SiC Layer Microstructure in AGR-1 and AGR-2 TRISO Fuel Particles and the Influence of Its Variation on the Effective Diffusion of Key Fission Products

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Abstract: Tristructural isotropic (TRISO) coated particle fuel is a promising fuel form for advanced reactor concepts such as high temperature gas-cooled reactors (HTGR) and is being developed domestically under the US Department of Energy's Nuclear Reactor Technologies Initiative in support of Advanced Reactor Technologies. The fuel development and qualification plan includes a series of fuel irradiations to demonstrate fuel performance from the laboratory to commercial scale. The first irradiation campaign, AGR-1, included four separate TRISO fuel variants composed of multiple, laboratory-scale coater batches. The second irradiation campaign, AGR-2, included TRISO fuel particles fabricated by BWX Technologies with a larger coater representative of an industrial-scale system. The SiC layers of as-fabricated particles from the AGR-1 and AGR-2 irradiation campaigns have been investigated by electron backscatter diffraction (EBSD) to provide key information about the microstructural features relevant to fuel performance. The results of a comprehensive study of multiple particles from all constituent batches are reported. The observations indicate that there were microstructural differences between variants and among constituent batches in a single variant. Insights on the influence of microstructure on the effective diffusivity of key fission products in the SiC layer are also discussed.

Keywords: Silicon carbide, TRISO, coated particle fuel, EBSD, microstructure

1. Introduction

Tristructural isotropic (TRISO) coated particle fuel is a promising fuel form for advanced reactor concepts such as high temperature gas-cooled reactors (HTGR), molten salt reactors, and as accident tolerant fuel for light water reactor designs. The current focus of US efforts is on a version of the TRISO fuel particle containing a 350 or 425- μm -diameter kernel that is a mixture of uranium carbide and uranium oxide (UCO), surrounded by four successive isotropic layers: a 100- μm -thick porous carbon “buffer” layer, a 40- μm -thick dense inner pyrolytic carbon layer (IPyC), a 35- μm -thick chemical vapor deposition (CVD) silicon carbide (SiC) layer, and a second 40- μm -thick dense outer pyrolytic carbon layer (OPyC). The individual particles are overcoated with a resinated graphite powder and formed into half-inch-diameter, one-inch-long cylindrical compacts. The use of all graphite or refractory materials in the design of the fuel and core allows for higher operating temperatures, increases efficiency, and provides an inherent safety factor because fuel components remain intact in the case of coolant loss [1].

In the TRISO fuel design for HTGR applications, each component performs various functions during operation that contribute to the overall fuel integrity and retention of the nuclear fuel and radioactive isotopes generated by the fission process. Most fission products are retained within the kernel, and the UCO kernel chemistry is designed to optimize this retention while controlling CO production that can be detrimental to the particle integrity at higher burnup [2, 3]. The TRISO layers are designed to retain most metallic and gaseous fission products released from the kernel and play an important role in the stability of the kernel by preventing the release of gaseous reaction products. Having the greatest strength and irradiation stability, the SiC layer is the primary structural component and primary barrier to release of fission products that escape

the kernel and can diffuse through the carbon layers. As such, the integrity and properties of the SiC layer are paramount to TRISO fuel performance.

The current US TRISO fuel research effort is part of the US Department of Energy's Advanced Gas Reactor (AGR) Fuel Development and Qualification Program. The program plan includes a series of irradiation experiments conducted in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL) to demonstrate fuel performance and fabrication capabilities from the laboratory to commercial scale [1]. The first irradiation campaign, AGR-1, included laboratory-scale TRISO fuel, with kernels fabricated by BWX Technologies (BWXT) that were coated and compacted at Oak Ridge National Laboratory (ORNL). The irradiation test was completed in the ATR in November of 2009 [4]. The AGR-2 campaign included TRISO particles fabricated by BWXT to demonstrate industrial-scale coating, compacted at ORNL, and irradiated in the ATR. The AGR-2 irradiation was completed in October of 2013 [5]. All compacts in the AGR-1 experiment were composed of TRISO particles with UCO kernels. The AGR-2 experiment included separate tests of both UCO-TRISO and UO_2 -TRISO to explore the performance variations between the two kernel types.

The AGR-1 and AGR-2 campaigns include extensive post-irradiation examination (PIE) at INL and ORNL to understand the fuel evolution and performance. One area of the PIE effort is targeted towards understanding individual particle performance, which focuses on fission product retention and layer integrity [6],[7]. PIE efforts have indicated the release of select fission products, such as europium, silver, and strontium from intact TRISO particles [6]. In general, silver shows the greatest fractional release from particles with intact SiC, and AGR-1 PIE results have shown particle-to-particle and compact-to-compact variation in measured $^{110\text{m}}\text{Ag}$ retention, with the in reactor temperature believed to be the primary driver for the

observed release behaviors [6]. Ultimately, release of fission products is expected to be predominantly dependent on in-reactor variables, however, physical variables associated with the TRISO fuel construction and layer microstructures are also expected to influence release.

The SiC microstructure is hypothesized to impact the release of silver, and grain boundaries (GBs) are expected to facilitate transport across the SiC layer [8],[9],[10]. Understanding the SiC microstructural differences in TRISO particles can provide insight on the observed variation in $^{110\text{m}}\text{Ag}$ release, because GBs are expected to serve as transport pathways. The microstructural variables that can influence effective diffusion in polycrystalline materials include grain size, grain shape or aspect ratio (AR), and grain boundary character distribution (GBCD). Here GBCD is defined as the distribution of specific GB classes: low angle grain boundaries (LAGB), low- Σ coincidence site lattice (CSL) GBs, and high-angle grain boundaries (HAGB).

The AGR-1 irradiation experiment included four separately processed and tested TRISO fuel composites, each fabricated with intentional but minor variations in the coating conditions. Multiple TRISO-coated particle batches from the ORNL 50-mm-diameter fluidized-bed CVD coater were used to accumulate the particle inventory to make each of the four separate composites [11]. The TRISO particles for each AGR-2 fuel type were fabricated from a single, upgraded TRISO coating batch produced in the BWXT 150-mm-diameter coater [12]. Comprehensive microstructural information on the SiC layer of all coating batches included in the AGR-1 irradiation experiment and the two distinct batches from the AGR-2 irradiation experiment was obtained using Scanning Electron Microscopy (SEM) coupled with Electron Backscatter Diffraction (EBSD). This data provide a reference for TRISO fuel fabrication, PIE, and modeling efforts, and contribute to the understanding of the relationships between SiC microstructure and TRISO fuel performance.

2. Materials and Methods

2.1 Particles

2.1.1 AGR-1 Particles

The four fuel lots in AGR-1 consist of a baseline particle variant and three unique variants with different TRISO particle processing conditions to influence the IPyC and SiC layers [11]. The AGR-1 lots were identified as Baseline (AGR-1 B), Variant 1 (AGR-1 V1), Variant 2 (AGR-1 V2), and Variant 3 (AGR-1 V3). TRISO layers for the AGR-1 campaign were deposited using a laboratory scale 50-mm-diameter fluidized-bed coater. For AGR-1 B, AGR-1 V1, and AGR-1 V2 the CVD SiC layer was deposited by a fluidized-bed CVD process through the decomposition of methyltrichlorosilane (MTS) in hydrogen, using H₂ as a carrier gas and a coating bed temperature of ~1500°C. The fuel lots of AGR-1 V1 and AGR-1 V2 were considered IPyC variants as the IPyC deposition conditions were modified relative to AGR-1 B. The SiC process conditions were varied for AGR-1 V3 particles to modify the SiC microstructure, as Ar was added to the MTS + H₂ and the deposition was carried out at a bed temperature of ~1425°C[11]. The addition of Ar to the system improves fluidization and alters reaction kinetics and renucleation resulting in a finer SiC microstructure [11]. *Table 1* summarizes the standard key process parameters used to fabricate the AGR-1 fuel lots while *Table 2* identifies the process variations in the four AGR-1 fuel lots [11, 13].

Table 1: Key standard process parameters for AGR-1 TRISO particle fabrication, reproduced from ref. [13]

<i>Parameter</i>	<i>Buffer</i>	<i>IPyC</i>	<i>SiC</i>	<i>OPyC</i>
<i>Deposition Temperature</i>	1450°C	1265°C	1500°C	1290°C
<i>Coating Gas Ratio</i>	NA	$0.85 \frac{C_3H_6}{C_2H_2}$	NA	$0.85 \frac{C_3H_6}{C_2H_2}$
<i>Coating Gas Fraction^a</i>	$0.61 \frac{C_2H_2}{TGF}$	$0.30 \frac{C_2H_2+C_3H_6}{TGF}$	$0.012 \frac{MTS \text{ in } H_2}{TGF}$	$0.30 \frac{C_2H_2+C_3H_6}{TGF}$
<i>Deposition Rate^b</i>	21 µm/min	3.2 µm/min	0.25 µm/min	4.1 µm/min

^a TGF = total gas flow

^b Deposition rate is reported as an average rate over each coating step; the actual deposition rate varies with increasing surface area.

Table 2: Variation in standard process parameters for the four AGR-1 fuel lots, reproduced from ref. [13]

<i>Particle Lot</i>	<i>IPyC Temperature</i>	<i>IPyC Coating Gas Fraction</i>	<i>SiC Temperature</i>
<i>AGR-1 B</i>	1265°C	0.30	1500°C
<i>AGR-1 V1</i>	1290°C	0.30	1500°C
<i>AGR-1 V2</i>	1265°C	0.45	1500°C
<i>AGR-1 V3^a</i>	1265°C	0.30	1425°C

^a AGR-1 V3 SiC layer was deposited using 50:50 mix of Ar:H₂ for the fluidization gas.

Each AGR-1 lot was a composite of multiple batches to build up the necessary particle inventory for fuel qualification and testing. Table 3 identifies the unique batches associated with each fuel composite. In the remainder of the paper, each batch will be denoted by the AGR-1 composite descriptor (B, V1, V2, V3) followed by the unique batch number (##T), for example V1-43T, refers to a batch from the composite AGR-1 V1. A total of five individual TRISO particles from each of the thirteen batches comprising the fuel variants used in the AGR-1 test

program were investigated by SEM-EBSD to characterize the microstructure of the SiC layer. Multiple particles from each batch were investigated to obtain a representative data set and to understand composite variations and possible batch-to-batch and/or particle-to-particle variations within each composite.

Table 3: AGR-1 composite make-up

<i>Composites</i>	<i>AGR-1 B</i>	<i>AGR-1 V1</i>	<i>AGR-1 V2</i>	<i>AGR-1 V3</i>
<i>Batches</i>	<i>21T</i>	<i>29T</i>	<i>22T</i>	<i>24T</i>
	<i>27T</i>	<i>43T</i>	<i>40T</i>	<i>25T</i>
	<i>35T</i>	<i>44T</i>	<i>42T</i>	<i>37T</i>
	<i>45T</i>			

2.1.2 AGR-2 Particles

The two fuel lots in AGR-2 consisted of upgraded TRISO fuel particles each from a single batch of coated particles, one made with UCO kernels and one with UO₂ kernels. The TRISO coating process was conducted in a commercial-scale 150-mm-diameter fluidized bed coater and utilized similar SiC deposition conditions as the AGR-1 V3 composite, that is, MTS+H₂+Ar at a bed temperature of ~1425°C [14]. The two AGR-2 particle lots are denoted as “AGR-2 UCO” for the UCO kernel particles and “AGR-2 UO₂” for the UO₂ kernel particles in the remainder of the manuscript. The particles from each AGR-2 batches were “upgraded” from the initial coating lots by sorting with a roller-micrometer to remove undersized and faceted particles. Multiple particles from each batch were investigated to obtain a more representative data set for each variant. Discussions of the coating processes, variables, and differences between AGR-1 and AGR-2 coatings can be found in references [11, 12],[14].

2.2 Sample Preparation

The sample preparation step is critical to conduct accurate EBSD analysis. The data acquisition is sensitive to the sample surface integrity, so maintaining a reproducible, flat, damage-free surface is necessary to ensuring quality EBSD analysis. Individual mounts for each AGR-1 composite were fabricated containing fifteen TRISO particles from each batch. Particles from each composite were confined to a single mount, while particles from an individual batch were placed in a four by four array in a unique quadrant leaving one position empty to serve as a fiducial mark for batch determination. Individual mounts for each AGR-2 lot were fabricated containing eighteen to nineteen TRISO particles from the single batch with the particles arranged in a hexagonal array and a disruption in the pattern to identify unique particle locations. While multiple particles from each batch were prepared for analysis only five particles were analyzed from each unique batch. The samples were mounted in a one and a quarter inch diameter mount in a blend of IsoFast and 15-wt% graphite powder using a Struers automated hot press system. The mounted particles were then taken to midplane using alcohol-based 6- μm diamond suspension on a Struers MD-Allegro pad with Struers Blue DP-Lubricant followed by a rinse in isopropanol. The isopropanol rinse was repeated after each successive step and the kernels were removed before the next polishing step was commenced. It should be noted that the actual time to midplane, typically 10 min, varied from mount to mount based on the number of particles and condition of the grinding surface. The first polishing step was 2 min of alcohol-based 3- μm diamond suspension on a Struers MD-Dac pad with lubricant, followed by 5 min of alcohol-based 1- μm diamond suspension on a MD-Dac pad with lubricant. Final polishing was completed with a 0.05- μm colloidal silica Struers OP-S solution on Struers MD-Chem pad for 30 min followed by a 0.02- μm colloidal silica solution from Allied on Struers MD-Nap for 45 min. The OP-S is an active oxide polishing suspension with a pH of 9.8 and provides

chemical/mechanical polishing during the final polishing steps to yield a high integrity surface ideal for EBSD. The samples were then sonicated in deionized water for 1 min, rinsed in isopropanol, and dried.

2.3 EBSD Analysis

The EBSD analysis was conducted on a JEOL 6500F FEG-SEM equipped with EDAX's TSL/OIM analysis system. SEM conditions utilized for analysis were a 15-kV accelerating voltage, 20-mm working distance, at a 70° tilt. EBSD scans were performed on the SiC layer using a hexagonal grid array, with a step size of 0.1 μm , four by four binning, and background subtraction and normalized intensity histogram corrections. The 0.1- μm step size was selected for efficiency, as the resolution for EBSD is expected to be on the same order. Additionally, these conditions ensure accurately resolved details would not be lost due to resolution concerns [15]. Each particle was analyzed using a single scan. The average scan area was 25 μm by 40 μm , which ensured greater than 50 unique grains were included to achieve accurate statistical sampling when determining grain size [16]. Scan area was limited by the acquisition time per scan, which is a function of EBSD camera exposure time, step size, and analysis area. Acquired EBSD patterns were indexed against the β -SiC (diamond-cubic) polytype as previously reported. EBSD analysis has identified the TRISO SiC layer to be primarily β -SiC [17],[18],[19]. Raw data was processed by a single grain dilatation step with less than 10% of the acquired points processed according to the ASTM standard for grain size analysis determination by EBSD [20]. GBs were defined by a minimum adjacent misorientation of $<5^\circ$, and grains with less than four points were excluded from the analysis. Microstructural information was obtained from partitioned data sets by excluding low pattern image quality (IQ) points, $\text{IQ} < 1000$, to remove data from the IPyC and OPyC layers, where IQ is a quality metric related to the quality of the

acquired diffraction pattern [21]. Backscattering electron (BSE) micrographs of select scan areas were recorded along with the EBSD scan area. The conditions for the BSE analysis were optimized to enhance the channeling contrast to highlight the local variations in crystallography and provide a qualitative microstructural comparison with the acquired EBSD microstructures. The SEM conditions for BSE microstructural analysis were 6–7 mm WD, 3-kV accelerating voltage, and 0° tilt.

3. Results

The comparison between EBSD and SEM imaging analysis serves to provide a check on the accuracy of the EBSD analysis. An example of the EBSD/SEM comparison is shown in *Figure 1*, where the representative EBSD image shows the inverse pole figure (IPF) map overlaid on the IQ grayscale map identifying unique grain orientation, and the BSE image shows the channeling contrast for the SiC layer of an AGR-1 B particle. The microstructure in the EBSD IPF/IQ maps show good qualitative agreement with the channeling contrast BSE micrographs as corresponding features such as grain size, grain shape, and twinning are identified in each analysis technique. In *Figure 1* corresponding grains are identified in the complimentary micrographs produced by both techniques as denoted by the arrows marking select corresponding grains. These observations imply accurate implementation of EBSD analysis

Figure 1: Comparison of IPF/IQ map (bottom) with BSE micrograph (top) of AGR-1 B-21T indicating qualitative agreement between measured microstructures. Arrows indicate corresponding microstructural features.

Figure 2 shows the IPF/IQ map for representative particles from each variant included in AGR-1 and AGR-2. From Figure 2, a qualitative difference in microstructure is evident between the grouping of AGR-1 B, AGR-1 V1, and AGR-1 V2, which possess a larger grain size relative to the grouping of AGR-1 V3, AGR-2 UCO and AGR-2 UO₂. These groupings follow the microstructural differences expected from the change in processing conditions. The addition of Ar to the system and lower bed temperatures, for AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂ is expected to produce a finer microstructure, as the Ar increases renucleation while the lower bed temperatures reduces surface diffusion. The addition of Ar to the system also aids in improving fluidization of the particles during coating deposition. For all variants a non-equiaxed grain shape is observed, with an elongated axis along the radial growth direction.

With EBSD analysis, grain size is often defined by the average grain diameter and the grain diameter in the TSL/OIM analysis software is determined by assuming the two dimensional grain cross-section is a circle and then calculating the diameter of the circle from the

measured area of each uniquely defined grain. However, the non-equiaxed nature of the actual microstructure suggests the grain diameter measurement does not appropriately represent the grain size. In this work, grain size is reported in terms of the major and minor axis length, where the major and minor axes are determined from fitting an ellipse to each unique grain to account for the non-equiaxed shape of the observed columnar SiC microstructure. In the following analysis, grain size is also provided in terms of the less-appropriate grain diameter using the definition above to allow for comparison with previous studies of the SiC in TRISO fuel particles [17, 18]. The average reported statistics are weighted by area fraction [17] to account for non-uniform distributions in grain size. Additionally, a two-dimensional sample cross-section does not fully represent the aggregate microstructure; grain size analysis of the cross-section provides a distribution in measured grain sizes ranging from the maximum value to a value of zero. This constraint leads to an under-representation of the true grain size and yields only an apparent grain size.

Figure 2: IPF/IQ map of unique grain orientation for representative particles from each batch: (a) AGR-1 B-21T, (b) AGR-1 V1-44T, (c) AGR-1 V2-40T, (d) AGR-1 V3-25T, (e) AGR-2 UCO and (f) AGR-2 UO₂.

β -SiC is a material with a high probability of twin formation [22] where twins represent the dominant CSL boundary [17],[18]. Twins are the lowest energy Low- Σ CSL boundaries and are expected to serve as limited percolation pathways for GB diffusion [23, 24]. Describing the microstructure size in terms of “twins” and “no twins” provides a metric to visualize the effect of GBCD on the release of fission products through the SiC layer under the assumption of GB diffusion dominated release. The “no twins” analysis reports the SiC microstructure without the low diffusivity twin boundaries, such that the microstructure is defined by the higher diffusivity pathways contributing to GB diffusion. For the β -SiC system the contributions of the twin

boundaries, $60^\circ \langle 111 \rangle$ and $38.9^\circ \langle 110 \rangle$, are removed in the “no twins” data sets describing the SiC microstructure (grain size and grain shape) which are presented in the following sections.

3.1 Grain Size and Grain Shape

3.1.1 Average SiC Microstructure

Figure 3a shows the average major and minor axis distributions for all batches included in the AGR-1 and AGR-2 campaigns. The data in *Figure 3* is presented with twin boundaries contributing to the reported SiC microstructural data. The major and minor axis measurements of the batches comprising AGR-1 B, AGR-1 V1, and AGR-1 V2 present similar microstructures with deviations near the reported error. One exception was an outlier batch in the AGR-1 B composite. The batches from AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂ presented a finer grain size. Again, this difference in measured grain size is expected based on the changes in processing conditions, specifically the introduction of Ar to the system and the lower bed temperatures for the variants with the finer microstructure. The reported uncertainty in major and minor axis length was calculated based on one standard deviation of the mean.

As noted, the batch-to-batch comparison indicates consistent groupings for each batch comprising AGR-1 V1, AGR-1 V2, and AGR-1 V3, however AGR-1 B indicates a batch-to-batch discrepancy in grain size (note, AGR-2 lots were formed from a single batch so batch-to-batch comparisons are not applicable). The AGR-1 B, B-45T batch possessed a finer grain size than the remaining batches, B-21T, B-27T, and B-35T. *Figure 3b* shows the particle-to-particle distribution of average major and minor axes for B-45T and B-21T, where B-21T provides a reference to a representative AGR-1 B batch. The grain size reported for each particle are clustered around the average for each batch and confirm that the observed difference in grain size for B-45T is not due to an individual particle anomaly. The particle-to-particle comparison

in *Figure 3* also indicates some variation among particles, particularly for the larger-grain-size variants, which is likely due to sampling effects from a limited analysis area and highlights the need for multiple EBSD analysis sites per batch.

Figure 3: (a) Average major and minor axis distribution for all batches with twins included, (b) average major and minor axis distribution for individual particles from AGR-1 B-21T and AGR-1 B-45T with twins included (large data point represents batch average value).

Representative IPF/IQ maps of B-45T and B-21T are shown in *Figure 4* to visually represent the batch-to-batch grain size variation. From *Figure 4* it is apparent B-45T possesses a finer microstructure than the representative B-21T batch, corroborating the grain size measurements presented in *Figure 3*. Ultimately, the average grain size of AGR-1 B-45T appears more similar to the variants deposited with Ar at lower bed temperatures. The cause of the difference in microstructure between B-45T and the other AGR-1 B batches is not explicitly known, however slight differences in processing conditions were noted concerning MTS gas delivery during SiC deposition [11]. Methyltrichlorosilane is a liquid at room temperature and is delivered to the fluidized-bed reactor using a bubbler system in which hydrogen flowing through

the liquid reagent is saturated with MTS and subsequently carried to the furnace. Vapor concentration in a bubbler-based system is a function of liquid temperature, carrier gas flow, and contact time in the fluid. For the SiC deposition of AGR-1 B-45T, the liquid level in the bubbler was low and it is speculated this led to splashing and thus uncontrolled spikes or fluctuations in MTS concentration. The variations likely caused disruptions in the deposition process and thus the discontinuous SiC microstructure for this batch of particles. To account for the batch-to-batch microstructural differences in the AGR-1 B composite, the average microstructural statistics are reported separately for the AGR-1B-45T batch, while the remaining three similarly-structured batches are reported as a unit henceforth denoted as AGR-1 B*.

Figure 4: IPF/IQ map comparison of Baseline batches AGR-1 (a) B-21T and (b) B-45T.

Table 4 reports the average major and minor axis length, AR, and grain size in grain diameter for all variants. Here AR is defined by the ratio of the major to minor axes which define the grain ellipse. The same trends are observed in the reported grain size by diameter as are observed in the grain size by major and minor axis, that is, the grouping of AGR-1 B-45T, AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂ possesses a finer grain size relative to the grouping of AGR-1 B*, AGR-1 V1, and AGR-1 V2. This trend in grain diameter follows the differences

in processing conditions for the respective variants, with the exception of AGR-1 B-45T. The different groupings in grain diameter for the AGR-1 variants has been reported previously by *Kirchhofer et al.* [17], however, the behavior of the outlier batch AGR-1 B-45T was not identified because sampling was restricted to the mixed composites and only a few particles were analyzed. All samples had non-equiaxed microstructures as confirmed by the AR greater than one. However, a trend is noted where the larger grain size samples also appear to have a more columnar microstructure as identified by larger ARs associated with larger average grain size.

Table 4: Average microstructural data for all AGR-1 and AGR-2 variants.

Variant	Major Axis (μm)		Minor Axis (μm)		AR		Diameter (μm)	
	Twins	No Twins	Twins	No Twins	Twins	No Twins	Twins	No Twins
AGR-1 B*	2.41 $\pm$ 0.20	5.82 $\pm$ 0.87	0.71 $\pm$ 0.05	1.34 $\pm$ 0.15	2.76 $\pm$ 0.05	3.32 $\pm$ 0.38	2.56 $\pm$ 0.20	5.49 $\pm$ 0.78
AGR-1 B-45T	1.33 $\pm$ 0.18	2.78 $\pm$ 0.44	0.47 $\pm$ 0.06	0.78 $\pm$ 0.10	2.40 $\pm$ 0.31	2.75 $\pm$ 0.09	1.55 $\pm$ 0.20	2.85 $\pm$ 0.39
AGR-1 V1	2.39 $\pm$ 0.24	5.10 $\pm$ 1.24	0.71 $\pm$ 0.05	1.23 $\pm$ 0.07	2.82 $\pm$ 0.10	3.27 $\pm$ 0.19	2.53 $\pm$ 0.21	4.93 $\pm$ 1.09
AGR-1 V2	2.14 $\pm$ 0.16	5.29 $\pm$ 0.83	0.65 $\pm$ 0.05	1.19 $\pm$ 0.11	2.76 $\pm$ 0.26	3.32 $\pm$ 0.34	2.31 $\pm$ 0.17	4.92 $\pm$ 0.49
AGR-1 V3	0.71 $\pm$ 0.10	1.29 $\pm$ 0.17	0.31 $\pm$ 0.04	0.45 $\pm$ 0.06	2.12 $\pm$ 0.03	2.37 $\pm$ 0.06	0.92 $\pm$ 0.11	1.49 $\pm$ 0.19
AGR-2 UCO	0.89 $\pm$ 0.14	1.67 $\pm$ 0.25	0.35 $\pm$ 0.05	0.56 $\pm$ 0.08	2.25 $\pm$ 0.27	2.53 $\pm$ 0.36	1.09 $\pm$ 0.15	1.88 $\pm$ 0.26
AGR-2 UO₂	1.19 $\pm$ 0.18	2.37 $\pm$ 0.32	0.45 $\pm$ 0.06	0.77 $\pm$ 0.10	2.34 $\pm$ 0.30	2.61 $\pm$ 0.39	1.42 $\pm$ 0.20	2.62 $\pm$ 0.34

As noted by the values listed in *Table 4*, the exclusion of twin boundaries increases the magnitude of the major and minor axes as well as the magnitude of the AR and diameter. The exclusion of twin boundaries also enhances the observed differences between the larger-grained grouping of AGR-1 B*, AGR-1 V1, AGR-1 V2 and the finer-grained grouping of AGR-1 B-45T, AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂. The major axes are a factor of 2.13-2.47 larger for the no twins analysis of AGR-1 B*, AGR-1 V1, AGR-1 V2 compared to 1.80-2.09 larger for AGR-1 B-45T, AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂. While the minor axes are a

factor of 1.73-1.88 larger for the no twins analysis of AGR-1 B*, AGR-1 V1, AGR-1 V2 compared to 1.48-1.71 larger for AGR-1 B-45T, AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂.

3.1.2 Dependence of SiC Microstructure on Distance from IPyC/SiC Interface

The previous work by *Kirchhofer et al.* identified the SiC microstructure to vary as a function of distance from the IPyC/SiC interface with an approximately linear increase in grain diameter moving from the IPyC/SiC interface to the SiC/OPyC interface [17]. In this work, the microstructural dependence as a function of distance into the SiC layer from the IPyC/SiC interface was determined for all particles investigated. Each unique grain was assigned an x,y coordinate associated with the central point of the ellipses which defines the grain during OIM/TSL data processing. The distance from the grain ellipse central point perpendicular to the IPyC/SiC interface was used to determine the position of a unique grain relative to the interface. The IPyC/SiC interface was defined by a linear fit of the grains at the IPyC/SiC interface. The effect of the interfacial curvature is expected to be minimal and was not considered in the analysis, as the chord height defined by the arc length of the IPyC/SiC interface associated with the EBSD scan was minimal. The dependence of the major and minor axes as well as AR on the distance from the IPyC/SiC interface was determined for all fuel lots.

Figure 5 shows a comparison of major axis length with twins and with no twins included for all variants included in the AGR-1 and AGR-2 experiment. The major axis length analysis as a function of distance from the IPyC layer corroborates the observation of fine grains at the IPyC/SiC interface and shows an increase in grain size across the SiC with a decrease in grain size at the SiC/OPyC layer. This trend is more pronounced for the larger-grained variants deposited without Ar (AGR-1 B*, AGR-1 V1, and AGR-1 V2) with a significant increase in major axis length in the middle of the SiC layer, while the variants deposited with Ar (AGR-1

V3, AGR-2 UCO, and AGR-2 UO₂) have a relatively constant grain size across the thickness of the SiC layer. The fine grains at the IPyC/SiC interface are due to initial nucleation and growth of the SiC layer. This is visualized by the higher relative frequency of fine grains at the IPyC/SiC interface (*Figure 2*), and also confirmed by transmission electron microscopy analysis by *Kirchhofer et al.* [17]. The reduction in grain size, relative to the mid-layer, at the SiC/OPyC interface is due to the termination of the layer growth and is observed by the use of discrete binning of the major axis data. The results of the grain size dependence as a function of distance from the IPyC layer further confirm the similarity in microstructures among the grouping of the variants deposited without Ar of AGR-1 B*, AGR-1 V1, and AGR-2 and the grouping of the variants deposited with Ar of AGR-1 V3, and AGR-2 UCO and AGR-2 UO₂.

Figure 5: Major axis dependence on distance from the IPyC layer for all variants with twins included (red circles) and no twins included (blue squares). Data is binned every 5 μm from 0–40 μm .

The influence of twinning, twins versus no twins, on the dependence of the grain size (major axis length) across the thickness of the SiC layer from the IPyC/SiC interface to SiC/OPyC interface is visualized in Figure 5. For this analysis, the same trends noted in Table 4 concerning the influence of twins on the average microstructural features are reflected, in that the

increase in grain size, as defined by major axis length, is more pronounced for the larger-grained variants deposited without Ar relative to the variants deposited with Ar when the contributions from twins are not included in the microstructure. Additionally, the dependence of grain size on distance from the IPyC layer is exaggerated in the “no twins” data set for the variants deposited without Ar while variants deposited with Ar maintain a relatively constant microstructure across the thickness of the SiC layer, further confirming the influence of twinning is more prevalent in the variants deposited without Ar. For the outlier batch of AGR-1 B45-T, the “no twins” dependence of grain size on distance from the IPyC layer shows a distance dependence similar to the batches deposited without Ar where there is a maximum grain size in the center of the SiC layer, however, it still possesses a finer grain size than the other batches deposited without Ar. This behavior is particularly highlighted in the “no twins” analysis and differentiates the outlier AGR-1 B-45T batch from the variants deposited with Ar which show a relatively constant grain size after the initial nucleation at the IPyC/SiC interface. This observation indicates the AGR-1 B-45T batch experienced different SiC growth kinetics relative to the other batches included in this study.

Figure 6 shows a comparison of AR with twins and with no twins for all variants included in the AGR-1 and AGR-2 experiment. The observed relationship indicates the magnitude of the AR is lower at the IPyC/SiC interface, increases toward the center of the SiC layer, and decreases again at the SiC/OPyC interface, however, this trend is not strongly visualized for AGR-1 V3. This trend indicates more equiaxed grains at the PyC/SiC interfaces and more columnar grains in the center of the SiC layer. The binning suggests a rapid increase in AR moving out from the IPyC/SiC interface in the growth direction, indicating initial grains are more-equiaxed at the nucleation surface.

The comparison between the twins and no twins data show the exclusion of twins exaggerates the grain shape leading to more columnar grains along the growth direction relative to twins included. The AR dependence on distance from the IPyC layer shows the same differing behaviors for the two groupings. The larger-grained grouping from the variants deposited without Ar shows an increase in AR magnitude in the center of the SiC layer relative to the PyC/SiC interfaces, while the finer-grained grouping from the variants deposited with Ar show relatively constant AR. The AR dependence on the distance from the IPyC layer for AGR-1 B-45T follows the trend of a maximum AR at the center of the SiC layer relative to the PyC/SiC interfaces, which is in line with the variants deposited without Ar, however, the magnitude of the AR in the center of the SiC layer is statistically lower than in AGR-1 B*. The nature of the AR distance dependence for AGR-1 B-45T is limited in the twins included data set and more prevalent in the no twins analysis. The limited dependence of distance from the IPyC layer for grain size and AR observed for the finer-grained, SiC grouping deposited with Ar suggests near-constant growth conditions were likely reached during the deposition process.

Figure 6: Aspect ratio (AR) dependence on distance from the IPyC layer for all variants for twins included (red circles) and no twins included (blue squares). Data is binned every 5 μm from 0–40 μm .

3.2 Grain Boundary Character Distribution

The GBCD is a measure of the distribution of GB types in the SiC layer. The nature of the misorientation between two adjacent grains serves as a descriptor of the GB's local structure, which can influence the properties of the unique grain boundary. As noted, the GBCD is

described by the distribution of LAGB, HAGB, and low- Σ CSL GBs. LAGBs are defined as GBs with less than 15° misorientation between two adjacent grains, but for analysis purposes in this work, LAGBs are defined by GBs with misorientations of $5^\circ \leq \text{angle} \leq 15^\circ$ in the EBSD data processing. The lower limit of 5° was selected because a 5° misorientation was used to define the minimum adjacent grain misorientation to identify GBs comprising unique grains for the grain statistics. Additionally, using a 5° lower limit was preferred over the default 2° lower limit as the 2° lower limit on misorientation increased the fraction of LAGBs by introducing multiple GBs within the interior of unique grains. *Figure 7* shows a comparison of IPF maps with overlaid GB misorientation for minimum GB misorientation angles of 2° and 5° . The 2° minimum misorientation map shows numerous small GB segments in the interior of a grain that are not associated with a GB defined by a unique grain. These GB segments are not as frequently observed in the 5° minimum misorientation map. It is assumed that a majority of the GBs with misorientations 2° to 5° are analysis artifacts, possibly due to localized strain or SiC defect structures. This adds additional validation to the LAGB selection criteria of $5^\circ \leq \text{angle} \leq 15^\circ$. The magnitude of the LAGB artifacts contained in the 2° to 5° misorientation range is noted by the reported LAGB fraction, where the range in average LAGB fraction observed for AGR-1 and AGR-2 variants was 0.03-0.05 for the $5^\circ \leq \text{angle} \leq 15^\circ$ definition compared to 0.10-0.14 for the $2^\circ \leq \text{angle} \leq 15^\circ$ definition. The minimum and maximum LAGB fraction for an individual scan was reported to be 0.03 and 0.08 for the $5^\circ \leq \text{angle} \leq 15^\circ$ definition and 0.05 and 0.19 for the $2^\circ \leq \text{angle} \leq 15^\circ$ definition. This variation in LAGB fraction highlights the potential influence of EBSD processing artifacts on the GBCD.

Figure 7: IPF map of with a GB misorientation overlay: (a) minimum LAGB 5°, (b) minimum LAGB 2°. Blue GB segments represent lowest misorientation angle GBs, while red GB segments represent the highest misorientation angle GBs, the arrow identifies an example of a LAGB artifact.

HAGBs are generally described as GBs with misorientation greater than 15° and have the highest GB energies relative to the other GB types. CSL GBs are a subset of HAGBs and possess a higher than normal degree of symmetry and localized fit across the GB. The CSL is defined by the atomic positions, which coincide across the GB, as constructed from the two adjacent crystalline lattices transposed across the GB. CSL GBs are described by the Σ notation and defined by the Brandon criteria [25]. The Σ value is the reciprocal of the fraction of the area of the standard unit cell to the unit cell from the CSL lattice. Low- Σ CSL GBs, $\Sigma 3 \leq \Sigma 29$, are generally defined as a unique class as they behave more “bulk-like” compared to other GB classes [26]. The twin boundaries discussed earlier, 60° $\langle 111 \rangle$ and 38.9° $\langle 110 \rangle$, represent $\Sigma 3$ and $\Sigma 9$ CSL GBs respectively, and represent the lowest energy CSL GBs.

Figure 8 shows the distribution of low- Σ CSL GBs for all variants. The distribution is typical of diamond-cubic, low-stacking fault materials, where $\Sigma 3$ and $\Sigma 9$ CSL GBs are the dominant CSL GB [27]; however, there is also a notable fraction of $\Sigma 29a$ GBs. The relatively large fraction of $\Sigma 29a$ GBs is questionable as the energy of $\Sigma 29a,b$ GBs is not expected to be low relative to twin CSL GBs in diamond-cubic systems and the fraction of unique Low- Σ CSL GBs

is inversely proportional to the GB energy [28]. Considering this relationship, $\Sigma 29a$ fractions greater than $\Sigma 9$ fractions should not be observed. The inset in *Figure 8* shows an IPF/IQ maps with $\Sigma 29a,b$ GBs overlaid. The locations of these GBs appear to be irregular and define small sub-grains within an individual grain, suggesting $\Sigma 29a$ GBs are likely an analysis or processing artifact. To compensate for this artifact when describing the SiC layer microstructure, a subset of the Low- Σ CSL GBs is defined as “Special” CSLs from the range $\Sigma 3 \leq \Sigma 29$. Here $\Sigma 3$, 5, 9, 11, 17, 19, and 27 are categorized as “Special” CSL GBs as they are expected to possess low GB energies relative to the “Other” CSL GBs in diamond-cubic systems [27-29]. In this work, the “Special” CSL GBs are dominated by the fraction of twin variants, which comprise 86 to 91% of the total measured CSL GBs ($\Sigma 3 \leq \Sigma 29$) for all fuel lots investigated.

Figure 8: Low- Σ CSL GB fractions for all variants showing frequency of each CSL GB; (inset) IPF/IQ map with a GB misorientation overlay, minimum LAGB 5° , red lines indicate distribution of $\Sigma 29a$ and $\Sigma 29b$ GBs. The map is of a high $\Sigma 29a,b$ fraction scan (0.13 GB Fraction) from AGR-1 V2.

Table 5 shows the GBCD for all variants and breaks out “Special” CSL GBs from “Other” CSL GBs. From the data in *Table 5* it is apparent that two groupings exist and the differences are visualized by the ratio of “Special” CSL GBs to HAGBs. The variant grouping deposited without Ar of AGR-1 B*, AGR-1 V1, and AGR-1 V2 possess a higher fraction of “Special” CSL GBs to HAGBs, relative to the variant grouping deposited with Ar of AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂. The outlier batch of AGR-1 B-45T has a “Special” CSL GBs to HAGBs ratio within the range reported for the variants deposited with Ar. These same groupings are consistent with those observed for the average grain size and grain shape analysis. The higher fraction of “Special” CSL GBs for the larger-grained SiC microstructure grouping explains the observed exaggerated increase in grain size and shape when comparing microstructures with twins and with no twins.

Table 5: GBCD data for all AGR-1 and AGR-2 variants.

Variant	AGR-1 B*	AGR-1 B-45T	AGR-1 V1	AGR-1 V2	AGR-1 V3	AGR-2 UCO	AGR-2 UO ₂
LAGB ($5^\circ \leq \text{Angle} < 15^\circ$)	0.05±0.01	0.04±0.00	0.05±0.01	0.05±0.01	0.03±0.00	0.03±0.00	0.04±0.00
“Other” CSL	0.10±0.02	0.11±0.03	0.10±0.02	0.12±0.03	0.08±0.01	0.05±0.02	0.05±0.02
“Special” CSL	0.43±0.02	0.37±0.01	0.44±0.02	0.43±0.02	0.37±0.01	0.41±0.01	0.43±0.01
HAGB (Angle > 15°)	0.41±0.01	0.48±0.03	0.42±0.01	0.41±0.01	0.52±0.01	0.51±0.01	0.49±0.01
Ratio: “Special” CSL to HAGB	1.05±0.06	0.76±0.06	1.05±0.06	1.05±0.08	0.72±0.02	0.81±0.04	0.87±0.05

4. Discussion

4.1 Microstructural Variations

EBSD analysis of the SiC layer from all thirteen batches comprising the four laboratory scale composites of the AGR-1 fuel experiment and two commercial scale-up lots from AGR-2

showed that significant variations in SiC microstructure exist among the investigated batches. The results confirm that the deposition at lower bed temperatures with the addition of Ar to improve fluidization and enhance renucleation yields a finer-grained microstructure as AGR-1 V3, AGR-2 UCO, and AGR-2 UO₂ had a statistically smaller grain size compared to AGR-1 B*, AGR-1 V1 and AGR-1 V2. Analysis indicated different trends in grain size and shape across the SiC layer as a function of distance from the IPyC with the batches deposited without Ar having a maximum grain size and AR in the center of the SiC layer, while the variants deposited with Ar indicated a consistent microstructure across the thickness of the SiC layer with the exception of the initial nucleation at the IPyC/SiC interface.

The batch-to-batch microstructural analysis indicated the presence of a batch outlier in AGR-1 B-45T. The microstructure of AGR-1 B-45T, had average grain size, grain shape, and GBCD similar to the variants deposited with Ar, however, the dependence of these attributes on distance from the IPyC was more representative of the variants deposited without Ar implying different growth kinetics. The batch-to-batch variations highlight the benefit of comprehensive analysis when defining the aggregate microstructure for a TRISO fuel variant. In addition to the batch-to-batch variation, particle-to-particle variations were also observed. The particle-to-particle variation identifies the need for sufficient sampling to develop a data set that more accurately defines the microstructure associated with a TRISO fuel variant.

The EBSD analysis showed a correlation between grain size, grain shape, and GBCD. The dependence of the microstructure on distance from the IPyC for the coarse-grain and fine-grain groupings indicated the finer grain microstructure yielded a more-equiaxed grain shape and the AR appeared to be related to grain size. Dependence of grain size and shape is most apparent in *Figure 3a* which shows the average major and minor axis for all batches. A

linear fit of the average data generates the relationship, $y=4.26x-0.63$ ($y = \text{major axis}$, $x = \text{minor axis}$) with a coefficient of determination of 0.993. This indicates the grain size and shape dependence of the SiC layer follows a relationship when similar processing conditions are used to attain high density SiC layers in TRISO fuel.

The finer-grained SiC microstructures in the variants with Ar included in the system that were deposited at lower bed temperatures, showed an overall higher fraction of HAGBs. This was likely due to deposition at a lower temperature which limited surface diffusion, while the Ar in the coating gas promoted nucleation resulting in a higher number of competing grains. In contrast, the higher bed temperature of the larger-grained grouping deposited without Ar lead to increased surface diffusion promoting the growth of lower energy boundaries in the CVD-SiC layer.

As previously discussed, the specific cause of the batch-to-batch variation in composite AGR-1 B, is not explicitly known. While the deposition conditions were intended to be held constant for all batches of AGR-1 B, it is speculated that fluctuations in MTS concentration may be responsible for the difference in measured SiC microstructure for AGR-1 B-45T. Despite the measured difference in microstructure, the SiC layer met all fuel fabrication specifications and no variation in fuel performance or fission product retention was observed. Specifically, no noticeable variation between the fission product retention behaviors was observed between the heterogeneous-SiC microstructure fuel variant of AGR-1 B and the homogenous-SiC microstructure fuel variant of AGR-1 V1. AGR-1 PIE efforts measured individual particle fission product inventories and identified a broad distribution in fission product retention behavior for ^{110m}Ag in a single compact and this behavior was observed for all variants in the AGR-1 fuel irradiation. Post-irradiation examination of the SiC microstructure of high release

particles and low release particles in AGR-1 B compacts indicated no obvious correlation to the outlier fine-grained batch of AGR-1 B-45T [30].

4.2 Influence of EBSD Acquisition and Processing

The grain size analysis from this EBSD investigation of the AGR-1 variants yielded similar results to the previous study by *Kirchhofer et al.* [17] in that the AGR-1 B, AGR-1 V1, and AGR-1 V2 composites had a larger grain size than AGR-1 V3. In the work by *Kirchhofer et al.*, the average grain size, in grain diameter, ranged from 1.95–2.25 μm for AGR-1 B, AGR-1 V1, and AGR-1 V2 respectively and $\sim 0.75 \mu\text{m}$ for AGR-1 V3. For comparison, the average grain size in grain diameter reported for this work for AGR-1 B*, AGR-1 V1, AGR-1 V2, and AGR-1 V3 was $2.56 \pm 0.20 \mu\text{m}$, $2.53 \pm 0.21 \mu\text{m}$, $2.31 \pm 0.27 \mu\text{m}$ and $0.92 \pm 0.11 \mu\text{m}$, respectively. The average grain size was consistently larger in this study for all compared variants. In the *Kirchhofer et al.* work a step size of $0.05 \mu\text{m}$ was employed for the data acquisition compared to $0.1 \mu\text{m}$. To understand the effect of step size on the reported results EBSD analysis was performed on identical areas using steps sizes of $0.1 \mu\text{m}$ and $0.05 \mu\text{m}$. The use of a $0.05\text{-}\mu\text{m}$ step size resulted in an average refinement of the grain diameter by $0.12 \mu\text{m}$. The scatter observed in the batch-to-batch and particle-to-particle analysis of AGR-1 B-21T and B-45 (*Figure 3a and 3b*) also implies particle sampling may have contributed to the reported differences. For context, the range in grain diameter from all particles analyzed from AGR-1 B*, was $2.00 \pm 0.08 \mu\text{m}$ to $3.24 \pm 0.20 \mu\text{m}$, this range accounts for the observed differences between the two studies and highlights the benefit of sampling. In the *Kirchhofer et al.* [17] work a single particle from a deconsolidated compact was investigated, as such the specific batch could not be verified and the limited particle statistics may have affected the reported magnitude of the grain diameter.

The influence of processing on GBCD is also notable, especially considering the presence of system artifacts. The total fraction of LAGBs varied by up to 0.15, when changing the definitions of LAGB from $2^\circ \leq \text{angle} \leq 15^\circ$ to $5^\circ \leq \text{angle} \leq 15^\circ$. Similar artifacts may have influenced the previously reported work, which defined LAGB by the range of $2^\circ \leq \text{angle} \leq 15^\circ$ and indicated a LAGB fraction of 0.24, 0.04, 0.02, and 0.21 for AGR-1 B, V1, V2, and V3, respectively [17]. The LAGB fractions of 0.21-0.24 may be indicative of scan areas with a high artifact density, as noted in section 3.2.

One aspect that may also have influenced the reported microstructural analysis results between the two studies is the different sample preparation techniques employed, as the integrity of the sample surface influences the quality and accuracy of the acquired data. The study by *Kirchhofer et al.* [17] utilized focused ion beam milling to provide a fresh surface for EBSD analysis, while in this study metallographic sample preparation was used. The potential influence of these different techniques is unknown without systematic study, however, the potential effects should not be completely disregarded.

The impact of defined user parameters in the EBSD analysis concerning scan properties and data analysis definitions (grain and grain boundary) highlights the issues in describing the SiC layer microstructure. The reported differences between the *Kirchhofer et al.* [17] work and this work identify the sensitivity in selecting the appropriate analysis and acquisition conditions for comparing results from across studies. An understanding of the complete acquisition and processing routine is necessary to confidently compare and report EBSD results.

4.3 SiC Permeability to Fission Products

The permeability of a polycrystalline material to an impurity species via a diffusional process is dependent on the overall microstructure of the diffusion substrate. As discussed

earlier, GB diffusion is expected to play a role in fission product transport through the SiC layer, as such, the SiC microstructure is likely to effect the transport of fission product species across the layer. The influence of specific GB types is due to the prohibited GB diffusion kinetics of low- Σ CSL and LAGBs compared to HAGBs which indicates the impact of GBCD on the effective diffusivity of a material [31]. This observation implies that microstructures with a higher fraction of HAGBs would promote fission product release along the faster diffusing HAGBs. The grouping of AGR-1 V3, AGR-2 UCO and AGR-2 UO₂ has a higher fraction of HAGBs than the AGR-1 B*, AGR-1 V1, and AGR-1 V2 grouping. Considering these traits, the larger-grained, SiC deposited without Ar grouping possesses an ideal GBCD to limit fission product release, as the high relative fraction of low- Σ CSL GBs limit the percolation pathways, however, grain size and shape must be considered.

The effective diffusion coefficient ($\bar{D}$) for diffusion in polycrystalline materials can be represented by the Maxwell-Garnett equation (Equation 1), which considers the effect of grain size and shape on the effective diffusivity [32]. In Equation 1, D_{gb} is the grain boundary diffusion coefficient, D is the lattice diffusion coefficient, and g is a geometric factor dependent on the microstructure. This geometric factor, g , is defined in Equation 2 where d is a term describing the grain size, q a constant to describe the grain shape (1 for parallel grains, 3 for square grains) and δ is the GB width.

Equation 1:
$$\bar{D} = \frac{D_{gb}[(3-2g)D + 2gD_{gb}]}{gD + (3-g)D_{gb}}$$

Equation 2:
$$g = \frac{q\delta}{d}$$

For the TRISO relevant case of Ag diffusion in SiC the condition of $D \ll D_{gb}$, is expected [33],[34]. This leads to a limiting condition for $\bar{D}$ where it is dominated by D_{gb} and is effectively $\bar{D} \approx 2gD_{gb}/3$ [32]. The g term is analogous to the grain boundary volume fraction, but also

considers the orientation and shape of the grains along the diffusion direction. *Figure 9* shows the influence of the SiC microstructure on estimated $\bar{D}$ with twins ($\bar{D}_{twins}$) and with no twins ($\bar{D}_{no\ twins}$) defining the SiC microstructures, where $\bar{D}$ was calculated for each variant using $\bar{D} \approx 2gD_{gb}/3$. Here q is estimated from the measured AR by the relationship $q = C/AR$, where C is an unknown constant, while d is equivalent to the minor axis length. The minor axis length is used in the analysis because it is representative of the grain boundary spacing normal to the diffusion direction. The data is normalized to the magnitude of $\bar{D}_{twins}$ for AGR-1 B* ($\bar{D}_{twins}^{B*}$), as D_{gb} is dependent on the unique diffusion species and is unknown. As such, the constant $(2g/3)$ leading D_{gb} is indicative of the influence of microstructure on the magnitude of $\bar{D}$. The differences between the unique variants is then visualized in the ratio of $\bar{D}/\bar{D}_{twins}^{B*}$, where $\bar{D}_{twins}^{B*}$ represents a reference point for the comparison of $\bar{D}$ between the SiC microstructures “with twins” and with “no twins” considered in the analysis and normalizes the data to account for the unknown parameters in the system. The comparison between the $\bar{D}$ values becomes essentially dependent on the minor axis length and the relative magnitude of AR for the different variants as the constants $(2/3)$ and C , δ , and D_{gb} are common across the different variants. The analysis in *Figure 9* indicates the finer-grained SiC microstructure promotes transport by presenting a higher $\bar{D}$. Here, the variants deposited without Ar have similar $\bar{D}$ magnitudes for both $\bar{D}_{twins}$ and $\bar{D}_{no\ twins}$, with the $\bar{D}$ of AGR-1 V1 and AGR-1 V2 ranging ~1.0–1.1 times the magnitude of $\bar{D}_{twins}^{B*}$ ($\bar{D}_{twins}/\bar{D}_{twins}^{B*}$) and the same range present for $\bar{D}_{no\ twins}$ for both twins and no twins analysis analysis ($\bar{D}_{no\ twins}/\bar{D}_{twins}^{B*}$). The magnitudes of $\bar{D}$ for the variants deposited with Ar are 1.9–3.0 and 2.2–4.1 times larger than the $\bar{D}_{twins}^{B*}$ and $\bar{D}_{no\ twins}^{B*}$ analysis, that is $\bar{D}_{twins}/\bar{D}_{twins}^{B*}$ and

$\bar{D}_{twins}/\bar{D}_{twins}^{B*}$ respectively, with AGR-1 V3 having the highest $\bar{D}$ of all the variants. The outlier deposited without Ar of AGR-1 B-45T has a $\bar{D}$ between the two groupings.

Figure 9: Comparison of $\bar{D}/\bar{D}_{twins}^{B*}$ indicating influence of microstructure on $\bar{D}$, error bars represent the range in $\bar{D}$ based on the dependence of the microstructure on distance from the IPyC layer (left axis). Ratio of $\bar{D}_{twins}/\bar{D}_{twins}^{B*}$ (right axis). Arrows indicate the appropriate axis for each data set.

The relative influence of GBCD on $\bar{D}$ is found in the ratio of $\bar{D}_{twins}/\bar{D}_{twins}^{B*}$ in Figure 9. The difference in magnitude generally follows the trends in the reported GBCD (Table 5) for each variant. Here the variants deposited without Ar (AGR-1 B*, AGR-1 V1, and AGR-1 V2) which have the largest fraction of “Special” CSL grain boundaries have the greatest relative decrease in $\bar{D}$ when excluding twins from the microstructure. However, the difference in magnitude does not directly correlate with GBCD indicating the influence in grain shape and size when considering the different microstructures including twins and with no twins.

The difference in estimated $\bar{D}$ between the SiC microstructures suggest that CVD deposition process variations can be utilized to achieve SiC microstructures with different diffusional properties. Under the assumption that the “no twins” microstructure is representative of the active diffusion microstructure, $\bar{D}$ can be further influenced through GB engineering approaches by targeting ideal GBCDs. The $\bar{D}$ analysis, utilizing Equation 1 and *Equation 2*, implies a microstructure with a high fraction of “Special” CSL GBs, large grain size, and high AR is ideal to limit fission product transport, by minimizing the term, g , in *Equation 2*.

4.4 Irradiated TRISO fuel performance

Significant PIE efforts at ORNL and INL have been conducted on AGR-1 fuel compacts [30], with PIE on AGR-2 fuel compacts being initiated in 2015. PIE analysis of irradiated AGR-1 fuel indicated compacts containing particles from all AGR-1 variants released ^{110m}Ag . This included the AGR-1 V1 compact which had a $\bar{D}$ approximately 3.0 and 4.1 times lower than AGR-1 V3 for twins and no twins analysis respectively. This implies no particular microstructure completely mitigated silver release. However, insights on microstructural performance in-reactor are difficult to ascertain, as direct comparison of individual compact behavior is challenging because each compact experienced different time-dependent irradiation and thermal histories. Further complications are experienced as individual particles within a single compact also saw different irradiation conditions, most notably, temperature differences due to thermal gradients across the compact.

Compacts composed of AGR-1 B particles, which contained the three similar larger-grained batches (AGR-1 B*) and one unique finer-grained batch (AGR-1 B-45T), present an interesting opportunity for gaining insight on the effect of SiC microstructures on silver retention from PIE on irradiated compacts, as particles with varying SiC microstructures were

present in a single compact. During AGR-1 PIE, individual particle fission product inventories were determined by gamma spectroscopy to determine the activity of specific isotopes present in particles deconsolidated from the compact matrix and the retention behavior was calculated by ratioing the measured fission product activity to the average calculated activity for the specific isotope. This measured-to-calculated (M/C) ratio is indicative of the retention behavior of the specific isotope of interest in an individual particle. The analysis indicated particle-to-particle variation in the retention of fission products with large ^{110m}Ag M/C variations for select compacts. The fission product retention in the AGR-1 B compacts did not display obvious release behaviors that correlated to the fraction of particles with different SiC microstructure. For instance, one AGR-1 B compact exhibited a bimodal distribution in the measured ^{110m}Ag inventory that clearly indicated different silver retention behavior, with one subset of particles having a ^{110m}Ag inventory near or below the detection limit of the system and another subset centered around a higher M/C value. However, the fraction of particles in each subset did not correspond to the fraction of AGR-1 B particles with larger or smaller microstructure. [30, 35, 36]. Similar bi-modal particle-to-particle silver release variations were also noted for AGR-1 V1, which had a homogenous SiC layer microstructure [6, 37]. It is possible that the expected variation in $\bar{D}$ (~ 1.6 – 2.1 times larger for AGR-1 B-45T than for AGR-1 B*) is not sufficient to demonstrate a distinct difference in silver release behavior. It is probable that the release observed across all compacts in-pile was driven primarily by in-reactor variables. The likely dominant in-reactor variable driving fission product release during the AGR-1 experiment was temperature, where temperature varied spatially across compacts and temporally during the irradiation by up to $\sim 300^\circ\text{C}$ [38]. This temperature variation across the compact provides the most direct explanation of the particle-to-particle difference in release behavior under a

diffusion-driven release mechanism. In other words, none of the SiC had a sufficiently-low diffusion constant to limit release at certain temperatures. Additionally, while the impact of microstructure on release wasn't significant enough to observe in the AGR-1 experiment, it does not mean microstructure did not influence the release behavior in reactor to some degree.

As part of the AGR-1 campaign, out-of-pile safety testing of individual compacts was performed to understand fuel performance at accident conditions and to test the margins of fuel performance. Individual compacts were exposed to temperatures ranging 1600–1800°C for exposure times of 300 hours or more while periodically measuring the release of select fission products, notably $^{110\text{m}}\text{Ag}$, ^{154}Eu , and ^{90}Sr [39]. These fission products were selected as they are radiologically relevant ($^{110\text{m}}\text{Ag}$), biologically relevant (^{90}Sr), or have convenient gamma spectra for analysis purposes ($^{110\text{m}}\text{Ag}$ and ^{154}Eu). Here, ^{154}Eu may serve as an indicator for the behavior of other carbide forming lanthanides not readily measured by gamma spectroscopy.

During irradiation some fission products (e.g. $^{110\text{m}}\text{Ag}$, ^{154}Eu , and ^{90}Sr) were released through intact SiC layers into the OPyC and surrounding matrix. Some of what was released was retained in the OPyC and matrix at the end of the irradiation. The fraction of retained $^{110\text{m}}\text{Ag}$ varied from compact to compact, while ^{154}Eu and ^{90}Sr were almost completely held up in the carbonaceous material outside of the SiC layer.

The safety testing results reported by *Morris et al.* [39] indicate limited release of fission products for exposures at 1600°C and 1700°C. A time-dependent release of fission products ($^{110\text{m}}\text{Ag}$, ^{154}Eu , and ^{90}Sr) was noted, however, it was hypothesized that this release was due to fission products retained outside of the SiC layer. The initial release of $^{110\text{m}}\text{Ag}$ was rapid and was shown to typically occur during the heat-up and a few hours after reaching temperature with no

additional or limited release during the high-temperature soak. Slower release of ^{154}Eu and ^{90}Sr was also observed [39].

A limited number of margin tests were conducted at 1800°C. When the compacts were heated to 1800°C the initial releases were similar to the lower temperature safety testing. In these tests, an indication of release through intact SiC layers was observed in some cases. The time-dependent normalized inventory release rates from AGR-1 B, AGR-1 V1, and AGR-1 V3 compacts are shown in *Figure 10* and *Figure 11* for $^{110\text{m}}\text{Ag}$ and ^{154}Eu , respectively. The concentration of each species of interest in the compact and outside of the SiC layer varied depending on the irradiation conditions, so to simplify comparison between the four compacts in *Figure 10* and *Figure 11*, the magnitude of the inventory release rates were normalized to the release rates measured at approximately ~27–29 hours after reaching 1800°C. For the carbide formers of strontium and europium, the release from the matrix was over the entire test period. For $^{110\text{m}}\text{Ag}$, the fission products present outside of the SiC layer were liberated from the compact rapidly due to high diffusivity in the compact matrix, where the $^{110\text{m}}\text{Ag}$ release observed later in the test was less impacted by the $^{110\text{m}}\text{Ag}$ released during irradiation. For an in depth discussion of the safety testing PIE methodology refer to *Morris et al.* [39].

For the AGR-1 B and AGR-1 V1 compacts, the release rate of $^{110\text{m}}\text{Ag}$ (*Figure 10*) remained relatively constant and at a minimum rate after the initial burst of release observed during heat up, while for AGR-1 V3 compacts, the release rate of $^{110\text{m}}\text{Ag}$ reached a minimum rate after the initial burst of release, but then increased noticeably after an incubation period of ~70–100 hours. This observation indicates that additional $^{110\text{m}}\text{Ag}$ was being released through intact TRISO particles in AGR-1 V3 compacts and not AGR-1 B and AGR-1 V1 compacts. The release rate of ^{154}Eu from all four compacts (*Figure 11*) indicated an initial slow release from the

compact matrix, but a difference in release behavior was observed between the AGR-1 B and AGR-1 V1 compacts and the AGR-1 V3 compacts later in the test. The release rates for AGR-1 B and AGR-1 V1 compacts ultimately reduced over the course of the thermal exposure, suggesting depletion of the ^{154}Eu diffusion source in the matrix. In AGR-1 V3 compacts, the ^{154}Eu release rate increased after an ~140–155 hour incubation time, suggesting the source of europium in the matrix was increasing due to diffusion out of the SiC layer. The release behavior for ^{90}Sr was similar to ^{154}Eu and suggested simultaneous release of ^{90}Sr initially present outside of the SiC layer followed by release through intact SiC for AGR-1 V3 compacts after an incubation period [39].

Figure 10: Normalized inventory release rates of ^{110m}Ag from AGR-1 Compacts exposed to 1800°C for 300 hours, Compacts 4-4-1 and 4-3-2 were AGR-1 V3, while Compacts 3-2-3 and 5-1-3 were AGR-1 B and AGR-1 V1, respectively, and showed a difference in release behavior.

Figure 11: Normalized inventory release rates of ^{154}Eu from AGR-1 Compacts exposed to 1800°C for 300 hours, Compacts 4-4-1 and 4-3-2 were AGR-1 V3, while Compacts 3-2-3 and 5-1-3 were AGR-1 B and AGR-1 V1, respectively, and showed a difference in release behavior.

Further confirmation of release of $^{110\text{m}}\text{Ag}$ and ^{154}Eu through intact SiC in AGR-1 V3 was obtained from a longer exposure of individual particles at 1800°C for ~650 hours. Here the individual particle fission product inventories were tracked before and after thermal exposure, which clearly showed significant release of $^{110\text{m}}\text{Ag}$ and ^{154}Eu through intact SiC layers [40]. Note that in this loose particle test, the individual particle inventory of ^{90}Sr was not measured because the β -emission could not be measured from intact particles.

The only reported safety-tested compacts to indicate thermally driven release through the SiC layer were AGR-1 V3 compacts at 1800°C [39]. This observation confirms microstructure influences transport across the SiC layer and supports the expectation that the finer-grained microstructure of AGR-1 V3, which also contains the highest fraction of HAGBs, has a higher effective diffusivity relative to AGR-1 B and AGR-1 V1 under conditions where grain boundary diffusion is expected to dominate. This trend also reaffirms the expected trend on increasing magnitude of $\bar{D}$ based on finer grain size and higher fractions of HAGBs. These results also

suggests grain boundary diffusion may be responsible for the observed fission product release, notably ^{110m}Ag , in safety-tested compacts at 1800°C. Additionally, the comparison gives merit to the hypothesis that grain boundary diffusion is responsible for thermally driven silver release observed in historical out-of-pile release tests, which have been used to determine silver diffusion coefficients in SiC [9, 41, 42].

5. Summary

EBSD analysis was conducted on all constituent batches from the AGR-1 and AGR-2 irradiation campaigns. The ability to acquire statistically relevant microstructural data on the SiC layer of individual particles was demonstrated by utilizing a chemical/mechanical materialographic procedure to obtain high quality surfaces. The ability to qualify the observed microstructural data from EBSD analysis by comparison with micrographs from BSE channeling contrast was also demonstrated. Comparison with literature data corroborated the reported trends of the average SiC microstructure. Particle to particle analysis also identified the importance of comprehensive batch analysis to define the composite microstructure as noted by the batch discrepancy with AGR-1 B-45T and the importance of sampling. The analysis of the microstructure as a function of distance from the IPyC layer showed a variation in grain size and grain shape moving across the SiC layer for large-grained variants and an approximately constant microstructure for the fine-grained variants beyond the area adjacent to the IPyC/SiC interface. The analysis also highlighted the influence of user-defined parameters and artifacts in EBSD analysis; this influence was most notable when discussing the magnitude of the reported microstructural statistics of grain size and GBCD and should be accounted for when comparing literature data.

The description of the grain size by major and minor axis captured the SiC microstructure concerning the non-equiaxed grain shape and size of the SiC layer. A discussion of the grain size with twins and with no twins presented a comparison of the pathways for grain boundary dominated fission product transport, where the no twin microstructure may best represents the active GB diffusion microstructure. The difference in $\bar{D}$ expected from the unique microstructures in AGR-1 and AGR-2 variants demonstrated how microstructural tailoring is possible by changing processing conditions. Such tailoring may provide a microstructure which reduces the effective diffusivity of fission products in the SiC layer under an assumption of a GB diffusion dominated mechanism. Additionally, the difference in calculated $\bar{D}$ and the difference in fission product release behaviors for the fine-grain and larger-grain SiC microstructures at 1800°C suggests GB diffusion can become significant at elevated temperatures.

This data set presented in this paper provides insight on the variations in the as-fabricated SiC layer of AGR-1 and AGR-2 TRISO fuel particles. It highlights the processing conditions impact on the SiC microstructure and it provides fundamental microstructural insights necessary to understand the complex behavior of TRISO fuel in reactor. Comparison of the unique microstructures from the individual constituent batches with further study of the fission product release behavior in TRISO particles from the AGR-1 and AGR-2 irradiation experiments, notably ^{110m}Ag , may illuminate the responsible release mechanism and opportunities to improve TRISO fuel performance.

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