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Irradiated AGR 5/6/7 Compact 5-6-2 Examination Plan

April 2023

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Idaho National Laboratory

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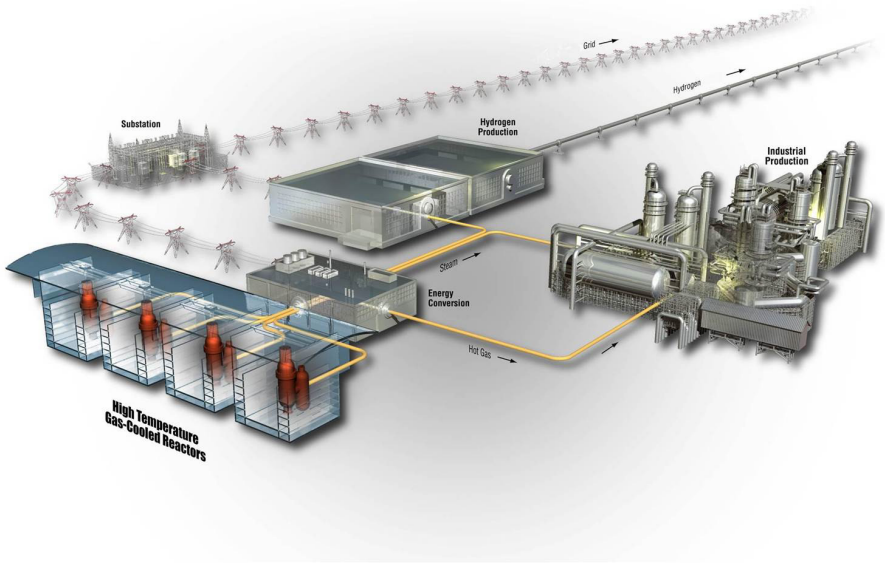
Irradiated AGR-5/6/7 Compact 5-6-2 Examination Plan



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ACRONYMS

AGR	Advanced Gas Reactor
CCCTF	Core Conduction Cooldown Test Facility
FIMA	fissions per initial metal atom
IMGA	Irradiated Microsphere Gamma Analyzer
INL	Idaho National Laboratory
LBL	leach-burn-leach
OPyC	outer pyrolytic carbon (coating layer)
ORNL	Oak Ridge National Laboratory
PIE	post-irradiation examination
SiC	silicon carbide (coating layer)
TA_{max}	time-average maximum (compact irradiation temperature)
TA_{min}	time-average minimum (compact irradiation temperature)
TAVA	time-average, volume-average (compact irradiation temperature)
TRISO	tristructural isotropic (coated particles)
UCO	uranium carbide and uranium oxide (kernels)

1. INTRODUCTION

This plan describes the post-irradiation examination (PIE) activities to be performed by Oak Ridge National Laboratory (ORNL) on irradiated Compact 5-6-2 taken from the Advanced Gas Reactor (AGR) experiment, AGR-5/6/7. This work will be performed in accordance with the general objectives outlined in the AGR-5/6/7 PIE Plan[1] and guidance in the ORNL PIE Statement of Work.[2]

2. FUEL COMPACT DESCRIPTION

The fuel specimen contains tristructural isotropic (TRISO)-coated particles with kernels containing both uranium carbide and uranium oxide (UCO) and was irradiated in Capsule 5 of the AGR-5/6/7 test train in the northeast flux trap of the Advanced Test Reactor at Idaho National Laboratory (INL).[3] Table 1 shows some properties and irradiation conditions for AGR-5/6/7 Compact 5-6-2.

Table 1. Identification and irradiation conditions for AGR-5/6/7 Compact 5-6-2.

Compact ID [a]	Container ID	Compact Batch ID	Average Particle Count	Average Burnup [c] (% FIMA [b])	Fast Fluence [c] ($\times 10^{25}$ n/m^2)	Irradiation Temperature (°C) [d]		
						TA_{min} [e]	TAVA [e]	TA_{max} [e]
AGR-5/6/7 5-6-2	AGR567-562	14155A	3392	6.75	1.67	467	634	741

a. The X-Y-Z naming convention denotes the location in the irradiation test train: capsule-level-stack.
b. Fissions per initial metal atom (FIMA).
c. Based on physics calculations.[4]
d. Based on thermal calculations.[5]
e. Time-average minimum (TA_{min}), time-average, volume-average (TAVA), time-average maximum (TA_{max}) temperature.

3. EXPERIMENTAL OBJECTIVES

- Evaluate the time-dependent release behavior of gaseous and condensable fission products during safety testing at high temperatures in pure helium in the Core Conduction Cooldown Test Facility (CCCTF). Fission products to be analyzed include ^{85}Kr, ^{90}Sr, ^{104}Pd, ^{106}Ru, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, and ^{155}Eu. Special emphasis will be on monitoring krypton and cesium release as an indicator of coating failure in individual particles, where krypton release can indicate a failure of all gas-retentive TRISO layers and cesium release can indicate a failure of the silicon carbide (SiC) layer when one or more gas-retentive pyrocarbon layers remains intact.
- Measure the inventory of uranium and fission products outside of intact SiC layers but retained in the compact matrix or outer pyrolytic carbon (OPyC). These measurements will be done by electrolytic deconsolidation of the compact followed by acid leaching of the particles and matrix debris using a leach-burn-leach (LBL) process, as described in Section 4.4. Particles and particle fragments will be separated from the matrix for examination prior to burn-leach.
- Examine individual particles deconsolidated from the safety-tested compact with the Irradiated Microsphere Gamma Analyzer (IMGA), as described in Section 4.5, to quantify retention of specific gamma-emitting fission products, including ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu, and to identify anomalous particles if present, especially those with a below-average cesium inventory indicative of SiC failure. If krypton or cesium release in the CCCTF indicates the presence of particles with coating failure, all particles recovered from the compact will be surveyed with a scan time appropriate for the measurement of ^{144}Ce and ^{137}Cs. This survey may also be completed if the additional analysis is requested by the INL fuels PIE technical lead. Short-duration gamma scanning of every particle recovered from the compact will provide ^{144}Ce data to identify particles with a below-average cerium inventory indicative of acid leaching from exposed kernels or the rare

occurrence of TRISO-coated abnormal kernels. Short-duration gamma scanning will also provide ^{137}Cs data, and the ^{137}Cs:^{144}Ce ratio will be used to identify and sort particles with a below-average cesium inventory indicative of SiC failure. An important objective of identifying these low-cesium particles and measuring their remaining ^{134}Cs inventory is to determine if their missing ^{134}Cs inventory is sufficient to account for the ^{134}Cs measured on the CCCTF's internal components and in the compact matrix. This ^{134}Cs accounting is helpful in enumerating particles with failed SiC.[6] Regardless of cesium release in the CCCTF, a multi-hour count of about 45–60 randomly selected particles will be performed to examine average fission product retention characteristics. Any abnormal particles with low ^{137}Cs and/or ^{144}Ce will also be counted with this longer duration prior to further analysis.

- Perform microanalysis on selected particles and recovered particle fragments, as described in Section 4.6, to better understand the correlation of particle microstructure with fission product retention. Of particular interest is the microanalysis of particles with below-average ^{144}Ce, indicative of failed TRISO (or potentially abnormal kernels), or a below-average ^{137}Cs:^{144}Ce ratio, indicative of failed SiC, and such particles should have the highest priority for examination. Representative particles with average fission product retention and particles with a below-average retention of specific fission products (e.g., ^{110m}Ag or ^{154}Eu), but otherwise average retention of ^{137}Cs and ^{144}Ce will also be examined. Based on the specific results and discussions with the INL fuels PIE technical lead, particles may be sent to INL for additional microanalysis.
- Archive remaining particles for possible later use. Maintain the identity of abnormal particles identified during visual inspection or IMGA survey, as well as individual particles subjected to multi-hour gamma counting.

4. SCOPE OF WORK

4.1 Receipt Inspection

The compact shipping drum will be unpacked, and the individual aluminum compact storage container removed. The compact will be removed from the storage container and inspected for any damage prior to proceeding with subsequent analysis. The condition of the compact and any features of interest will be photographed.

4.2 Furnace Testing

The compact will be heated in high-purity helium in the CCCTF to a maximum temperature of 1600°C; Figure 1 and Table 2 show the temperature profile for the safety test. The temperatures in the profile will be maintained within the accuracy limits of the CCCTF furnace thermocouples and control software. The hold duration at 1600°C will be based on observed fuel performance. A shorter test may be called for if significant fuel failure is observed early in the test, which is indicated either by online fission gas (^{85}Kr) release measurements or a preliminary gamma measurement of the released cesium activity on the CCCTF deposition cups. In the absence of significant early fuel failure, the hold time at 1600°C will be about 300 h. Radioisotope release will be monitored throughout the test, and the amount measured will help determine the duration of the test. Changes to final heating times or any other test changes will be discussed with the fuels PIE technical lead.

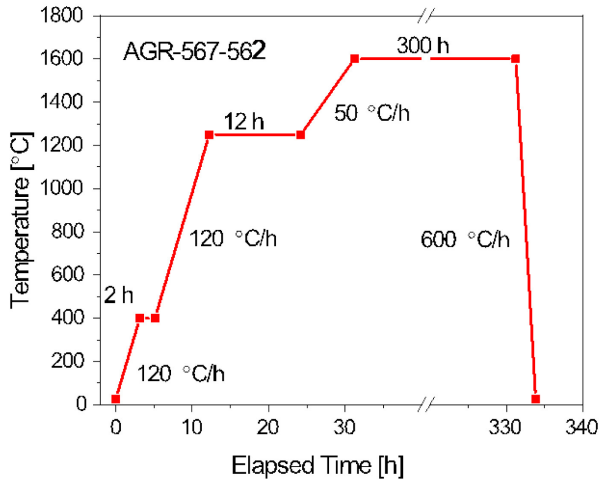

Figure 1. Temperature profile for the safety test of AGR-5/6/7 Compact 5-6-2. Elapsed time and ramp rates are approximate.

Table 2. Temperature profile for the AGR-5/6/7 Compact 5-6-2 safety test.

Approximate Elapsed Time (h)[a]	Temperature (°C)
0.0	30
3.1	400
5.1	400
12.2	1250
24.2	1250
31.2	1600
331.2[a]	1600
333.8[a]	30

a. Actual hold time may be modified depending on test results.

The first deposition cup will be exchanged near the end of the 1250°C hold cycle. A second cup will be exchanged once the furnace temperature reaches the target of 1600°C. The deposition cups will then be exchanged approximately once every 12 h for the next 36 h (i.e., three more cups after the first cup change at 1600°C). After ~36 h (i.e., four cup changes at 1600°C), the cups will be changed once every 24 h for the remainder of the safety test, or more frequently if rapid releases warrant shorter collection times. The actual exchange interval may deviate from this schedule based on results from the early test stages, but it should not be less than 6 h or more than 18 h for the initial 36 h at 1600°C or more than 30 h for the remainder of the test.

The cups will be examined to determine the inventory of deposited gamma-emitting radionuclides (such as ^{106}Ru, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, and ^{155}Eu) and any other radionuclides detected. In practice, a preliminary analysis will be conducted by performing a gamma scan as soon as possible after the cup is removed from the furnace to provide rapid feedback about the progress of the safety test. This feedback will allow changes to be made to the test if necessary. After completing the test, the cups may undergo additional gamma analysis prior to leaching. An analysis of the leach solutions includes the measurement of ^{90}Sr and ^{104}Pd and may include additional analyses for gamma-emitting isotopes.

4.3 Analysis Of Graphite Holder and Tantalum Liner

Significant fractions of some of the fission products (such as europium and strontium) that exit the compact are retained in the graphite holder or held up on the tantalum liner and gas inlet line prior to reaching the deposition cups. Therefore, the fraction deposited on the deposition cups, also called the collection efficiency of the deposition cups, must be determined to estimate the total time-dependent release of each fission product throughout the safety test based on the fission products collected on the periodically exchanged deposition cups. After the graphite fuel holder containing the compact has been removed from the furnace and returned to the main hot cell, the fuel compact will be removed from the holder and temporarily stored in a labeled container until deconsolidation and acid leaching is performed. The holder will be oxidized in air, and the resultant ash will be leached in boiling nitric acid to identify and determine the amount of any fission products retained in the holder. The tantalum liner and tube will be initially gamma scanned, leached with acid (optionally augmented with ball milling), and gamma scanned again after leaching. If needed, a gamma analysis of the leachant will be scaled by the tantalum leaching efficiency calculated from the pre- and post-leach gamma scans to determine the amount of each fission product collected on the tantalum. The combined analysis of all furnace internals will be used to estimate the average deposition cup collection efficiency for a final determination of fission product release during the safety test (i.e., the cumulative inventories collected on all deposition cups will be divided by the cumulative inventories collected on the graphite holder, Ta internal components, and all deposition cups).

4.4 Compact Deconsolidation and Acid Leaching

After safety testing, the fuel compact will be electrolytically deconsolidated in nitric acid to break up the matrix material and free the TRISO fuel particles. Particles and matrix debris will then be subjected to an LBL process. Unless the traditional hot cell deconsolidation and preburn leach process is modified as discussed below, particles and matrix debris will be collected in a Soxhlet thimble, two 24-hour hot nitric acid leaches in a Soxhlet extractor will be performed, and the leachates will be analyzed for actinides and fission products. After the two preburn leaches, a further digestion of the particles and matrix debris in boiling acid will be performed to help remove any matrix residue from the TRISO particles and break up the matrix debris further. The matrix debris will be separated from the particles by washing through a sieve stack. The sieve stack will include a sieve slightly larger than the expected particle size to collect particle clusters, a sieve slightly smaller than the expected particle size to collect individual particles, and a sieve suitable for collecting particle fragments while passing the majority of the matrix debris to the bottom sieve pan. Recovered particles and any recovered particle fragments will be rinsed, dried, and transferred to the hot cell cubicle housing the IMGA, where they will undergo visual inspection and a gamma survey as described in Section 4.5.

The separated matrix debris will be dried by distilling off the acid and rinse water. The dry matrix debris will be heated at 750°C in air to burn off the carbon and oxidize metallic fission products (some metal carbides have low solubility in nitric acid). The residual ash and burn vessel will be subjected to two postburn nitric acid leaches, and the leachates will be analyzed for uranium and fission products.

The process described above may require modifications to address the unique properties of the AGR-5/6/7 compacts. The previous deconsolidation of two Capsule 2 AGR-5/6/7 fuel compacts resulted in a greater number of large matrix fragments than was observed after the deconsolidation of irradiated AGR-1 and AGR-2 compacts, which used a different matrix formulation. Many of these large matrix fragments were similar in size to the TRISO particles, which made separation using a sieve impractical. If large matrix fragments are generated and too numerous to be easily separated using the particle micro-manipulator in the IMGA cubicle, an additional sink-float separation technique will be applied after performing the deconsolidation, preburn leaching, and digestion steps. This technique involves selectively floating the matrix debris in a sodium polytungstate solution diluted with high-purity water to a density between 2.2 and 2.3 g/cm^3. Fuel particles placed in the solution sink to the bottom of the separation funnel because their density is higher than 2.3 g/cm^3, while matrix debris stays at the top because its density is less than 2.2 g/cm^3. After the sink-float separation, the matrix debris will be combined with the sieve-separated debris and subjected to burn-leach, as described above, while the particles will be rinsed, dried, and subjected to an IMGA survey.

After completing the IMGA counting as described in Section 4.5, an archive sample of about 10% will be riffled out, and the remaining 90% of the TRISO particles will be returned to the main hot cell for particle burn-leach. The actual fraction held back will be determined by photographing and counting the individual particles so that the burn-leach data may be scaled appropriately. Particles will be loaded back into the Soxhlet thimble used for preburn leaching. Similar to the matrix burn-leach, particles will be heated at 750°C in air to remove the exposed carbon and then leached twice in the Soxhlet extractor. An analysis of the particle burn-leach solutions will be performed to detect actinides and fission products not leached before the burn, including those from any exposed kernels that might be associated with particles with failed SiC not separated out during the IMGA analysis. After burn-leach, the burned-back particles will be washed, dried, and archived.

4.5 Particle Inspection and Gamma Analysis

Prior to particle burn-leach, deconsolidated particles will be inspected and imaged using the particle micro-manipulator in the IMGA cubicle to assess overall condition, identify features of interest such as cracked coatings or coating fragments, and obtain a particle count. Abnormal particles or coating fragments may be culled out for targeted examination.

The need for a full gamma survey with IMGA of all particles recovered from the compact will be based on the preliminary safety test results. If krypton release indicates the probable presence of particles with failed TRISO (i.e., failure of all three dense layers) or cesium release indicates the probable presence of particles with failed SiC, then a full IMGA survey will be performed to find failed particles that remained in one piece. A full survey may also be performed with the approval of the INL fuels PIE technical lead if it is determined appropriate and beneficial to the post-safety test analysis. If a full gamma survey is performed, a short counting time (typically 50–100 s, depending on activity and counting statistics) will be used to measure ^{137}Cs and ^{144}Ce and sort out particles with a low ^{144}Ce content or a low ^{137}Cs:^{144}Ce ratio. A below-average ^{144}Ce content may indicate abnormal kernels or general fission product loss that may indicate a failed TRISO particle. A below-average ^{137}Cs:^{144}Ce ratio is indicative of significant cesium release during irradiation or safety testing that could be due to a failed SiC layer.

After completing the full IMGA survey (or after particle inspection if the full IMGA survey is not performed), an archive sample of about 10% of the deconsolidated TRISO particles will be riffled out with a chute splitter, and the remaining 90% of the deconsolidated TRISO particles will be returned to the main cell for burn-leach analysis, as described in Section 4.4. Unless present in excessive numbers, all particles sorted out due to an abnormal ^{137}Cs or ^{144}Ce inventory will undergo a longer count time (typically 3–6 h, depending on activity and counting statistics) to more accurately measure their radionuclide inventory (e.g., ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu) and determine distributions for ^{154}Eu and ^{110m}Ag, which have previously exhibited diffusive release from normal TRISO particles under certain conditions. Long-count IMGA analysis will also be performed on a random riffled sample of about 45–60 particles taken from the 10% archive sample. All long-counted particles will be labeled with unique identifiers and retained to support subsequent analyses or future testing.

It is preferable to perform the IMGA counting on TRISO-coated particles prior to removing the OPyC layer during the LBL burn phase, as these particles provide a better source for follow-on analyses, and it is less likely that particles with failed TRISO will be recovered after the burn-leach. However, if excessive matrix debris on the surface of the OPyC layer makes automated handling with the IMGA vacuum needle impractical, then particles may be subjected to burn-leach prior to the IMGA survey.

4.6 Microstructural Analysis

After IMGA analysis, particles of interest will be selected for x-ray imaging and/or materialography, especially any particles with a below-average ^{137}Cs:^{144}Ce ratio or low overall radioisotopic inventory that may be indicative of failed SiC or failed TRISO, respectively. Materialography may include optical microscopy, electron microscopy, and/or elemental analysis of the polished cross-sections. Based on the specific results and discussions with the INL fuels PIE technical lead, individual particles may be sent to INL for additional microanalysis.

In addition to identifying and studying particles with failed SiC or failed TRISO, another goal of this PIE effort is to identify any mechanisms that may explain why some particles release silver and europium to a greater extent than others. Features of interest will include structure of the coatings and kernels, particularly changes induced by radiation. Also of interest are the location and composition of actinides or fission products accumulated within the coating layers, primarily in the inner pyrolytic carbon and SiC layers.

X-ray imaging with tomographic reconstruction will be used to achieve a nondestructive examination of the internal structure of individual particles. Particles identified with the IMGA to have a below-average ^{137}Cs:^{144}Ce ratio or low overall radioisotopic inventory will be subjected to x-ray tomography prior to any destructive analysis unless present in large numbers, in which case representative samples will be taken from the population of failed particles to provide a sufficient survey of the failure mechanisms and microstructures. The three-dimensional visualization of the x-ray tomography data will provide important insight to complement and guide further materialographic examination.

Particles selected for materialography will be mounted in epoxy and the mounts polished to inspect particle cross-sections using microscopic methods. Optical microscopy can be used to inspect the overall condition of kernels and coatings. While most materialographic mounts will only include 1–3 particles to promote high-quality polishing and removal from the hot cell for electron microscopy, some mounts will also be produced that contain about 40 particles each to provide a better optical microscopy survey of the general post-irradiation TRISO particle microstructure. Sufficient sampling for categorization of the buffer layer behavior is of particular interest.

Scanning electron microscopy can be used to perform higher resolution inspections of kernel and coating layer microstructures. Energy-dispersive x-ray spectroscopy can be used to characterize the elemental distributions within the kernel and coating layers, as well as actinide and/or fission product interaction with the SiC layer (palladium and uranium accumulation and interaction with the SiC are of particular interest). Focused ion beam milling can be used in conjunction with scanning electron microscopy and energy-dispersive x-ray spectroscopy to reveal profiles perpendicular to the polish plane or to sequentially image multiple planes to produce tomographic datasets of buried volumes that yield additional insight on the coating microstructure, as well as actinide and/or fission product distribution.

4.7 Data Acquisition, Analysis, and Reporting

A compact PIE report will be prepared and will include a description of the experiments performed and all relevant data acquired. Overall data to be reported in the compact PIE report will include:

- Analysis results of furnace internals and a final deposition cup efficiency determination
- A compact fractional inventory of fission products released during the safety test, based on as-run inventory calculations
- Compact fractional fission product inventories outside of SiC, as determined by the deconsolidation and acid leaching and based on as-run inventory calculations[4]
- Results of particle inspection and gamma analysis of individual particles
- X-ray and materialographic images, including detailed analyses of particles with low cesium retention
- Discussion of any unusual particle, kernel, or coating behavior that may be linked to fission product releases.

An interim report containing only the results from the CCCTF testing (e.g., fission gas release and activities measured on the deposition cups and furnace internals) could be produced prior to and separately from the report documenting the post-safety-test compact destructive exams.

5. QUALITY ASSURANCE

PIE activities performed at ORNL shall be performed in accordance with the AGR-5/6/7 PIE Plan, applicable ORNL procedures, and the ORNL Quality Assurance Plan for Nuclear Research and Development Activities[7] to meet the INL quality assurance requirements specified in Inter-Entity Work Order #164133.

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