Comparative Study of Laboratory-Scale and Prototypic Production-Scale Fuel Fabrication Processes and Product Characteristics

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Abstract – An objective of the High Temperature Gas Reactor fuel development and qualification program for the United States Department of Energy has been to qualify fuel fabricated in prototypic production-scale equipment. The quality and characteristics of the tristructural isotropic (TRISO) coatings on fuel kernels are influenced by the equipment scale and processing parameters. The standard deviations of some TRISO layer characteristics were diminished while others have become more significant in the larger processing equipment. The impact on statistical variability of the processes and the products, as equipment was scaled, are discussed. The prototypic production-scale processes produce test fuels meeting all fuel quality specifications.

I. INTRODUCTION

The United States Department of Energy (US-DOE) has chartered the Very High Temperature Reactor Technology Development Office (VHTR-TDO) at Idaho National Laboratory (INL) in Idaho, USA to manage the Advanced Gas Reactor (AGR) fuel development and qualification program.

Two of the primary objectives of this program are to establish the capability to fabricate high-quality, tristructural isotropic (TRISO) coated particle fuel in the commercial sector, using prototypic production-scale equipment, and to qualify that fuel for use in a High Temperature Gas Reactor (HTGR) [1][2]. These have been met.

Early fuel fabrication process development activities were performed in laboratory-scale equipment and processes, primarily at Oak Ridge National Laboratory (ORNL) in Tennessee, USA.

Technology and methods from laboratory-scale fuel fabrication processes employed at ORNL have been scaled up to prototypic production-scale processes deployed in facilities owned by Babcock & Wilcox Nuclear Operations Group – Lynchburg (B&W NOG-L), in Virginia, USA. Laboratory-scale technologies were critically reviewed during the transfer of process technology and methods to B&W NOG-L to determine if alternative processes and equipment might offer significant advantages in productivity, footprint, and safety of operation. The successful design of the prototypic production-scale processes is attributed to the engineering efforts of B&W NOG-L.

Changes to TRISO particle fabrication equipment and processes are discussed along with the impact of the changes on product variability for TRISO particle coating.

The AGR fuel development and qualification program is subdivided into multiple irradiation experiments [2]. Three fuel fabrication campaigns supporting the irradiation experiments illustrate the progression from laboratory-scale to prototypic production-scale equipment; namely, shakedown fuel (AGR-1), performance fuel (AGR-2), and qualification fuel (AGR-5/6). The AGR-1 fuel fabrication campaign used laboratory-scale equipment for all processes other than kernel fabrication, which was performed in prototypic equipment from program inception. The AGR-2 fuel fabrication campaign used a prototypic coater to deposit the TRISO layers on the kernels, but overcoating and compacting remained on a laboratory scale. The fuel fabrication campaign for AGR-5/6 is being performed solely in prototypic production-scale equipment.
II. TRISO PARTICLE COATING

II.A. Equipment Description

Laboratory-scale TRISO particle coatings were applied at ORNL in a one-piece, 5.0-cm diameter, conical-cylindrical retort with a single gas inlet, which was screwed onto a water-cooled gas line [3]. Accretions of carbon and silicon carbide (SiC) formed volcano-shaped protrusions surrounding the gas inlet, allowing for some particle stagnation between the nozzle accretion and the retort wall and necessitating the use of a new retort for each coating run. A typical charge mass of approximately 64 grams of 350 μm uranium carbide/uranium oxide (UCO) kernels was used for the coating runs. All four TRISO coating layers were normally applied without unloading the particle bed. Buffer and IPyC layer density data could only be obtained by interrupting a coating run after the respective layer had been applied. Therefore, the actual buffer and IPyC densities of the fuel used in the first fuel irradiation experiment (AGR-1) were inferred by process control and previously measured values from interrupted runs.

TRISO coatings are applied in a 15.2-cm diameter retort at B&W NOG-L, using an INL design with a replaceable, multi-port gas distribution nozzle. The nozzle is a dome that protrudes 1 cm above the retort base with seven gas ports; one central and six satellite ports (Fig. 1). The nozzle and retort converging section form an annular gas inlet at the bottom of the retort through which inert diluent gas is passed to fluidize the particles at the bottom of the retort, cool the gas distributor nozzle, and dilute the reactive gases. A typical kernel charge mass for the prototypic coater is 1.5 kg – 2.0 kg. All four TRISO coatings are sequentially applied without discharging the bed. The furnace is equipped with two dip samplers used to collect particles from the hot bed for buffer and IPyC density analyses, enabling these data to be obtained on every TRISO coated particle batch.

Fig. 1: Rendering of the gas distributor nozzle design.

The AGR-2 UCO and the AGR-5/6 qualification runs used control thermocouples in different locations. AGR-2 UCO runs used a Type C thermocouple mounted externally about mid-way up the coater retort to control process temperatures whereas the AGR-5/6 qualification runs used a Type C thermocouple mounted externally, about 20 cm lower, near the elevation of the particle bed after inner coatings are deposited.

II.B. TRISO Particle Upgrading Methods

With the small quantities of TRISO produced during a coating run in a 5.0-cm-diameter coater (~190 g), several TRISO particle upgrading technologies are viable. ORNL sorted by shape on an inclined, vibrating sorting table and sorted by size using a roller micrometer. Efficient particle sorting with the inclined sorting table and the roller micrometer necessitates slow particle feed rates onto these devices. Use of the sorting table and roller micrometer were abandoned for the larger TRISO particle batches from the 15.2-cm-diameter coater (≤ 4.5 kg) at B&W NOG-L. The sorting table operation was inefficient and resulted in a substantial loss of product with little measureable improvement in product aspect ratios. The roller micrometer was efficient in sorting the particles, but the length of time to process a batch was prohibitively long for an industrial process. B&W NOG-L has found standard sieving technology to be sufficient for rejecting undersized particles and oversized debris that may break off from nozzle accretions [4].

II.C. TRISO Coating Qualification Runs

A series of five consecutive coating runs were performed to qualify the coating process parameters for the upcoming AGR-5/6 fuel fabrication. No intentional process changes were made to coating parameters, except in response to changes in kernel properties (diameter and density) that altered the total bed surface area of the kernel charge. The required quantity of precursor gases and the rate that they are fed to the coater are dependent on the surface area of the underlying substrate and the desired layer properties. Coating run 93158 had kernels with a slightly different bed surface area than the previous four runs, which necessitated some minor adjustments to the gas flow rates. Objectives for the qualification runs were to quantify product variability, demonstrate process controllability, and to reproducibly meet fuel specifications. Average process temperatures and gas flow rates are given in Table 1 for the five qualification runs along with associated standard deviations (rounded to the indicated precision).
Table 1. Time-averaged parameters and standard deviations for the AGR-5/6 TRISO coating process qualification runs.

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* Some parameters adjusted for a change in kernel properties.

Process data were recorded at 15-second intervals during layer depositions and layer property data were collected on product samples. These data show a remarkably well controlled process. Dosing of methyltrichlorosilane (MTS) for the silicon carbide layer is achieved in a system that atomizes and fully vaporizes MTS into the hydrogen carrier gas stream.

II.D. TRISO Layer Thickness Variability

The particle bed in a coating furnace primarily operates in a pulsatory spouted bed regime [5] due to the kernel/particle properties consistent with a Geldart Group B powder and fluidization outside of stable spouting conditions [6][7].

The precursor and fluidizing gases enter the bed from the gas inlets and partition between a stream of gases forming a chain of rising bubbles in the center (i.e., spout) and interstitial gases percolating up through the subsiding annulus of loosely packed particles. The gases heat up rapidly upon entering the bed, and the precursor gases decompose into condensable species that deposit on the particles. The rate of deposition and the density of the localized deposits on the particles likely differ between the coating regions in the spout and the annulus. This is evident from the banding that can be seen in layers of sectioned particles. The more frequently a particle passes through regions dense in coating gases, the more rapidly the layer thickness grows. It is intuitive that the number of passes a particle will make in a small coater with a limited charge mass can be significantly more than a particle in a larger coater and a larger bed mass.

It is reasonable to expect the probability distribution for the number of passes that particles make through coating zones would be broader (i.e., more disperse) for a large coater than for a small coater. This is manifested as a larger standard deviation in the coating layer thicknesses within a coating batch. This phenomenon is seen in Fig. 2. AGR-1 TRISO fuel was coated in the 5.0-cm coater with a kernel charge mass of 64 g. All subsequent TRISO fabrication data were in the 15.2-cm coater. AGR-2 UCO TRISO fuel was coated with a kernel charge mass of 1.3 kg, AGR-2 UO TRISO with a kernel charge mass of 1.7 kg, and the AGR-5/6 qualification runs had a kernel charge mass of 2.0 kg.

![Fig. 2: Mean population layer thickness standard deviation.](image-url)
The lowest average standard deviation for layer thicknesses was attained in the laboratory-scale coater. The AGR-2 and AGR-5/6 runs used kernels with nominally 425 µm diameters and densities near 11.0 g/cm³, but different charge masses. The influence of charge mass is best seen by comparing these two data sets. The average coated particle batch standard deviations increased as the charge mass increased.

However, run-to-run variability in the mean layer thicknesses is less sensitive to the coater diameter and more sensitive to the methods used to calculate and control the addition of coating layer precursor chemicals into the bed. Despite the larger batch standard deviations for layer thicknesses, the variability from run-to-run in the mean layer thickness is much improved with time and experience (Fig. 3). The data shown in Fig. 3 are from the same coating runs as used for Fig. 2.

Substantial improvement in the run-to-run variability for AGR-5/6 qualification runs is attributed mostly to the practice of calculating the gas flow rates and durations (i.e., dosing) based on the total bed surface area. Another contributing factor was the replacement of the methyltrichlorosilane (MTS) vaporizer for the SiC layer after AGR-2 fuel particles had been coated, which resulted in a notable improvement in the run-to-run variability in the SiC layer thickness. The new vaporizer does not rely on a pool of MTS, but rather atomizes the MTS into a warmed carrier gas stream to ensure total vaporization and uniform feed rate control of MTS to the coater furnace.

The average particle-to-particle standard deviations in the layer thicknesses for the AGR-5/6 qualification runs are comparable to those from historical reference fuels from the USA, German, and Japanese programs (Fig. 4) despite the fact that the fuels were fabricated in coaters with differing designs and kernel charge masses [8][9][10]. The standard deviations of layer thickness data for the AGR program indicate that the process control is comparable to that of the reference fuel processes. The improvement in the population standard deviations for layer thicknesses within the USA programs is attributed to improved measurement methods and improved process control.

II.E. TRISO Layer Density Variability

The variability in layer densities is another aspect that can be affected by changes in coater retort dimension and kernel charge mass as they directly influence particle velocities in various regions of the bed, bed circulation rates, and residence times in the coating zones. The mean layer densities are controlled by the processing parameters (such as bed temperature and linear deposition rate) and vary insignificantly with changes in the coating retort dimension or charge mass. The measure of dispersion about the mean (standard deviation), however, may be susceptible to such changes, because sustaining a linear layer deposition rate necessitates changing the superficial gas velocity with changes in kernel charge mass, which may impact bed fluidization and particle circulation rates.

The densities for the inner and outer pyrocarbon layers and SiC layer are measured by placing layer fragments into a density gradient column and measuring the densities of the individual fragments in the population “cloud.” Fig. 5 shows the average standard deviations in the density measurements for individual layers. The data suggest that little if any change was observed when the coater retort was scaled up and the charge mass was 1.3 kg, but an increase in the dispersion width was observed for the inner pyrocarbon (IPyC) layer when the kernel charge mass was increased to 2.0 kg.
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The calculated values for the pyrocarbon layer Bacon Anisotropy Factor (BAF\(_0\)), based on ellipsometry diattenuation values, is shown in Fig. 7 for the three AGR campaigns.

Both pyrocarbon layers appear to have been adversely affected by the dimensional changes to the coater retort and possibly to the increase in charge mass for the last data set. The AGR-1 and AGR-2 anisotropy data sets were measured at ORNL and the AGR-5/6 qualification run data were measured at B&W NOG-L, using similar equipment. As noted earlier, the AGR-2 and AGR-5/6 data sets were collected from particles coated with the control thermocouple at different locations. Insufficient information exists to attribute the increase in anisotropy between the latter two campaigns to the increase in the charge mass because of the concurrent change in the location of the control thermocouple and analytical laboratory. The effect of changing the control thermocouple location is expected to be minor.

Particle-to-particle variability in the BAF\(_0\) is shown in Fig. 8. The data displayed in this figure suggest that the consistency from particle-to-particle within a batch may have improved somewhat with the larger retort. The subsequent increase in the variability for the AGR-5/6 qualification runs

**II.F. Pyrocarbon Anisotropy Variability**

Pyrocarbon deposits are inherently anisotropic. It is fortuitous that the nearly random orientation and spin on the particles within a spouted-bed coater allow for nearly isotropic pyrocarbon layer properties. Both density and the anisotropy are inversely related to the process temperature. Careful selection of the processing temperatures was required for the AGR program coating development runs to ensure compliance with both fuel specifications.
suggests that increasing the charge mass may have had a detrimental effect. The particle-to-particle variability should be relatively immune to the location of the control thermocouple because uniformity is more a function of bed fluidization and particle circulation.

The run-to-run variability in the batch mean anisotropies is shown in Fig. 9. The run-to-run data show a stark difference between AGR-2 UCO data and data from the other campaigns. It is noteworthy that the mean particle and run-to-run standard deviations are less for the AGR-2 campaign despite the AGR-2 fuel exhibiting greater anisotropy than the AGR-1 fuel. The data in Figures 8 and 9, collectively, indicate that the larger coater has better fluidization during pyrocarbon deposition than the smaller coater, especially for the AGR-2 campaign, which had the smaller kernel charge mass.

Figure 10: Mean layer aspect ratio.

Because the SiC thickness is very uniformly deposited on any given particle, the increase in the SiC aspect ratio suggests that the faceting began with the underlying substrates. Kernel aspect ratios have been very consistent and do not correlate with the increase in aspect ratio for the AGR-5/6 qualification runs. Therefore, the increase in aspect ratio is linked to the buffer and IPyC layers, which has been verified by visual inspection of mounted and sectioned particles.

The change in control thermocouple location is unlikely to have contributed to the increase in the aspect ratio, so the primary contributing factor is the charge mass, which establishes the bed depth and influences particle fluidization and particle residence times in the annular and spouted regions.

The particle-to-particle standard deviation of the aspect ratio, shown in Fig. 11, also shows an increase in population dispersion associated with an increased charge mass in the prototypic coater, which supports the hypothesis that particle fluidization and residence times had been adversely affected.
Fig. 11: Mean particle standard deviations in layer aspect ratio.

Run-to-run dispersion in the aspect ratio is about $1/6^\text{th}$ the particle-to-particle dispersion and exhibits no sensitivity to the charge mass. The low run-to-run dispersion indicates that the coating process is well controlled concerning this property.

III. CONCLUSIONS

The AGR TRISO fuel particles meet the USA fuel specifications for all layer properties and the coating process is well controlled and reliable. Program objectives for fuel quality and reproducibility have been met in the prototypic coating process.

Although changes in individual properties or the standard deviations (both particle-to-particle and run-to-run) generally do not give strong indications of the causal factors associated with the changes, collectively they point to charge mass as a stronger influence in the properties and the standard deviations than coater retort scale.

The run-to-run variabilities in layer thickness and density are favorably influenced by a more massive bed as slight variations in gas flow rates are less significant relative to the larger gas flows required with the larger charge masses. Variability in the buffer density after AGR-1 fuel may be attributable to differences in the analytical methods and equipment used in the measurement and may not be reflective of an actual change associated with increasing the retort dimensions.

The anisotropy of the pyrocarbon layers may have been adversely impacted by the scale of the coater and the bed mass. Superficial (axial) gas velocities do not remain constant as the bed is scaled. Bed mass increases nearly with the cube of the retort diameter whereas the cross sectional area increases with the square; resulting in higher superficial gas velocities to maintain a constant deposition rate in larger coaters. This leads to lower coating efficiencies and altered bed fluidization. These factors appear to influence the pyrocarbon deposition mechanisms that control the pyrocarbon isotropy.

The AGR program has maintained fixed precursor gas concentrations and linear deposition rates for the pyrocarbon layers by adjusting the total gas flows during scale-up or changes in the kernel charge mass to ensure compliance with fuel specifications.

The aspect ratio of the coated particles did not degrade with the transition from laboratory to prototypic-scale coater equipment, but some degradation is possibly associated with increasing the kernel charge mass in the prototypic coater.

REFERENCES


