103.5 µm
IPyC thickness	39.4 µm
SiC thickness	35.3 µm
OPyC thickness	41.0 µm
Particles/compact	4145
Packing fraction	37%
U contamination	< 5 × 10 ⁻⁷

2. FURNACE OPERATION

Prior to conducting a run, the CCCTF furnace is rebuilt with a new tantalum inner can, gas feedline, and fuel holder so that the system begins at a very low contamination level. Following a safety test, and prior to performing a new test, the FACS furnace is heated to $T \geq 1600^\circ\text{C}$ with no fuel specimen for a duration of at least 24 h, while exchanging several condensation plates. The purpose of this procedure is to reduce the levels of fission products in the furnace hot zone from the preceding test. Analysis of the condensation plates provides verification that residual activity from the previous test does not contribute significantly to the activity detected in the subsequent test.

The general test program proceeds by ramping the furnace to 400°C under flowing helium and holding for two hours to bake out water or air that may be present in the fuel compact and furnace components. Next, the furnace is ramped to 1250°C and held for 12 h to complete the baking process and to allow the fuel to come to equilibrium near its irradiation temperature. A deposition cup/plate is usually changed at the end of this period. The furnace is then ramped from 1250°C up to the test temperature at a rate of $\sim 50^\circ\text{C/h}$ and a second deposition cup/plate exchange is performed within 1 h after reaching the test temperature. The next 2–3 cups/plates are changed at around 12 h intervals and then the intervals are increased to around 24 h for the remainder of the test.

Both furnaces have dual thermocouples so that operation can be continued if one should fail; comparison of the two readings provides an indication of any drift. In addition, the furnace power control signal is monitored for changes that would indicate a substantial temperature drift due to a malfunctioning or drifting thermocouple. The thermocouple temperature measurement uncertainty is $\pm 1\%$ of the reading.

During the entire test, the fission gas traps are continuously monitored for ^{85}Kr , with counting periods of 2–4 h. Unless there is a problem, the trap inventory is allowed to build up during the test; the traps are not emptied until the test is over, at which time the ^{85}Kr in the trap is sometimes removed for optional offline analysis. Dual traps allow the test to continue should one become plugged.

The isotopic inventories deposited on the cups/plates are measured by gamma counting followed by acid leaching for mass spectrometry of stable or long-lived isotopes and to obtain solutions for measuring the beta emitting ^{90}Sr isotope. The fraction of each isotope released from the compact that is collected on the cups/plates, versus being deposited elsewhere in the furnace, is estimated; and the measured isotopic inventory on each cup/plate is divided by this "collection efficiency" to estimate the actual time-dependent release from the compact. The change in compact inventory cannot be directly measured because the release is only a small fraction of the total inventory; therefore, a mass balance between the release measured in the furnace system and the loss from the compact is not possible.

For the CCCTF, the Ta liner and gas inlet tube are gamma counted and leached and the graphite components are ashed and leached; this provides an accounting of the fission products that escaped the compacts but did not reach the deposition cups. Before and after gamma counting of the leached Ta components allows an estimation of Ta component leaching efficiency and controls uncertainty on a case-by-case basis. Generally, gamma counting has a direct uncertainty of 10%, while beta/gamma counting from leaching has a higher uncertainty of ~25%. Strontium presents the greatest difficulty because leaching efficiency cannot be estimated with a reasonable degree of certainty due to the lack of a gamma signature. For the CCCTF at 1600°C, the collection efficiencies of the deposition cups are typically: Ag ~ 98%, Cs ~ 84%, Eu ~ 8%, Sr ~ 25%. The efficiencies increase as the test temperature increases and there is considerable run-to-run variation. The CCCTF deposition cup collection efficiencies are separately determined for each test by measuring the isotopic inventories on them and the other furnace components.

For the FACS, internal components are not measured after each test and collection efficiency is based on prior testing. Condensation plate collection efficiencies for the FACS furnace were determined at 1600°C by dedicated calibration tests during furnace development using surrogate samples (described in Demkowicz et al., 2012). The efficiencies determined in these tests, and used to determine the compact releases reported here, were: Ag 62% ($\pm 4.4\%$), Cs 66% ($\pm 21\%$), Eu 51% ($\pm 16\%$) and Sr 55% ($\pm 10\%$).

3. REPRESENTATIVE 1600 °C TESTS

In the 1600 °C test range, two tests are of particular interest since they illustrate the release behavior of cesium at this test temperature. The result of the Compact 6-2-1 test is shown in Fig. 3; this compact did not release significant cesium. In contrast, Compact 4-1-2 (Fig. 4) released significant cesium during its test due to a SiC failure, as was later verified by compact deconsolidation and identification of the failed particle with IMGA (Hunn et al., 2015). Test results are plotted as the cumulative fraction of the total calculated compact inventory (uncertainty estimated to be 7-15% depending on isotope) generated by the three-year irradiation that was released from the compact during the furnace test. Compact 6-2-1 ^{134}Cs release (a better indicator of trace levels of cesium release than ^{137}Cs because of lower hot cell contamination due to the much shorter half-life of ^{134}Cs limiting its accumulation) was below 5×10^{-6} ; while in the test of Compact 4-1-2, the collected cesium was an appreciable fraction of one particle (one particle is equivalent to a compact fraction of 2.4×10^{-4}). Neither compact released significant radioactive gas during its furnace test (^{85}Kr was near or below detection levels). This indicates that at least one pyrocarbon layer remained intact to retain gaseous fission products in the Compact 4-1-2 particle with failed SiC and no other particles in either compact had failed TRISO. Fig. 3 also illustrates that the other measured isotopes do not generally track the Cs behavior, with many particle's worth of Ag, Eu, and Sr released from Compact 6-2-1 even though the Cs release was negligible. This suggests a source of release for these elements independent of SiC containment failure, as discussed in the next section.

Fig. 3. Compact 6-2-1 test results. Cesium release was very low; higher ^{137}Cs accumulation, compared to ^{134}Cs , indicates hot cell contamination.

Fig. 4. Compact 4-1-2 test results. Cesium release above 10^{-4} indicates that this compact contained a particle whose SiC layer was not intact.

4. AGGREGATE 1600 °C TESTS

A total of eight runs at 1600 °C have been conducted to date. A major observation is that the fission products were released as the compacts came up to temperature and unless a SiC failure occurred, the Ag and Cs releases quickly tapered off with time while the Sr and Eu releases tended toward a relatively constant rate. Composite results for krypton release are not reported here because they were near or below ^{85}Kr detection levels for the entire 1600 °C test series, corresponding to a release fraction of $<5 \times 10^{-6}$ for all tests.

The overall cesium release results for the 1600 °C test series are shown in Fig. 5, where ^{134}Cs is presented because it is less influenced by hot cell contamination than ^{137}Cs , as previously discussed. Cumulative ^{134}Cs fractional release for five out of the eight compacts tested at 1600 °C was below 10^{-5} (corresponding to a few percent of a single particle's inventory). Significant cesium release was connected with failed SiC; the three compacts that released cesium each had one particle with failed SiC. The cesium releases from Compacts 4-1-2 and 3-3-2 were investigated in detail to locate the cesium-releasing particles (Hunn et al., 2015).

Fig. 5. Cesium release results for the 1600 °C test series. Note the large difference between the three compacts that released cesium due to failed SiC.

Silver release was much higher than cesium, with compacts releasing the equivalent of 10–1000 particle's ^{110m}Ag inventory (Fig. 6). Silver release did not correspond to the presence of particles with failed SiC; for example, one of the lowest Cs releasing compacts, Compact 4-3-3, had the highest ^{110m}Ag release. It should also be noted that although the release fractions are based on the calculated inventory, many compacts started the test with a reduced inventory, due to silver release during irradiation (Demkowicz et al., 2015-1).

Fig. 6. Silver release results for the 1600 °C test series. Note the large range in measured release. The abnormal Compact 3-3-2 two-step release will be explained in the next section.

Silver and cesium demonstrated a relatively rapid release behavior. When releases occurred, either as compacts were heated to 1600 °C or when SiC failures occurred early in the testing, cumulative values quickly approached their maximum and then plateaued. This is indicative of fast release from the carbonaceous material outside of intact SiC rather than slower diffusion through intact SiC. It cannot be determined if the comparatively small gradual accumulation later in the test was due to release through intact SiC or simply from residual inventory in the matrix and OPyC. The less volatile elements, europium and strontium, displayed a release behavior with a much longer rise time as compared to silver and cesium (Fig. 7 and Fig. 8).

In six of the eight 1600 °C tests, Eu releases were equivalent to 3–12 particle's inventory, even though two of those compacts (Compacts 3-3-2 and 6-4-1) only had single-particle SiC failures and the other four compacts had none. In the remaining two tests, where each compact released closer to one particle's Eu inventory, only Compact 4-1-2 had a SiC failure. The greatest Eu release was from Compact 6-2-1, which did not contain any particles with failed SiC. Thus, Eu release was not strongly connected with SiC failure.

Sr release approximately paralleled Eu release, but at a lower level (compacts that released more Eu also tended to release about the same fraction more Sr). Six compacts had releases either slightly below or significantly above one particle's worth of Sr, but only three of those (3-3-2, 4-1-2, and 6-4-1) were found to have single SiC failures. As for Eu, the greatest release was from Compact 6-2-1; thus, Sr release also was not obviously coupled to SiC failure. Recovering and measuring ^{90}Sr from furnace components was difficult so part of the wider range of releases in Fig. 8 may be due to incomplete ^{90}Sr recovery or uncertainties in the estimated deposition cup collection efficiency.

Post-test DLBL showed that europium and strontium were distributed throughout the OPyC, matrix, graphite holder, and tantalum furnace components; this observation, in conjunction with the gradual accumulation on the water-cooled collectors, suggests slow release of europium and strontium from the carbonaceous material.

As discussed above, while any significant Cs release typically corresponded to the number of particles with failed SiC, other isotopic releases (Ag, Eu, and Sr) did not appear to be related to the same mechanism. In addition, the combined cumulative heating test releases and residual inventories detected by DLBL were comparable to measured inventories in the matrix and OPyC of as-irradiated compacts (Demkowicz et al.,

2015-1). This leads to a hypothesis that, in the absence of SiC failures, the dominate source of fission products in these tests is from the inventory that has accumulated in the matrix and OPyC from diffusive release through intact SiC during irradiation. The hypothesis is strongest for Ag and Cs because of their rapid movement at temperature and very low residual post-heating inventories, but weaker for Sr and Eu because they are less volatile at temperature and had higher post-heating inventories, making it difficult to separate a modest SiC release during heating from the gradual release of a relatively large starting inventory in the matrix and OPyC.

Fig. 7. Europium release results for the 1600 °C test series. Note the relatively narrow range of values across all tests.

Fig. 8. Strontium release results for the 1600 °C test series.

5. TEMPERATURE CYCLING AND SILVER RELEASE

Some furnace problems were encountered during early testing in the CCCTF and these problems led to the discovery of some unusual silver release behavior. Fig. 9 shows one example, where it was noted during Compact 3-3-2 testing that temperature cycling at the beginning of the test led to the additional release of silver even after the initial release had plateaued. The effect was only seen with Ag and does not appear to be connected to the single-particle with failed SiC that released cesium around 100 hours into the test.

Temperature cycling during the Compact 3-3-2 test was due to furnace power supply problems at the beginning of the test and intentional thermal cycling at the end of the test to further study the effect and see if thermal cycling still resulted in silver release after the long 1600 °C soak. Even though negligible ^{110m}Ag had been released over a period of 250 h, additional silver release from thermal cycling was still observable, albeit at a much lower level (Fig. 10 shows the ^{110m}Ag on a linear scale). Further studies are being conducted to explore this effect. Two possibilities under consideration are that silver could be coming through intact SiC in a temperature range less than 1600 °C and that temperature changes or some other dynamic process that may exist during thermal ramping could be enhancing silver release through intact SiC.

Fig. 9. Temperature cycling and enhanced silver release from Compact 3-3-2. Note the increase in ^{110m}Ag at 200 h, even though it had plateaued during the initial soak at 1600 °C; the release was many times more than would come from the particle that released Cs during the initial 1600 °C soak.

Fig. 10. Silver data from plotted on a linear scale, showing enhanced silver release from Compact 3-3-2 due to temperature cycling.

6. SCOPING TESTS AT 1700 °C

Two margin tests were conducted at 1700 °C to determine if there was a significant change in release behavior compared to 1600 °C and overall behavior was found to be similar. Cesium release results are shown in Fig. 11. In the absence of failed SiC, cesium release from Compact 4-4-3 was very low, similar to the 1600 °C tests. This shows that intact SiC continues to retain cesium well at this temperature (the slight increase in cesium release at the end of the Compact 4-4-3 test was still low compared to possible hot cell contamination).

Cesium release from Compact 3-3-1 equivalent to the inventory in ~1.5 particles indicated multiple particles released cesium. IMGA survey detected two particles with failed SiC, and DLBL suggested the existence of two more that did not remain intact through the deconsolidation and leaching process to be available for survey (Hunn et al., 2015). So whereas the average cesium retention at 1700 °C appeared to be as good as it was at 1600 °C, the likelihood for individual particle failure appeared to be slightly higher. Fractional release of ^{85}Kr was again very low for this series ($< 5 \times 10^{-6}$) indicating that the particles with failed SiC retained at least one intact pyrocarbon layer throughout the furnace test. Silver, europium, and strontium also exhibited release behavior similar to that seen at 1600 °C (Fig. 12). Compact aggregate releases exceeded any extra release from the four particles with SiC failures in Compact 3-3-1 (a fractional release of 10^{-3} is about equivalent to the inventory in four particles). Like Compact 3-3-2, the heating test for Compact 3-3-1 was somewhat unusual in that there were two power interruptions during the 1250 °C soak period which resulted in additional thermal cycling. Additional silver release from thermal cycling was again observed, with each thermal cycle resulting in a modest, but measurable $^{110\text{m}}\text{Ag}$ release (Fig. 13).

Fig. 11. Cesium release results for the 1700 °C test series. Compact 3-3-1 had an estimated four particles with failed SiC.

Fig. 12. Silver, europium, and strontium release results for the 1700 °C test series.

Fig. 13. Release results for Compact 3-3-1 at 1700 °C. Note the temperature cycling during the 1250 °C phase and the increasing silver release.

7. 1800 °C TESTS

A test series was run at 1800 °C to increase the likelihood for detecting diffusive release and to further explore the SiC performance margin at this very high temperature. All four tests resulted in several particles with failed SiC. In addition, the release behavior for silver, europium, and strontium was different in the latter part of the tests from what was observed at the lower test temperatures. All of the 1800 °C-tested compacts released significant amounts of cesium due to multiple particles with failed SiC; and, in this limited sampling, the Variant 3 compacts (4-3-2 and 4-4-1), which had a finer-grained SiC microstructure (Phillips et al., 2010), released cesium later in the heating cycle (Fig. 14).

Fig. 15 shows significant ^{85}Kr release from what appears to be two Compact 4-3-2 particles with none of the three outer TRISO layers intact, and much lower, but detectable ^{85}Kr , from what might be diffusive release through particles with failed SiC and intact pyrocarbon. Note that the krypton release in the latter part of the Compacts 4-3-2 test was extremely rapid, and totaled approximately 100% of the predicted single-particle inventory in both cases. Additional details of the krypton and cesium release from this compact are discussed in another paper (Demkowicz et al., 2015-2).

Fig. 16 and Fig. 17 show the ^{154}Eu and ^{90}Sr releases from the four compacts tested at 1800 °C; two patterns are evident. As observed in the lower temperature furnace tests, europium and strontium releases from a given compact tended to track together. Total releases for these four compacts were similar and greater than the 1600 °C total releases, but the release curves start at a high rate and decrease for Baseline and Variant 1 fuel (Compacts 3-2-3 and 5-1-3, respectively) and do the opposite for the two Variant 3 compacts (implying SiC diffusion in these compacts). Although this behavior has not yet been directly connected to the SiC microstructure, it is interesting given that the Baseline and Variant 1 SiC layers were deposited using the same deposition conditions (methyl-trichlorosilane with hydrogen) and the Variant 3 SiC layers were deposited with the addition of argon gas to produce a finer grain structure (Phillips et al., 2010).

Fig. 14. Cesium release for 1800 °C tests. Note the linear scale for this and the following graphs. Variant 3 compacts showed a later release.

Fig. 15. Krypton release in the 1800 °C test series. Note that Compact 4-3-2 was releasing Cs (~100 h) before its first TRISO failure at 210 h.

Fig. 16. Europium release for the 1800 °C series. Variant 3 compacts showed increasing release rate in latter half of test.

Fig. 17. Strontium release for the 1800 °C series. Release patterns were similar to europium in Fig. 16.

Interpreting the release patterns in Fig. 16 and Fig. 17 is complicated by the possible variation in the amount of ^{154}Eu and ^{90}Sr released through intact SiC during irradiation and trapped in the compact matrix and OPyC, as it was available for early release and would be more mobile at this higher temperature. The 1800 °C heating releases were low enough that depletion of the inventory inside the particles would not be significant, and thus this would not be a reason for the reduction in the release rate from Compacts 3-2-3 and 5-1-3. One possible explanation for the decreasing release rates from Compacts 3-2-3 and 5-1-3 versus the increasing release rates from Compacts 4-3-2 and 4-4-1 would be if Compacts 3-2-3 and 5-1-3 had more Eu and Sr in the compact matrix and OPyC and less release through intact SiC, while Compacts 4-3-2 and 4-4-1 had less Eu and Sr in the compact matrix and OPyC and more release through intact SiC. Considerable variation in the europium and strontium inventory outside of intact SiC has been observed in as-irradiated compacts (Demkowicz et al., 2015-1) and the grain structure of the Variant 3 fuel is different (Phillips et al., 2010); however, further work will be required to determine if these aspects impacted the 1800 °C release patterns.

Silver release patterns also showed a difference between the Variant 3 compacts and the compacts with coarser-grained SiC (Fig. 18). The increasing release rates in the latter half of the Variant 3 furnace tests implies that diffusive release through intact SiC may have been occurring at a high enough rate that it was not obscured by contribution from $^{110\text{m}}\text{Ag}$ trapped in the OPyC and matrix at the end of irradiation. This suggests that the coating diffusive behavior at 1800 °C is significantly higher than at 1600 °C and that the two SiC variations may have different behavior at extreme temperatures.

Fig. 18. Silver release for the 1800 °C series. Like Eu and Sr, Variant 3 compacts showed an increasing Ag release rate in the latter half of the test.

8. PARTICLES WITH FAILED SiC

As already discussed, cesium releases at all test temperatures were close to background levels or dominated by individual particles with failed SiC. Locating and examining these particles were an important part of post-test analysis, and details are reported in another paper (Hunn et al., 2015). For compacts tested at ORNL, the IMGA was used to locate particles low in cesium by examining each particle's $^{137}\text{Cs}/^{144}\text{Ce}$ ratio (where ^{137}Cs is released from particles with failed SiC, while ^{144}Ce is mostly retained). Furnace releases were combined with the IMGA analysis of cesium inventory and post-test DLBL to estimate the number of particles that released cesium in each test. However, sometimes particles from 1700°C and 1800°C testing did not survive the process of electrolytic deconsolidation, acid leaching, and sieving used to separate the particles from the compact matrix prior to IMGA survey, making it difficult to account for all the cesium release and determine the exact number of particles with failed SiC in these tests. Particles with failed SiC did not release all of their cesium and what they did release varied, so furnace release results alone were not sufficient for determining the number of particles with failed SiC. Particles with SiC failures were studied by x-ray imaging and materialographic examination (Hunn et al., 2015). The two particles that released cesium during 1600 °C testing in the CCCTF were both traced to as-fabricated coating defects, while all the cesium-releasing particles recovered from the 1700 and 1800 °C CCCTF tests were found to be cases where irradiation-induced cracks in the IPyC led to palladium build up at the IPyC/SiC interface that eventually corroded through the layer when the particles were held at high temperature during furnace testing. Particles that released cesium in the FACS tests were not subjected to IMGA and post-test microstructural analysis.

9. SUMMARY

An extensive series of high-temperature inert-atmosphere testing was undertaken using irradiated compacts from the AGR-1 experiment (Petti et al., 2010). Results from furnace tests on fourteen compacts (~58,000 particles) were summarized in this paper. Three UCO fuel types with minor coating variations in the IPyC or

SiC were tested from 1600–1800 °C. All compacts performed well at these temperatures for testing periods of 300 or more hours.

Krypton release above a small fraction of a single particle's inventory was only detected in one of the compact tests, where essentially 100% of the inventory was released from two particles at 1800°C. These particles are presumed to have had failed TRISO coatings that allowed escape of the fission gas, in addition to other radioisotopes.

Cesium retention by intact SiC was excellent, even up to 1800°C; cumulative cesium releases in the absence of failed SiC were less than 5×10^{-6} and were often difficult to measure above background contamination. The only significant cesium releases were associated with individual particles with failed SiC (Hunn et al., 2015). At 1600 °C, data indicates that 3 out of ~33,100 particles experienced failed SiC layers during safety testing. At least two of these failures were associated with fracture of the SiC layer in particles with fabrication defects; the defective SiC in these two particles remained intact during irradiation, but fractured when heated to 1600°C. Significant krypton was not released from these particles because the OPyC remained intact. At 1700 °C, ~4 out of ~8300 particles and at 1800 °C, ~23 out of ~16,500 particles had failed SiC layers; except for the two fabrication defects, all other analyzed particles that showed failures were caused by palladium attack after exposure of the inside of the SiC layer by irradiation-induced fracture of the IPyC.

A working hypothesis is that europium and strontium release appeared to be dominated by the slow release of these elements from the OPyC and compact matrix, having been previously released through intact SiC during irradiation and retained outside of the SiC by these carbonaceous components. Diffusion through SiC at the test temperatures cannot be ruled out, but if it is occurring, it appears to be overwhelmed by this other component. This is an area for future work. One exception was that two of the compacts tested at 1800 °C (both with Variant 3, fine-grained SiC) showed indications of increased release rates for europium and strontium that could possibly be linked to their diffusion through the SiC layer, but further study is required to investigate this possibility.

As might be expected, ^{110m}Ag release was the most prevalent of the radioisotope releases tracked in this study, given that it is known to often exhibit high fractional release during irradiation (Demkowicz et al., 2015-1). In most cases, silver was rapidly released during the ramp up to test temperature and relatively little additional silver was released through the remainder of the test; this can be explained by rapid evaporation of silver residing in the OPyC and matrix at the beginning of the furnace test, due to previous in-pile release. A secondary release of ^{110m}Ag was observed in the two Variant 3 compacts heated to 1800 °C (similar to the increased europium and strontium release observed from those compacts); this appears to be most likely related to diffusion of silver through intact SiC. Unusual secondary release of ^{110m}Ag was also observed from a couple of compacts after thermal cycling from elevated temperatures down to room temperature and then back to the test temperature. This effect is currently being studied by additional furnace testing to explore possible mechanisms.

The fission product release behavior observed in this series of heating tests is qualitatively similar to that reported for a heating test performed on loose TRISO-coated UCO particles from the HRB-15B irradiation experiment (Bullock, 1984). Most of the kernel and coating properties were similar to the AGR-1 fuel, with the exception of a lower-density SiC that was noted to have a laminar microstructure which may have impacted diffusion. After irradiation to 21.7% FIMA at 860 °C, three sets of ten particles each were heated at 1200, 1350, and 1500 °C for over 10,000 h and accumulated release of cesium, europium, cerium, and silver was monitored as a function of time. No cesium release was detected from the UCO fuel particles (two other fuel types included in that study released cesium due to SiC failure); this result is consistent with the current study where the only significant cesium releases were associated with individual particles with failed SiC.

In the HRB-15B loose particle 1500 °C heating test, cumulative europium release from UCO fuel reached 37%; the release was noted to be uniform from all particles in the batch and conform to classical diffusion theory. Only at 1800 °C were there indications of diffusive europium behavior in the current study, while at 1600 °C, contribution from europium in the matrix and OPyC appeared to dominate the release. Perhaps longer test times would have resulted in measurable diffusion through the SiC in the AGR-1 UCO fuel particles, given the observation of significant diffusion at 1500 °C over several thousand hours in the HRB-15B particle test. Also in that study, europium release was noted at 1200 °C, with a cumulative release of 6% after 10,000 h; this is only slightly higher than the typical TAVA compact temperature over the >14,000 h AGR-1 irradiation period and could explain how the matrix acquired its europium inventory.

Significant cerium release from the compact was not observed in the AGR-1 heating tests, but post-test DLBL showed a small amount of cerium in the compact matrix ($<1 \times 10^{-3}$). This cerium release from the AGR-1 particles included both in-pile and any possible heating test releases, making it difficult to estimate the heating test release separately. Cerium release in the longer HRB-15B loose particle 1500 °C heating test was 1–2%, and approximately half of that was retained by the thin-walled graphite crucible that held the particles. This relatively lower release and strong retention of cerium by graphite is consistent between both experiments.

Silver release from the UCO particles in the HRB-15B 1500 °C heating test was anomalously low and exhibited a very long (>8000 h) breakthrough time; this behavior was conjectured to have been associated with the laminar SiC microstructure. TRISO-coated UO₂ fuel particles included in that study, with higher-density columnar-grained SiC, exhibited shorter but still relatively long breakthrough periods (>800 h) compared to the duration of the AGR-1 heating tests. This supports the hypothesis that silver release from compacts in the AGR-1 heating tests was mostly due to rapid evaporation of silver residing in the OPyC and matrix at the beginning of the test. Like europium, quantitative analysis of diffusive silver release (if any) from AGR-1 compacts would likely require a longer testing period, except possibly in cases that duplicate the secondary silver release observed at 1800 °C and during temperature cycling.

10. ACKNOWLEDGMENTS

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