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Irradiated AGR 2 Compact 6 2 2 Examination Plan

John Stempein

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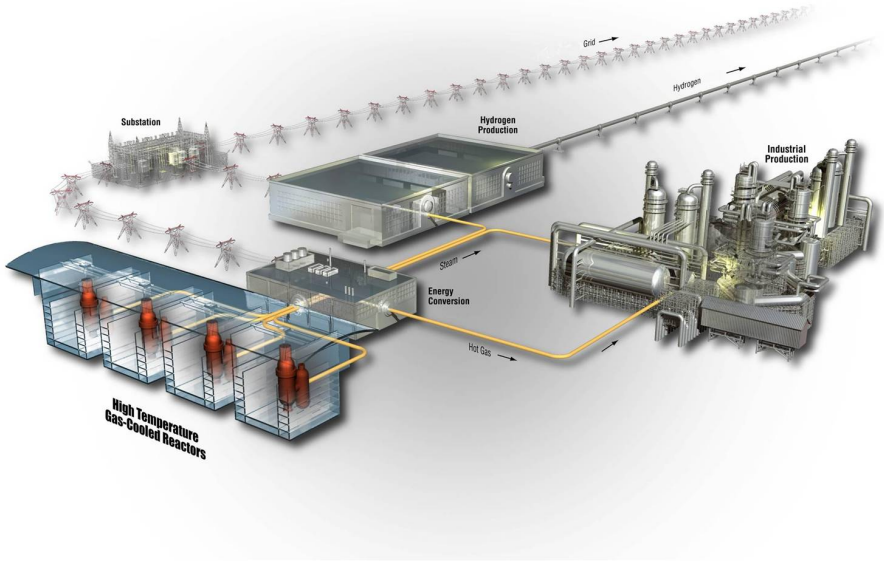
Irradiated AGR-2 Compact 6-2-2 Examination Plan



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REVISION LOG

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ACRONYMS

AGR	Advanced Gas Reactor
AGR-2	Second AGR program irradiation experiment
CCCTF	Core Conduction Cooldown Test Facility
IMGA	Irradiated Microsphere Gamma Analyzer
INL	Idaho National Laboratory
LBL	leach-burn-leach
ORNL	Oak Ridge National Laboratory
PIE	post-irradiation examination
QA	quality assurance
SiC	silicon carbide (coating layer)
TRISO	tristructural isotropic (coated particles)
UCO	uranium oxycarbide (mixture of uranium carbide, uranium dioxide)

1. INTRODUCTION

This plan describes post-irradiation examination (PIE) activities (safety testing and post-safety test analyses) to be performed by Oak Ridge National Laboratory (ORNL) on irradiated Compact 6-2-2 taken from the Advanced Gas Reactor (AGR) experiment, AGR-2. This work will be performed in accordance with general objectives outlined in the AGR-2 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 a mixture of uranium carbide and uranium oxide and was irradiated in Capsule 6 of the AGR-2 test train in the B-12 position in the Advanced Test Reactor at Idaho National Laboratory (INL).[3] Table 1 provides the identifying information for AGR-2 Compact 6-2-2.

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

Compact ID[a]	Compact Container ID	Fabrication ID	Fuel Type	Average Burnup (% FIMA[b])[c]	Fast Fluence $\times 10^{25}$ (n/m^2)[c]	Irradiation Temperature (°C)[d]
AGR-2 6-2-2	AGR247	LEU09-OP2-Z056	UCO	10.19	2.61	1129

[a] The X-Y-Z naming convention denotes the location in the irradiation test train: capsule-level-stack.[1]
[b] FIMA = fissions per initial metal atom.
[c] Based on physics calculations.[4]
[d] Time-averaged, volume-averaged temperature based on thermal calculations.[5]

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 failure of all gas-retentive TRISO layers and cesium release can indicate 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 after the safety test. These measurements will be done by electrolytic deconsolidation of the compact and acid leaching of the particles and matrix debris using the leach-burn-leach (LBL) process described in Section 4.4.1.
- Examine individual particles deconsolidated from the safety-tested compact with the Irradiated Microsphere Gamma Analyzer (IMGA) to quantify retention of specific gamma-emitting fission products (including ^{106}Ru, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu) and to identify anomalous particles, especially those with below-average ^{144}Ce or ^{137}Cs/^{144}Ce ratio indicative of failed TRISO or failed SiC, respectively. If krypton or cesium release in the CCCTF indicates the presence of particles with coating failure, then all particles recovered from the compact will be surveyed with a scan time appropriate for 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. 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 50 randomly selected particles will be performed to examine average fission product retention characteristics.

- Perform microanalysis on selected particles to better understand the correlation of particle microstructure with fission product retention. Microanalysis of particles with below-average ^{144}Ce or $^{137}Cs/^{144}Ce$ ratio is of particular interest and such particles will have the highest priority for examination. Particles with below-average retention of other fission products (e.g., ^{110m}Ag or ^{154}Eu) and particles that are representative of average fission product retention may 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.
- Temporarily archive remaining particles for possible later use until the closeout of the project.

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 photographically documented.

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 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 hours. Radioisotope release will be monitored throughout the test and the amount will help determine the duration of the test. Changes to final heating times or any other test changes will be discussed with the INL Fuels PIE Technical Lead.

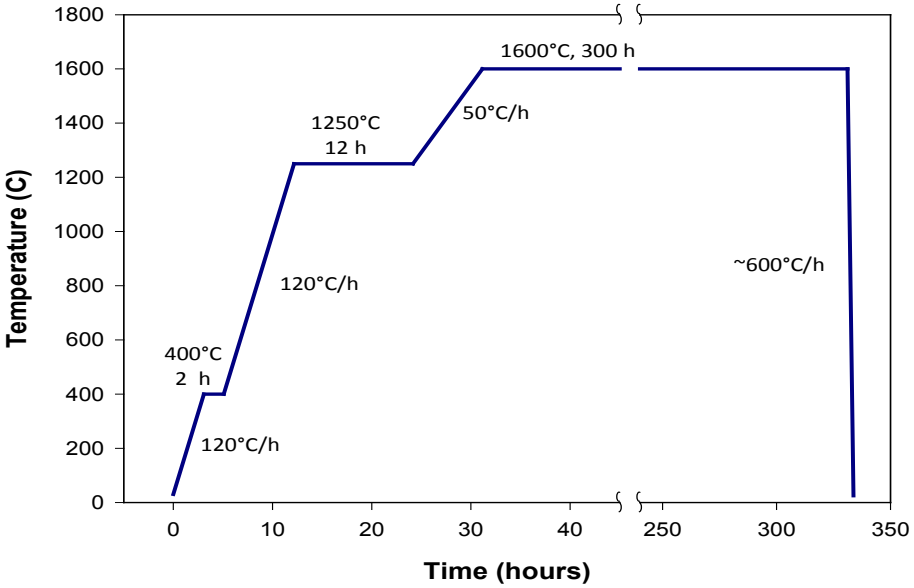

Figure 1. Temperature profile for the safety test of Compact 6-2-2. Elapsed time and ramp rates are approximate.

Table 2. Temperature profile for safety test of AGR-2 Compact 6-2-2.

Approximate Elapsed Time (hours)[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 hours for the next 36 hours (i.e., three more cups after the first cup change at 1600°C). After 36 hours (i.e., four cup changes at 1600°C), the cups will be changed once every 24 hours 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 should not be less than 6 hours or more than 18 hours for the initial 36-hour period at 1600°C or more than 30 hours 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, 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 progress of the safety test. This feedback will allow changes to be made to the test if necessary. After completion of the test, the cups may undergo additional gamma analysis prior to leaching. Analysis of the leach solutions includes measurement of ^{90}Sr and ^{104}Pd, and may include additional analysis 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 leaching is performed. The holder will be oxidized in air and the resultant ash leached 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, 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 fraction of each fission product collected on the tantalum. The combined analysis of all furnace internals will be used to estimate deposition cup collection efficiency for final determination of fission product release during the safety test.

4.4 Post-Safety Test Examination

Post-safety test examination of the fuel compact and TRISO particles will involve deconsolidation and leach-burn-leach, gamma scanning with the IMGA, x-ray tomography to examine the internal microstructure of selected particles, and materialographic examination of particle cross sections with optical and scanning electron microscopes, as described in Sections 4.4.1–4.4.3.

4.4.1 Deconsolidation and Acid Leaching

The 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. Particles and matrix debris will be collected in a Soxhlet thimble, two 24-hour nitric acid leaches in a Soxhlet extractor performed, and the leachates analyzed for uranium and fission products. After the two pre-burn leaches, 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. Recovered particles will be rinsed and dried and transferred to the hot cell cubicle housing the IMGA, where they will undergo inspection and gamma analysis as described in Section 4.4.2.

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 post-burn nitric acid leaches and the leachates analyzed for uranium and fission products.

After completion of the inspection and gamma analysis as described in Section 4.4.2, an archive sample of about 10% will be riffled out, and the remaining 90% of the TRISO particles will be returned to the main cell for particle burn-leach. Particles will be loaded back into the Soxhlet thimble used for pre-burn 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. Analysis of the particle burn-leach solutions will be performed to detect uranium and fission products not leached before the burn, including any exposed kernels from particles with failed SiC not separated out during IMGA analysis. After burn-leach, the burned-back particles will be washed, dried, and archived.

4.4.2 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 count the particles, assess the overall condition, and identify features of interest such as cracked coatings or coating fragments. Abnormal particles or coating fragments may be selected for further 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. A full survey may also be performed with the approval of the 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 (i.e., typically 50–150 seconds) will be used to measure ^{137}Cs and ^{144}Ce and sort out particles with a low cesium-to-cerium ratio or low-cerium content. A below average ^{137}Cs/^{144}Ce ratio is indicative of significant cesium release due to failed SiC or failed TRISO. Below average ^{144}Ce content may indicate abnormal kernels or general fission product loss that could possibly be related to failed TRISO.

After completion of the full IMGA survey (or after particle inspection if the full IMGA survey is not performed), an archive sample of about 10% of the particles will be riffled out and the remaining 90% of

the deconsolidated TRISO particles will be returned to the main cell for burn-leach analysis, as described in Section 4.4.1. Unless present in excessive number, all particles sorted out due to abnormal ^{137}Cs or ^{144}Ce inventory will undergo a longer count time (typically 3–6 hours) to more accurately measure radioisotopic inventory (^{106}Ru, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu) and determine distributions for ^{154}Eu and ^{110m}Ag. Long-count IMGA analysis will also be performed on a random riffled sample of about 50 particles taken from the 10% archive sample.

4.4.3 Microstructural Analysis

After IMGA analysis, particles of interest will be selected for x-ray imaging and/or materialography, with highest priority for 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. Materialography may include optical imaging, scanning electron microscopy imaging, and/or elemental analysis of 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 particles that released cesium due to failed SiC, another goal of this PIE is to identify any mechanisms that may explain why some particles with normal cesium retention indicative of intact SiC release fission products 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 location and composition of fission products or actinide inclusions within the coating layers, primarily in the inner pyrolytic carbon and SiC layers.

X-ray imaging with tomographic reconstruction will be used to achieve non-destructive 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. Three-dimensional visualization of the tomographic data can provide important insight to complement and guide materialographic examination.

Particles 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. Scanning electron microscopy can be used to perform higher resolution inspections of kernel and SiC microstructures. Energy-dispersive and wavelength-dispersive x-ray spectroscopy can be used to characterize the elemental distributions within the kernel and coating layers, as well as fission product attack of the SiC layer, where palladium or uranium clustering and interaction with the SiC are of particular interest.

4.5 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 will include the following, as applicable:

- 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
- Results of particle inspection and IMGA examination of individual particles
- X-ray and materialographic images, including detailed analysis of particles with low-cesium retention

- Discussion of any unusual particle, kernel, or coating behavior that may be linked to fission product releases.

5. QUALITY ASSURANCE

Activities performed at ORNL shall be performed in accordance with applicable ORNL procedures and the ORNL Quality Assurance (QA) Plan for Nuclear Research and Development Activities[7] to meet the INL QA requirements specified in Inter-entity Work Order #150293.

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