

Safety Testing of AGR-2 UCO Compacts 6-4-2 and 2-3-1



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Fusion and Materials for Nuclear Systems Division

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CONTENTS

Contents	iii
Revision Log	iv
List of Figures	v
List of Tables	vi
Acronyms	vii
Acknowledgments.....	viii
1. Introduction.....	1
2. Results of Safety Testing.....	3
2.1 Compact 6-4-2 versus Compact 5-2-2	3
2.2 Compact 2-3-1 versus Compact 2-2-2	7
3. Conclusion	10
4. References.....	11

REVISION LOG

Revision	Date	Affected Pages	Revision Description
0		All	Initial issue

LIST OF FIGURES

1. Release of fission products from Compact 6-4-2 during safety testing to 1600°C.	3
2. Release of fission products from Compact 5-2-2 during safety testing to 1600°C.	4
3. Ratio of $^{110\text{m}}\text{Ag}$ retained in 43 Compact 6-4-2 particles after safety testing to 1600°C.	5
4. Ratio of $^{110\text{m}}\text{Ag}$ retained in 55 Compact 5-2-2 particles after safety testing to 1600°C.	5
5. Rate of fission product release from Compact 6-4-2 during safety testing to 1600°C.	6
6. Rate of fission product release from Compact 5-2-2 during safety testing to 1600°C.	6
7. Release of fission products from Compact 2-3-1 during safety testing to 1600°C.	7
8. Release of fission products from Compact 2-2-2 during safety testing to 1600°C.	8
9. Rate of fission product release from Compact 2-3-1 during safety testing to 1600°C.	9
10. Rate of fission product release from Compact 2-2-2 during safety testing to 1600°C.	9

LIST OF TABLES

1. Irradiation conditions for AGR-2 UCO Compacts safety tested at 1600°C2

2. Radioactive isotope distribution on furnace internal components after the
Compact 6-4-2 safety test3

3. Radioactive isotope distribution on furnace internal components after the
Compact 5-2-2 safety test4

4. Radioactive isotope distribution on furnace internal components after the
Compact 2-3-1 safety test8

5. Radioactive isotope distribution on furnace internal components after the
Compact 2-2-2 safety test8

6. Cumulative releases of radioactive isotopes from AGR-2 UCO TRISO
1600°C safety tests10

ACRONYMS

AGR	Advanced Gas Reactor (Fuel Development and Qualification Program)
AGR-1	First AGR program irradiation experiment
AGR-2	Second AGR program irradiation experiment
BWXT	BWX Technologies
CCCTF	Core Conduction Cooldown Test Facility
CO	Carbon monoxide
DLBL	Deconsolidation leach-burn-leach
FACS	Fuel Accident Condition Simulator
FIMA	Fissions per initial metal atom
HTGR	High Temperature Gas-cooled Reactor
ID	Identification
IFEL	Irradiated Fuels Examination Laboratory (hot cells)
IMGA	Irradiated Microsphere Gamma Analyzer
INL	Idaho National Laboratory
LBL	Leach-burn-leach
OPyC	Outer pyrolytic carbon (TRISO layer)
ORNL	Oak Ridge National Laboratory
PIE	Post-irradiation examination
SiC	Silicon carbide (TRISO layer)
TAVA	Time-averaged/volume-averaged temperature
TRISO	Tristructural-isotropic (coated particles)
UCO	Uranium carbide/uranium oxide mixture (fuel kernels)
UO ₂	Uranium dioxide (fuel kernels)

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1. INTRODUCTION*

Post-irradiation examination (PIE) and elevated-temperature safety testing are being performed on tristructural-isotropic (TRISO) coated-particle fuel compacts from the Advanced Gas Reactor (AGR) Fuel Development and Qualification Program second irradiation experiment (AGR-2). Details on this irradiation experiment have been previously reported [Collin 2014]. The AGR-2 PIE effort builds upon the understanding acquired throughout the AGR-1 PIE campaign [Demkowicz et al. 2015] and is establishing a database for the different AGR-2 fuel designs.

The AGR-2 irradiation experiment included TRISO fuel particles coated at BWX Technologies (BWXT) with a 150-mm-diameter engineering-scale coater. Two coating batches were tested in the AGR-2 irradiation experiment. Batch 93085 had 508- μm -diameter uranium dioxide (UO_2) kernels. Batch 93073 had 427- μm -diameter UCO kernels, which is a kernel design where some of the uranium oxide is converted to uranium carbide during fabrication to provide a getter for oxygen liberated during fission and limit CO production. Fabrication and property data for the AGR-2 coating batches have been compiled [Barnes and Marshall 2009] and compared to AGR-1 [Phillips, Barnes, and Hunn 2010]. The AGR-2 TRISO coatings were most like the AGR-1 Variant 3 TRISO deposited in the 50-mm-diameter ORNL lab-scale coater [Hunn and Lowden 2006]. In both cases, the hydrogen and methyltrichlorosilane coating gas mixture employed to deposit the SiC was diluted with argon to produce a finer-grain, more equiaxed SiC microstructure [Lowden 2006; Gerczak et al. 2016]. In addition to the fact that AGR-1 fuel had smaller, 350- μm -diameter UCO kernels, notable differences in the TRISO particle properties included the pyrocarbon anisotropy, which was slightly higher in the particles coated in the engineering-scale coater, and the exposed kernel defect fraction, which was higher for AGR-2 fuel due to the detected presence of particles with impact damage introduced during TRISO particle handling [Hunn 2010].

Irradiation test compacts containing AGR-2 fuel particles were compacted at ORNL with the same resinated-graphite blend used to make AGR-1 compacts and a modified pressing process that utilized a die heated to 65°C and a new computer-controlled servo-press. Two compact lots were produced and qualified for the AGR-2 irradiation test: lot LEU09-OP2-Z contained the UCO TRISO particles [Hunn, Montgomery, and Pappano 2010a] and lot LEU11-OP2-Z contained the UO_2 fuel [Hunn, Montgomery, and Pappano 2010b]. Compared to the AGR-1 compacts, which were compacted at room temperature using a manual press, the modified AGR-2 compacting process produced compacts with reduced variability in length and higher matrix density (1.6–1.7 g/cc for AGR-2 versus 1.2–1.3 g/cc for AGR-1). Compilations of the properties data for the particles and compacts are available in pre-irradiation characterization summary reports for the AGR-1 [Hunn, Savage, and Silva 2012] and AGR-2 [Hunn, Savage, and Silva 2010] fuel composites.

The *AGR-2 Post-Irradiation Examination Plan* [Demkowicz 2013] 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 fuel microstructure, individual particle coating failure, and overall fission product[†] retention. The safety tests involve heating compacts in flowing helium to maximum temperatures of 1600, 1700, or 1800°C and holding at these temperatures for approximately 300 h. The standard test temperature of 1600°C is the expected maximum temperature during a high-temperature gas-cooled reactor (HTGR) depressurization conduction-cooldown event, while 1700°C and 1800°C tests explore the safety margin and provide additional data on mechanisms for particle coating

* Background content in this introduction section has appeared in a previous AGR-2 PIE report [Hunn et al. 2016a].

[†] In this report, the term “fission product” is used in a general sense to refer to all the post-fission isotopes remaining at the end of the irradiation test. These include: isotopes directly generated by the fission process, isotopes generated by neutron activation, isotopes generated by radioactive decay, and residual uranium.

failure, fission product diffusion, and other fission product interactions with the TRISO coatings. The first two CCCTF AGR-2 safety tests were performed on AGR-2 UO₂ Compacts 3-3-2 and 3-4-2; both were heated to 1600°C in flowing helium for 300 h and results were summarized in a previous report [Hunn et al. 2015a]. These UO₂ Compacts both exhibited multiple particle failure at 1600°C due to CO corrosion, which is in sharp contrast to the performance of the UCO compacts that have been safety tested thus far [Morris et al. 2016].

In this report, new results of 1600°C safety testing on AGR-2 UCO Compacts 6-4-2 and 2-3-1 are reported and compared to previously reported results [Hunn et al. 2016a] from the other two 1600°C safety tests on AGR-2 UCO compacts completed thus far (Compacts 5-2-2 and 2-2-2). Table 1 shows the calculated burnup in percent fissions per initial metal atom (FIMA), the fast neutron fluence (neutron energies > 0.18 MeV), and the average compact temperatures during irradiation for these four compacts. Compact 5-2-2 represents the baseline AGR-2 irradiation condition. Compact 6-4-2 was irradiated to a lower irradiation dose and temperature. Compacts 2-3-1 and 2-2-2 were irradiated under similar conditions, but at notably higher temperature than the baseline condition.

Table 1. Irradiation conditions for AGR-2 UCO Compacts safety tested at 1600°C

Compact ID ^a	Fabrication ID ^b	Fuel Type	Average Burnup ^c (%FIMA)	Fast Fluence ^c (n/m ²)	Temperature ^d (°C)
AGR-2 6-4-2	LEU09-OP2-Z049	UCO	9.26	2.21×10 ²⁵	1018
AGR-2 5-2-2	LEU09-OP2-Z128	UCO	12.34	3.39×10 ²⁵	1141
AGR-2 2-3-1	LEU09-OP2-Z125	UCO	12.63	3.42×10 ²⁵	1296
AGR-2 2-2-2	LEU09-OP2-Z075	UCO	12.55	3.39×10 ²⁵	1287

^a The X-Y-Z compact identification (ID) convention denotes the location in the irradiation test train: Capsule-Level-Stack.

^b Physical properties data for individual compacts are available and tabulated based on fabrication ID [Hunn, Montgomery, and Pappano 2010a, pages 60–69].

^c Burnup [Sterbentz 2014, table 6] and fast fluence [Sterbentz 2014, table 12] are based on physics calculations.

^d Time-averaged, volume-averaged (TAVA) irradiation temperature [Hawkes 2014, table 4] is based on thermal calculations.

Safety testing in the CCCTF furnace was accomplished with the same methods used for AGR-1 safety testing [Baldwin et al. 2012]. Compacts were placed in a graphite holder that positions the compact in the furnace and simulates the graphite that surrounds the compacts in a prismatic-block reactor. A water-cooled deposition cup located near the top of the tantalum-lined furnace chamber collected vaporized metallic elements that escaped from the compact and surrounding graphite holder. Deposition cups were periodically removed and replaced with a new cup using a maximum exchange interval of ~24 h and shorter exchange intervals for the first few cups removed after heating up to the test temperature. The cups were monitored with gamma spectrometry to track safety test progress, with particular emphasis on collected cesium inventory that would indicate SiC failure [Hunn et al. 2014a]. Gaseous fission products were collected from the helium sweep gas as it passed through a liquid-nitrogen-cooled trap that was monitored for ⁸⁵Kr because significant and rapid krypton release would indicate complete failure of a TRISO coating [Morris et al. 2014]. After completion of each safety test, additional analysis was performed to measure fission products on the deposition cups and other CCCTF furnace internals (graphite fuel holder, tantalum furnace liner, and tantalum gas inlet line). This allowed for the determination of an average deposition cup collection efficiency for each detected fission product, and this efficiency value was used to adjust the time-dependent deposition cup data to estimate the time-dependent fission product release from the compact.

2. RESULTS OF SAFETY TESTING

2.1 COMPACT 6-4-2 VERSUS COMPACT 5-2-2

Figure 1 shows the fractional release of key isotopes from AGR-2 UCO Compact 6-4-2 during safety testing at 1600°C. This figure summarizes the overall estimated time-dependent fission product release from the safety test. The plotted data points represent the measured amount of each isotope collected on a deposition cup divided by the calculated amount of that isotope expected to be present as a result of the irradiation test [Sterbentz 2014] and adjusted for the deposition cup collection efficiency by dividing by the cumulative fraction of that isotope collected on the cups throughout the test (Table 2, Row 1). Cesium-137 is not reported because the released values were too low to accurately resolve from the contributions of hot cell contamination, which is high in the Irradiated Fuels Examination Laboratory (IFEL) hot cells due to six decades of release of this abundant and long-lived isotope. Results are presented below in the same manner for the other 1600°C safety tests. Note that deposition cup collection efficiencies were significantly higher for the more recent safety tests of Compacts 6-4-2 and 2-3-1. Collection efficiencies for the other two tests were abnormally low due to CCCTF operational problems during those runs, where the helium gas flow direction was inadvertently reversed.

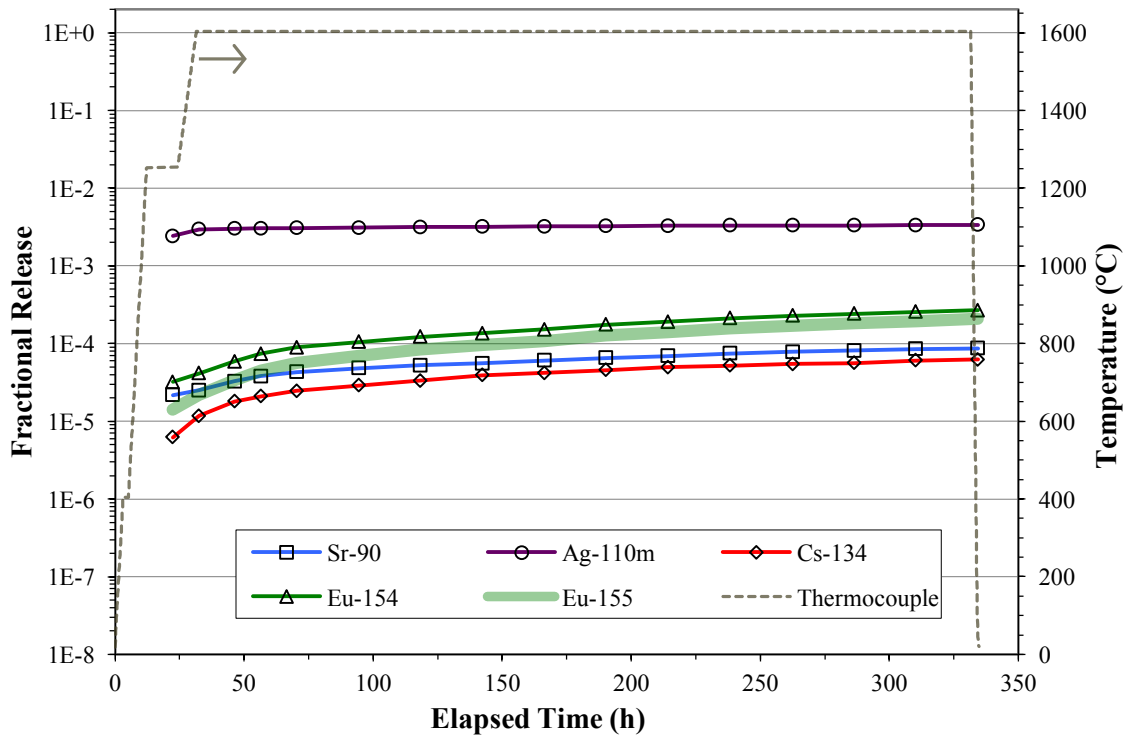


Figure 1. Release of fission products from Compact 6-4-2 during safety testing to 1600°C.

Table 2. Radioactive isotope distribution on furnace internal components after the Compact 6-4-2 safety test

Component	⁹⁰ Sr	^{110m} Ag	¹³⁴ Cs	¹⁵⁴ Eu	¹⁵⁵ Eu
Deposition cups	94.5%	100%	62.5%	59.5%	46.2%
Tantalum parts	1.9%	~0%	18.1%	~0%	~0%
Graphite holder	3.6%	~0%	19.4%	40.5%	53.8%

¹³⁷Cs is not reported for Compact 6-4-2 because it was too low to measure above background contamination.

Figure 2 and Table 3 show the previously reported results for Compact 5-2-2 [Hunn et al. 2016a]. Both tests showed the same type of silver-release behavior; namely, the typical rapid release as compacts were brought to 1600°C of silver previously released through intact SiC during irradiation and held up in the graphite matrix and outer pyrocarbon (OPyC), and then relatively negligible additional release during the 300-h hold. Compact 6-4-2 released a smaller fraction of ^{110m}Ag, which is presumably due to the lower temperature irradiation resulting in less silver release through intact SiC. Lower silver release during irradiation is supported by compact gamma analysis with the INL Precision Gamma Scanner (PGS), which showed higher fractional release from Compact 5-2-2 (78.2% versus 33.5% of the calculated inventory [Harp et al. 2016]), and particle gamma scanning with the ORNL Irradiated Microsphere Gamma Analyzer (IMGA), which showed much higher fractional retention of ^{110m}Ag in the particles from Compact 6-4-2 (Figure 3 and Figure 4).

Cesium release from Compact 6-4-2 was moderately elevated compared to Compact 5-2-2, whose ¹³⁴Cs release was less than 10⁻⁵. A ¹³⁴Cs release less than 10⁻⁵ was typical for compacts with no SiC failure during AGR-1 safety testing [Morris et al. 2014, figure 5]. However, the release from Compact 6-4-2 was lower than observed during 1600°C safety testing of AGR-1 compacts with SiC failure and only equivalent to about 20% of the average ¹³⁴Cs in a single particle, leaving it difficult to conclude from these results alone whether the ¹³⁴Cs release observed during the Compact 6-4-2 safety test was due to a particle with SiC failure or from contamination outside the SiC.

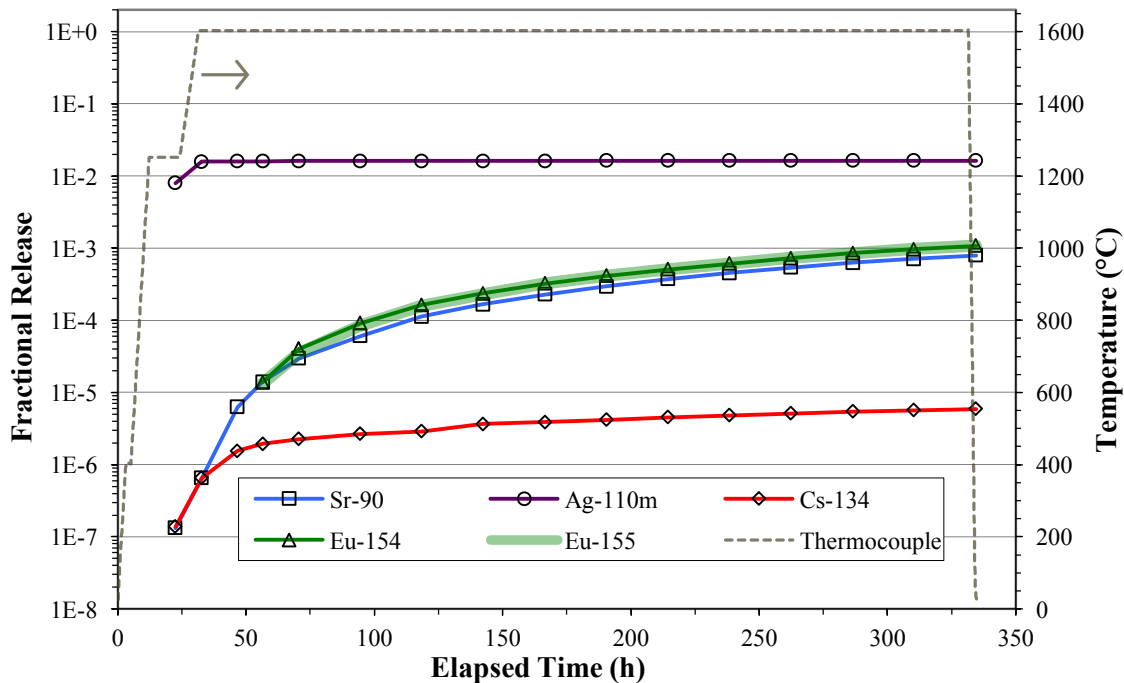


Figure 2. Release of fission products from Compact 5-2-2 during safety testing to 1600°C.

Table 3. Radioactive isotope distribution on furnace internal components after the Compact 5-2-2 safety test

Component	⁹⁰ Sr	^{110m} Ag	¹³⁴ Cs	¹⁵⁴ Eu	¹⁵⁵ Eu
Deposition cups	4.8%	32.9%	18.6%	2.1%	1.9%
Tantalum parts	22.3%	67.1%	81.4%	9.6%	9.1%
Graphite holder	72.9%	~0%	~0%	88.3%	89.1%

¹³⁷Cs is not reported for Compact 5-2-2 because it was too low to measure above background contamination.

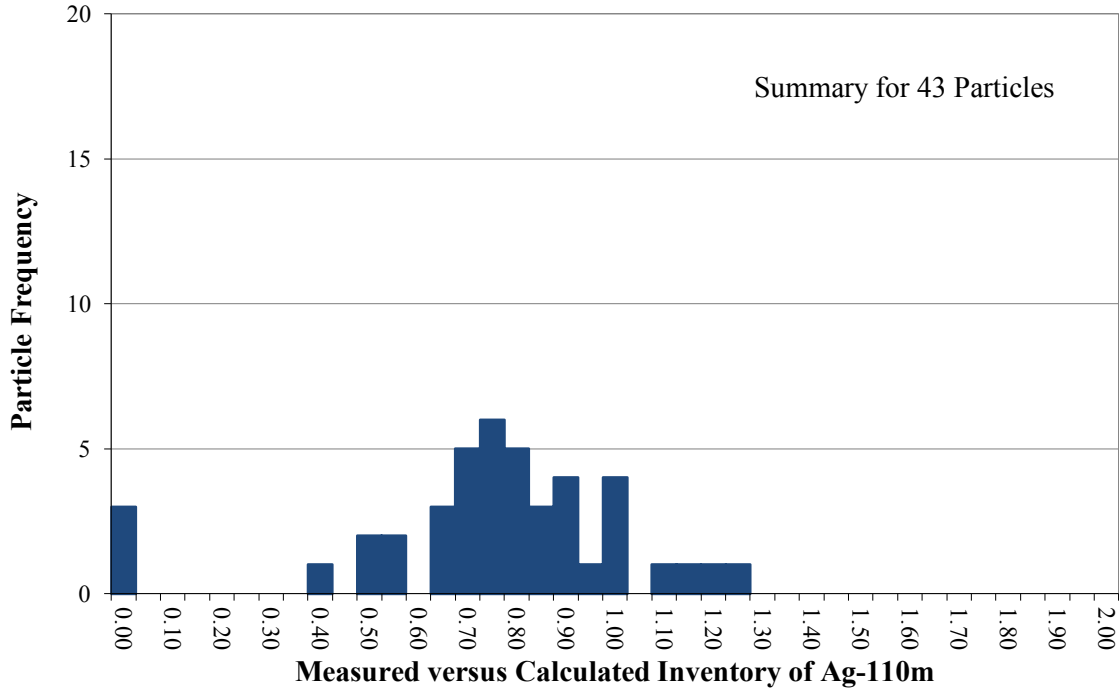


Figure 3. Ratio of ^{110m}Ag retained in 43 Compact 6-4-2 particles after safety testing to 1600°C versus the calculated inventory, adjusted for variation in fissionable material and burnup with the measured ^{137}Cs activity. Particles plotted as "zero" had a measured to calculated ratio of $M/C \approx 33\%$. The symmetric peak around M/C of 0.75 indicates the ^{110m}Ag inventory calculated value was probably higher than the true value.

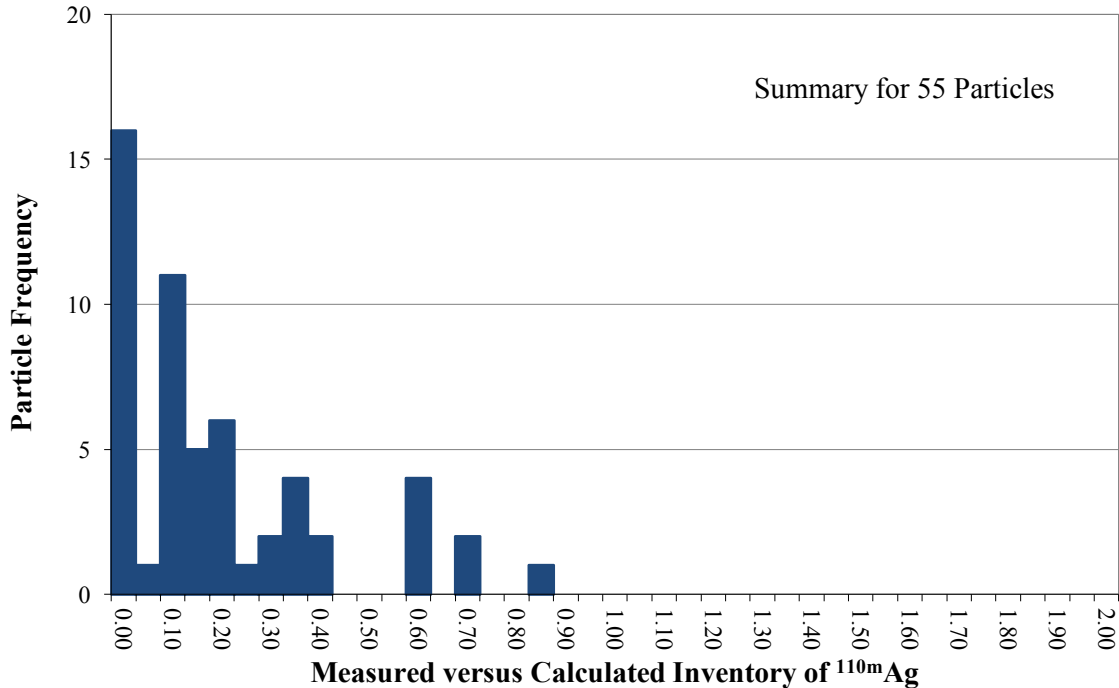


Figure 4. Ratio of ^{110m}Ag retained in 55 Compact 5-2-2 particles after safety testing to 1600°C versus the calculated inventory, adjusted for variation in fissionable material and burnup with the measured ^{137}Cs activity. Particles plotted as "zero" had $M/C \approx 7.0\%$.

Figure 5 and Figure 6 show the same data for Compacts 6-4-2 and 5-2-2 as Figure 1 and Figure 2, respectively, with the data plotted in terms of average release rate to each deposition cup. These rate plots highlight the early silver release presumed to come from previous release under irradiation. Deposition cups taken at the end of the 1250°C, 12-hour-hold and 1 h after the ramp to 1600°C had one to two orders of magnitude higher ^{110m}Ag collection rates than later cups. Typically, a failed-SiC particle would similarly result in a clearly discernable peak in the ¹³⁴Cs release rate, but this was not evident in the Compact 6-4-2 data.

After safety testing, Compact 6-4-2 was subjected to deconsolidation leach-burn-leach (DLBL) analysis as described in Hunn et al. 2013. Only 5.8% of one particle's inventory of ²³⁸U was detected outside of intact SiC by this analysis. If a particle with failed SiC was present in this compact, there should have been close to a whole particle's inventory of ²³⁸U detected in the post-burn leaches. This indicates that the observed cesium release during safety testing probably came from contamination outside the SiC, rather than a particle with failed SiC.

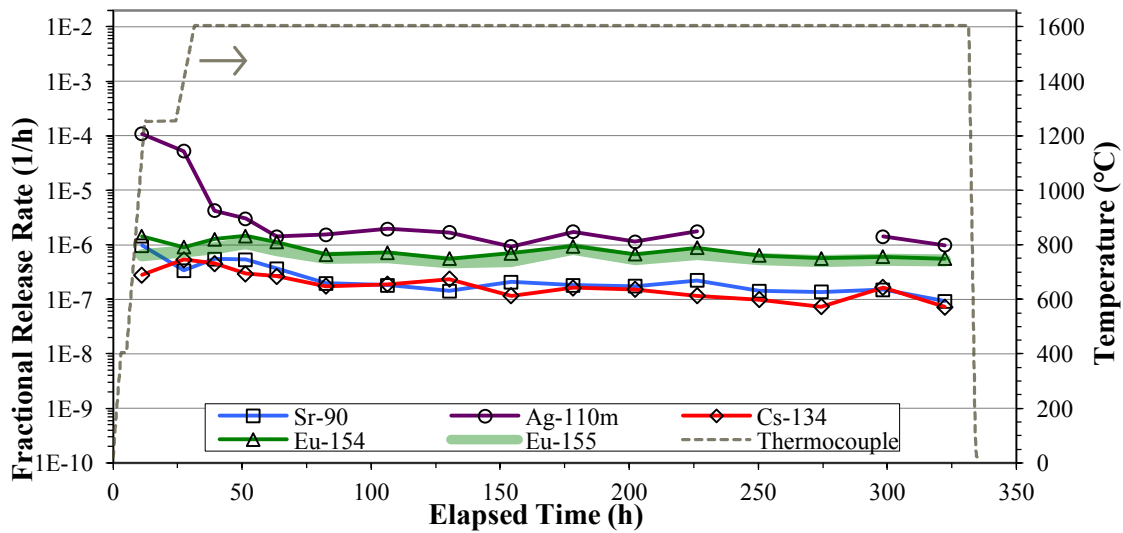


Figure 5. Rate of fission product release from Compact 6-4-2 during safety testing to 1600°C.

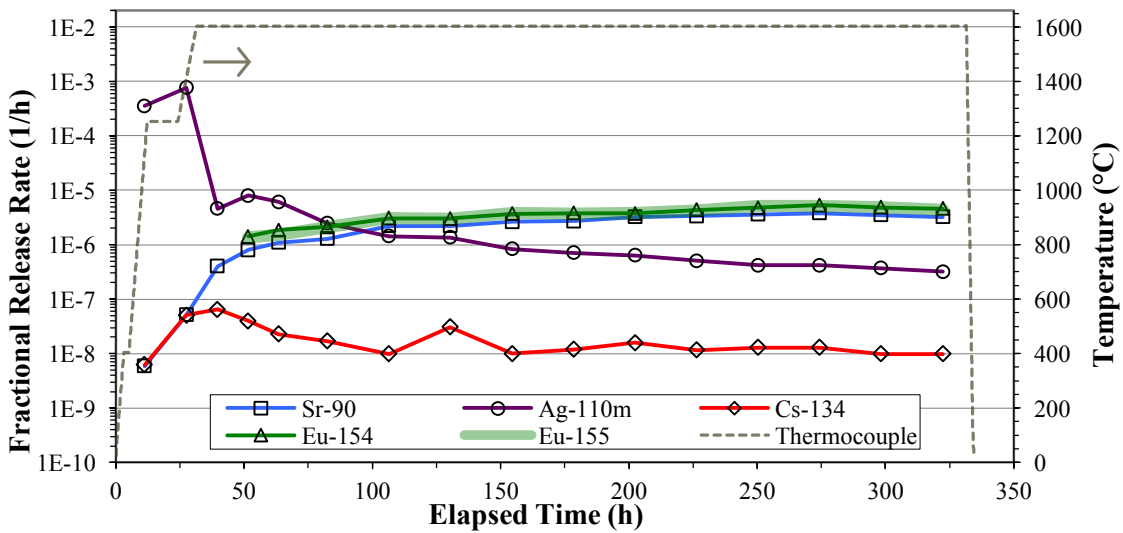


Figure 6. Rate of fission product release from Compact 5-2-2 during safety testing to 1600°C.

2.2 COMPACT 2-3-1 VERSUS COMPACT 2-2-2

Figure 7 shows the fractional release from 1600°C safety-tested Compact 2-3-1 and Figure 8 shows Compact 2-2-2 for comparison. Relative distribution of the isotopes to the furnace internals is provided in Table 4 and Table 5. Cesium release from these compacts was very low, indicating there were no particles with failed SiC. Release of silver during the ramp to 1600°C was followed by negligible additional release, while europium and strontium exhibited a roughly constant release rate after an initial period to reach equilibrium (Figure 9 and Figure 10). The primary explanation for differences in the magnitude of europium and strontium release during these two safety tests, compared to Compacts 6-4-2 and 5-2-2, is the higher irradiation temperature for the Capsule 2 compacts (Table 1), which resulted in greater quantities of europium and strontium being released during irradiation. Deconsolidation and LBL of as-irradiated compacts [Hunn et al. 2016b] has shown there was a dramatically-higher inventory of europium and strontium outside of intact SiC in the Capsule 2 compacts at the end of irradiation, compared to compacts irradiated at lower temperature in other AGR-2 capsules or in AGR-1 [Demkowicz et al. 2015].

As discussed in a previous report [Hunn et al. 2017], in the absence of coating failure, the release behavior of silver, europium, and strontium during AGR-1 safety testing was attributed to release of fission products trapped in the compact matrix graphite and OPyC that had been previously released through intact SiC during the three-year irradiation [Morris et al. 2014]. This same mechanism is the most likely explanation for the observed release of these elements from AGR-2 compacts. The relatively rapid release of silver can be explained by its low-retention in the carbonaceous OPyC and matrix at elevated temperatures. The relatively slow release of europium and strontium, along with the initial delay in its collection on the deposition cups, can be explained by the fact that these elements diffuse through the OPyC, matrix, and graphite holder at a much slower rate than silver or cesium. Table 2–Table 5 show that a large fraction of the europium and strontium released from the compacts was still in the graphite holder at the conclusion of the 1600°C safety tests.

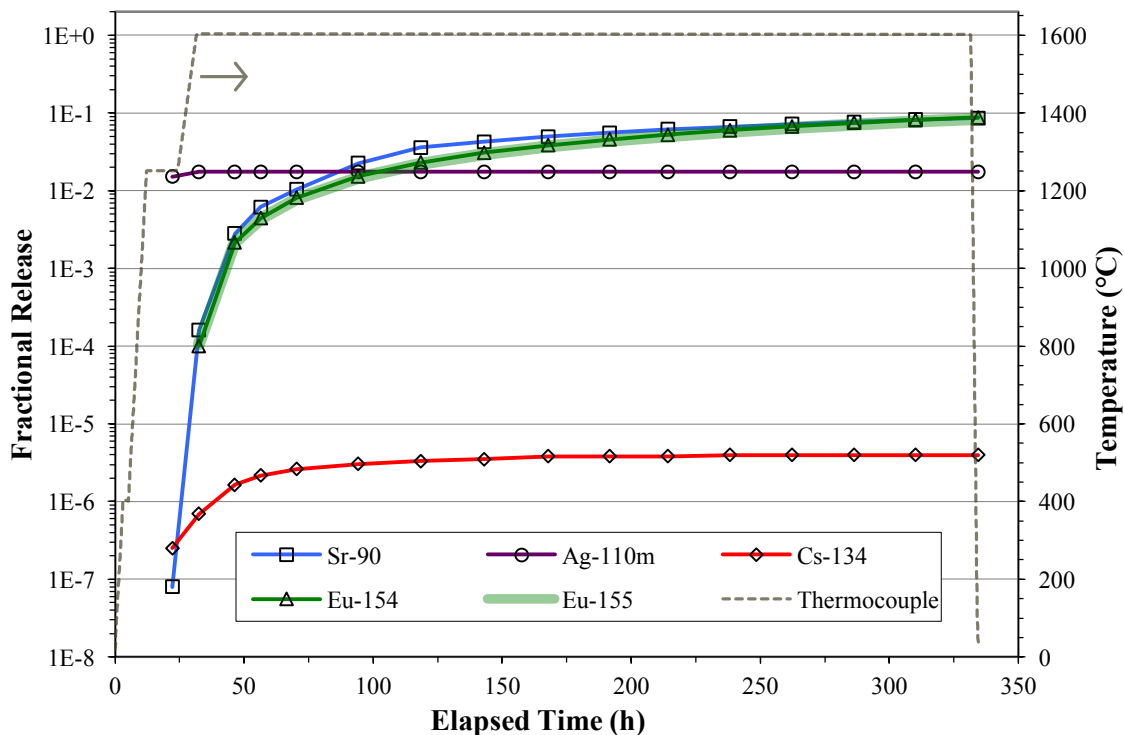


Figure 7. Release of fission products from Compact 2-3-1 during safety testing to 1600°C.

Table 4. Radioactive isotope distribution on furnace internal components after the Compact 2-3-1 safety test

Component	⁹⁰ Sr	^{110m} Ag	¹³⁴ Cs	¹⁵⁴ Eu	¹⁵⁵ Eu
Deposition cups	50.6%	100%	80.6%	22.1%	19.4%
Tantalum parts	12.7%	~0%	19.4%	12.5%	13.1%
Graphite holder	36.7%	~0%	~0%	65.5%	67.6%

¹³⁷Cs is not reported for Compact 2-3-1 because it was too low to measure above background contamination.

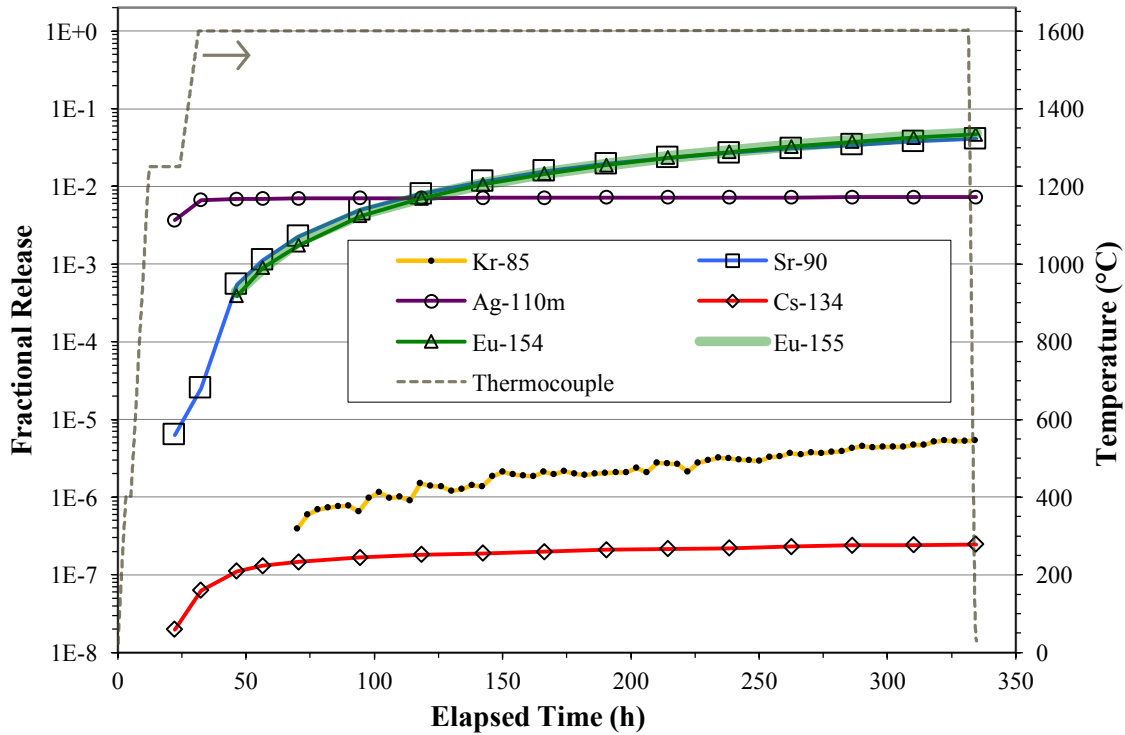


Figure 8. Release of fission products from Compact 2-2-2 during safety testing to 1600°C. The ⁸⁵Kr release was too low to have come from a particle with failed TRISO and may have been from uranium in the matrix [Hunn et al. 2016a].

Table 5. Radioactive isotope distribution on furnace internal components after the Compact 2-2-2 safety test

Component	⁹⁰ Sr	^{110m} Ag	¹³⁴ Cs	¹⁵⁴ Eu	¹⁵⁵ Eu
Deposition cups	0.09%	23.8%	84.9%	0.1%	0.1%
Tantalum parts	45.9%	76.2%	~0%	19.4%	20.3%
Graphite holder	54.0%	~0%	15.1%	80.5%	79.6%

¹³⁷Cs is not reported for Compact 2-2-2 because it was too low to measure above background contamination.

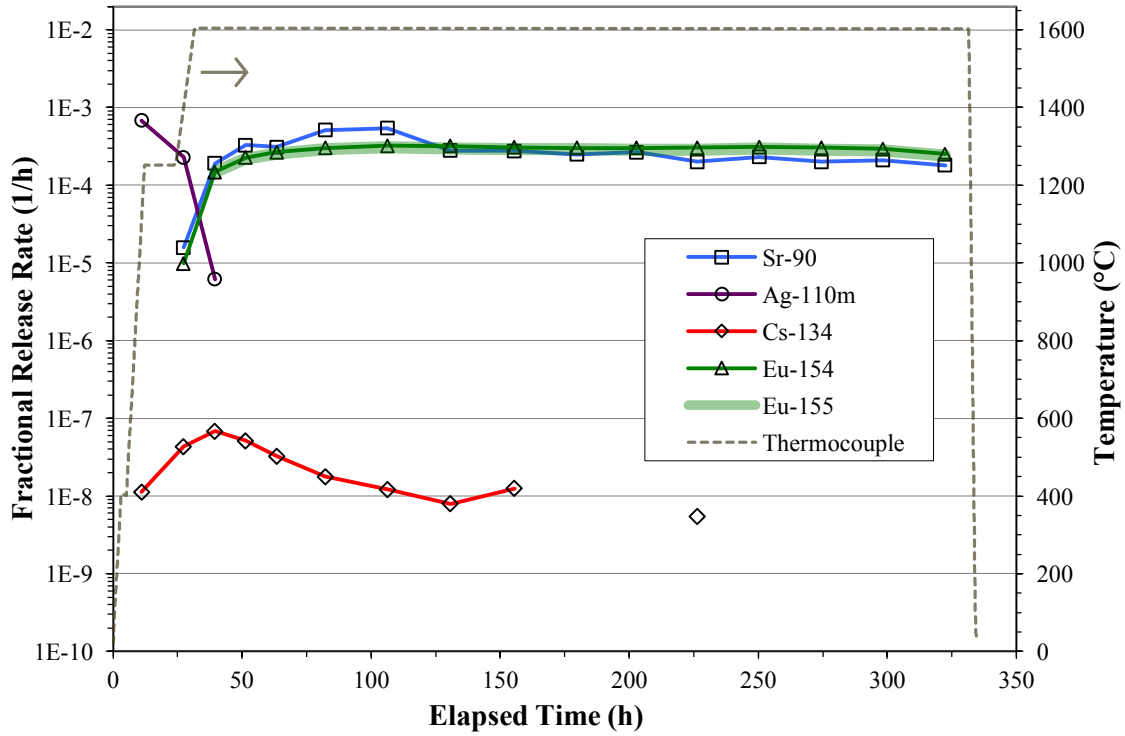


Figure 9. Rate of fission product release from Compact 2-3-1 during safety testing to 1600°C (data points with no measurable release rate are not plotted).

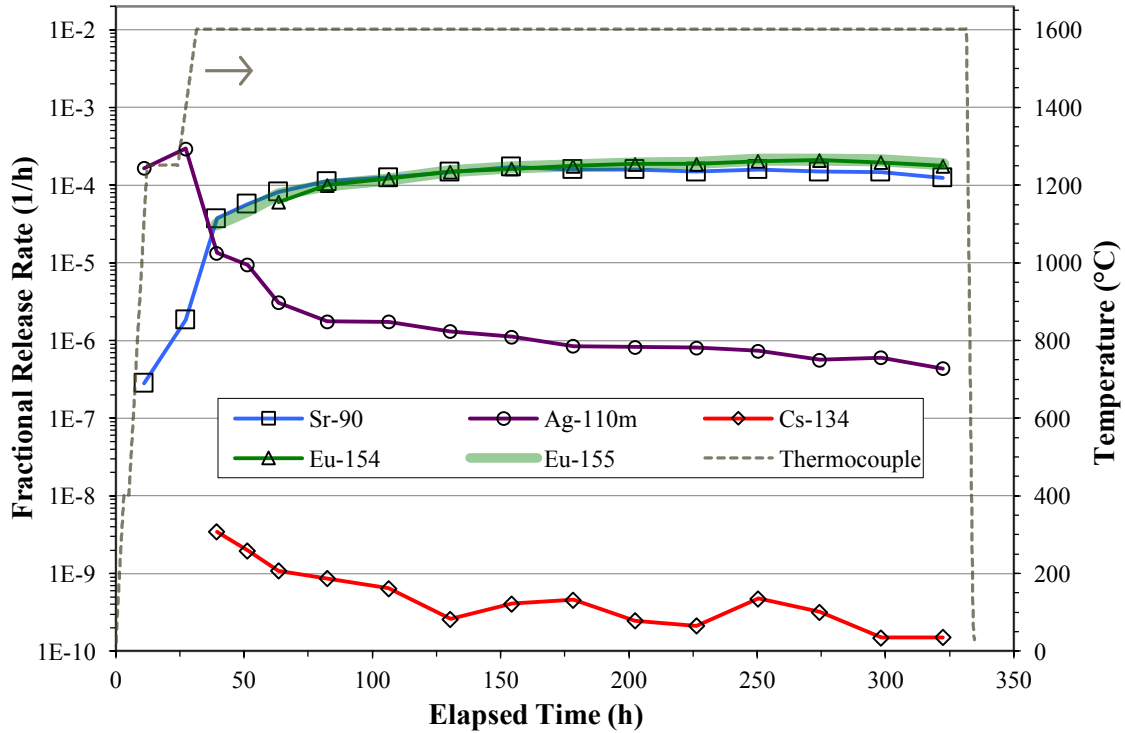


Figure 10. Rate of fission product release from Compact 2-2-2 during safety testing to 1600°C.

3. CONCLUSION

Safety testing at 1600°C was completed on AGR-2 UCO Compacts 6-4-2 and 2-3-1. Overall fission product release behavior was consistent with that observed for two previous AGR-2 UCO compact 1600°C safety tests [Hunn et al. 2016a] and safety tests on AGR-1 UCO compacts [Morris et al. 2014; Demkowicz et al. 2015]. Table 6 is a summary of the cumulative release from the four 1600°C safety-tested AGR-2 UCO compacts. In general, there were no clear indicators for TRISO failure (sudden ^{85}Kr release or cumulative release comparable to a particle inventory) or failed SiC (sudden Cs release comparable to a few tenths or more of a particle inventory). A small amount of ^{85}Kr release during Compact 2-2-2 safety testing was too low to have come from a particle with failed TRISO and may have been from uranium in the matrix [Hunn et al. 2016a]. The cumulative ^{134}Cs release from Compact 6-4-2 reached 20% of one particle's inventory by the end of the safety test. This cesium release is a factor of 10 greater than expected from a compact with no SiC failure, but below what is typical for a compact with a failed-SiC particle. No exposed kernels were detected in Compact 6-4-2 during post-safety test DLBL and this led to the conclusion that the relatively-gradual release of cesium from Compact 6-4-2 was more likely related to uranium contamination in the matrix.

Table 6. Cumulative releases of radioactive isotopes from AGR-2 UCO TRISO 1600°C safety tests

Isotope	Compact 6-4-2		Compact 5-2-2		Compact 2-3-1		Compact 2-2-2	
	Compact fraction	Particle equivalent	Compact fraction	Particle equivalent	Compact fraction	Particle equivalent	Compact fraction	Particle equivalent
^{85}Kr	$<9\times 10^{-7}$	<0.002	$<7\times 10^{-7}$	<0.002	$<7\times 10^{-7}$	<0.002	5.4×10^{-6}	0.017
^{90}Sr	8.7×10^{-5}	0.28	7.9×10^{-4}	2.5	8.6×10^{-2}	270	4.1×10^{-2}	130
$^{110\text{m}}\text{Ag}$	3.4×10^{-3}	11	1.6×10^{-2}	52	1.8×10^{-2}	56	7.3×10^{-3}	23
^{134}Cs	6.2×10^{-5}	0.20	5.9×10^{-6}	0.019	4.0×10^{-6}	0.013	2.5×10^{-7}	0.00078
^{154}Eu	2.7×10^{-4}	0.85	1.1×10^{-3}	3.4	8.4×10^{-2}	280	4.7×10^{-2}	150
^{155}Eu	2.1×10^{-4}	0.66	1.1×10^{-3}	3.4	8.6×10^{-2}	270	4.8×10^{-2}	150

Europium and strontium releases from Compact 6-4-2 were low enough that they may have also been impacted by this presumed uranium contamination. Overall, the measured fission product release at 1600°C appeared to be dominated by release of radioisotopes previously released under irradiation and subsequently retained in the carbonaceous matrix and OPyC until the compacts were heated above the irradiation temperature, as was observed for AGR-1 [Morris et al. 2014]. Release of europium and strontium from AGR-2 UCO Compacts 2-3-1 and 2-2-2 was very high but still consistent with release from the matrix and OPyC, as the higher Capsule 2 irradiation temperatures resulted in significantly-higher release of these isotopes [Hunn et al. 2016b]. Leach-burn-leach analysis is currently in progress on as-irradiated Compact 2-2-1, results from which a direct comparison can be made to the Compact 2-2-2 safety test releases, as these two compacts were irradiated to near-identical conditions.

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