SAFETY TESTS ON IRRADIATED AGR-1 COMPACTS 5-3-3 AND 5-1-3

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Introduction

Post-irradiation examination (PIE) is in progress on coated particle fuel compacts from the first Advanced Gas Reactor (AGR) Fuel Development and Qualification irradiation experiment (AGR-1). The *AGR-1 Post-Irradiation Examination Plan* (INL/PLN-2828, Rev. 1) includes safety testing of the irradiated compacts in the Oak Ridge National Laboratory (ORNL) Core Conduction Cooldown Test Facility (CCCTF) and the Idaho National Laboratory (INL) Fuel Accident Condition Simulator (FACS) to evaluate the effect of elevated temperature on the fuel microstructure and fission product retention. Safety tests involve heating compacts to maximum temperatures of 1600–1800°C for typically 300 hours, where 1600°C is the expected maximum temperature during a high-temperature gas-cooled reactor (HTGR) depressurization conduction cooldown event, and 1800°C is the expected temperature at which the performance of the SiC layer will begin to degrade.

ORNL completed the first AGR-1 post-irradiation compact safety test in FY2011, and four more safety tests were completed in FY2012. Three safety tests were completed during the first three quarters of FY2013, and two additional safety tests were completed in the last quarter of FY2013 and first quarter of FY2014 (Table 1). The first five safety tests at ORNL were performed on AGR-1 Baseline fuel from AGR-1 Irradiation Test Capsules 3 and 6. The first three FY2013 safety tests involved compacts from Capsule 4, which contained AGR-1 Variant 3 fuel, a coating variant which employed the addition of an argon diluent to the hydrogen and methyltrichlorosilane (MTS) used to deposit the SiC layer. The addition of argon results in a finer-grained SiC microstructure that is deemed to be desirable for optimum fuel performance; this coating variation has been adopted as the standard for AGR production-scale coating. The two safety tests discussed in this report were on compacts from Capsule 5, which contained AGR-1 Variant 1 fuel. The AGR-1 Variant 1 particles were similar to AGR-1 baseline, with the exception that the inner pyrocarbon layer was deposited at ~25°C higher temperature; this process change resulted in a lower IPyC sink/float density (1.853 g/cm^3 compared to 1.907 g/cm^3).

Table 1. Summary of safety tests in ORNL CCCTF

<table>
<tr><th>ORNL Test</th><th>Compact ID</th><th>Composite</th><th>Safety Test Temperature</th><th>Safety Test Summary Report</th><th>PIE Summary Report</th></tr>
<tr><td>1</td><td>6-4-3</td><td>Baseline</td><td>1600°C</td><td>HTR2012-3-027</td><td rowspan="2">ORNL/LTR-2012/397</td></tr>
<tr><td>2</td><td>3-3-2</td><td>Baseline</td><td>1600°C</td><td rowspan="4">ORNL/LTR-2012/396</td></tr>
<tr><td>3</td><td>3-2-2</td><td>Baseline</td><td>1600°C</td><td>ORNL/LTR-2012/0928</td></tr>
<tr><td>4</td><td>6-2-1</td><td>Baseline</td><td>1600°C</td><td rowspan="3">ORNL/LTR-2013/291</td></tr>
<tr><td>5</td><td>3-3-1</td><td>Baseline</td><td>1700°C</td><td>ORNL/LTR-2012/0926</td></tr>
<tr><td>7</td><td>4-4-1</td><td>Variant 3</td><td>1800°C</td><td rowspan="3">ORNL/LTR-2013/290</td></tr>
<tr><td>6</td><td>4-1-2</td><td>Variant 3</td><td>1600°C</td><td rowspan="4">TBD</td></tr>
<tr><td>8</td><td>4-4-3</td><td>Variant 3</td><td>1700°C</td></tr>
<tr><td>9</td><td>5-3-3</td><td>Variant 1</td><td>1600°C</td><td rowspan="2">ORNL/LTR-2013/603</td></tr>
<tr><td>10</td><td>5-1-3</td><td>Variant 1</td><td>1800°C</td></tr>
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The first four ORNL safety tests were all performed at 1600°C, and the fifth was conducted at 1700°C. FY2013 safety tests involved maximum temperatures of 1600°C, 1700°C, and 1800°C; the two Variant 1 compact safety tests discussed in this report were performed at 1600°C and 1800°C. The standard heating profile specified in each compact examination plan called for a controlled ramp of the furnace to the safety test temperature, with soaks at 400°C and 1250°C, a hold at maximum temperature for 300 hours, and then a controlled ramp back down to room temperature.

During the safety tests, condensable fission products (metallic elements) were collected by the CCCTF in-furnace cold finger and gaseous fission products were collected from the helium sweep gas as it passed through liquid nitrogen-cooled cold traps. Deposition cups attached to the in-furnace cold finger were periodically removed and analyzed by gamma spectrometry and the sweep gas traps were constantly monitored for gamma activity throughout each run. As originally designed, the cooled-deposition cups have a high efficiency for collection of silver and cesium, which are highly volatile at 1600–1800°C, and the sweep gas trap system has a good sensitivity to release of ^{85}Kr. Time-dependent monitoring of europium and strontium (not originally part of the CCCTF design) includes a larger uncertainty, due to the fact that these elements do not readily release from the graphite and tantalum furnace internals; analysis of the CCCTF furnace internals at the end of each test is important for accurate assessment of the total cumulative release of these elements. For the 1600°C safety tests, the fraction of europium that eventually accumulated on the deposition cups has typically been 10–13%. For the 1700 and 1800°C tests, a larger fraction of europium was collected on the cups, up to 80% for the latest 1800°C test of Compact 5-1-3.

After completion of each safety test, additional analysis is performed on the deposition cups and other CCCTF furnace internals (graphite fuel holder, tantalum furnace liner, and tantalum gas inlet line) to measure the activity from the radioactive fission products that had been transferred from the compact to these various furnace components. An average deposition cup collection efficiency is determined for each fission product by measuring the total release from the compact (summing the inventories measured on the deposition cups, graphite holder, and tantalum furnace internals), and calculating the fraction of the total that went to the cups. The average collection efficiency is used to adjust the time-dependent deposition cup data to estimate the time-dependent fission product release from the compact. The accuracy of this estimation is impacted by how much time it takes for each element to migrate to the deposition cup, compared to the cup change interval; therefore, apparent trends in the estimated time-dependent release of europium and strontium may not be representative of the actual time-dependent release from the compact, due to hold up in the graphite and tantalum furnace internals.

Overall results of AGR-1 safety testing show that the coated particle fuel continues to retain fission products generated during the three-year irradiation test, as designed. No significant gaseous ^{85}Kr release has been detected by the sweep gas analysis system in any of the safety tests completed to date. Release of an appreciable amount of ^{85}Kr (typically more than half a particle's inventory) would be indicative of a breach through all three gas-tight outer layers of the tristructural isotropic (TRISO) coating, and is the primary indicator for catastrophic particle failure during the heating test. However, some radioactive fission products have been released from the compacts during safety testing. In terms of the fraction of the calculated compact inventory at the end of irradiation (assuming no loss during irradiation), the largest fractional release of all the radioisotopes detected on the deposition cups has been ^{110m}Ag; smaller, but still significant, releases of ^{154}Eu, ^{155}Eu, and ^{90}Sr have also been typically observed. During the Compact 5-3-3 safety test at 1600°C, a very low activity from ^{134}Cs was detected on the deposition cups, and the ^{137}Cs was too low to be determined above background contamination levels. This indicates that the SiC layers in all the particles in this compact remained intact. During the Compact 5-1-3 safety test at 1800°C, ^{134}Cs and ^{137}Cs were detected at levels typical for the release from several particles with breached SiC; however, the fact that these particles did not release krypton indicates one or more pyrocarbon layer in each particle remained intact.

Compact 5-3-3 Safety Test

AGR-1 Compact 5-3-3 was irradiated at the INL Advanced Test Reactor (ATR) to an average calculated burnup of 16.9% fissions per initial metal atom (FIMA) and an average calculated fast fluence of $3.65 \cdot 10^{25}$ n/m^2, at a calculated time-averaged and volume-averaged temperature of approximately 1042°C. The Compact 5-3-3 safety test began on May 28, 2013, and ended on June 11, 2013. The compact was taken to a maximum test temperature of 1600°C and held there for 300 hours. The overall time-dependent results of the Compact 5-3-3 furnace test are shown in Figure 1. The plotted fractional release values are the cumulative amount of each fission product collected on the deposition cups,

adjusted for radioactive decay and average collection efficiency, and divided by the predicted compact inventory for each fission product at the end of the irradiation. The test was completed without interruption, and observed fission product release was similar to other 1600°C tests. Cesium release was very low; only the ^{134}Cs isotope is reported because measured ^{137}Cs activities were below the detection limit for this longer-lived isotope, which has higher background interference from several decades of accumulated hot cell contamination.

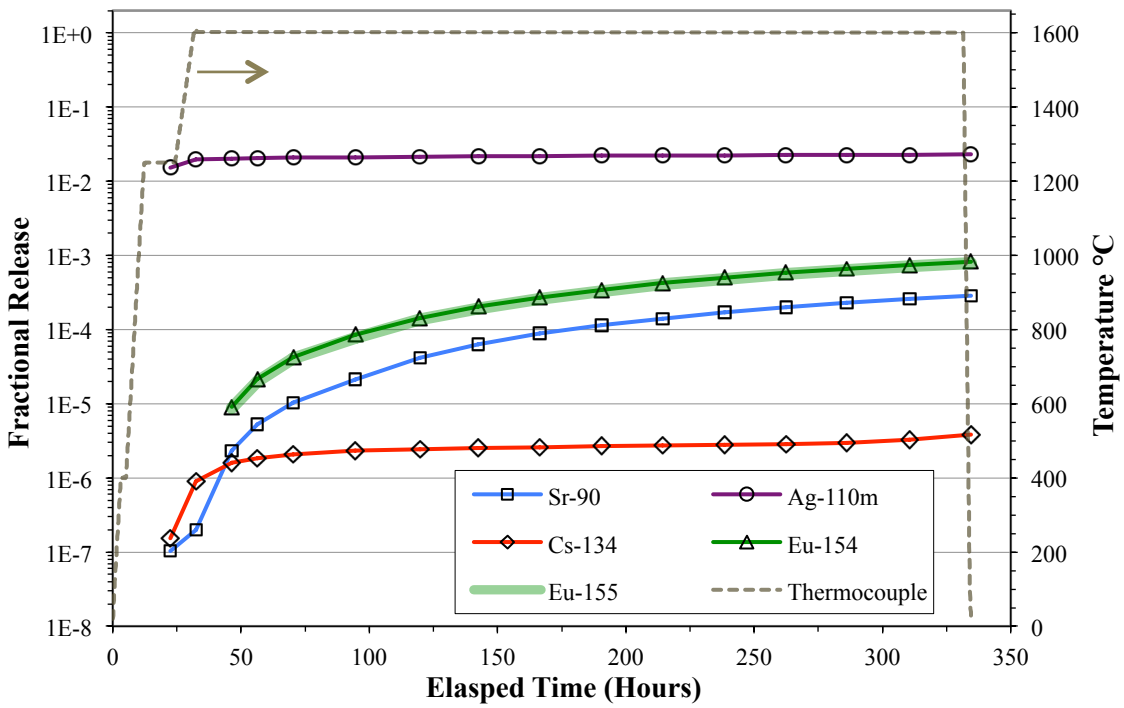

Figure 1. Release of fission products from Compact 5-3-3 during safety testing to 1600°C.

Table 2 lists the total furnace test releases for each detected fission product. Cesium releases were insignificant, indicating there were no particles in the compact with through-layer defects in the SiC; krypton release was also insignificant. Europium and strontium were released at low, but detectable levels throughout the 1600°C soak, and the two europium isotopes were in excellent agreement. Gradual release of europium and strontium throughout the 1600°C safety test is typical for AGR-1 fuel, and is probably due to slow release from the outer pyrocarbon and matrix graphite. A few percent of the irradiated compact inventory of ^{110m}Ag was released during the initial heat up to 1600°C. Constant exposure at 1600°C for 300 hours did not result in any significant additional silver release, even though subsequent PIE has shown a large fraction of the ^{110m}Ag inventory is still contained in most of the particles. This is similar to the results of other AGR-1 safety tests, and consistent with the conclusion that the silver that comes out during a normal safety test is probably already outside of intact SiC layers.

Table 2. Cumulative fission product release from Compact 5-3-3 safety test

Isotope	Fraction of Calculated Compact Inventory Released	Equivalent Particle Inventory
^{85}Kr	< 1E-6	< 4E-3
^{90}Sr	2.89E-4	1.2
^{110m}Ag	2.30E-2	95
^{134}Cs	3.80E-6	0.016
^{137}Cs	< 1E-5	< 0.04
^{154}Eu	8.28E-4	3.4
^{155}Eu	7.87E-4	3.3

Note: One particle is equivalent to a compact inventory fraction of 2.41E-4.

Compact 5-1-3 Heating Test

AGR-1 Compact 5-1-3 was irradiated at the INL ATR to an average calculated burnup of 18.2% FIMA and an average calculated fast fluence of $3.82 \cdot 10^{25}$ n/m^2, at a calculated time-averaged and volume-averaged temperature of approximately 1042°C. The Compact 5-1-3 safety test began on August 28, 2013, and ended on September 11, 2013. The compact was taken to a maximum test temperature of 1800°C and held there for 300 hours; this was the second 1800°C AGR-1 safety test (AGR-1 Variant 3 Compact 4-4-1 was tested at 1800°C in FY2013). The final heating profile and overall time-dependent release results of the Compact 5-1-3 furnace test are shown in Figure 2.

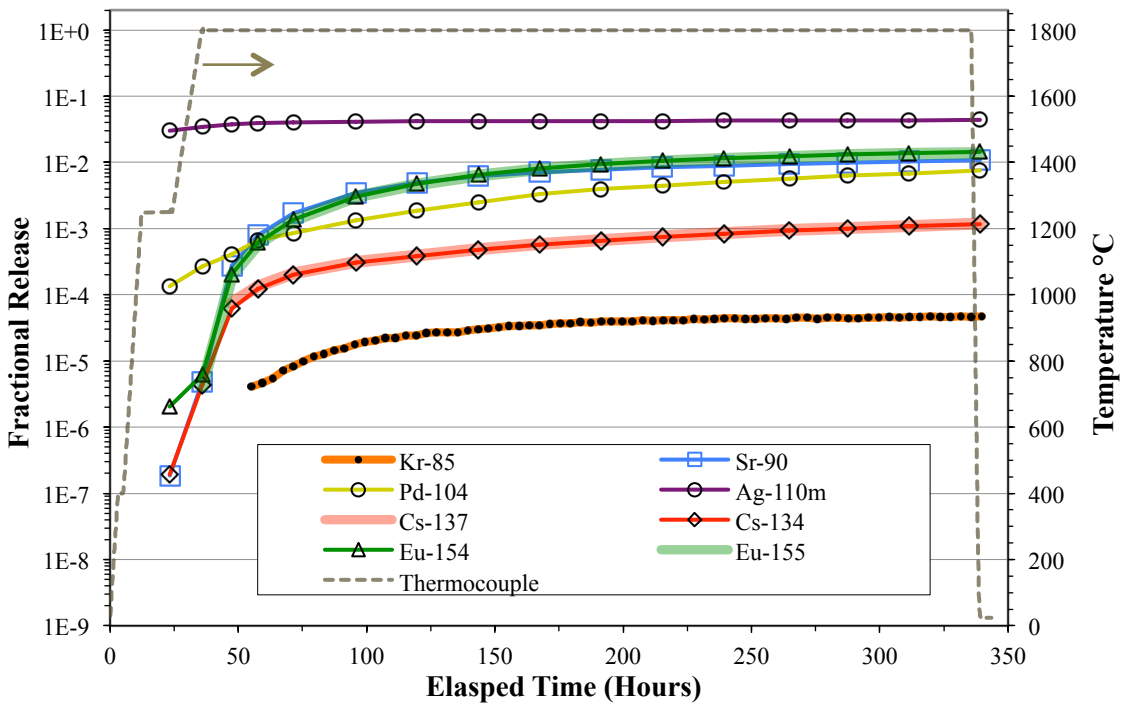

Figure 2. Release of fission products from Compact 5-1-3 during safety testing to 1800°C.

Table 3. Cumulative fission product release from Compact 5-1-3 safety test

Isotope	Fraction of Calculated Compact Inventory Released	Equivalent Particle Inventory
^{85}Kr	4.76E-5	0.20
^{90}Sr	1.07E-2	45
^{104}Pd	7.52E-3	31
^{110m}Ag	4.35E-2	181
^{134}Cs	1.17E-3	4.9
^{137}Cs	1.18E-3	4.9
^{154}Eu	1.43E-2	59
^{155}Eu	1.39E-2	58

Note: One particle is equivalent to a compact inventory fraction of 2.41E-4.

Table 3 lists the total furnace test releases for each detected fission product. Again, several percent of the irradiated compact inventory of ^{110m}Ag was released during the initial heat up, and this represented about 90% of the total ^{110m}Ag released during the test. This silver release behavior was similar to that exhibited during the 1600°C safety test of Compact 5-3-3, and probably indicates that the silver was already outside of intact SiC layers. Compact 5-1-3 released about twice as much ^{110m}Ag, compared to Compact 5-3-3, but this difference is well within the variation that has been observed for the fraction of

^{110m}Ag retained in the matrix of AGR-1 compacts after irradiation. There was no significant increase in the ^{110m}Ag release rate like that observed during the 1800°C test of Compact 4-4-1 after about 100 h at 1800°C (see Figure 3 taken from ORNL/LTR-2013/290).

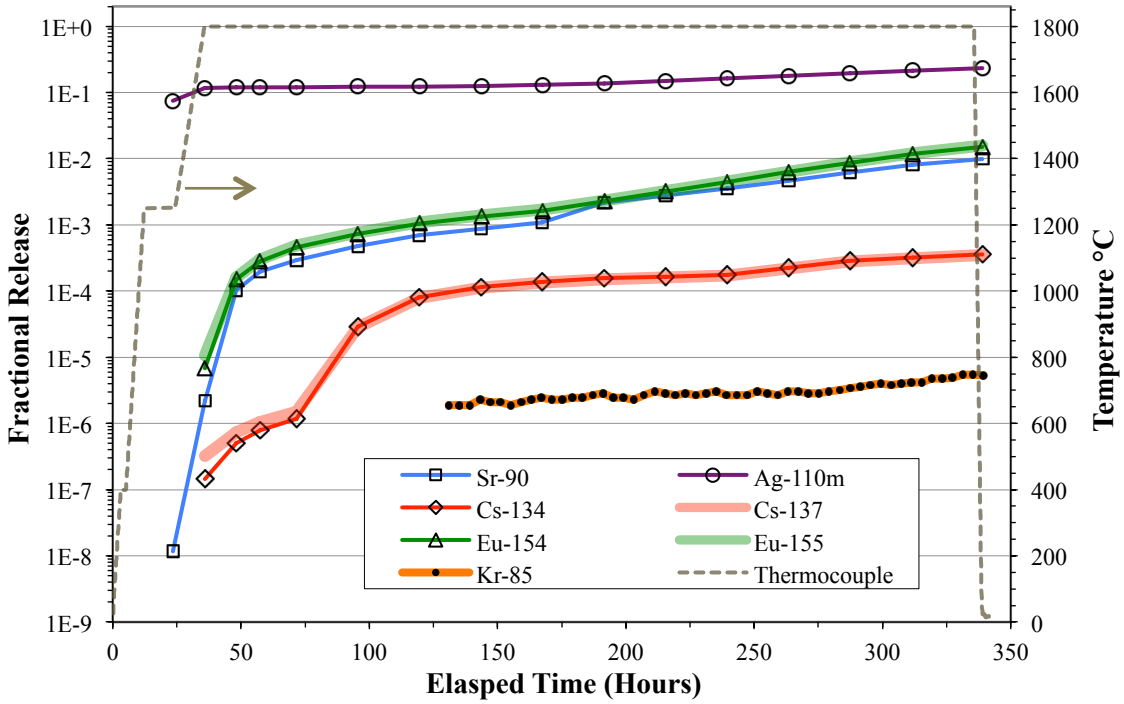

Figure 3. Release of fission products from Compact 4-4-1 during safety testing to 1800°C.

The Compact 5-1-3 safety test also did not exhibit an increase in the release rate of europium or strontium midway through the safety test, like that observed during the Compact 4-4-1 test. However, comparison between Figure 2 and Figure 3 shows that the cumulative europium and strontium release from Compact 5-1-3 was about the same as that from Compact 4-4-1 by the end of the test. Compact 5-1-3 exhibited a higher release rate earlier in the test that gradually dropped off as the test progressed. Further study is required to understand the consistent trend for higher europium and strontium release during 300-hour safety testing at 1800°C; 1800°C tests of Compacts 4-3-2 and 3-2-3 also resulted in 1–2% europium release (strontium data is not yet available for these two tests). The amount of europium and strontium released during safety testing at 1600 and 1700°C has typically been an order of magnitude lower, and post-safety test acid leaching has not shown enough of these elements remaining in these compacts (outside of intact SiC) to account for the difference. Therefore, it is possible that europium and strontium are passing through the SiC at 1800°C. An 1800°C test of individual TRISO particles is being planned to isolate the europium and strontium release from inside of particles with intact SiC shells from what may be coming from the compact matrix and OPyC during compact safety testing, due to previous release from the particles in the compact during irradiation.

A significant amount of cesium was released during the Compact 5-1-3 safety test (equivalent to the cesium contained in five particles). This indicates that a number of particles had breached SiC layers. Five of these particles have been identified by cesium inventory survey with the ORNL Irradiated Microsphere Gamma Analyzer (IMGA). The amount of cesium missing from these five particles only accounts for ~60% of the cesium released in the CCCTF, so several unrecovered particles may have also released cesium. Because of the relatively large number of particles with SiC breaches, the cesium release was spread out over the entire 1800°C test period. Figure 4 shows how the cesium release rate varied; several peaks can be resolved in the figure. The cesium released in the first peak, immediately following the ramp up to 1800°C, was probably associated with three or four particles. Subsequent increases in the cesium

release rate observed after this initial peak were probably associated with additional particles experiencing SiC coating failure.

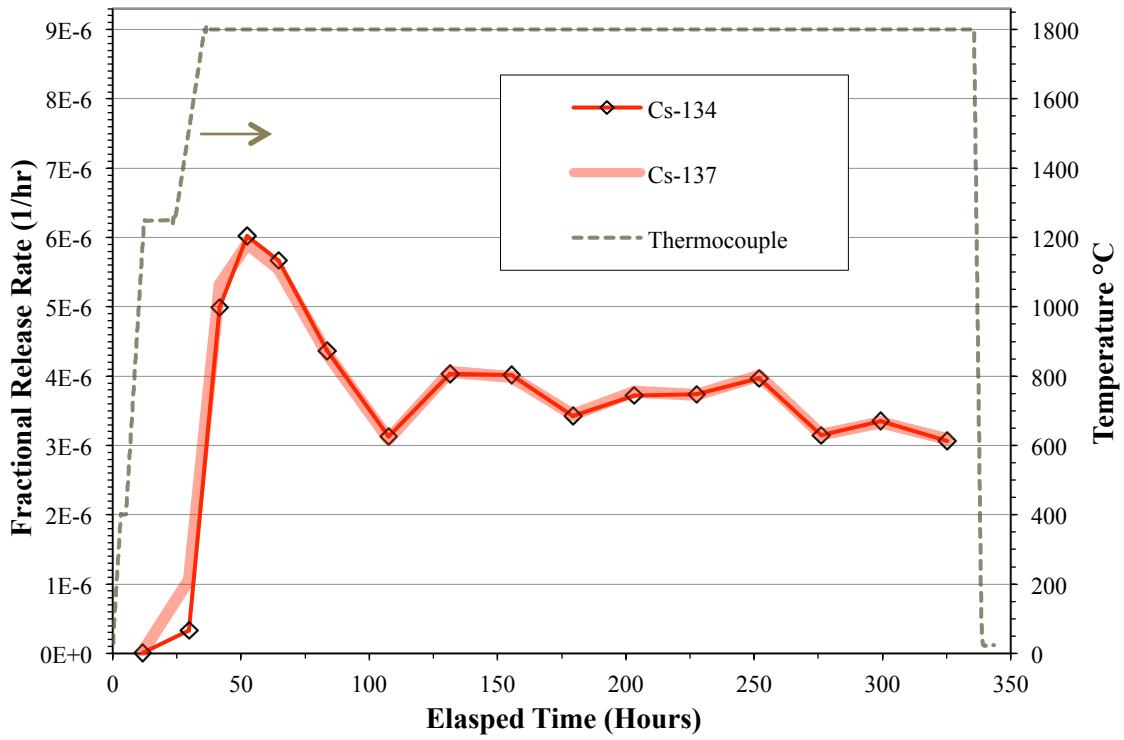

Figure 4. Release rate of cesium from Compact 5-1-3 during safety testing to 1800°C.

Figure 2 shows that there was a slow build up throughout the test of a small amount of ^{85}Kr in the sweep gas trap (20% of one particle's inventory). This is the most ^{85}Kr that has been detected in the CCCTF out of the first ten ORNL AGR-1 safety tests. The amount of ^{85}Kr released was not enough to have come from a particle with a complete TRISO-coating failure (based on historical CCCTF tests, typically >50% of one particle's inventory is rapidly released when a particle fails at safety test temperatures), so it is reasonable to assume that the particles that had breached SiC also had at least one intact dense pyrocarbon layer that could at least partially retain the ^{85}Kr. The slow release of ^{85}Kr was probably due to slow diffusion through the intact pyrocarbon of these particles.

AGR-1 PIE has found that palladium can also pass through intact SiC, and Figure 2 includes data showing a significant fractional release of ^{104}Pd during the safety test of Compact 5-1-3. Palladium release has been detected on the deposition cups and other furnace internals of previous safety tests, but quantitative results have not been available because of the difficulty separating the normally analyzed ^{105}Pd isotope from a $^{65}Cu^{40}Ar^{+}$ interference that forms in the mass spectrometer plasma (the CCCTF deposition cups are copper coated). For the Compact 5-1-3 test, additional palladium isotopes were included in all relevant analyses. Leach-burn-leach (LBL) of as-irradiated compacts has shown a good agreement between the measured compact fraction of ^{104}Pd and ^{105}Pd detected at various stages of the analysis, supporting the use of the ^{104}Pd isotope to monitor safety test release of palladium.

Conclusion

Ten safety tests have been completed in the ORNL CCCTF. The results of the latest two tests on AGR-1 Variant 1 compacts from Capsule 5 have shown results that are in general agreement with previous tests. Cesium appears to be retained well when compacts are heated during post-irradiation safety testing, even up to 1800°C, except when particles have through-wall defects in the SiC layer. The lack of significant ^{85}Kr release into the sweep gas indicates the absence of any catastrophic failures in the TRISO coatings as a whole, with the pyrocarbon layers serving as a secondary line of defense for those few particles with

defective or failed SiC. Significant quantities of silver, palladium, europium, and strontium have been observed to be released from the compacts during safety testing in the CCCTF, far beyond what can be attributed to through-wall defects in the SiC layer. For the two compacts discussed in this report, most of the released silver came out of the compacts immediately upon heating, probably coming from outside of intact SiC, due to previous irradiation-related release through the SiC and subsequent retention by the OPyC and compact matrix at the lower irradiation test temperatures. In the 1800°C test of Compact 5-1-3, the collection of palladium, europium, and strontium on the deposition cups occurred over a longer time period, and some of the observed releases may have come from inside of intact SiC coatings. Further PIE is in progress on the two safety tests summarized in this report, and results of this additional analysis will be provided in future reports.

Acknowledgments

This work was supported by the U.S. Department of Energy, Office of Nuclear Energy, under the Very High Temperature Reactor Technology Development Office Advanced Gas Reactor Fuel Development and Qualification Program. Hot cell activities were supported by the staff of the ORNL Irradiated Fuels Examination Laboratory, and analysis of deposition cups and internal furnace component leach solutions was provided by the ORNL Nuclear Analytical Chemistry & Isotopics Lab.