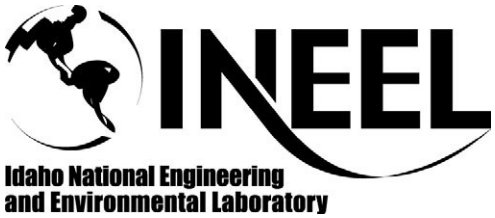

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The DOE Advanced Gas Reactor (AGR) Fuel Development And Qualification Program

David Petti
Gary Bell
AGR Team

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The DOE Advanced Gas Reactor (AGR) Fuel Development and Qualification Program

David Petti[a], Gary Bell[b], and the AGR Team

[a]*David A. Petti*
Idaho National Laboratory
P.O. Box 1625
Idaho Falls, ID 83415 U.S.A.
(208)526-7735
David.Petti@inl.gov

[b]*Gary Bell*
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37381 U.S.A
(865)241-4400,
bellgl@ornl.gov

Abstract

The Department of Energy has established the Advanced Gas Reactor Fuel Development and Qualification Program to address the following overall goals:

- Provide a baseline fuel qualification data set in support of the licensing and operation of the Next Generation Nuclear Plant (NGNP). Gas-reactor fuel performance demonstration and qualification comprise the longest duration research and development (R&D) task for the NGNP feasibility. The baseline fuel form is to be demonstrated and qualified for a peak fuel centerline temperature of 1250°C.
- Support near-term deployment of an NGNP by reducing market entry risks posed by technical uncertainties associated with fuel production and qualification.
- Utilize international collaboration mechanisms to extend the value of DOE resources.

The Advanced Gas Reactor Fuel Development and Qualification Program consists of five elements: fuel manufacture, fuel and materials irradiations, postirradiation examination (PIE) and safety testing, fuel performance modeling, and fission product transport and source term evaluation.

An underlying theme for the fuel development work is the need to develop a more complete fundamental understanding of the relationship between the fuel fabrication process, key fuel properties, the irradiation performance of the fuel, and the release and transport of fission products in the NGNP primary coolant system. Fuel performance modeling and analysis of the fission product behavior in the primary circuit are important aspects of this work. The performance models are considered essential for several reasons, including guidance for the plant designer in establishing the core design and operating limits, and demonstration to the licensing authority that the applicant has a thorough understanding of the in-service behavior of the fuel system. The fission product behavior task will also provide primary source term data needed for licensing. An overview of the program and recent progress will be presented.

I. INTRODUCTION

In the coming decades, the United States, the other industrialized countries, and the entire world will need energy supplies and an upgraded energy infrastructure to meet growing demands for electric power and transportation fuels. The Generation IV initiative identified reactor system concepts for producing electricity that excelled at meeting the goals of superior economics, safety, sustainability, proliferation resistance, and physical security. One of these reactor system concepts, the Very High Temperature Gas Cooled Reactor System (VHTR), is also uniquely suited for producing hydrogen without the consumption of fossil fuels or the emission of greenhouse gases. DOE has selected this system for the Next Generation Nuclear Plant (NGNP) Project, a project to demonstrate emissions-free nuclear-assisted electricity and hydrogen production by 2015.

The NGNP reference concept will be a helium-cooled, graphite moderated, thermal neutron spectrum reactor with a design goal outlet temperature of 900-1000°C. The reactor core could be either a prismatic graphite block type core or a pebble bed core; the final selection of a reference core concept will be made following completion of the pre-conceptual designs for each. The NGNP will be able to produce both electricity and hydrogen. The process heat for hydrogen production will be transferred to the hydrogen plant through an intermediate heat exchanger (IHX). The reactor thermal power (about 600 MWt) and core configuration will be designed to assure passive decay heat removal without fuel damage during hypothetical accidents. The fuel cycle will be a once-through very high burnup low-enriched uranium fuel cycle.

The fuel for the NGNP builds on the potential of the TRISO-coated particle fuel design demonstrated in high temperature gas-cooled reactors in the UK, U.S., Germany, and elsewhere. The TRISO-coated particle is a spherical-layered composite about 1 mm in diameter. It consists of a kernel of uranium dioxide (UO_2) or uranium oxycarbide (UCO) surrounded by a porous graphite buffer layer that absorbs radiation damage and allows space for fission gases produced during irradiation. Surrounding the buffer layer are a layer of dense pyrolytic carbon called the inner pyrolytic carbon (IPyC), a silicon carbide (SiC) layer, and a dense outer pyrolytic carbon layer (the OPyC). The pyrolytic carbon layers shrink under irradiation and create compressive forces that act to protect the SiC layer, which is the primary pressure boundary for the microsphere. The inner pyrolytic carbon layer also protects the kernel from corrosive gases present during deposition of the SiC layer. The SiC layer provides the primary containment of fission products generated during irradiation and under accident conditions. Each microsphere acts as a mini pressure vessel, a feature intended to impart robustness to the gas reactor fuel and plant safety system.

The baseline fuel kernel for the NGNP is low-enriched (about 15% U-235 in the prismatic block reactor version of the NGNP and about 8% in the pebble bed version) UCO instead of UO_2, owing to performance issues associated with the UO_2 fuel at high power, temperature, and burnup. At the high power densities expected in the NGNP (>6 W/cm^3), the associated large thermal gradients can drive kernel migration in UO_2-coated particles. Migration of the kernel through the buffer and inner pyrocarbon layers and subsequent contact with the SiC layer can result in damage to the SiC layer. Furthermore and more importantly, at the high burnups proposed for NGNP (15 to 20% FIMA), the CO and fission product gas pressure in a UO_2 fuel particle can be substantial, resulting in particle failure, especially under accident conditions. The high NGNP fuel temperatures (maximum time averaged temperature ~1250˚C) increase the effect of both of these mechanisms. UCO was selected because the mixture of carbide and oxide components precludes free oxygen from being released due to fission. As a result, no carbon monoxide is generated during irradiation, and little kernel migration (amoeba effect) is expected. Yet, like UO_2, the oxycarbide fuel still ties up the lanthanide fission products as immobile oxides in the kernel, which gives the fuel added stability under accident conditions.

For the pebble bed version of a NGNP, the coated particles are over-coated with a graphitic powder and binders. These over-coated particles are then mixed with additional graphitic powder and binders and then molded into a 50-mm-diameter sphere. An additional 5-mm fuel free zone layer is added to the sphere before isostatic pressing, machining, carbonization, and heat-treating.

For the prismatic version of the NGNP, a similar process is envisioned, where the over-coated particles are mixed with graphitic powder and binders to form a cylindrical compact about 50 mm long and 12.5 mm in diameter. After final heat treatment, these compacts are inserted into specified holes in the graphite blocks. Figure 1 shows a sketch of a TRISO-coated fuel particle and photographs of fuel particles, compacts, and fuel elements (prismatic blocks of graphite with fuel compacts and coolant channels) used in the high-temperature gas reactor at Fort St. Vrain. The Advanced Gas Reactor Fuel Development and Qualification (AGR) Program is currently focusing on the prismatic fuel form.

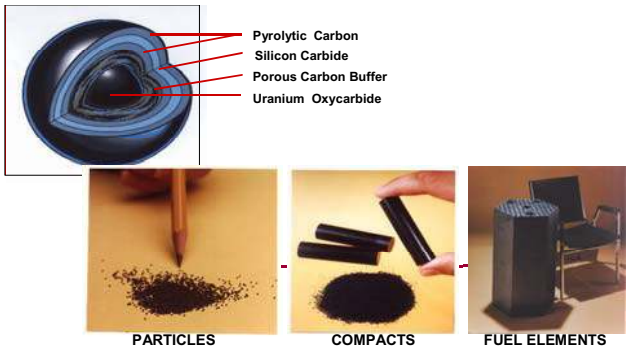

Figure 1. Cutaway of TRISO-coated fuel particle and pictures of prismatic fueled high temperature gas reactor fuel particles, compacts, and fuel elements.

The goals of the program are:

- Provide a baseline fuel qualification data set in support of the licensing and operation of the NGNP. Gas-reactor fuel performance demonstration and qualification comprise the longest duration research and development (R&D) task for NGNP feasibility. The baseline fuel form is to be demonstrated and qualified for a peak fuel centerline temperature of 1250°C.
- Support near-term deployment of an NGNP by reducing market entry risks posed by technical uncertainties associated with fuel production and qualification.
- Utilize international collaboration mechanisms (such as through the IAEA) to extend the value of DOE resources.

There are five elements in the Advanced Gas Reactor Fuel Development and Qualification Program: fuel manufacture, fuel and materials irradiations, postirradiation examination (PIE) and safety testing, fuel performance modeling, and fission product transport and source term evaluation. These are discussed in detail in the following sections.

An underlying theme for the fuel development work is the need to develop a more complete fundamental understanding of the relationship between the fuel fabrication process, key fuel properties, the irradiation performance of the fuel, and the release and transport of fission products in the NGNP primary coolant system. Fuel performance modeling and analysis of the fission product behavior in the primary circuit are important aspects of this work. The performance models are considered essential for several reasons, including guidance for the plant designer in establishing the core design and operating limits, and demonstration to the licensing authority that the applicant has a thorough understanding of the in-service behavior of the fuel system. The fission product behavior task will also provide primary source term data needed for licensing.

II. ADVANCED GAS REACTOR (AGR) PROGRAM JUSTIFICATION AND NEED

A recent review [1] concludes that there has historically been a difference in the quality of U.S. and German high temperature gas reactor fuel [2]. This fact is illustrated in Figure 2 where the krypton release rate to birth rate (R/B) measurements from most of the U.S. and German TRISO coated fuel irradiation experiments are plotted versus fast fluence. The U.S. data from individual experiments is shown as lines whereas the yellow band in Figure 2 shows the range of the German data. This difference has been traced to technical differences in the fabrication processes used in Germany and the United States, as well as differences in the irradiation and testing programs in the two countries. Review of the fabrication processes used in Germany and the United States to make coated particle fuel indicates that the scale of fuel fabrication and development efforts in the last 25 years have been quite different. German fabrication was at an industrial/production scale supporting the German AVR and THTR reactors and providing an established infrastructure for additional production of high quality fuel in support of HTR-Modul development. Only about 100 defects were measured in the German high quality fuel among 3.3 million particles produced in support of HTR-Modul development. The post-Fort St. Vrain U.S. program has been a mixture of laboratory- and larger-scale fabrication. The initial defect levels varied greatly and were much greater than those produced in Germany. Also, the U.S. program was scattered and disjointed, and multiple variables were "attacked" in each irradiation experiment, leading to a situation where it was not always possible to isolate the cause for poor results.

Comparison of the U.S. and German fabrication processes has revealed many differences. Three specific technical differences in the TRISO fuel coating layers produced by the respective fabrication processes have important impacts in terms of performance under irradiation and accident conditions: pyrocarbon anisotropy and density, IPyC/SiC interface structure, and SiC microstructure.

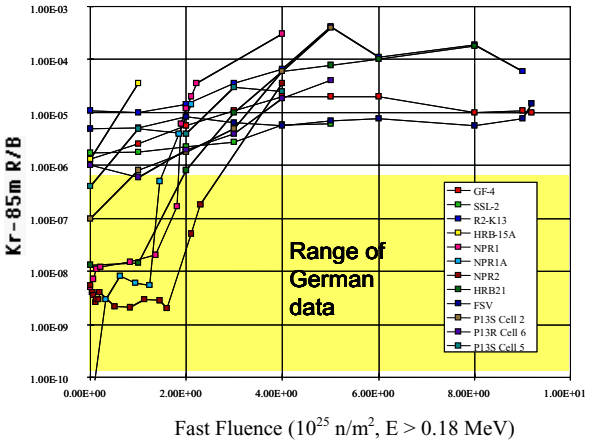

Figure 2. Krypton release to birth ratios versus fast fluence from a variety of U.S. and German fuel irradiation experiments showing the better performance of the German fuel.

II.A. Pyrocarbon Coating Rate

The density and anisotropy of the pyrocarbon layers of the TRISO fuel particle is determined by the conditions in the coater [3]. The German pyrocarbon was deposited at a higher coating gas concentration, which in turn results in a higher coating rate (~4-6 µm/minute) than generally used in the U.S. The German pyrocarbon was very isotropic and thus survived irradiation quite well. However, the German fabrication conditions appear to lead to somewhat greater surface porosity than in U.S. pyrocarbon, possibly leading to increased permeability to chlorine gas during the SiC coating process, and reaction with the kernel. U.S. pyrocarbon has been coated under a variety of conditions. In many cases, it was coated at very low coating gas concentrations, which results in a lower coating rate (2-4 µm/minute), and leads not only to a very dense and impermeable IPyC layer, which is important to preventing attack of the kernel by chlorine during deposition of the SiC layer, but also to excessive anisotropy, which can cause cracking of the pyrocarbon under irradiation.

A plot of the irradiation-induced strain as a function of coating rate is shown in Figure 3. This plot indicates that strains induced in irradiated pyrocarbon are much greater for pyrocarbon coated at very low coating rates. Post-irradiation examination of many of the U.S. capsules indicate shrinkage cracks in the inner pyrocarbon layer, which has been shown [4,5,6] to lead to stress concentrations in the SiC layer and subsequent failure of the SiC layer. Furthermore, anisotropy measurements on pyrocarbon have not adequately correlated processing parameters to pyrocarbon isotropy, and have not yet proven to be a reliable predictor of in- reactor pyrocarbon failure. More reliable methods of anisotropy characterization are needed to ensure a link between acceptable coating processing parameters and satisfactory pyrocarbon in-reactor behavior.

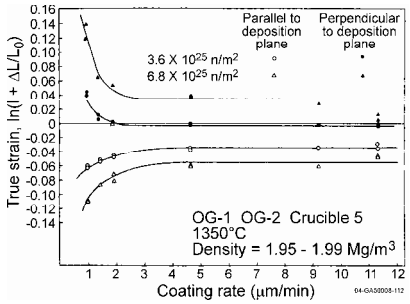

Figure 3. Irradiation-induced strains in PyC as a function of PyC coating rate.

II.B. Nature of the OPyC/ SiC Interface

Differences in the microstructure and surface porosity between the German and U.S. IPyC also led to differences in the nature of the bond that existed between the layers. Photomicrographs of the IPyC/SiC interface in German and U.S. fuel are shown in Figure 4. The figure shows that the interface in German fuel is more tightly bonded because the SiC was deposited into the IPyC, which has apparently greater surface porosity. The U.S. fuel's denser, less porous IPyC surface resulted in a smoother interface, having a lower strength bond. The TRISO coating on the German fuel never exhibited debonding under irradiation, whereas irradiation results indicated that the TRISO coating on the U.S. fuel debonded frequently. The debonding is believed to be related to the strength of the IPyC/ SiC interface and can lead to stress intensification in the SiC layer, which may cause failure.

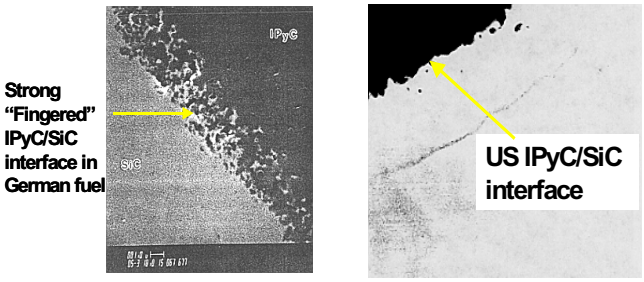

Figure 4. Comparison of SiC/IPyC interface in German (left) and U.S. fuel.

II. C. SiC Microstructure

The microstructures of German and U.S. SiC were different, as illustrated in Figure 5. The German process resulted in small equiaxed grains, whereas the U.S. process produced larger columnar (sometimes thru-wall) grained SiC. This difference in microstructure is believed to be primarily a function of the temperature used during the SiC coating phase in the coaters, with the U.S. coater producing SiC at a higher temperature in some or all regions of the coater compared to the German process. These differences are important from a performance perspective because the smaller-grained German SiC, with its higher tortuosity, should in principle retain metallic fission products better than the large thru-wall columnar U.S. SiC with more direct grain boundary pathways through the layer.

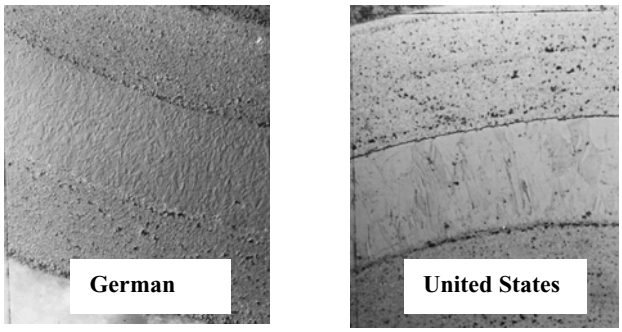

Figure 5. Comparison of the structure of German and U.S. produced SiC.

II. D. Irradiation Testing

Review of the U.S. and German irradiation programs over the last 25 years indicates that the irradiation programs were implemented differently, with vastly different results. The focus of the German program was on UO_2-TRISO fuel for the pebble bed reactors AVR and THTR and all future pebble bed reactor designs, such as the HTR Modul design. The U.S. program examined many different variants (different coatings, different kernels) with apparently few lessons learned from one irradiation to the next or feedback to the fabrication process. The most striking is that the on-line gas release measurements indicated that the German fuel exhibits about a factor of 1000 less fission gas release under irradiation than the U.S. fuel under a broad range of irradiation conditions (temperature, burnup, fluence; see Figure 6). Furthermore, the post-irradiation examination of the U. S. fuel confirmed the more extensive gas release data.

In summary, the German fuel was excellent. Of about 340,000 particles tested, there were no in-pile failures and only a few "damaged" particles from experimental anomalies. The fission gas release that did occur was attributed only to as-manufactured defects and heavy metal contamination. The U.S. fuel did not perform very well. There were relatively high numbers of failures of individual layers of the TRISO coated U. S. fuel and, in many cases, a significant fraction (~1 to 10 percent) of the total particles completely failed, (see Figure 7, note that individual layer failure fractions are plotted, not TRISO coated particle failure fractions). A variety of failure mechanisms were noted relating to effects of accelerated irradiation and attributes of the fabrication process.

This comparison strongly supports the need for process improvement studies for fuel manufactured using traditional U.S. methods and potential scoping irradiations to demonstrate the effectiveness of any changes in the process.

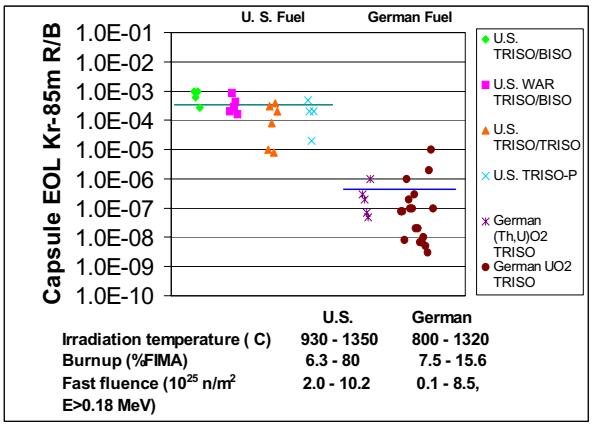

Figure 6. Comparison of end-of-life Kr-85m R/B from historic German and U.S. irradiations.

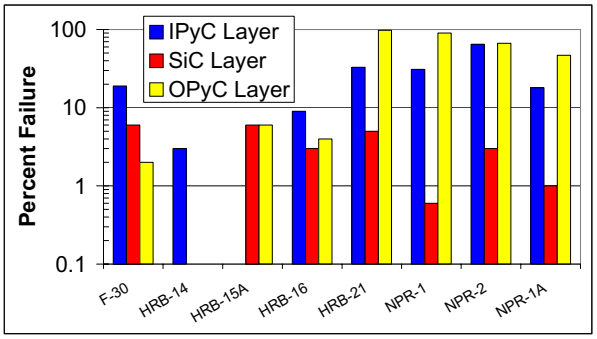

Figure 7. Individual layer failures observed during post-irradiation examination of U.S.-coated particle fuel over the past 25 years.

III. ADVANCED GAS REACTOR PROGRAM STRUCTURE

Development and qualification of TRISO-coated low-enriched uranium fuel is a key R&D activity associated with the NGNP Program. The work is being conducted in accordance with the *Technical Program Plan for the Advanced Gas Reactor Fuel Development and Qualification Program* [7]. The AGR Program includes work on improving the kernel fabrication, coating, and compacting technologies, irradiation and accident testing of fuel specimens, and fuel performance and fission product transport modeling. The primary goal of these activities is to successfully demonstrate that TRISO-coated fuel can be fabricated to withstand the high temperatures, burnup, and power density requirements of a prismatic block type NGNP with an acceptable failure fraction. It is assumed that TRISO fuel that is successful in a block reactor will also be successful in a pebble-bed

reactor since the particle packing fraction and the fuel temperatures are somewhat lower in pebble-bed reactors than in block reactors. In addition, commercialization of the fuel fabrication process, to achieve a cost-competitive fuel manufacturing capability that will reduce entry-level risks, is a secondary goal of the project.

The project is co-managed by the Idaho National Laboratory (INL) and the Oak Ridge National Laboratory (ORNL) against a resource loaded, critical path schedule with three levels of key milestones. This schedule clearly defines the activities and deliverables required and determined feasible through early schedule and cost analysis.

Implementation of the quality assurance requirements delineated in the Technical Program Plan will be in accordance with DOE quality assurance requirements specified in 10 CFR 830 "Nuclear Safety Management," Subpart A, "Quality Assurance Requirements" and in DOE Order 414.1B, "Quality Assurance." In addition, all activities that have direct input to irradiation test specimen fabrication and irradiation campaigns will be conducted in accordance with the national consensus standard ASME NQA-1-2000, "Quality Assurance Requirements for Nuclear Facility Applications," published by the American Society of Mechanical Engineers (ASME).

III. A. Fuel Manufacture

This program element addresses the work necessary to produce coated-particle fuel that meets fuel performance specifications and includes process development for kernels, coatings, and compacting; material characterization and quality control methods development; scale-up analyses; and process documentation needed for technology transfer. The effort will produce fuel and material samples for characterization, irradiation, and accident testing as necessary to meet the overall goals. There will also eventually be work to develop automated fuel fabrication technology suitable for mass production of coated-particle fuel at an acceptable cost; that work will be conducted during the later stages of the program in conjunction with cosponsoring industrial partners. Fuel manufacture development is guided by a detailed fuel product specification established based on historical U.S. and international experience.

Near-term activities focus on production of UCO kernels, coating of particles in a continuous process using a small (2-inch) laboratory-scale coater, production of fuel compacts, and characterization of the resulting materials. The goal of the kernel studies is to better define the operating window that will produce kernels meeting all specifications. For example, studies in early 2005 demonstrated carbon dispersion parameters that would result in adequate sintered kernel density. Following fabrication of the AGR-1 kernels, additional kernel development studies are needed to further define the operating envelope for both broth and sintering parameters relative to the fuel specification and other properties such as kernel strength and friability, and surface reactivity.

The goal of the initial coating studies is to produce coatings like those produced by the German program in the late 1980s. All three layers were coated in a continuous manner in the German process, whereas in the U.S. process, the fuel particles were unloaded after each coating layer to perform quality measurements. Additional coating variants are planned that will confirm understanding of the historical coating fabrication database and enhance the prospects for one or more successful outcomes, and the baseline and selected variants will then be irradiated in the first irradiation test, AGR-1. Recommended coating rates and temperatures for the coating variant candidates planned for the AGR-1 fuel fabrication campaign are listed in Table 1 (these conditions may be adjusted based on understanding gained from early fuel production and characterization).

Table 1. Candidate coating variants for AGR-1.

Variant	IPyC Conditions	SiC Conditions	Comment
1	1300 ºC; 4.5 µm/min	1510 ºC; 0.2-0.25; µm/min	German baseline
2	1300 ºC; 4.5 µm/min	1580 ºC; 0.2-0.25 µm/min	Higher SiC deposition temperature
3	1300 ºC; 3.0 µm/min	1510 ºC; 0.2-0.25 µm/min	Low IPyC coating rate (anisotropic)
4	1300 ºC; 3.0 µm/min	1580 ºC; 0.2-0.25 µm/min	Low IPyC coating rate (anisotropic)
5	1300 ºC; 6 µm/min	1510 ºC; 0.2-0.25 µm/min	High IPyC coating rate
6	1300 ºC; 6 µm/min	1580 ºC; 0.2-0.25 µm/min	Higher SiC deposition temperature
7	1300 ºC; 4.5 µm/min	1510 ºC; 0.2-0.25 µm/min	Interrupted variant of Case 1
8	1300 ºC; 4.5 µm/min	~ 1300 ºC with Argon	

Coating conditions are planned that span the range from producing highly anisotropic/high density PyC to highly isotropic/low density PyC. Two different SiC coating temperatures are planned to determine an acceptable window for producing the desired fine-grained SiC. An interrupted run is also planned to more quantitatively characterize fuel produced in both interrupted and uninterrupted modes. In addition, a variant in which argon gas is used during SiC coating is planned, since the UK Dragon project and current microelectronics production has demonstrated that good SiC can be produced at much lower temperatures when this gas is used.

The second phase of coating development involves scaleup of the continuous coating process to production size (e.g., 6-inch) coaters. The goal is to produce high quality coatings for performance demonstration and, ultimately, qualification.

The laboratory scale coating development work includes the development of an extensive coating process model to support small coater process development and the transition from laboratory scale coaters to production scale coaters. A major challenge is to account for the effects of the turbulent gas-solids interactions in the fluidized bed reactor on the rate controlling processes and the final product quality of the chemical vapor deposition. The modeling team will make use of the latest computational fluid dynamics computer codes and correlations available for simulating the hydrodynamics, heat and mass transfer, and chemical reaction kinetics on the particle surfaces. In addition, experimental validation will be needed at each stage of development to ensure that the model predictions are consistent with the actual physics and chemistry. The latter is critical to the implementation of successful scale-up from the laboratory to production prototype.

Coated particles will then be over-coated and molded into cylindrical compacts using a matrix of graphite flour and carbonized resin. The thermosetting resin based matrix and warm pressing compacting process selected for the program is similar to processes used in Germany and Japan, and a substantial departure from the thermoplastic matrix injection process used previously in the U.S. Development work is required to adapt the process to the U.S. fuel compact specifications. Although the matrix is similar to the German matrix, the ratio of matrix to particles is quite different, approximately 72:28 versus 90:10 for the Germans. Also, the German pebbles were isostatically pressed into spheres while the AGR compacts will be compression molded (via the warm pressing step) into cylindrical compacts. Being that the fuel particles are non-compressible, this reduction in amount of matrix and change in molding technique requires a consistent particle overcoat thickness and careful pressing of the particles into compacts. A primary objective of the compacting development is to limit particle damage to the very low levels required by the fuel product specifications, with allowance for a low level of defects in the coated particles used to form the compacts.

Parameters needed to establish a uniform overcoat have been optimized using surrogates, and compacts have been warm pressed and carbonized (see Figure 8). Future plans include optimizing the final heat treatment and compacting uranium bearing coated particles.

Figure 8. Compacts produced using ORNL thermosetting resin process.

In parallel with the fuel fabrication, additional effort is being expended in the area of fuel characterization, with the goal of providing feedback to fabrication process development, demonstrating compliance with product specifications, and establishing more advanced and more robust techniques to measure key attributes of the fuel that can be integrated into a continuous production-scale coating process. Initial activities focus on reestablishing conventional characterization procedures and developing improved anisotropy and optical image measurement and analysis techniques. Advanced tomography techniques to measure layer thickness and densities are also planned and ORNL is acquiring a high resolution (1-2µm) x-ray inspection system to support this effort.

Computer controlled sample positioning and digital imaging plus ORNL-developed image analysis software is used to quickly and easily analyze 1000's of particles for size and shape with a 2-µm resolution. The system is also capable of quickly and easily analyzing 100's of particle cross-sections with 1 µm resolution and providing copious data from which particle dimensions, layer

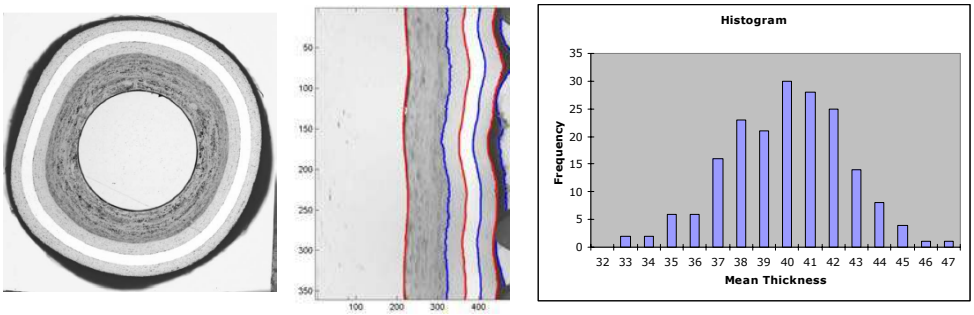

Figure 9. Example information from the ORNL computer automated optical characterization system. An IPyC histogram is shown on the right.

thickness and particle shape can be obtained. Figure 9 displays results from the computer automated optical characterization equipment developed at ORNL.

ORNL has also developed an advanced optical method to measure pyrocarbon anisotropy. The degree and direction of pyrocarbon crystallite orientation is measured by a scanning ellipsometry technique called the 2-MGEM (2-modulator generalized ellipsometry microscope). Figure 10 shows typical results from that equipment. Recent data indicates a 2 μm spot size has been achieved providing new information on the variation in pyrocarbon properties within a layer for both archived U.S. and German fuel as well as material produced by the Program.

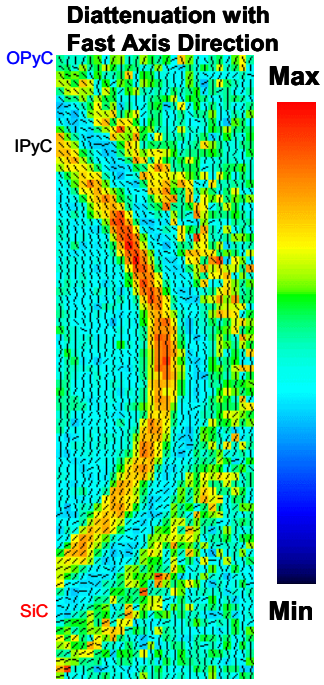

Figure 10. Typical results from the ORNL equipment for measuring pyrocarbon anisotropy.

III. B. Fuels and Materials Irradiation

The fuel and materials irradiation activities will produce data on fuel performance under irradiation as necessary to support fuel process development, to qualify fuel for normal operating conditions, and to support development and validation of fuel performance and fission product transport models and codes. The irradiations will also produce irradiated fuel and materials as necessary for post-irradiation examination and ex-core high-temperature furnace safety testing.

A total of eight irradiation capsules will be used to obtain the necessary data and sample materials. Each capsule will be irradiated for approximately two years. Details on each irradiation are listed in Table 2. The purpose of AGR-1 is to shakedown the new multi-cell capsule design, fabrication, and operation to reduce the chances of capsule failures in subsequent irradiation tests. If successfully taken to a substantial fraction of design burnup and fast fluence, the test will yield key irradiation performance data from a number of early fuel variants produced under different processing conditions from laboratory-scale coating equipment, as discussed above. AGR-2 will be a performance demonstration irradiation with fuel fabricated from a production-scale coater. Feedback to the fabrication process is expected following both AGR-1 and AGR-2. AGR-3 is devoted to obtaining data on fission gases and fission metals under normal irradiation conditions. AGR-4 will study fission product behavior in fuel compact matrix and graphite materials.

Table 2. Planned AGR irradiation capsules.

Capsule	Task	Proposed Start Date
AGR-1	Shakedown and early fuel	9/2006
AGR-2	Performance test fuel	10/2007
AGR-3	Fission product transport -1	4/2008
AGR-4	Fission product transport -2	7/2008
AGR-5	Fuel qualification -1	9/2010
AGR-6	Fuel qualification -2	9/2010
AGR-7	Fuel performance model validation	12/2010
AGR-8	Fission product transport -3	12/2010

Given the statistical nature of coated particle fuel, a large number of fuel specimens are needed to fully qualify the fuel and demonstrate compliance with the fuel failure specification. AGR-5 and AGR-6 are identical irradiations that will be used to qualify the fuel for the NGNP. AGR-7 and AGR-8 are irradiations designed to provide data with which to verify and validate fuel performance and fission product transport models.

A schematic of the test train to be used for AGR-1 is shown in Figure 11. Each AGR capsule will be a highly instrumented multi-cell capsule capable of irradiating six different fuel forms with different thermal conditions, if required. Flux wires will be used to measure the thermal and fast neutron fluences. Thermocouples in graphite bodies surrounding the fuel will be used to monitor temperatures during the irradiation. The graphite bodies may contain boron carbide to control power generation during the irradiation and prevent large power swings historically experienced when irradiating fuel to high burnup. A low flow of inert sweep gas is used during irradiation to provide the correct thermal conductance to allow the fuel to be irradiated at the proper temperature. Usually, most of the sweep/thermal control gas is helium. Small amounts of neon are used to change the overall conductance to compensate for depletion of uranium due to burnup and still keep the fuel at the required temperature.

Planned AGR-1 irradiation conditions are a peak burnup of 18 to 20% FIMA, a volume average time average temperature of 1150 °C, a time average peak temperature of 1250 °C, and a fast neutron fluence of 5×10^{25} n/m^2 (E>0.18 MeV). The capsules will be irradiated in one of the large B positions at the Advanced Test Reactor at the Idaho National Laboratory. The large B position has a neutron spectrum very similar to that expected in a gas reactor. Preliminary calculations suggest that each capsule will be irradiated for 2.5 years to meet the requirements stated above, which will simulate a three to four year irradiation in the NGNP.

An important objective of the irradiation is to measure the fission gas release from the fuel and correlate it to the operating parameters in the irradiation. The sweep gas from each cell containing fuel specimens will be "sniffed" for fission gas. The sweep gas also transports any fission gases released from the fuel to a location outside of the reactor, where an NaI detector with enough sensitivity to indicate a single fuel particle failure (evident by a spike in its signal) will measure gross radiation in the line. The isotopic content of the gas in the line will be monitored on line using the state-of-the-art fission product monitoring system similar to that shown in Figure 12. This system consists of a gamma spectrometer for continuous measurement of the concentration of the various fission gas isotopes in the sweep gas. With this instrumentation, particle failures can be monitored and correlated to conditions in the cell. The isotope concentration data will be used to calculate the R/B ratio for various fission products, a key measure of fission product retention and fuel performance.

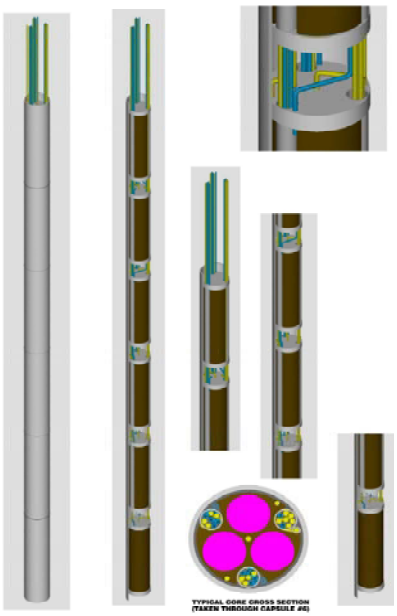

Figure 11. Schematic of AGR-1 multicell capsule.

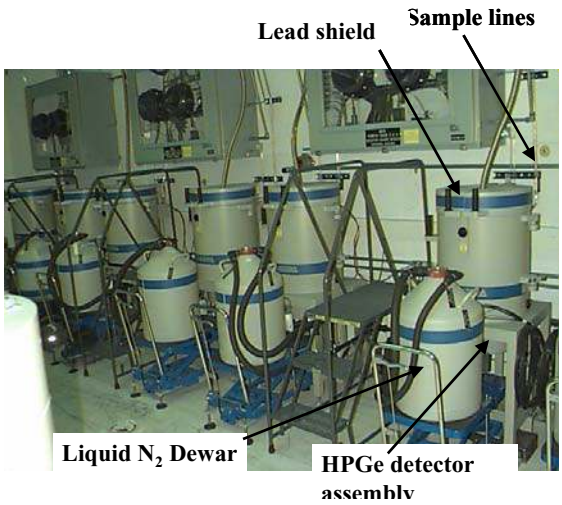

Figure 12. INL fission product monitoring system.

III. C. Post-irradiation Examination and Safety Testing

Data from the post-irradiation examination and safety testing will supplement the in-reactor measurements (primarily fission gas release-to-birth ratio measurements) as necessary to demonstrate compliance with the fuel performance requirements and support development and validation of the computational fuel performance models. This work will also support the fuel manufacture with feedback on the performance of kernels, coatings, and compacts.

III. C.1. Post-irradiation Examination

Post-irradiation examination is a collection of nondestructive and destructive techniques that can be used to characterize the state of the fuel either after irradiation or after safety testing. The different types of analyses or measurements that will likely be performed, the purpose of the measurements, and their value to the overall fuel qualification plan are discussed in the next few paragraphs.

Following removal of the irradiation test train from the reactor to the hot cell, a gamma scan of the entire test train will be performed. A collimated gamma spectrometer in the hot cell will traverse the capsule and record the gamma activity as a function of axial length. Such a measurement is generally qualitative and will provide information to determine whether any fuel compacts have broken or if a significant number of fission products have been released and moved within the capsule.

Following capsule disassembly and removal of the fuel element, the general condition of the fuel will be noted, specimens will be weighed, and dimensional measurements of the specimens will be performed to characterize the shrinkage or swelling that occurred during irradiation.

To examine the physical characteristics of irradiated fuel particle coatings, optical metallography will be performed on cross sections of the fuel pebble or fuel compact. These high magnification examinations offer excellent visual evidence of the condition of the fuel following testing. This technique will be used to investigate layer integrity, possible layer debonding, densification of layers (e.g., buffer) the degree of void formation due to fission gas, the extent of kernel migration and swelling, and the nature and extent of the fission product attack on the SiC. Use of bright field and polarized light and etching are useful techniques to reveal the microstructure of the SiC layer. With proper etching techniques, SiC grain orientation and sizes can be determined. Figure 13 is a photograph of optical metallography performed on German fuel following irradiation in the AVR. Development of a nondestructive tomographic x-ray inspection technique is also under consideration.

Figure 13. Photomicrograph of German AVR fuel after irradiation.

Gamma-scanning of capsule components (e.g., graphite bodies) or leaching and gamma counting of capsule components will be used to determine the identity, migration, and distribution of fission products following irradiation.

To identify where the fission products are located within irradiated fuel particles, the fuel element will be deconsolidated to obtain individual particles for examination by electron microscopy to reduce the radiation background. The radiation background is the issue here, not damage to particles or the release of fission products. The reduced background radiation from a single fuel particle is usually required for good measurements by electron microprobe, where one is looking for x-rays characteristic of specific fission products (measured by energy dispersive or wave length diffraction techniques). This technique looks for evidence of fission product accumulation at the IPyC/SiC interface, fission product attack of the SiC, and fission products outside the fuel particles.

For irradiations of fuel compacts or pebbles, there will be a need to measure fuel particle failure fraction independently of the on-line R/B measurements, due to the uncertainty in the R/B measurement for a few particle failures and the inability to measure metallic releases.

The most useful technique for fuel particle failure measurements, when the on-line R/B measurements suggest a failure fraction well under 1%, is leach-burn-leach. In this technique, the fuel compact or pebble is leached with acid to remove any fission metals (e.g., cesium) released from defective fuel particles and heavy metal contamination. The fuel element is then burned in air to remove all carbon matrix material, the OPyC layers, and also the IPyC/Buffer layers of any particles with failed SiC. Particles that remain are then leached with an acid solution to remove any exposed uranium that had been enclosed by an intact pyrocarbon layer. The measurement of the free uranium is then converted to a SiC defect fraction.

Another technique performed on coated particle fuel is the irradiated microsphere gamma analyzer (IMGA) developed at ORNL. With this technique, fuel particles following deconsolidation are analyzed individually by a gamma spectrometer and catalogued based on the ratio of mobile and immobile fission products measured in the particle. A histogram of such ratios is developed based on all the particles in a sphere or compact and compared to a normal distribution. Variations from normal can easily be seen with such a technique. Metallography following IMGA on the particles that depart from normal can be valuable to tie the microstructure of the anomalous particles to the fission product release. For high-quality fuel with low gas release, this technique may not be required, but for intermediate failure fractions of 10^{-4} to 10^{-2}, deconsolidation followed by IMGA is useful.

Traditional burnup analysis is also performed as part of the series of post-irradiation examinations. Following deconsolidation, a few particles can be sent for destructive radiochemical assay to determine the concentration of transuranics and minor actinides, from which burnup can be assessed.

III. C.2. Safety Testing

An important goal of this program is to determine the performance of the fuel under high-temperature accident conditions, since integrity of the coated particle to high temperature is a crucial part of the safety case for the NGNP. In particular, three environments are of interest: helium, air, and steam. The irradiated TRISO fuel will be exposed to these environments for up to 500 hours. The exact composition of these environments have not yet been defined, but assumptions are that the test will be run at atmospheric pressure, and steam and air concentrations will be in the range of 10,000 ppm. Some of the early German data of this type is plotted in Figure 14, which shows krypton fractional releases as a function of heating time at 1600 °C and burnup. Note that the lower burnup fuel (8-10% FIMA) had little release, but the higher burnup fuel, typical of the burnup expected in NGNP, had much higher releases. Although this data is not directly applicable to the NGNP because of differences in fabrication and particle size, it is illustrative of the need to test fuel at a variety of burnup levels.

The maximum temperature, including a 100 °C uncertainty, predicted for a core conduction cooldown accident in small modular gas cooled reactors is 1600 °C and is reached within ~50 to 100 hours after initiation of the event. Temperatures remain at ~1600 °C for about 25 to 50 hr, followed by a long, slow (hundreds of hours) cooldown. Traditionally, post-irradiation isothermal annealing at temperatures of 1600, 1700, and 1800 °C have been performed for several hundred hours, with continuous collection of released fission products.

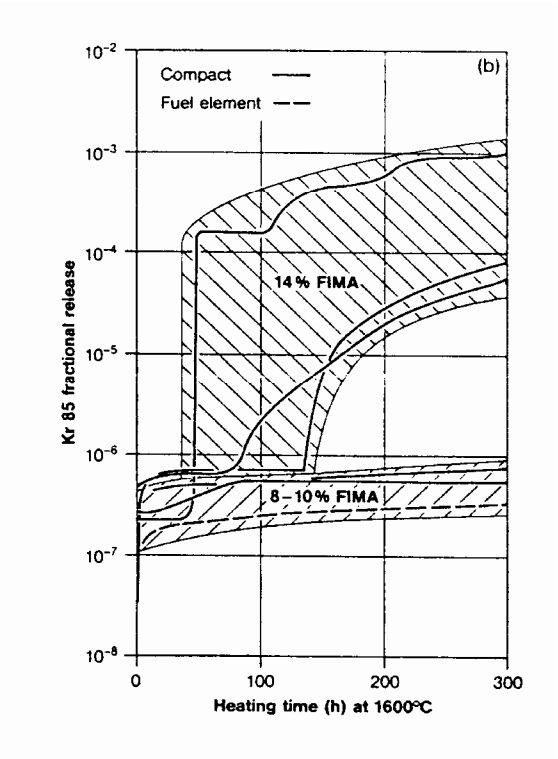

Figure 14. Krypton 85 fraction release data versus heating time at 1600° C and fuel burnup from German heating tests.

Isothermal tests are generally considered to be conservative relative to heatup transient tests, which follow more closely the time-temperature profiles calculated to occur in a core conduction cooldown transient, because more time is spent at the highest temperatures. Thermal gradients are not expected to be significant. Isothermal tests are also easier to analyze

than transient tests and, given the long thermal time constant associated with the transients, there is little new information to be gained by conducting transient tests. The experimental facility will consist of a furnace to maintain a fuel specimen at specified temperatures with a cold finger to trap the condensable fission products and a cold trap for trapping fission gases. The cold finger and cold traps are analyzed using traditional gamma spectroscopy. The data needed from safety testing are fission product release, TRISO coating layer integrity, and fission product distribution within fuel particles (corrosion likelihood) and fuel compacts.

The release behavior of the fission products is somewhat different than in other nuclear fuels. Silver (Ag-110m) is released first because of its greater mobility through the SiC coating on TRISO particle fuel. This is followed by Cs-134 and Cs-137, which can diffuse through the PyC and SiC layers after long times at these temperatures. Lastly, the fission gases Kr-85 are released.

Postheating test activities include characterization of the TRISO coating layer integrity by optical metallography including looking for evidence of SiC layer thinning and decomposition, chemical attack of the SiC, and the mechanical condition and microstructures of the SiC and PyC layers. Other procedures discussed earlier for irradiated fuel may also be applied. Detailed test matrices will be developed as the program evolves. Nondestructive x-ray tomography (if developed) will also be applicable.

III. D. Fuel Performance Modeling

The high temperature gas reactor TRISO coated fuel performance computer codes and models will be further developed and validated as necessary to support the fuel fabrication process development and the NGNP design and licensing activities. The fuel performance modeling will address the structural, thermal, and chemical processes that can lead to coated-particle failures. The models will address the release of fission products from the fuel particle and the effects of fission product chemical interactions with the coatings, which can lead to degradation of the coated-particle properties.

Compared to light water reactor and liquid metal reactor fuel forms, the behavior of coated-particle fuel is inherently more multidimensional. Moreover, modeling of fuel behavior is made more difficult because of statistical variations in fuel physical dimensions and component properties, from particle to particle and around the circumference of any given particle due to the nature of the chemical vapor deposition fabrication process. Previous attempts to model this fuel form have attacked different aspects of the problem. Simple one-dimensional models exist to describe the structural response of the fuel particle. Models or correlations exist to describe the fission product behavior in the fuel, though the database may not be complete owing to the changes in fuel design that have occurred over the last 25 years. Significant effort has gone into modeling the statistical nature of fuel particles. However, under pressure to perform over one million simulations with the computing power available in the 1970s and 1980s, the structural response of the particle was simplified to improve speed of calculation.

New models are currently being developed in the United States that represent a first-principles-based mechanistic, integrated, thermal-mechanical-physio-chemical-irradiation performance model for particle fuel, which has the proper dimensionality yet captures the statistical nature and loading of the fuel. The mechanistic model for coated-particle fuel considers both structural and physio-chemical behavior of a particle-coated fuel system during irradiation. The INL model, called PARFUME, includes the following important phenomena:

- Anisotropic response of the pyrolytic carbon layers to irradiation (shrinkage, swelling, and creep that are functions of temperature, fluence, and orientation/direction in the carbon).

- Failure of a SiC ceramic in the coating system (using the classic Weibull formulation for a brittle material), either by traditional pressure vessel failure or by mechanisms such as particle asphericity (see Figure 15), or pyrocarbon layer cracking (see Figure 16), or debonding and subsequent stress concentrations in the SiC layer.

- Chemical changes of the fuel kernel during irradiation (changes in carbon/oxygen, carbon/metal and/or oxygen/metal ratios, depending on the kernel fuel type, and production of CO/CO_2 gas) and its influence on fission product and/or kernel attack on the particle coatings.

- Thermo-mechanical response of the kernel and buffer as a result of buffer densification, kernel swelling, and gas generation (fission gases and CO), including development of gaps between the buffer and the TRISO-coating layers as a function of burnup, fast fluence, and temperature.

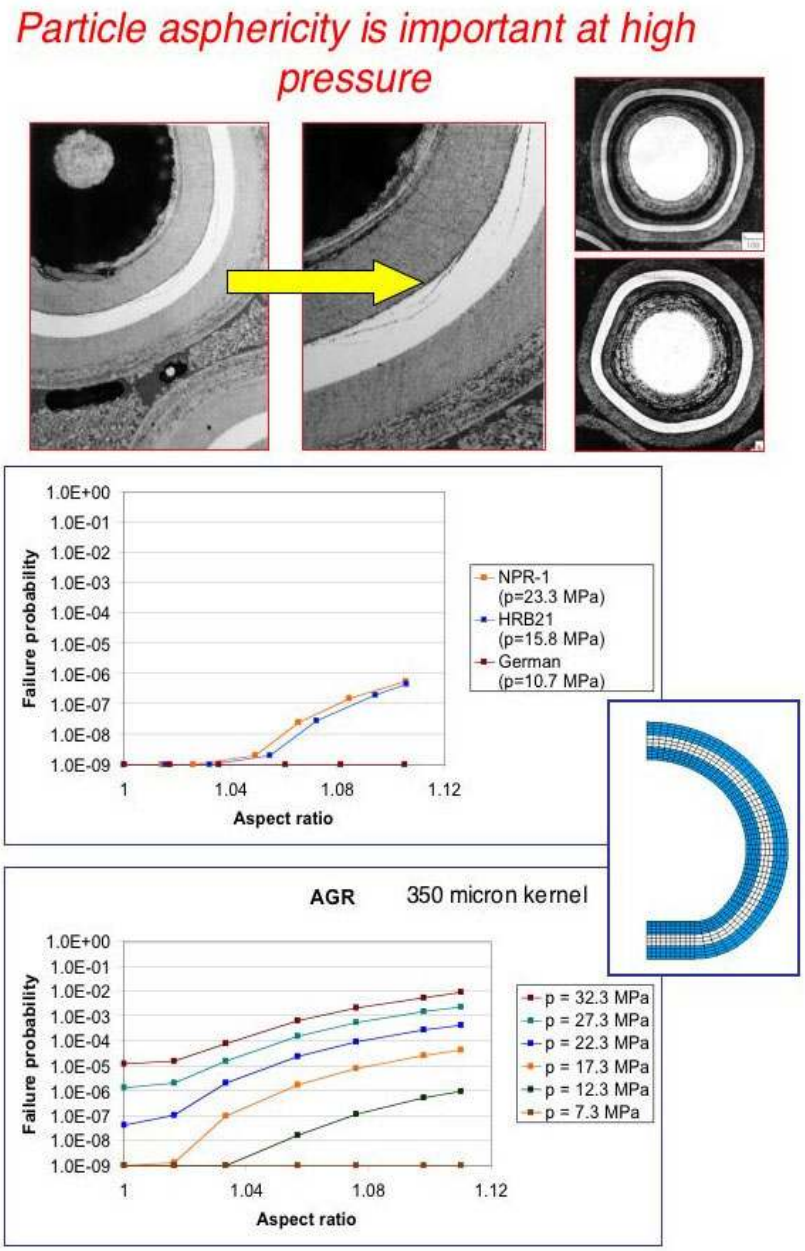

Figure 15. Effect of particle asphericity on failure probability.

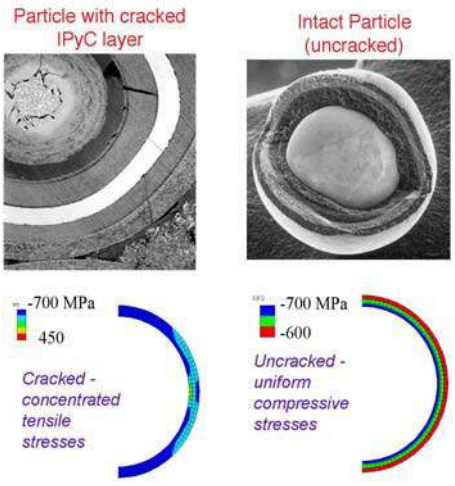

Figure 16. Cracked inner pyrolytic carbon layers could lead to SiC layer failure.

- Attack of the SiC layer by Pd and other fission products, and by kernel migration.

- Transport of key fission products (Kr, Ag, Sr, Cs) from the kernel and through each layer of the particle.

Statistical variations of key properties of the particle associated with the production process, requiring Monte Carlo analysis of a very large number of particles to understand the aggregate behavior. Fabricated particles will exhibit statistical distributions for not only the physical dimensions of the individual coatings but also for the mechanical properties.

These models have had some success in predicting fuel failure mechanisms and rates in the U.S. fuel tested over the last decade, thereby facilitating a better understanding of TRISO coated fuel behavior. Such a tool can be very useful for both pretest and posttest predictions for any experiment performed in this program.

In addition, sensitivity studies with the model can be used to identify critical materials properties data and constitutive relations whose uncertainty needs to be reduced because they drive the predicted performance of the coated fuel particle. Furthermore, use of piggyback cells (small encapsulated fuel samples outside the compacts) in the irradiation capsules can be used to study those key individual phenomena in coated particles that have high uncertainty (e.g., shrinkage and swelling of pyrocarbon, fission product release behavior in a purposely defective or initially failed particle).

Moreover, some of the post-irradiation examination techniques can provide maps of fission products through the particle, which can be compared with model predictions of fission product transport through the coatings. All of this type of data will eventually be needed to validate the overall TRISO coated fuel performance model. Such fuel performance models will eventually be needed to provide some understanding of fuel behavior inside the operations and safety envelope defined by the irradiation and safety testing (i.e., interpolation) and outside these envelopes where the margins of failure of the fuel may be approached (i.e., extrapolation). Finally, a validated fuel performance model can be used to help evaluate and guide potential future changes in the next-generation coated particle fuel.

The importance of fuel performance modeling has been recognized internationally. The United States is part of the IAEA Coordinated Research Project on coated particle fuel technology. A key task is associated with benchmarking coated particle fuel performance models under both normal and off-normal conditions. The fuel behavior models under development by the AGR program are part of the international benchmark.

III.E. Fission Product Transport and Source Term Modeling

Transport of fission products produced within the coated particles will be modeled to obtain a technical basis for source terms for advanced gas reactors under normal and accidental conditions. The design methods (computer models) will be validated by experimental data as necessary to support plant design and licensing.

The NRC will require validated computer models that accurately predict the following phenomena:

- Fission product release from the kernel

- Transport through failed coatings

- Deposition fraction of the released fission products in the compact or sphere matrix

- Deposition fraction of what gets through the compact on fuel element graphite (prismatic variant only)

- Deposition fraction of what gets out of the fuel element onto graphite dust and metallic surfaces in the primary circuit.

- Re-entrainment of deposited fission products during an elevated temperature accident, or depressurization event

- Transport of fission products on dust particles, and subsequent release to the environment if the primary circuit is breached.

Each of the phenomena listed above is complex and difficult to model. It is also difficult to design and conduct experiments that can cover the multitude of variables that affect the physical situation. The AGR program has developed a research and development plan that, when the work is successfully completed, will produce a technical basis for source terms under normal and accident conditions for advanced gas-cooled reactors. The program consists of irradiations to provide data on fission gas and fission metal release from the kernel and transport through failed coatings (AGR-3), fission product transport behavior in the fuel element matrix and graphite block

(AGR-4), out of pile experiments to characterize plateout, and reentrainment of fission products during accident conditions. The program also contains an irradiation (AGR-8) that will be used to validate computer models that describe the in-vessel gas reactor source term.

IV. Summary and Conclusions

The DOE NGNP AGR Fuel Development and Qualification Program consists of five elements:

- Fuel manufacture,
- Fuel and materials irradiations,
- Safety testing and post-irradiation examinations,
- Fuel performance modeling, and
- Fission product transport and source term modeling.

The goal is to qualify the fuel form for use in the NGNP to the following:

- Burnup of 15–20% FIMA,
- Volume average time average temperature of 1150 °C,
- Time average peak temperature of 1250°C, and
- Fast neutron fluence of 5×10^{25} n/m^2 (E>0.18 MeV),
- High fission product retentiveness for hundreds of hours at 1600 °C.

The fuel form is based on reference UCO, SiC TRISO particles bonded by a matrix of graphite flour and carbonized thermosetting resin, incorporating past German fabrication experience.

An underlying theme for the fuel development work is the need to develop a more complete fundamental understanding of the relationship between the fuel fabrication process, key fuel properties, the irradiation performance of the fuel, and the release and transport of fission products in the NGNP primary coolant system during both normal operation and any conceivable accident. The logic of the program is structured such that there are multiple feedback loops and opportunities for improvement in the fabrication process based on early results. The fuel performance modeling and analysis of the fission product behavior in the primary circuit are important aspects of this work. The performance models are considered essential for several reasons, including guidance for the plant designer in establishing the core design and operating limits, and demonstration to the licensing authority that the applicant has a thorough understanding of the in-service behavior of the fuel system. The fission product behavior task will also provide primary source term data needed for licensing.

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