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HIGH-TEMPERATURE SAFETY TESTING OF IRRADIATED AGR-1 TRISO FUEL

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Three irradiated AGR-1 UCO TRISO fuel compacts were tested using a time-varying temperature profile with a peak temperature of 1695 °C, simulating a conduction cooldown event in a gas-cooled reactor. Of the approximately 12,378 fuel particles in this test, less than a single particle's inventory of Cs, Eu, Kr or Sr was released during the test, indicating that all TRISO layers in all particles remained intact. The transient nature of the test did not stress the fuel beyond what has been observed in isothermal tests within the same temperature range. It is anticipated that tests of this nature will be repeated using fuel from subsequent irradiations (e.g. AGR-2, which has both UCO and UO_2 fuel compacts, and AGR-5/6/7, which has only UCO fuel).

I. INTRODUCTION

The purpose of the Advanced Gas Reactor (AGR) fuel development and qualification program is to design, fabricate, irradiate, analyze, and qualify tristructural isotropic (TRISO) fuel for use in high-temperature gas-cooled reactors (HTGRs) in the United States.[1] Extensive safety testing of the AGR-1 fuel compacts has been performed using primarily isothermal hold temperatures of 1600, 1700, or 1800°C, reported previously.[2] A recently-completed safety test utilized the Fuel Accident Condition Simulator (FACS) furnace at the Idaho National Laboratory (INL). In this test, three intact, irradiated fuel compacts were heated under a temperature transient characteristic of a core-conduction cool-down event in an HTGR. Releases of condensable fission products (Ag, Cs, Eu, Sb, and Sr) and fission product gases (Kr) were determined as a function of test time.

II. BACKGROUND AND SAMPLE SELECTION

During a core-conduction cool-down event, forced coolant circulation ceases, the system depressurizes, and heat removal from the system is primarily by conduction through the reactor vessel into the reactor cavity. The reactor core gradually heats up over tens of hours before decreasing after reaching a peak temperature. To-date, most safety tests in the AGR program have been isothermal tests, holding at a specific temperature for several hundred hours. Here, a time-varying temperature profile was applied in order to test the effects of a temperature transient and to compare the results with isothermal safety tests.

II.A. Historical temperature transient safety tests

A spherical, irradiated fuel element, designated AVR-91/31, was tested in a time-varying temperature profile in the cold finger apparatus (KÜFA) in the Federal Republic of Germany.[3] The TRISO-coated particle fuel in this sphere featured a UO_2 kernel, and prior to the test, the sphere had been irradiated to a burnup of 9.0% fissions per metal atom (FIMA). The heating profile used in this test followed the shape of a calculated design-basis core-conduction cool-down event; however, the curve had been shifted up to a maximum temperature of 1700 °C compared to 1600 °C for the design basis.

The AVR-91/31 test was particularly noteworthy in that approximately 20 TRISO failures occurred, based on the level of Kr release. This is a higher failure fraction than was observed during isothermal tests of similar (albeit lower burnup) AVR fuel. It is unknown if the transient nature of the test caused additional stress in the fuel, leading to increased failure rates compared to a 1700 °C isothermal test. Part of the motivation for the AGR safety test reported here was to determine if AGR fuel performed differently during a temperature transient than during high-temperature, isothermal exposure.

II.B. Compact Selection for the Transient Safety Test

Three compacts from the first AGR irradiation (AGR-1) were selected for simultaneous heating in the FACS furnace at INL. The AGR-1 irradiation featured TRISO-coated particles embedded in cylindrical graphitic matrix fuel compacts. All kernels were a heterogeneous mixture of uranium oxide and uranium carbide (UCO). The kernel carbon:uranium atomic ratio was 0.32, and the oxygen:uranium atomic ratio was 1.36 (Ref 4). Four combinations of coating formation process parameters were used to produce four TRISO coating variants.[4,5,6] Each particle variant was irradiated in separate compacts in separate capsules. Compacts selected for this test used "Variant 3" fuel particles with SiC layers deposited at lower temperatures in an argon-hydrogen mixture that produced a finer grain structure expected to reduce SiC

defects caused by uranium dispersion.[4,5] A summary of kernel and particle properties, as well as references to fuel characterization reports, can be found in Ref 4.

The three compacts selected for the test were from the same AGR-1 irradiation capsule. Each compact contained approximately 4,126 fuel particles (a volume packing fraction of 36 %) with a uranium enrichment of 19.7 wt %. The compacts had similar burnup and time-average volume average (TAVA) temperature. The Ag-110m inventory in the compacts was determined by gamma spectrometry and compared to predicted inventories to estimate the fraction of silver that was retained in each compact during irradiation.[7] The results indicated similar levels of silver retention in the three compacts. Properties for each of the three compacts used in the temperature transient safety test are summarized in Table I.

Table I. AGR-1 Compact irradiation properties.

Compact Name	Compact Burnup (% FIMA)	Compact Fast Neutron Fluence $\times 10^{25}$ (E > 0.18 MeV, n/m^2)	TAVA Irradiation Temperature (°C)	Retained Ag-110m Fraction
1-4-2	14.9	3.01	1045	0.76
1-1-3	15.3	2.86	1018	0.80
1-1-1	15.2	2.81	1017	0.79

III. EXPERIMENTAL SETUP

The FACS furnace is located in the main hot cell at the Hot Fuels Examination Facility (HFEF) at INL. Three compacts were heated simultaneously in the FACS furnace depicted schematically in Fig. 1. A helium sweep gas at a total flow rate of 1 L/min passed through the tantalum flow tube housing the samples. A tantalum sample holder held the three compacts. A water-cooled cold finger holds condensation plates which collect condensable fission products. The condensation plates are swapped at various points during the test. A fission gas monitoring system (FGMS) collects and counts Kr-85 throughout the test. Additional information on the FACS furnace is available in Ref. 8.

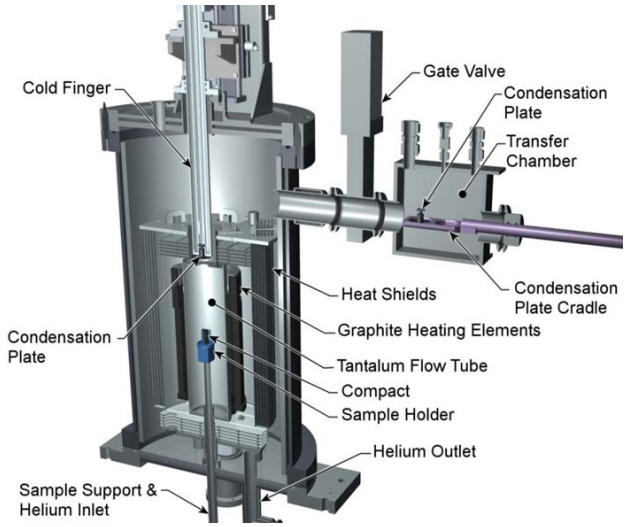

Fig. 1. FACS furnace schematic.

Fig. 2 shows the temperature profile programmed into the FACS furnace. This is the same profile used in the German AVR-91/31 test. Fig. 2 also shows the measured sample temperature during the test, and the time and temperature of condensation plate changes. The test began with a hold at 300 °C in order to drive off any adsorbed moisture from the compacts. From there, the temperature was raised and held at 857 °C for 70 hours to simulate a period of normal reactor operation. Note that the time on the x-axis was plotted such that the beginning of the rise in temperature from the hold at 857 °C is at t = 0. Between time t = 0 and t = 30 hours, the temperature was raised to a peak of 1695 °C. From the peak temperature attained at 30 hours, the temperature was gradually lowered to 1200 °C at 300 hours. At 302 hours, the test was terminated and the temperature dropped from 1200 °C to 20 °C in 2 hours.

The triangles in Fig. 2 denote condensation plate changes. Changes were made after each isothermal hold and at various other points throughout the test. Post-test analysis of the fission product activity on each plate was used to determine the time dependence of fission product releases.

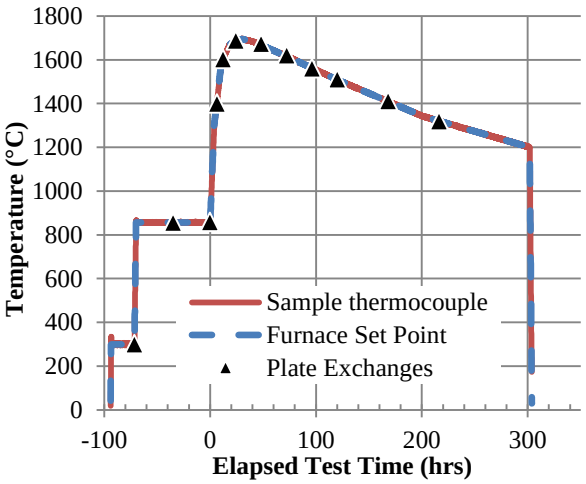

Fig. 2. Temperature profile and time and temperature of condensation plate changes.

Following completion of the test, each stainless steel condensation plate was analyzed with gamma spectrometry for gamma-emitting fission products. Common isotopes observed on the plates include Ag-110m, Cs-134/137, Eu-154/155, and Sb-125. Following gamma counting, the plates were stripped via electrolysis in nitric acid. A strontium separation was then performed on the stripping solution, and beta-emitting Sr-90 was detected via gas proportional counting. Liquid nitrogen-cooled charcoal traps in the FGMS collected Kr-85. The traps are counted throughout the test using high-purity germanium gamma detectors. All detected activities were decay-corrected back to the end-of-irradiation (EOI) plus one day. The measured activities of each isotope detected on condensation plates and in FGMS traps were compared to the activities predicted by physics calculations (Ref. 9) to exist in each compact at EOI plus one day. For the Kr-85 and the condensable fission products, the activities measured in the traps and on the condensation plates are adjusted according to previously-determined experimental collection efficiencies for the traps and the plates.

IV. RESULTS OF TRANSIENT SAFETY TEST

The plots in the following sections show the cumulative fractional fission product releases and release rates as a function of time for the transient test of AGR-1 Variant 3 Compacts 1-4-2, 1-1-3, and 1-1-1. Results from three previous isothermal safety tests in the FACS furnace[8] of individual AGR-1 Compacts 6-4-1 (Baseline Variant), 4-3-3 (Variant 3), and 4-3-2 (Variant 3) are also plotted for comparison, as these are somewhat representative of the results obtained from the AGR-1 isothermal tests. The transient test results are plotted such that time t = 0 marks the beginning of the transient rise in temperature. The previously reported isothermal test results are plotted such that time t = 0 represents the time at which the isothermal test temperature was reached. Refer to Ref. 8 for irradiation properties of these other compacts. Data from additional AGR-1 isothermal tests are available in Ref. 2. Since the transient test utilized three compacts, a single average particle is equivalent to a fraction of 8.08E-5 of the transient test fission product inventory. From the prior three single-compact tests, a single particle represents a fraction of 2.4E-4 of a single compact inventory. Table II summarizes the total fractional releases of fission products from the transient test. For all measured nuclides except silver, the release was less than the inventory in a single particle.

Table II. Total inventories released in transient test.

Isotope	Equivalent Particle Inventories Released	Total Fraction Released
Ag-110m	902	7.29E-2
Cs-134	0.006	4.80E-7
Cs-137	0.015	1.23E-6
Eu-154	0.46	3.69E-5
Eu-155	0.49	3.95E-5
Kr-85	0.039*	3.21E-6*
Sb-125	0.014¥	1.13E-6¥
Sr-90	0.011	2.61E-6

*Due primarily to in-leakage from hot cell.
¥Assuming release equal to the minimum detectable activity

IV.A. Silver behavior

Fig. 3 shows that about 7% of the total silver inventory from the three compacts was released (equivalent to the inventory from 902 average particles), and that this 7% release had been reached approximately 10 hours into the test. Nearly all of the silver released from this test came during the temperature ramp after the hold at 857 °C and prior to reaching the peak test temperature. This indicates relatively rapid, early release of silver that had accumulated in the outer pyrolytic carbon (OPyC) layer of the fuel particles and compact graphitic matrix during the preceding irradiation. This is consistent with the accompanying data from Compacts 4-3-2, 4-3-3, and 6-4-1 and the observations made in Ref. 8.

Fig. 4 shows the silver release rate throughout the transient test, and Fig. 5 highlights the release rate during the ramp to peak temperature. In Fig. 4, compared to the isothermal tests at 1600 and 1800 °C, the silver release rate varies considerably with the temperature of the transient

test. Between 24 hours and 166 hours of the transient test (when the temperature was between 1400 and 1650 °C), the transient test silver release rate was similar to those observed during the isothermal 1600 °C safety tests of Compacts 4-3-3 and 6-4-1. Fig. 5 shows that the peak silver release rate (1.6E-3 fraction/hr) occurred at about 6 hours and a test temperature of 1390 °C. After peaking at 6 hours, as the test temperature continued to increase to its peak of 1695 °C at 30 hours, the silver release rate decreased. Fig. 4 shows that the silver release rate continued to decrease as the test temperature decreased from its peak of 1695 °C at 30 hours to 1500 °C at 120 hours. From 168 hours to the termination of the test at 304 hours, the temperature continued to decrease; however, the silver release rate increased. The average release rate for the final three condensation plates exchanged in the test was 2.4E-5 fraction/hr, and the average temperature over these three plates was 1299 °C. Since the silver release rate decreased above 1390 °C during the ramp to peak temperature (see Fig. 5) and increased between 1500 and 1200 °C during the ramp down from peak temperature (see Fig. 4), this suggests a region where the silver release rate is inversely proportional to the test temperature.

This apparent inverse-temperature behavior is consistent with behavior noted in the safety test of AGR-1 Compact 4-2-2 (Ref. 10). In Ref. 10, the test temperature was varied between 1000 and 1600 °C, and the Ag-110m release rates were highest in the 1100 to 1300 °C range. From Ref. 10, the silver release rate during a hold at 1300 °C was 2.0E-5 (fraction/hr) compared to an average release rate of 2.4E-5 (fraction/hr) over the last three condensation plate exchanges in the test reported here. The peak silver release rate from this transient test occurs within this 1100 to 1400 °C range where silver release rates appear to be higher than they are in the 1400 to 1700 °C range. While the mechanism of silver transport at these intermediate temperatures is not yet understood, the increase in silver release rate in the 1800°C test of Compact 4-3-2 shown in Fig. 4 is believed to be from increased diffusive release from intact particles at the higher test temperature.

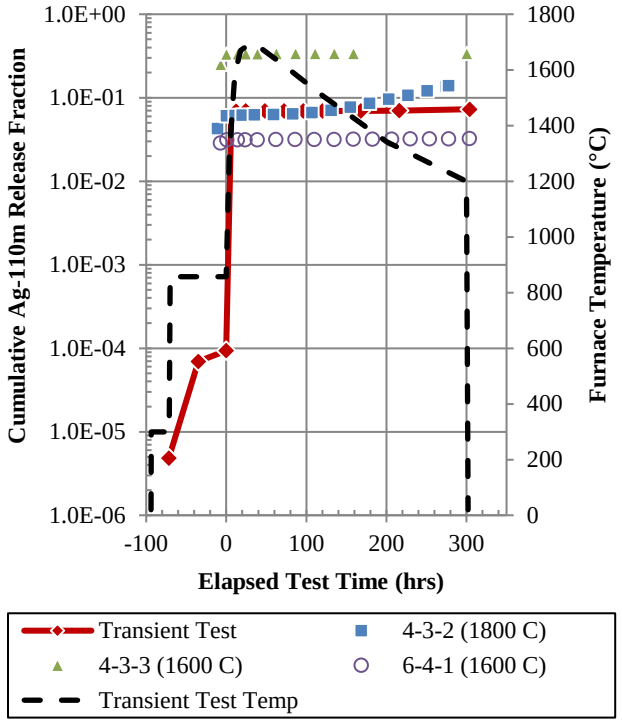

Fig. 3. Cumulative Ag-110m release fraction versus time for the transient test and three prior isothermal tests of individual compacts.

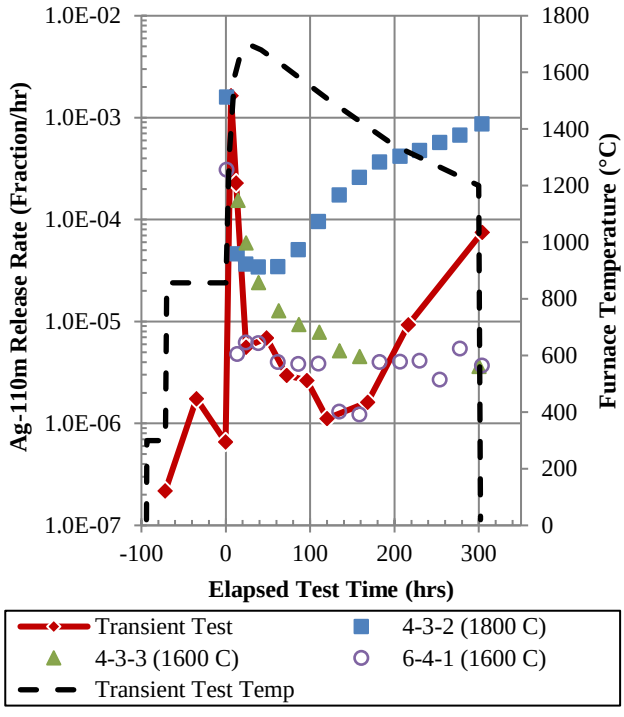

Fig. 4. Ag-110m release rate (fraction/hr).

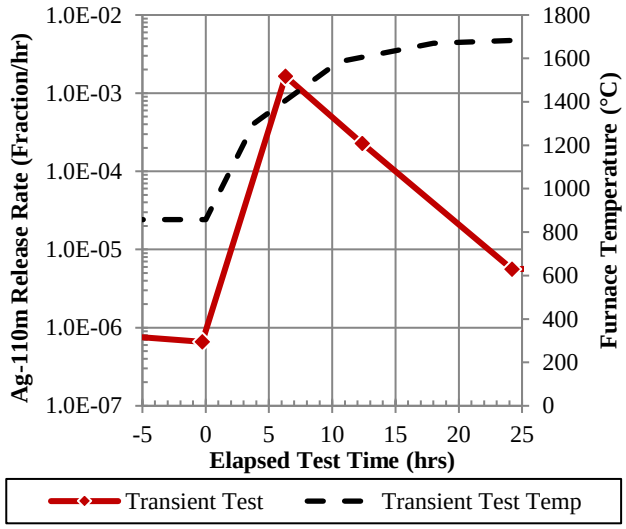

Fig. 5. Ag-110m release rate (fraction/hr) during heat-up.

IV.B. Cesium behavior

Fig. 6 shows that a fraction of 4.8E-7 of the EOI Cs-134 inventory was released from the compacts during the transient test. This is equivalent to 0.6% of the inventory from a single, average particle. Despite a peak temperature of 1695 °C, the transient test Cs-134 fractional release is similar to that of the isothermal 1600 °C test of Compact 4-3-3. This plot also shows that most cesium release occurred during the ramp to peak temperature following the hold at 857 °C. Indeed, Fig. 7 shows that the two highest cesium release rates are calculated from the two condensation plates exchanged immediately prior to the peak temperature. The highest rate of Cs-134 release was 3.8E-9 fraction/hr from the plate exchanged at 1600 °C, and the second highest rate was 2.7E-9 fraction/hr from the plate exchanged at 1690 °C. Since little cesium release occurred after peak test temperature, this indicates that cesium releases during the test were limited to cesium which had migrated out beyond the fuel particle SiC layers during irradiation. This type of behavior is seen even more clearly in the data for Compact 4-3-3 (which did not experience any layer failures) in Fig. 7. Compact 6-4-1 had a single SiC layer failure early in the test, and it is estimated that Compact 4-3-2 had three SiC layer failures (at 132 hours, 217 hours, and 377 hours).[8] These SiC failures account for the higher cesium release rates from Compacts 6-4-1 and 4-3-2.

Table II summarizes that, in total, 0.6% of a single, average particle's Cs-134 had been released and 1.5% of a single average particle's Cs-137 inventory had been released. This indicates that no SiC layer degradation occurred. If significant SiC layer degradation had occurred in a single particle, released cesium fractions ≥ 50% of an average particle's inventory would be reasonably expected.[11] Based on the ratio of Cs-134 and Cs-137 inventories on the plates, it appears that there may have been minor contamination of Cs-137 from the hot cell on the final two plates. It was determined that this did not significantly affect the results. The jumps in the cumulative release of Cs-134 from Compact 4-3-2 are due to SiC layer failures.[8]

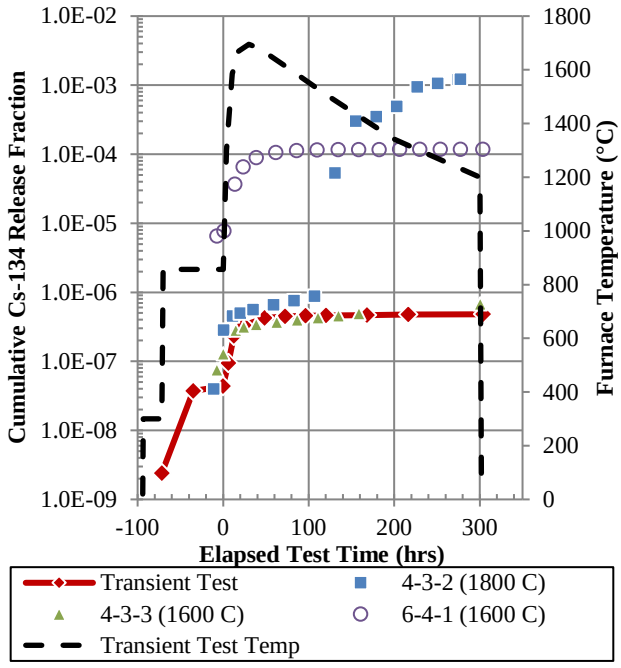

Fig. 6. Cumulative Cs-134 release fraction versus time.

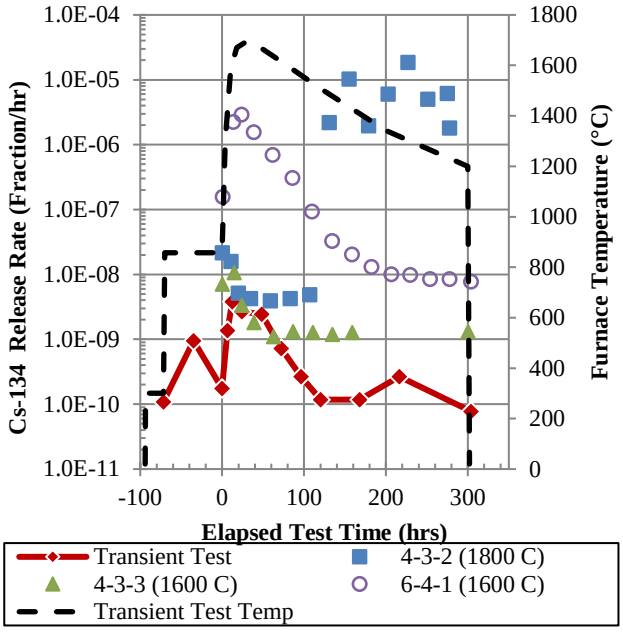

Fig. 7. Cs-134 fractional release rates.

IV.C. Europium behavior

Fig. 8 shows that the cumulative europium releases are proportional to the test temperatures. Fig. 9 shows that the release rates are also proportional to temperature, with the calculated Eu-154 release rate following the temperature profile from the transient test. The increase in the Eu-154 release rate after 114 hours for Compact 4-3-2 is a behavior observed only during 1800 °C tests of Variant 3 compacts and is due to diffusive release through intact particles which eventually overwhelms release of europium retained in the compact matrix during irradiation.[2]

IV.D. Strontium behavior

Strontium release behaviors are similar to those of europium. The cumulative releases (Fig. 10) and release rates (Fig. 11) are proportional to the test temperatures. The rates are relatively steady during the isothermal tests, but generally vary in proportion to the temperature profile used in the transient test. As with europium, diffusive releases of strontium through intact particles occur after roughly 114 hours during the 1800 °C safety tests of AGR-1 Variant 3 fuel (such as Compact 4-3-2).

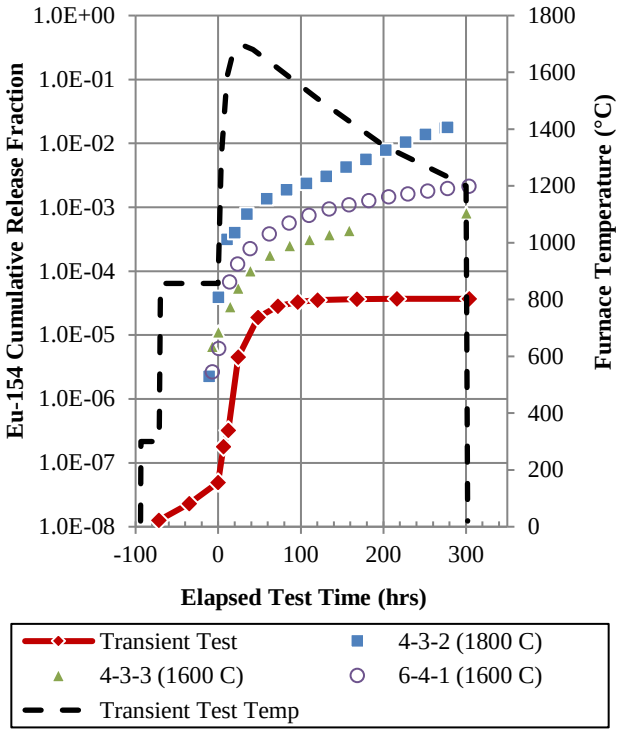

Fig. 8. Eu-154 cumulative release fractions.

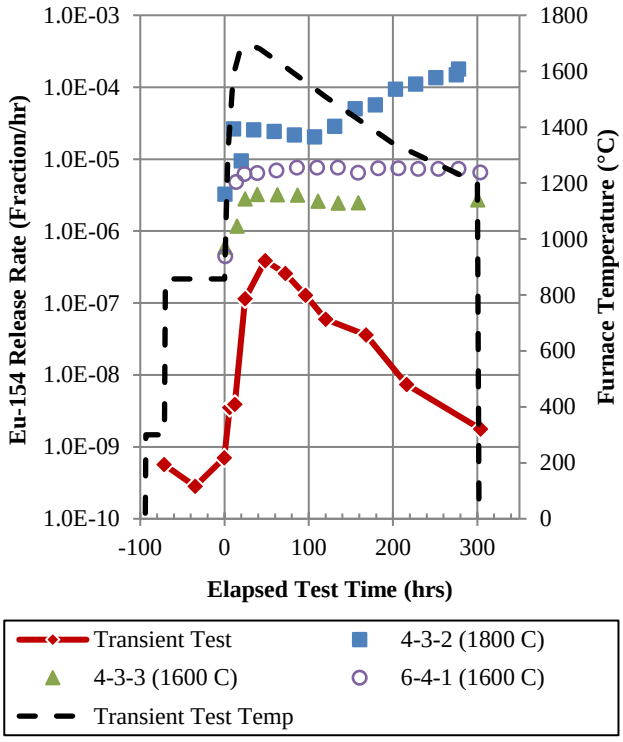

Fig. 9. Eu-154 fractional release rates.

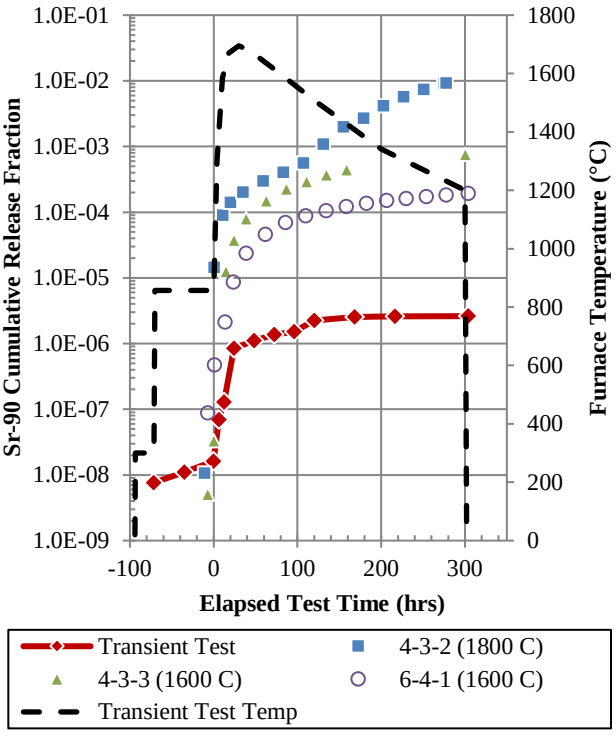

Fig. 10. Cumulative Sr-90 release fractions.

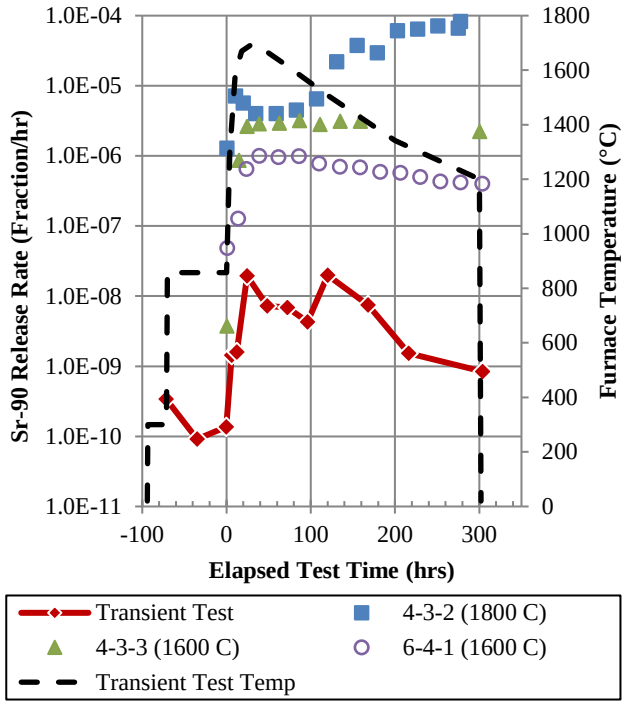

Fig. 11. Sr-90 fractional release rates.

IV.E. Krypton behavior

Fig. 12 shows the cumulative Kr-85 fractions collected in the FGMS traps for the transient test and three previous AGR-1 single-compact isothermal tests. Recall that a single average particle is equivalent to a fraction of 8.08E-5 of the transient test fission product inventory. From the prior three single-compact tests, a single particle represents a fraction of 2.4E-4 of a single compact inventory.

The word "collected" is used here with respect to the transient test because this krypton is most-likely due to minor leakage of the HFEF hot cell atmosphere (which contains Kr-85 from previous exams on other fuel specimens) into the FACS system and not from releases from the fuel. Prior to 196 hours, the measured Kr-85 activity versus time for the transient test represents a collection rate comparable to the rate measured during an unfueled (no sample) test of the software programming and temperature profile used for the transient test. In this un-fueled test, the Kr-85 collection rate was 18.8 Bq/ hr (0.50 nCi/hr) compared to 19.2 Bq/hr (0.52 nCi/hr, 5.6E-9 Kr-85 fraction/hr) for the transient test prior to 196 hours. After 196 hours, a distinctively different Kr-85 collection rate was measured at 44.4 Bq/hr (1.2 nCi/hr, 1.3E-8 Kr-85 fraction/hr). This is comparable to 51.8 Bq/hr (1.4 nCi/hr) measured for a different unfueled test (at ambient temperature). Since the rates of Kr-85 collection during the transient test are similar to those measured during two unfueled tests with no samples, it is reasonable to conclude that the majority of the Kr-85 measured during the transient test is from in-leakage of hot cell contamination. The two collection rates, 18.8 and 44.4 Bq/hr could be due to a change in the fitment of the O-rings used to seal the moveable cold finger that holds the condensation plates.

As summarized in Table II, even if all of the Kr-85 measured from the transient test was directly attributable to the fuel, it represents only 3.9% of a single particle's inventory and would not indicate any TRISO failures. If a TRISO failure had occurred, it is expected that a significant fraction of the Kr-85 would have been rapidly released for each particle with failed TRISO layers. This was the case for the 1800 °C safety test of Compact 4-3-2 where two TRISO failures occurred and Kr-85 activities corresponding to two particle inventories were measured. In Fig. 12, the jump in the released activity for Compact 4-3-2 at 132 hours is due to a single particle that experienced SiC failure. Additional increases were observed later in the 1800 °C test of Compact 4-3-2 due to two particles that experienced TRISO failure.[8]

Compared to previous tests, the average Kr-85 collection rate for the duration of the transient test (38.4 Bq/hr, 7.6E-9 fraction/hr) is a little higher than the average rates observed in the 1600°C isothermal tests of Compacts 6-4-1 (6.3 Bq/hr, 4.3E-9 fraction/hr) and 4-3-3 (4.5 Bq/hr, 2.3E-9 fraction/hour). As discussed above, the majority of the Kr-85 measured during the transient test is believed to be from in-leakage of hot cell contamination. None of these compacts had TRISO failures. The transient test average Kr-85 collection rate is about a factor of three lower than the rate (21.8 Bq/hr, 1.2E-8 fraction/hr) observed during the early part of the 1800°C test of Compact 4-3-2, prior to the failure of any coating layers in Compact 4-3-2.

This temperature transient test demonstrates that AGR-1 fuel does not experience the significantly higher TRISO failure rates observed during the temperature transient test of the German UO_2 sphere AVR-91/31. Furthermore, at 95% confidence, the transient test results are consistent with the relatively low layer failure rates observed in AGR-1 fuel during isothermal testing[1,2] at temperatures of 1600–1800°C. Future temperature transient tests utilizing irradiated AGR-2 UCO and UO_2 fuel may be performed.

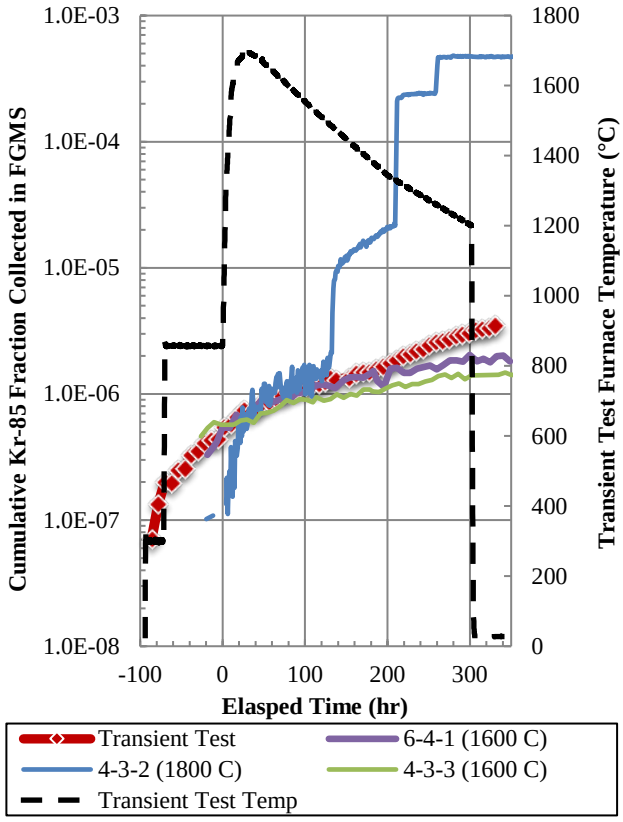

Fig. 12. Cumulative collected Kr-85 fraction.

V. CONCLUSIONS

Irradiated compacts with TRISO fuel containing UCO kernels irradiated to a burnup of approximately 15% FIMA were tested under a simulated core conduction cooldown event, and except for silver, very small fractions (less than a single particle inventory) of fission products were released. This demonstrates that the AGR UCO fuel retains fission products and TRISO layer integrity under design-basis temperature transients. The temperature variation did not stress the fuel any more than an isothermal test at 1600 or 1700 °C. AGR-1 fuel does not experience the higher TRISO failure rates observed during the temperature transient test of the German UO_2 sphere AVR-91/31. It is expected that additional tests of this nature will be performed on fuel from subsequent irradiation experiments (AGR-2 and AGR-5/6/7).

ACKNOWLEDGMENTS

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