Two decades of DOE investment lays the foundation for TRISO-fueled reactors

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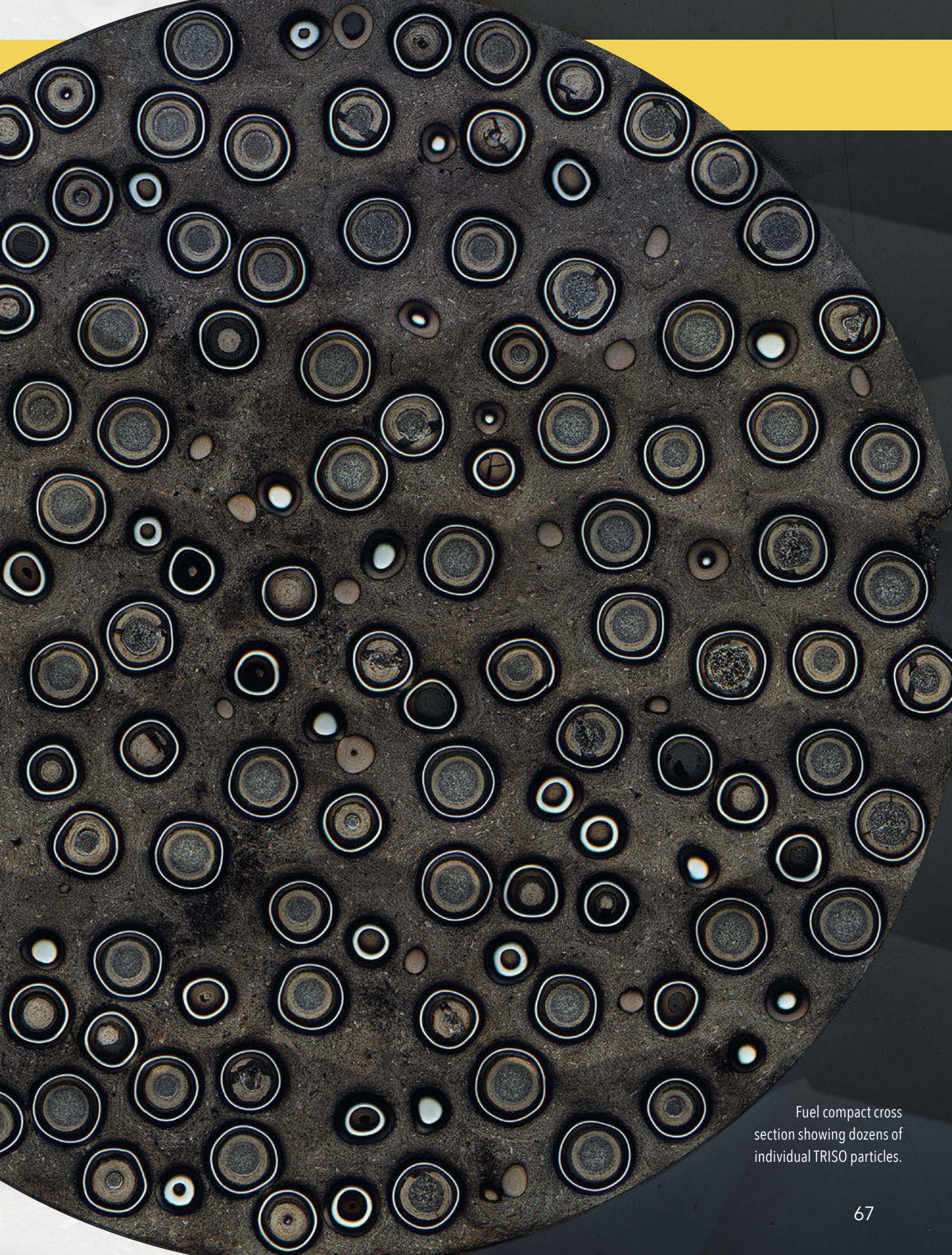
Tristructural isotropic (TRISO) coated particle fuel is a robust, microencapsulated fuel form developed originally for use in high-temperature gas-cooled reactors (HTGRs). The particles consist of a spherical fissile kernel surrounded by several layers of pyrocarbon and a silicon carbide (SiC) layer. The particles are formed into cylindrical or spherical fuel forms using a resinated graphite matrix material for insertion into an HTGR. The kernel and coating layers together act to retain fission products within the particle during normal reactor operation and during postulated accidents; TRISO particles can maintain structural integrity at extremely high temperatures, reaching as high as approximately 1,600 °C in limiting HTGR accidents. This limits the fission product activity circulating in the helium coolant and the activity released to the environment during accidents. Acceptable performance of TRISO particles is therefore essential for reactor safety.

The most common kernel types utilized in modern TRISO particles are uranium dioxide (UO_2) and a mixture of uranium oxide and uranium carbide, often colloquially referred to as uranium oxycarbide or UCO. Despite the origin and historic use of TRISO fuel in HTGRs, the conventional TRISO particle design is now being considered for other types of advanced, high-temperature reactors (HTRs) including the fluoride salt–cooled high-temperature reactor and microreactors. Particles and fuel forms with modified design—including more exotic kernel compositions and different matrix materials but retaining the fundamental TRISO coating structure—are being considered as accident-tolerant fuel for light-water reactors.



Fuel compact cross section showing dozens of individual TRISO particles.

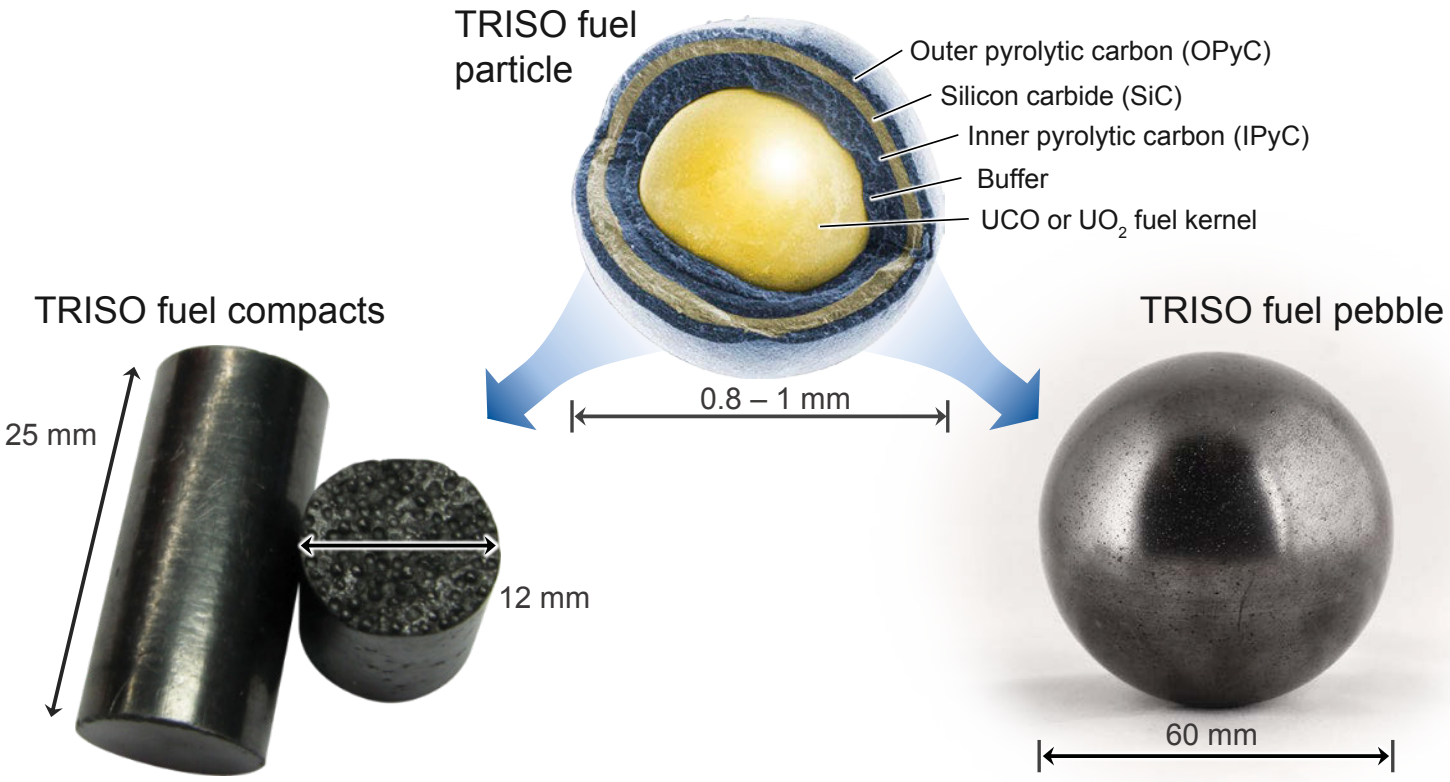


Figure 1. TRISO fuel particle, fuel compacts, and spherical fuel pebble. Fuel pebble image provided by X-energy LLC.

TRISO fuel development program

The U.S. Department of Energy renewed efforts to develop low-enriched uranium (LEU) UCO TRISO fuel by initiating the Advanced Gas Reactor (AGR) Fuel Development and Qualification Program in 2002. The objectives were to help establish a domestic, commercial TRISO fuel fabrication capability in the United States and to generate fuel performance data that can support high-temperature reactor design and licensing efforts. To accomplish these objectives, the program set out to demonstrate high-quality TRISO fuel fabrication at the pilot scale with low coating defect levels, and perform irradiation testing and post-irradiation safety testing to provide data on fuel performance under normal reactor operating conditions and anticipated accident conditions. This work has been performed in the framework of a program compliant with ASME NQA-1 quality standards to ensure the pedigree of generated data.

The AGR program was established initially to support the DOE Next Generation Nuclear Plant (NGNP) project, which sought to deploy a modular HTGR in the United States to take advantage of enhanced safety features of this Generation IV reactor design. Although the DOE chose not to pursue NGNP design in 2011, the AGR program continues currently under the DOE Office of Advanced Reactor Technologies to support the licensing of HTR designs now being pursued by U.S. commercial reactor designers.

Fuel fabrication

The program has focused on UCO TRISO fuel in cylindrical compact form (see Figure 1), based upon previous fuel development efforts for modular prismatic HTGRs in the United States. The selection of UCO kernels over UO_2 was based on performance benefits of this fuel type at higher burnups (>10 percent fissions per initial metal atom, or FIMA), a result of a significantly lower tendency of UCO kernels to form carbon monoxide gas within the particles during irradiation. Early work was dedicated to developing

fuel performance models to help evaluate past TRISO fuel performance deficiencies and establishing fuel property specifications that would result in acceptable particle performance. Initial fuel fabrication efforts concentrated on the production of high-quality fuel at the laboratory scale, using lab-scale coating and compacting equipment and processes at Oak Ridge National Laboratory (ORNL). Work then progressed in a phased approach to scale up fabrication to the pilot scale, first with TRISO coating, and finally with fuel compact fabrication by BWXT Nuclear Operations Group.

An important measure of as-fabricated TRISO fuel quality is the number of defects in the coating layers and the amount of dispersed uranium (i.e., uranium located in the fuel compact outside of the SiC layer of particles). A key defect type is called an "exposed kernel defect" because the coatings on such a particle are damaged or defective, such that fission gas is released from the particle during irradiation. The fraction of such defects was substantially less than 10^{-5} (less than one particle per 100,000) for the laboratory-scale fuel, and tended to be higher in the pilot-scale fuel used for AGR-2, with values of approximately 5×10^{-5}, highlighting one of the challenges of increasing the scale of fuel fabrication. Similarly, the dispersed uranium was extremely low in the laboratory-scale fuel (up to 4×10^{-7}) and higher by roughly a factor of 10 in the pilot-scale fuel. Information gained from this initial pilot effort can be used in follow-on efforts by commercial fuel fabricators to eliminate the causes of these observed defect fraction increases and develop more optimal large-scale processes.

Irradiation experiment overview

A series of irradiation tests have been executed to evaluate the performance of the fuel produced in each fabrication campaign. The objective was to test fuel under a broad range of service conditions, including time-average, volume-average temperature up to 1,250 °C, burnup to 20 percent FIMA, and fast neutron fluence to approximately 5×10^{25} n/m^2. In addition, a dedicated irradiation test has been performed to provide data on fission product transport in fuel and reactor core graphite materials. Essential features of all irradiations included the online monitoring of fission gas activity released from the fuel to assess the status of the particles, and computational physics and thermal modeling of the capsules to predict uranium depletion and fuel temperatures.

The irradiations have all been performed in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL); the first three are completed, and the fourth and final test will be completed in 2020. All the irradiation experiments involved multiple instrumented capsules with dedicated gas flows. In all experiments, the individual capsules were welded together into a single assembly or "test train" for insertion into the ATR; the experiments have contained between five and 12 capsules. This design approach maximized the axial length of the ATR core that could be used, providing a large test volume to irradiate fuel particles in compacts over a range of burnup and fast neutron fluence profiles while still allowing fuel temperatures in all compacts to be maintained within the desired ranges. The tests are summarized on the next page, and an accompanying table presents some of the key features of the four irradiation experiments.

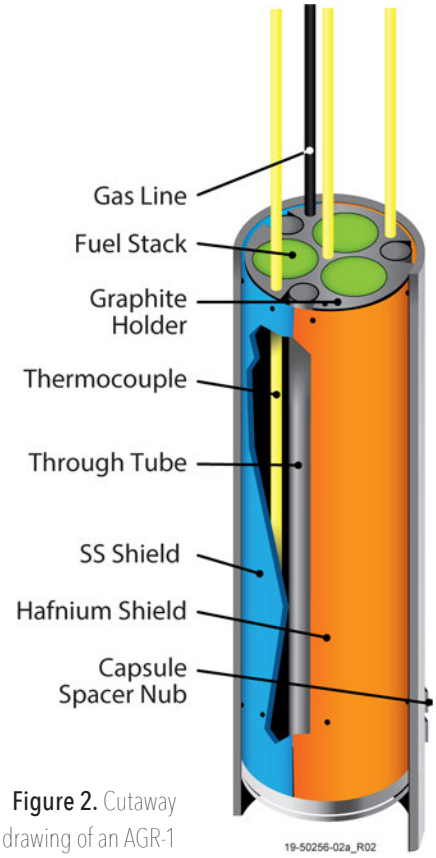


Figure 2. Cutaway drawing of an AGR-1 irradiation capsule.

■ **AGR-1:** This experiment represented a shakedown test of the multicapsule, instrumented test train design approach that would be used in all subsequent experiments and was meant to assess the performance of fuel fabricated at the laboratory scale. Kernels fabricated at BWXT were coated in a 50-mm-diameter cylindrical chamber and formed into compacts at ORNL. Several different fuel particle lots were included, involving variations in fabrication parameters for either the inner pyrolytic carbon layer or the SiC layer, to explore the impact of layer properties on fuel performance. Figure 2 shows a drawing of an AGR-1 irradiation capsule. The irradiation was performed from December 2006 to November 2009, achieving peak burnup of 19.6 percent FIMA.

■ **AGR-2:** This experiment was a performance demonstration for coated particles fabricated at BWXT in a pilot-scale process, using a 150-mm-diameter coating chamber. The test included particles fabricated from both UCO and UO_2 kernels. The UO_2 fuel was included to compare performance of the two kernel types and was driven by interest in UO_2-fueled pebble-bed reactors. Coated particles were compacted at ORNL using a laboratory-scale process similar to that used for AGR-1 fuel. The test train had a design very similar to AGR-1: three capsules contained 12 UCO compacts each, while a fourth contained 12 UO_2 compacts fabricated in the United States. The remaining two capsules contained UO_2 TRISO fuel supplied from CEA (French Alternative Energies and Atomic Energy Commission) in France and PBMR (Pebble Bed Modular Reactor, Ltd) in South Africa as part of the Generation IV collaboration. One of the U.S. UCO capsules—Capsule 2—was intentionally operated at significantly higher temperatures to test the upper temperature margin of fuel performance. Time-average peak temperature in this capsule was 1,360 °C. The irradiation was performed from June 2010 to October 2013.

■ **AGR-3/4:** This experiment was dedicated to studying fission product transport in fuel compact matrix material and reactor-grade graphite. This was accomplished by fabricating fuel compacts in which approximately 1 percent of the particles were "designed to fail" (DTF). During the irradiation, the singular, thin pyrocarbon coating on each DTF particle failed, and the particles released fission products, which migrated through the surrounding materials. The intact "driver" fuel particles were similar to the AGR-1 TRISO particles. The test train included 12 separate capsules, which allowed a large range of fuel temperatures (see table) and two different grades of graphite to be investigated. The experiment was irradiated from December 2011 to April 2014.

■ **AGR-5/6/7:** This is the final fuel qualification and performance margin irradiation experiment. It includes UCO coated particles and fuel compacts, all fabricated at BWXT in pilot-scale processes. The test train includes five separate capsules and approximately 570,000 particles. Originally planned as three separate irradiation experiments (designated AGR-5, AGR-6, and AGR-7) taking place in large B positions in the ATR, the experiments were combined into a single test designed for the much larger ATR northeast flux trap position. The central capsule (Capsule 3) comprises the AGR-7 portion of the experiment, which is a high-temperature fuel performance margin test designed to explore fuel behavior at temperatures significantly exceeding those expected during normal operation in an HTGR. The remaining capsules comprise the AGR-5/6 experiment. For this ambitious experiment, the peak burnup will be approximately 16 percent FIMA and the time-average maximum temperature target is 1,500 °C. The irradiation started in February 2018 and is expected to be completed in 2020.

	AGR-1	AGR-2	AGR-3/4	AGR-5/6/7
Description	Test of lab-scale coated particles and compacts	Performance test of pilot-scale coated particles in lab-scale compacts	Fission product transport experiment; includes 1% designed-to-fail particles to release fission products during irradiation	Fuel qualification and performance margin test (time-average peak temperatures up to 1,500 °C)
Kernel type	UCO	UCO UO_2	UCO	UCO
Average kernel diameter (μm) / enrichment (wt% ^{235}U)	350 / 19.7	427 / 14.0 508 / 9.6	357 / 19.7	426 / 15.5
Compacts/ particles	72 / 298,000	36 / 114,000 12 / 18,500	48 / 91,000	170 / 570,000
Fuel Temp (°C)[a]	1,069 – 1,197	1,080 – 1,360 1,072 – 1,105	865 – 1,418	~700 – 1,500[b]
Burnup (% FIMA)	11.3 – 19.6	7.3 – 13.2 9.0 – 10.7	4.9 – 15.3	~6 – 16[c]

a. Time-average peak fuel temperature range for all compacts
b. Targeted values; experiment temperature analysis is not complete
c. Projected end-of-life values

TRISO fuel performance

The key measure of TRISO fuel performance during irradiation is the magnitude of fission gas release. The basis of this approach is that intact TRISO layers retain fission gases extremely well, and therefore small increases in the total release can indicate TRISO failure (failure of all three dense coating layers). The fission gas release is measured by continuously monitoring the sweep gas exiting each capsule using gamma spectrometers. Fission gas release is subsequently evaluated using the release-to-birth (R/B) ratio, which compares the measured release rate for a specific isotope to the rate of generation in the fuel from fission.

The AGR-1 end-of-life ^{85m}Kr R/B values were 0.2–2 × 10^{-7} for the six capsules (note that a value of 10^{-7} indicates that one ^{85m}Kr atom is released from the fuel for every 10 million ^{85m}Kr atoms produced by fission). These low values indicated that no particles out of the approximately 300,000 in the experiment experienced TRISO failure during the irradiation. The ^{85m}Kr R/B values at the beginning of the AGR-2 irradiation were ~6 × 10^{-7} in the two capsules containing UCO at normal operating temperatures, and 10^{-6} in Capsule 2 with UCO particles operating at a higher irradiation temperature. The higher values in this experiment reflect higher uranium contamination outside of particle coatings in this fuel relative to AGR-1 fuel. Experimental issues during the AGR-2 irradiation resulted in failure of some of the gas lines and mixing of the gas streams from different capsules, which prevented determination of end-of-life R/B values from individual capsules.

Post-irradiation examination (PIE) further helps to elucidate in-pile fuel performance. PIE for the AGR-1 experiment is complete and the AGR-2 PIE is nearing completion. Two key aspects that hold the predominant focus of coated particle fuel performance evaluation are coating layer failure rates and fission product release rates, and a large volume of data on these as-

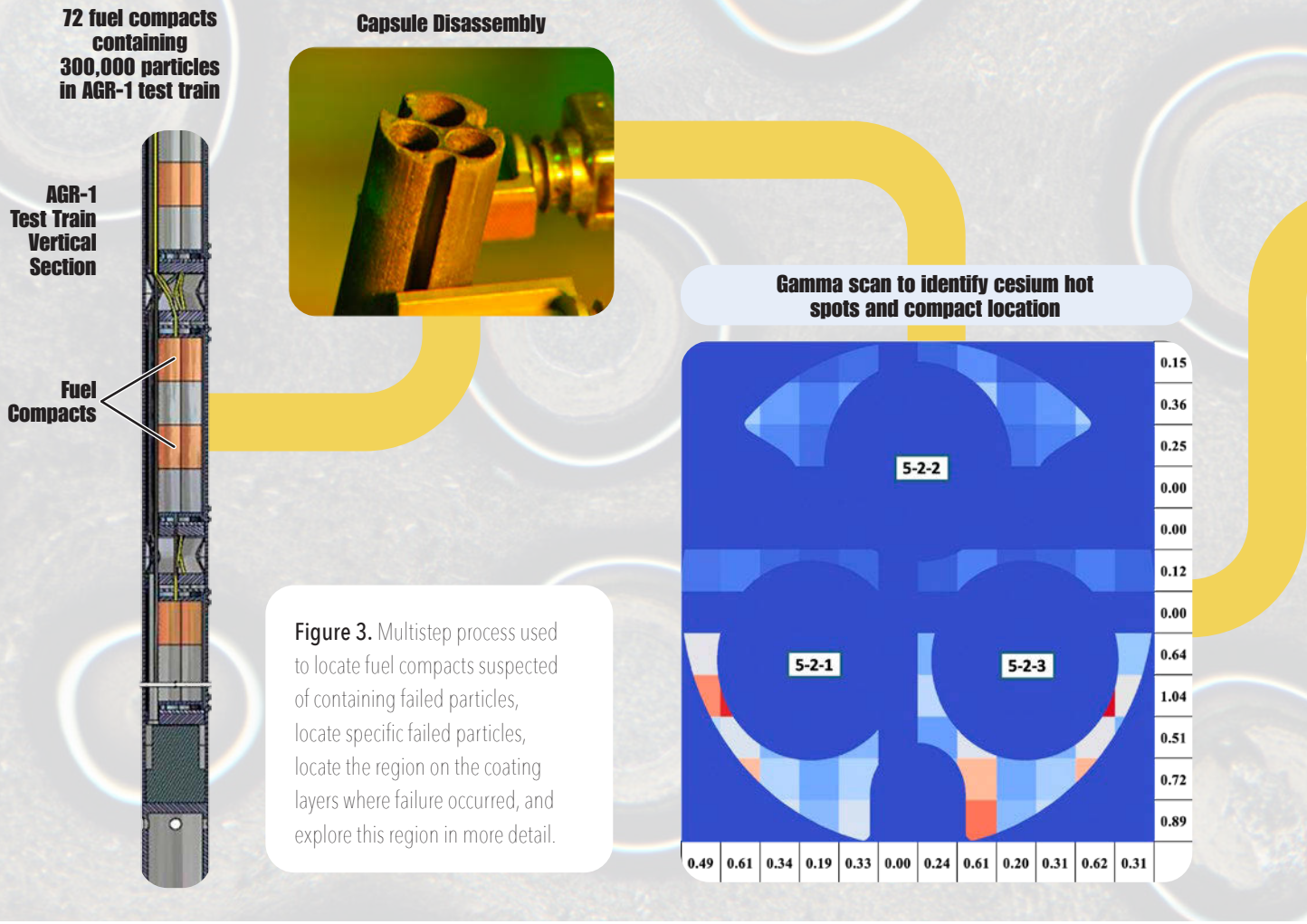


Figure 3. Multistep process used to locate fuel compacts suspected of containing failed particles, locate specific failed particles, locate the region on the coating layers where failure occurred, and explore this region in more detail.

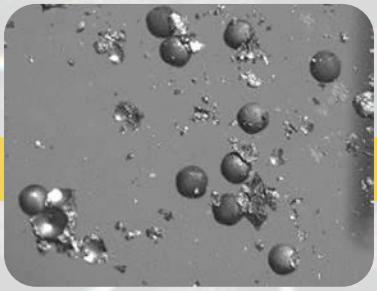
pects has been obtained from the AGR-1 and -2 irradiation and PIE campaigns. TRISO failure rates during irradiation were determined primarily by R/B data, as discussed above. Where fission gas release data during irradiation were inconclusive, additional information from PIE was used to help evaluate TRISO failure rates.

In addition to PIE, high-temperature safety tests were performed on the irradiated fuel specimens to assess fuel behavior at temperatures significantly exceeding those experienced during irradiation, and more representative of temperatures achieved during reactor accidents involving a loss of helium coolant flow. Tests were performed at temperatures of 1,500–1,800 °C for several hundred hours in pure helium while measuring the time-dependent release of fission products, including isotopes of silver, cesium, europium, strontium, and krypton. This provided critical information on the level of fission product release from the fuel as well as the impact of high temperatures on coating layer failure; elevated coating layer failure rates are to be expected because of thermally driven degradation of the coating layers at these extreme temperatures. The data obtained from these tests is an essential component of the safety evaluation of TRISO fuel.

Failure of the particle SiC layer (where at least one of the pyrocarbon layers remains intact and continues to retain fission gases) was determined by examining fission product cesium release from the fuel during PIE—an indication of SiC failure, since intact pyrocarbon alone poorly retains cesium—and studying suspect particles in detail. Advances in PIE methods in the AGR program have made this possible, and enabled researchers to find and isolate for analysis a small number of particles with failures out of hundreds of thousands of nonfailed particles

Identify particles with failed coatings

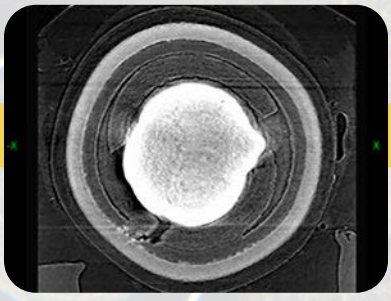
Deconsolidation to obtain ~4,000 particles from compact



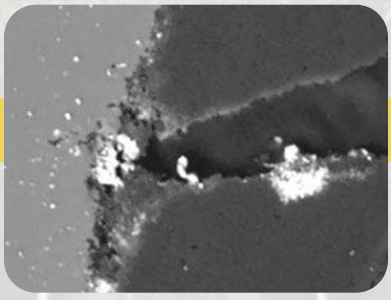
Gamma count to find particles with low cesium retention

Study particles with failed coatings

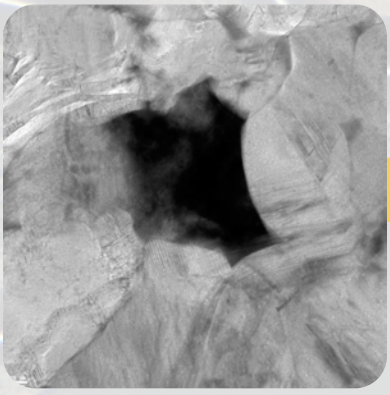
X-ray tomography to locate failures



Materialography to expose defective region for analysis



Advanced microscopy to study coating layers in detail



in the irradiation experiments.

The basic process for isolating and examining failed particles is outlined in Figure 3, and involves: gamma scanning the graphite components that retained the fuel compacts during irradiation to find "hot spots" of cesium that can indicate the proximity of particles with failed coatings; deconsolidating suspect compacts to obtain all the particles (up to ~4,000 particles in an AGR compact); gamma counting each particle to identify those with relatively low cesium, indicative of release; performing nondestructive examination of these particles with x-radiography to observe the morphology of the coatings; and focusing destructive microanalysis (e.g., scanning electron microscopy, transmission electron microscopy, elemental analysis) on the exact location where the coating layer(s) failed. The process is also applied following safety tests when the fission product release data indicate one or more particles experienced coating layer failures. As an example, it was determined that four particles out of approximately 300,000 in the AGR-1 irradiation experiment experienced SiC layer failure, and these particles were found and studied to better understand the failure mechanisms.

Figure 4 displays the experimentally determined coating failure rates during AGR-1 irradiation and subsequent high-temperature post-irradiation safety testing at various temperatures. This includes both TRISO failures and less-extreme SiC failures. The values on the plot are based on the combined results from the AGR-1 irradiation, PIE, and safety testing, and are the calculated upper limit on the failure rates at 95 percent confidence. Historic high-temperature gas-cooled reactor design specifications for allowable failure fractions during normal operation (2×10^{-4}) and during high-temperature accidents at temperatures up to 1,600 °C (6×10^{-4}) are shown on the plot for comparison. The results indicate significant safety margin.

Microanalysis of fuel particles following irradiation (and also after safety tests) has been performed to understand kernel and coating morphology evolution as a function of neutron irradiation and time at temperature, assess coating damage, and better understand fission product transport in the coating layers. This has included imaging of thousands of particle cross sections using optical microscopy and examining a subset of these particles with scanning electron microscopy and elemental analysis. Figure 5 shows typical particle cross sections before and after irradiation. More detailed analyses have also been performed using scanning transmission electron microscopy and related tools to understand fission product transport at the nanometer length scale. Nondestructive examination of irradiated particles using X-ray imaging and tomographic reconstruction has been an integral tool in the program to understand coating failure mechanisms and focus subsequent destructive ex-

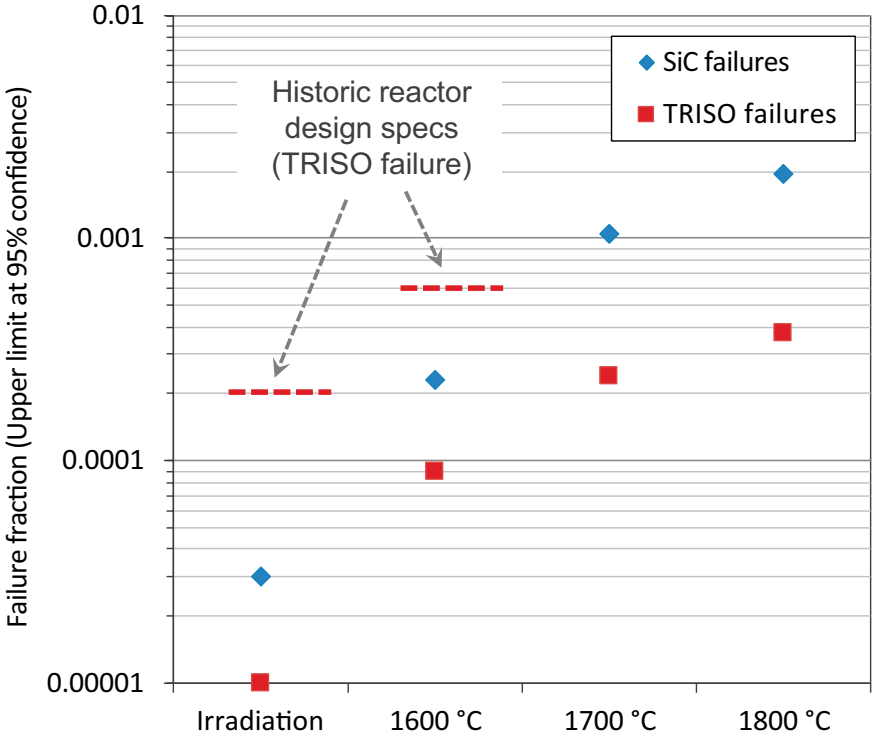


Figure 4. Failure fractions observed in AGR-1 irradiation and safety tests.

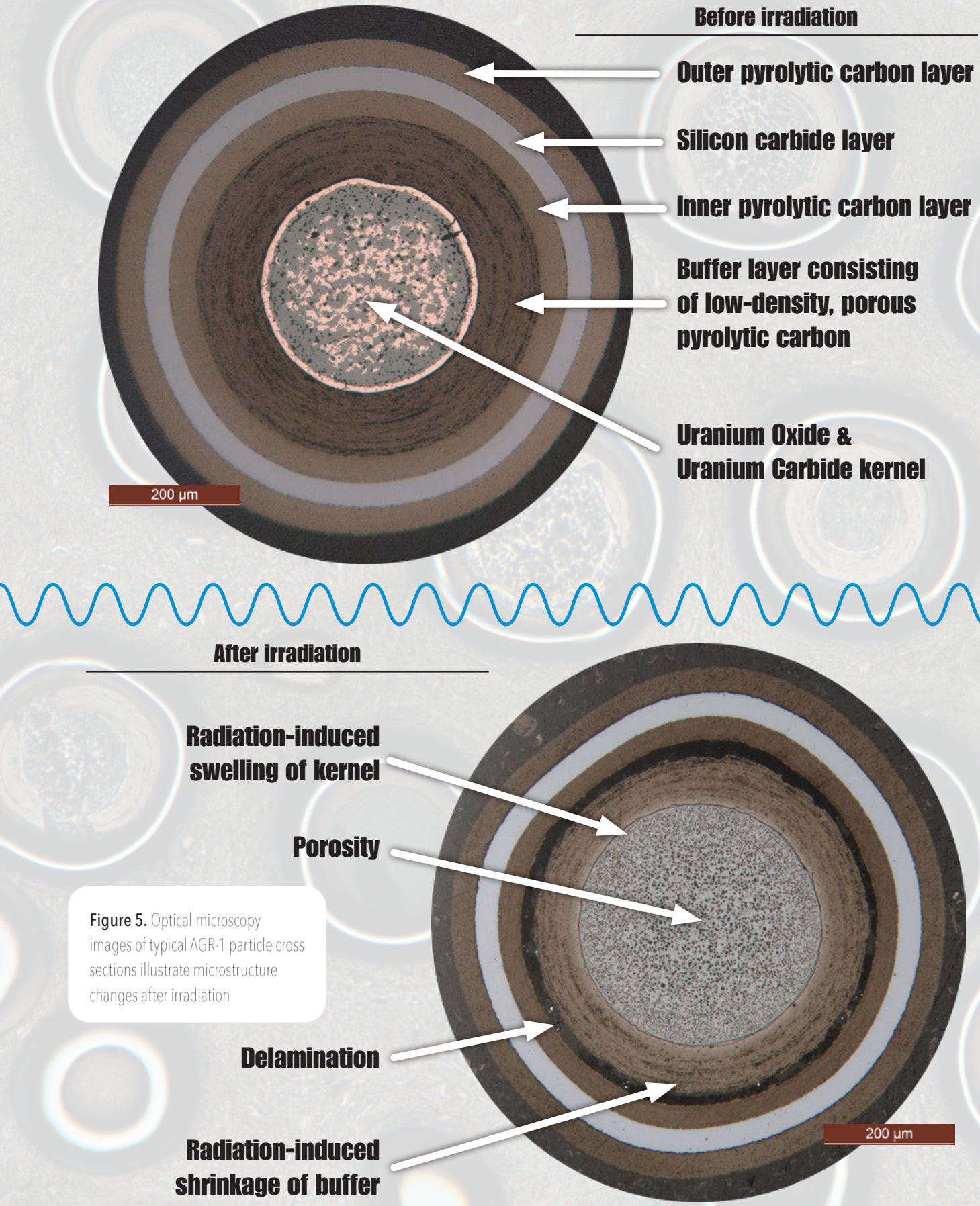


Figure 5. Optical microscopy images of typical AGR-1 particle cross sections illustrate microstructure changes after irradiation

ams to specific regions of interest.

The data from the AGR-1 and AGR-2 experiments demonstrate some broad trends in fission product release behavior of this fuel form. The particles retain fission gas exceptionally well when any of the dense coating layers remain intact and retain cesium nearly completely when the SiC layer remains intact. Hence the release of these fission products is dependent primarily on coating failure rates, which are very low, as discussed above. Europium and strontium are released in modest amounts from intact TRISO particles; the total release fraction from fuel compacts under normal HTGR operating temperatures is less than ~5×10^{-4}. As observed over decades of TRISO fuel irradiation experience, silver transports fairly readily out of intact TRISO particles at temperatures above 1,000–1,100 °C. Silver behavior in individual coated particles in these two irradiation experiments depended primarily on fuel temperature and ranged from nearly complete retention to nearly complete release.

The AGR-1 and AGR-2 fuel performance results were recently compiled and submitted to the Nuclear Regulatory Commission in a Topical Report by EPRI (see Topical Report EPRI-AR-1 [NP]), in partnership with INL and industry participants in the Nuclear Energy Institute's High-Temperature Reactor Technology Working Group ("TRISO fuel nears qualification," *Nuclear News*, September 2019). The NRC's review of the report is currently in the final stages, with a safety evaluation potentially to be issued in summer 2020. This safety evaluation will help accelerate licensing of advanced high-temperature reactor designs, by obtaining NRC review and approval of key AGR program data demonstrating TRISO fuel particle performance under high-temperature reactor conditions.

PIE of the AGR-3/4 irradiation experiment is still in progress. This focuses on examining the irradiated fuel compacts and matrix/graphite components of the capsules to measure the distribution of fission products silver, cesium, europium, and strontium within these specimens. Data on fission product migration during the irradiation will allow researchers to refine the fundamental parameters (including diffusivities) that govern the transport of these elements through the graphitic matrix and reactor core materials. This vital data will support development of fission product transport models used to predict the radiological source terms during reactor operation and accidents.

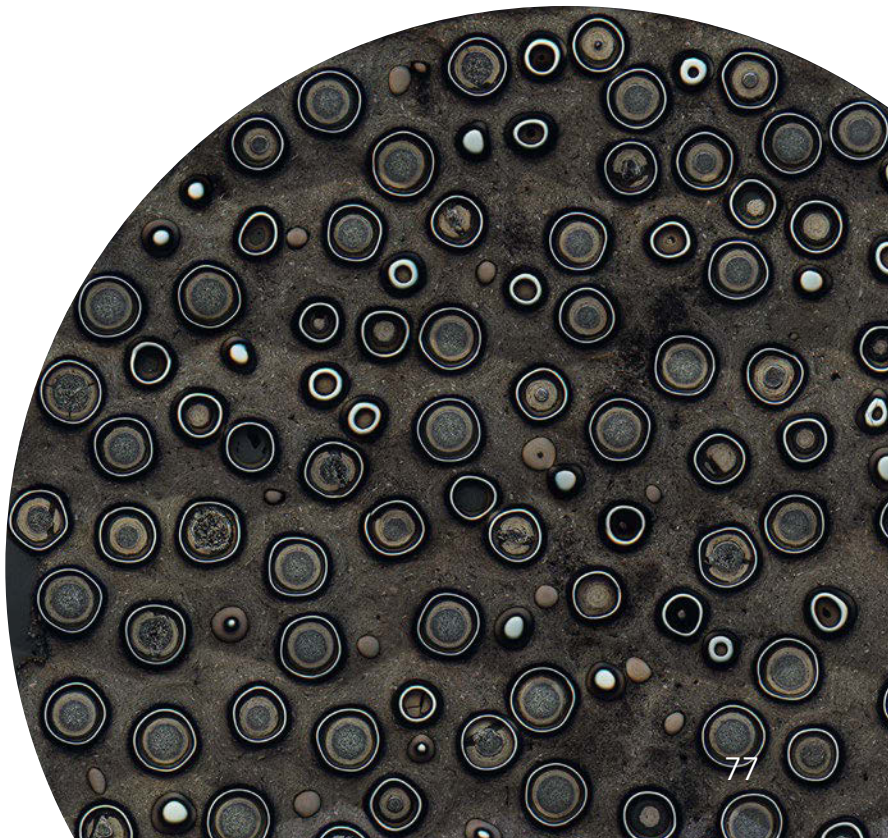
Looking ahead, the key remaining activities in the program are the completion of the AGR-5/6/7 irradiation, followed by PIE and safety testing. A critical part of this work—and a

Further reading:

- D. A. Petti et al., "The DOE Advanced Gas Reactor Fuel Development and Qualification Program," *JOM*, 62, 62 (2010); doi.org/10.1007/s11837-010-0140-5.
- P. A. Demkowicz, J. D. Hunn, D. A. Petti, and R. N. Morris, "Key Results from Irradiation and Post-Irradiation Examination of AGR-1 UCO TRISO Fuel," *Nucl. Eng. Des.*, 329, 102 (2018); doi.org/10.1016/j.nucengdes.2017.09.005.
- J. D. Hunn et al., "Post-Irradiation Examination and Safety Testing of US AGR-2 Irradiation Test Compacts," *Proc. 9th Int. Topl. Mtg. High Temperature Reactor Technology (HTR-2018)*, Warsaw, Poland, October 8–10, 2018.
- P. A. Demkowicz, B. Liu, and J. D. Hunn, "Coated Particle Fuel: Historical Perspectives and Current Progress," *J. Nucl. Mater.*, 515, 434 (2019); doi.org/10.1016/j.jnucmat.2018.09.044.
- H. D. Gougar et al., "The US Department of Energy's High Temperature Reactor Research and Development Program—Progress as of 2019," *Nucl. Eng. Des.*, 358, 110397 (2020); doi.org/10.1016/j.nucengdes.2019.110397.

component of the fuel qualification program that has not been addressed in previous experiment campaigns—is the evaluation of fuel performance at high temperatures in oxidizing environments. Work is currently underway to develop a capability for heating irradiated fuel specimens in the presence of various concentrations of oxygen or moisture while measuring the release of gaseous and condensable fission products. These tests are crucially important for assessing the behavior of the fuel in conditions that could exist in a high-temperature reactor during inadvertent ingress of oxidants (steam or air) into the core.

When complete, the data obtained from this qualification program will support advanced reactor licensing by demonstrating fuel performance under a range of operating conditions and by providing data to refine fission product transport models used in reactor design and safety analyses. In response to the widespread interest in TRISO fuel, two U.S. companies have announced that they are actively developing commercial TRISO fuel fabrication capabilities. BWXT has restarted its TRISO fuel production line in Lynchburg, Va., where it is using and expanding on capabilities applied in the pilot-scale fabrication of AGR-2 and AGR-5/6/7 fuel compacts. Also, with funding support from the DOE, X-energy LLC is pursuing development of pilot-scale fuel fabrication equipment and methods in a facility at ORNL and is working to establish and license its own TRISO fuel fabrication facility. Much of what has been developed and learned over the past two decades by the DOE AGR program, including aspects of fuel fabrication, optimization of TRISO particle design, modernization of characterization and inspection methods for quality control, instrumented irradiation testing, post-irradiation examination, and safety testing will be leveraged by these two companies and others interested in pursuing TRISO-based fuel technologies.



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