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Effect of Neutron Irradiation Damage on Fission Product Transport in the SiC Layer of TRISO Fuel Particles

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Abstract – *Metallic fission products are found to transport through the SiC layer of the tristructural isotropic (TRISO) fuel particle and various studies have been undertaken to understand the transport behavior. It has been recently discovered that the precipitation of intragranular palladium-containing fission products in the SiC layer of neutron-irradiated TRISO fuel particles occurs via a novel dual-step nucleation mechanism. Direct observations of Pd silicide imprinting into morphological templates of α-SiC precipitates in neutron-irradiated SiC layer of TRISO fuel will be discussed, with examples from selected particles from both the Advanced Gas Reactor (AGR)-1 (fabricated laboratory-scale at Oak Ridge National Laboratory) and AGR-2 (fabricated at pilot scale by an industrial vendor) experiments. As neutron-irradiation damage structures are expected to accelerate the intragranular diffusion kinetics of fission products, their size, shape, density and distribution patterns across the radial thickness of the SiC layer can be directly correlated with the nature of intragranular transport pathways of fission products. In this work, the distinction of neutron-irradiation damage structures as a function of fuel type, burnup level, and fabrication histories of TRISO coated particles from the AGR-1 and AGR-2 experiments has been studied. The five particles investigated, three AGR-1 and two AGR-2 particles, represent irradiation histories of four fuel types with burnup levels varying from 12.55 to 17.4% fissions per initial metal atom. In this study it was found that (1) voids and dislocation loops appear to segregate to intragranular α-SiC and Pd silicide precipitates, (2) an inverse relation of void size and density for AGR-1 and AGR-2 experiments; and (3) Ag-110m retention in SiC layer appears to have an inverse relation with void sizes.*

I. INTRODUCTION

Tri-isotropic (TRISO) coated nuclear fuel particle is a part of the Advanced Gas Reactor (AGR) Fuel development program that seeks to develop a safe and reliable nuclear fuel concept for the Very High Temperature Gas Cooled Reactor (VHTR). TRISO fuel kernels are spherical in shape, ~350-425 µm in diameter, and composed of a mixture of uranium oxide and uranium, carbide with a nominal stoichiometry consisting of $UO_{1.37}C_{0.35}$.3. The fuel kernel is surrounded by various structural layers deposited by fluidized bed chemical vapor deposition [1] [2]. The structural layers consist of a buffer (porous carbon layer), inner pyrolytic carbon (IPyC) layer, polycrystalline silicon carbide layer (SiC), and the outer pyrolytic carbon layer which is a dense and outermost carbon layer. The aforementioned fuel particles are dispersed within a graphite matrix in spheres or a prismatic compact as part of the fuel concept.

SiC was downselected as a structural material due to its enhanced neutron tolerance at high doses, strength retention, and chemical inertness at elevated temperatures [3]. During neutron irradiation on SiC, some of the dominant microstructural defects encountered are Frank loops [3], dominating at ~1,100°C until dislocation interactions become significant [4-5]. In other works, cavities have also been observed to form at grown-in stacking faults and were generally spherical [5-6]. Kondo reported both spherical and polygonal shaped (~triangular) cavities observed at neutron densities of 1.9×10^{25} n/m^2. Additionally, a preferential formation of smaller voids occurring at stacking faults was also observed in most specimens, indicating the preferential cavities nucleation at, or very close to, the stacking faults [5].

Black spot defects (BSD) were widely observed as areas of dark transmission electron microscopy (TEM) contrast, contributing to the reminder of the swelling on the SiC matrix [7]. Senor et al. [8] and Katoh et al. [4] reported black dots as a mix of small dislocation loops. BSDs were mostly circular or slightly oval exhibiting dark-contrast arising from lattice strains.

Neutron-irradiation damage was simulated by means of ion irradiation (Kr and C ions) on 4H-SiC by 3.15 MeV C^{2+} up to 0.4 dpa and 1 MeV Kr+ up to 0.4 and 0.8 dpa. For Kr ions and a constant ion irradiation damage level, the defect diameter increases consistently as a function of irradiation temperature. At 0.4 dpa, the number density remains constant but at 0.8 dpa, number density increases. Conversely, with increasing temperature, the average BSD size in C-irradiated samples was reported as unchanged while the number density drastically decreased. Additionally, it has been recently discovered that the irradiation induced accelerated diffusion in β-SiC leads to nanoscale α-SiC precipitation at structural defects such as stacking faults and Frank loops [9-10]. The α-SiC precipitate eventually serve as a template for reaction with fission products such as Pd [9]. Hence, the study of defect characteristics is vital to understand comprehensively the intragranular fission product transport in SiC.

This study, which spans both AGR-1 and AGR-2 experiments, focuses on defect structure analysis using TEM. The aim of this study is to understand the influence of various neutron irradiation parameters as well as structural integrity (i.e., buffer layer, IPyC etc.) of TRISO particles on microscopically observed defect characteristics in the SiC layer.

II. EXPERIMENT, SAMPLE DESCRIPTION AND METHODS

Post-irradiation TEM was carried out on TRISO Particles AGR1-131-066, AGR1-433-001, AGR1-532-SP01 belonging to AGR-1 experiment along with Particles AGR2-222-RS36 and AGR2-223-RS06 of the AGR-2 experiment. The irradiation conditions for these two programs are reported in Table 1. Samples for TEM were prepared by a dual-beam Quanta 3D focused ion beam instrument. STEM and conventional TEM were carried out on an FEI Tecnai F30 microscope operated at 300 kV. Gatan digital micrograph software was used for post-processing of TEM data.

Table 1: Neutron irradiation parameter of AGR-1 and AGR-2 experiments.

Compact	Burn up (% FIMA)	Fast Neutron Fluence (x 10^{25} n/m^2) E>0.18 MeV	Time-Average Volume-Average Temperature (°C)	Time-Average Peak Temperature (°C)	Ag Retention
AGR1-433-001*	18.6	4.16	1094	1179	66
AGR1-131-066	15.3	3.22	1092	1166	39
AGR1-523-SP01	17.4	3.77	1059	1141	16
AGR2-223-RS006	12.68	3.46	1296	1360	8
AGR2-222-RS036*	12.55	3.39	1287	1354	Not detectable

* Safety Tested -1600°C, 300 hrs

TEM image analyses were carried out to quantify the areas damaged by neutron-irradiation exposure. A total of 63 TEM micrographs were employed during the analyses. The micrographs were strategically selected along the SiC layer of the fuel. The different locations include an inner area, which is closer to the fuel kernel, a center area, and an outer area which is adjacent to the outer pyrolytic graphite layer in the sample.

TEM imaging was carried out either in overfocus or underfocus bright field conditions for better visibility of the defect structures. A total area of 3×10^6 nm^2 was analyzed with the image software ImageJ to quantify the damaged areas and provide relevant data regarding size (diameter), area fraction, and damaged area density in all samples. Due to the nature of circular shapes in the damaged areas (mostly voids or gas bubbles), the areas of interest were precisely selected in circular segments. The average diameter was calculated and compared among all samples to find trends on the area "size" at the different irradiation temperatures among the different samples.

Additionally, the void density was calculated by dividing the number of voids present per unit area (# void/nm^2) to understand the different concentrations of damage among the different sections of the SiC layer. The collected data were analyzed and tabulated in graphs to provide a more concise interpretation of the effects of neutron damage along the different sections of the SiC layer in the fuel.

III. RESULTS

III.A. Nature of irradiation-induced defects

In an unirradiated SiC-layer specimen [9] of TRISO fuel, no cavity, dislocation loops, or precipitates were observed. Fig.1 shows the preferential formation of cavities at stacking faults Fig. 2 shows a compilation of TEM images showing different types of irradiation-induced defects observed in the SiC layer in a range of TRISO fuel particles subjected to different irradiation condition. Fig. 2(a) shows very small cavities, with size about 1–2 nm with no specific crystallographic orientations in AGR1-433-001 particle. These have been referred as "black spots" in this study. Fig. 2(b) shows neutron irradiation induced cavities that range between 4 and 7 nm in size in all TRISO particles with strong crystallographic orientation in the cubic SiC matrix. The details of the orientation of these cavities are explained later. Apart from cavities, Fig. 2(c) shows the formation of Frank loops in SiC.. It is also observed the number density of cavities at faulted regions is high while their size is smaller when compared to that of cavities in the matrix.

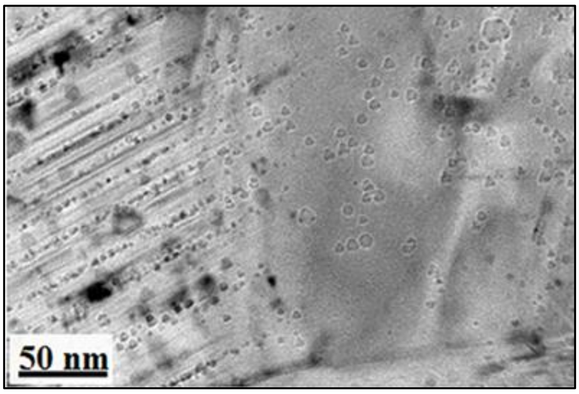

Fig. 1: A TEM image shows preferential high rate of nucleation of void at stacking faults compared to that in the uniform β-SiC matrix.

One objective of this study is to determine the nature of cavities at varied radial distances from the fuel kernel. Fig. 3(a) shows two locations (A and B) of Particle AGR2-223-RS006 chosen for study of defect sizes. The buffer layer adjacent to location A is intact while it is broken close to location B. Fig. 3(b) shows the comparison of average defect sizes (only polygonal cavities) across the SiC layer at three different radial distances from the kernel, referred as inner, center, and outer, for location A and B of Particle AGR2-223-RS036. It was observed that the defect size at the inner layer of location B is significantly larger when compared with that of location A. One of the possible reasons of variation of neutron damage can be the grain orientations of SiC at inner layers. Certain grains are possibly oriented to be more susceptible to neutron capture and thus damage. Further investigations are required to confirm the reason of variation of void sizes at different spots with same radial distance from kernel.

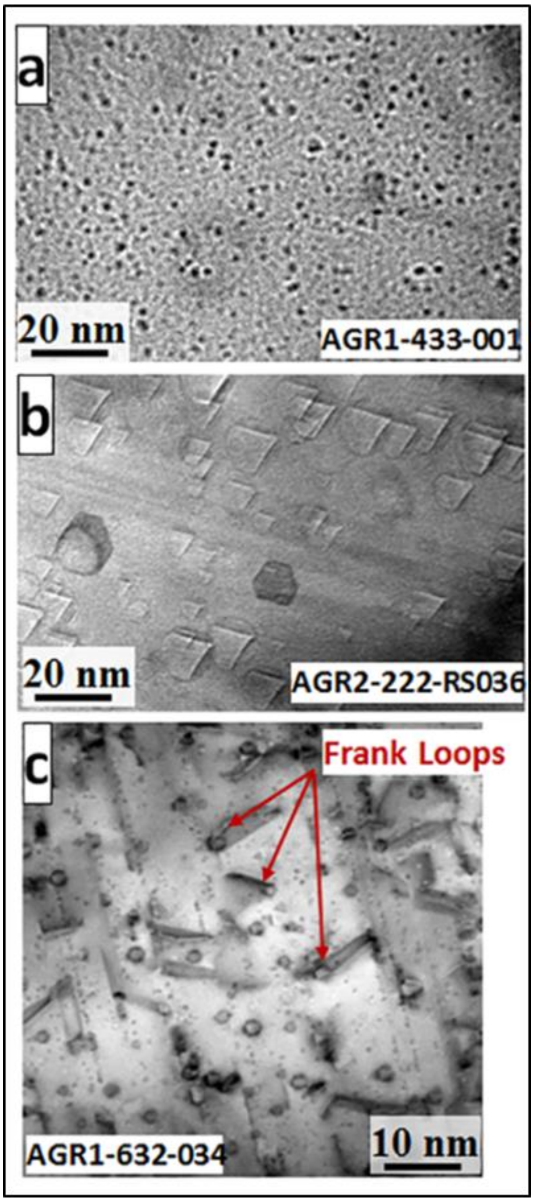

Fig. 2: TEM images show different types of neutron-irradiation–induced defects in β-SiC layer of TRISO coated fuel particles. (a) Cavities ranging from 1 to 2 nm in size with no crystallographic orientation, as found in particle AGR1-433-001, are referred as black spots. (b) Voids of 4–7 nm in size with sharp edges were observed in all particles except AGR1-433-001. (c) Irradiation-induced Frank loops are observed in STEM imaging. These dislocation loops often act as nucleation sites for α-SiC and, subsequently, Pd silicide as indicated by red lines.

Similar analysis was carried out for Particle AGR2-222-RS036, where two locations on SiC layer, namely A and B, were analyzed at three different radial distances (inner, center and outer). The locations A and B are nearly identical; however, there is a slight breakage in the buffer layer adjacent to location A is observed, as seen in Fig. 3(c). But, there was no apparent difference in defect size for location A and B in this safety tested particle, as shown in Fig. 3(d).

III.B. Neutron damage analysis

Except for Particle AGR1-433-001, all TRISO fuel particles evaluated show polygonal cavities. The representative nature of these polygonal cavities across the width of the SiC layer has been shown in Fig. 4 for Particle AGR2-222-RS036. While the defect size ranges between 6 and 7 nm (Fig. 4[d]), the nature of the defect has been determined by electron diffraction pattern along <011> direction of cubic β-SiC. Two sides of the triangular voids are parallel to $\{\bar{1}11\}$ and $\{\bar{1}1\bar{1}\}$ planes, respectively, while the other side is parallel to $<\bar{2}20>$ direction. A similar defect characteristics had been observed earlier by Katoh et al. [3].

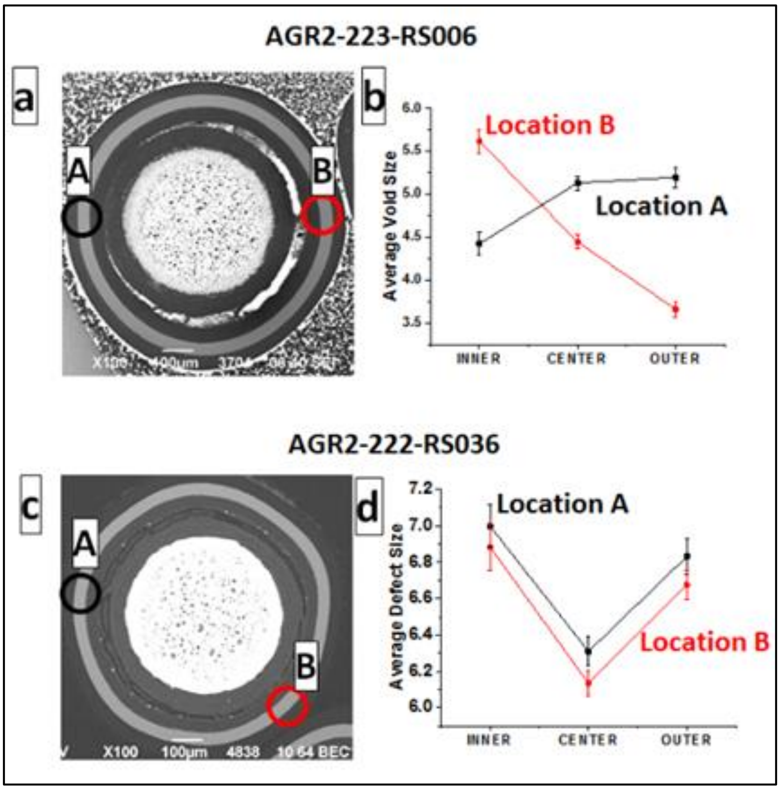

Fig. 3: The variation of void size along radial distance of SiC layer has been studied for two AGR-2 particles. (a-b) The analysis on two locations (A and B) by TEM shows larger void at inner region size at location B where there is a breakage in the buffer region. (c-d) Similar analysis on two locations on Particle AGR2-222-RS036 does not any apparent difference in void size at inner regions of location A and B.

Neutron-irradiation void characteristics for AGR-1 and 2 experiments are tabulated in Table 1, and although further interpretation is in progress, it is preliminarily observed that the void fraction for the two AGR-2 particles is larger than those of the three AGR-1 experiment particles. One notable difference between the AGR-1 and AGR-2 particle irradiation history is that the time-average peak temperatures for the AGR-2 particles are approximately 200°C higher than those for the AGR-1 particles. However, the actual irradiation temperature of the particles are probably ±150°C of the TAVA.

Table 2: Neutron-irradiation void characteristics for AGR-1 and AGR-2 experiments.

Compact	Void size (nm)	Void fraction (%)	Void number Density (voids/nm^3)
AGR1-433-001	2.12	8.42	0.023
AGR1-131-066	2.89	8.06	0.011
AGR1-523-SP01	4.03	7.44	0.005
AGR2-223-RS006	4.91	10.05	0.005
AGR2-222-RS036	6.71	11.70	0.003

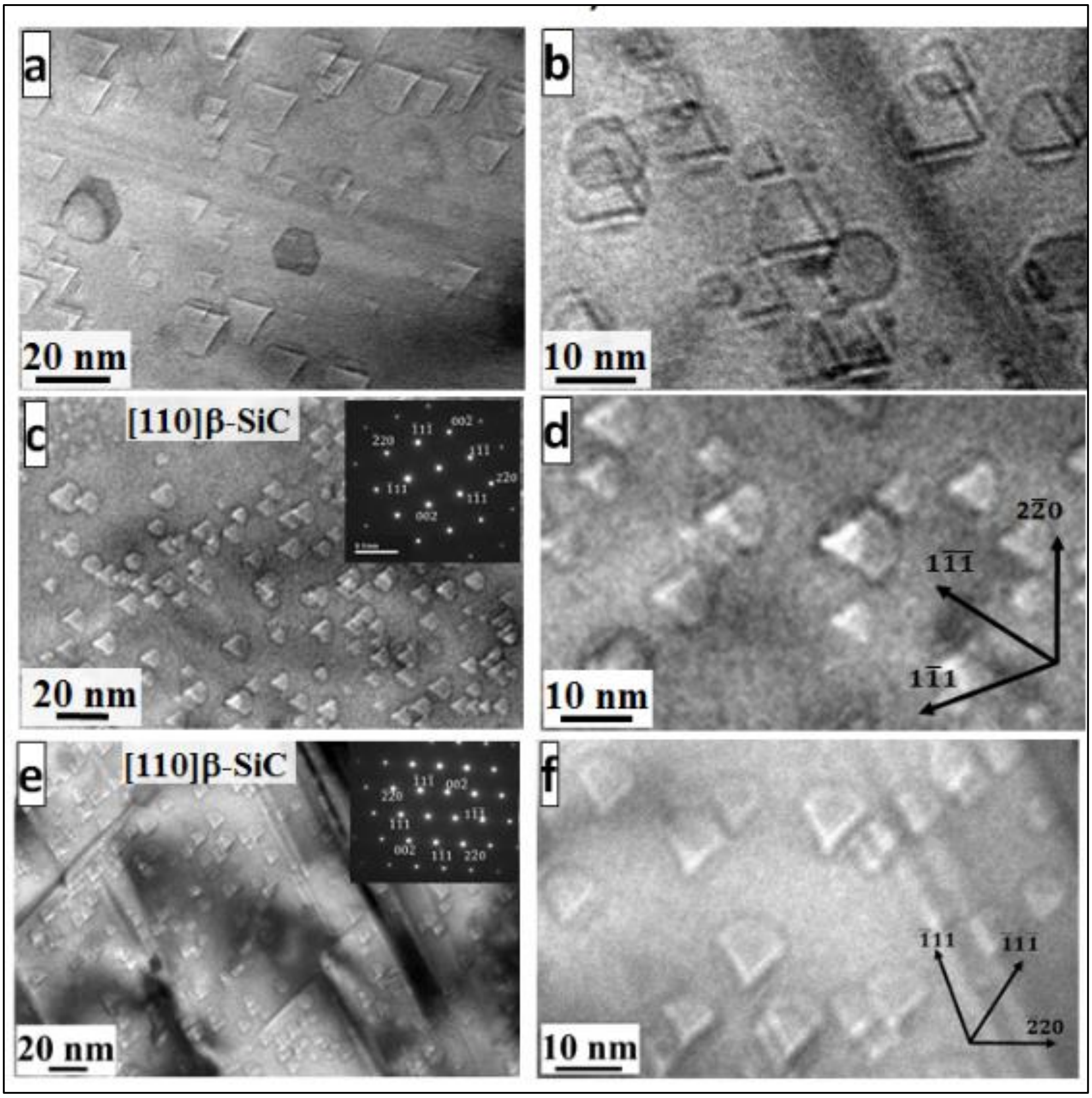

Fig. 4: The nature of the voids with sharp edges has been analyzed along the radial width of the SiC and is shown for the particle AGR2-222-RS036, location B for the inner (a, b), center (c, d) and outer (e, f) regions.

III.C. Neutron damage and intragranular precipitate relation

In both the AGR-1 and AGR-2 experiments, intragranular α-SiC and Pd silicide precipitates were observed [9]. In all of the particles analyzed in this study, the voids are not associated with these precipitates. Fig. 5(a) shows such a case in Particle AGR2-223-RS036, where the Pd precipitate is not associated with any void. But in the safety-tested Particle AGR2-222-RS036 (where the specimen was post-heat-treated at 1600°C for 300 hours), it was repeatedly observed that the voids are aligned in crystallographic fashion at the edges of the precipitates, as shown in Fig. 5(b-d). Because the heterogeneous nucleation of these intragranular precipitates usually occurs at stacking faults or Frank loops, it is highly unlike that the defects are energetically high enough to facilitate nucleation of these precipitates. Hence, the microscopically observed segregation of voids and loops to precipitate edge only in the safety tested condition is probably to reduce the energy associated with defects.

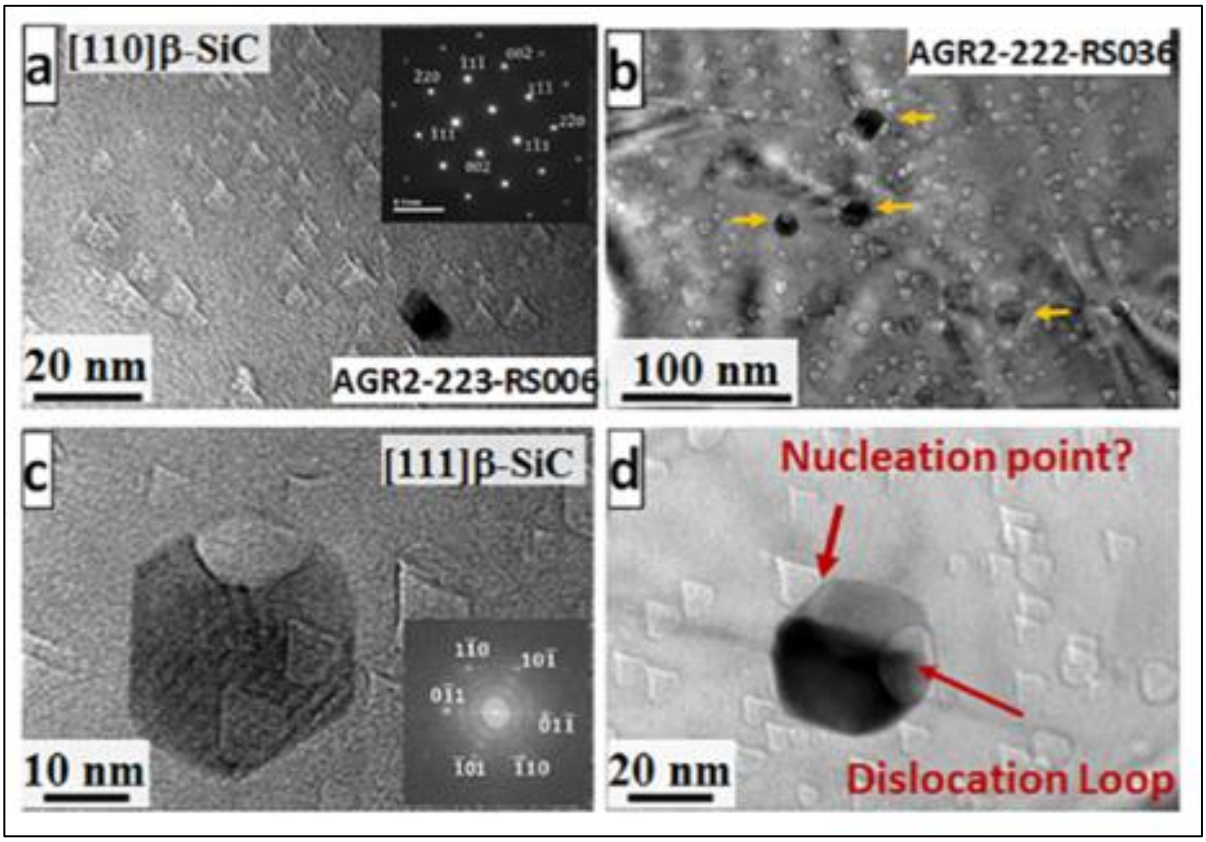

Fig. 5: A comparison of intragranular α-SiC and Pd silicide precipitates relation to irradiation-induced voids was carried out for both the safety-tested and non-safety tested conditions. (a) A TEM image reveals no apparent correlation of defects with nanoscale precipitates in non-safety tested Particle AGR2-223-RS006. (b - d) TEM images shows that dislocation loops and voids are associated with precipitates in the safety-tested condition of Particle AGR2-222-RS036.

III.D. Effect of temperature, Ag retention, and burnup

Fig. 6 is a radar diagram to analyze the possible factors that can influence the nature of irradiation-induced cavities for both AGR-1 and 2 experiments. Table 1 and 2 report irradiation-history and void-characteristic data that are used to create this radar diagram. From both Fig. 6 and Table 2, it was unambiguously observed that the defect size is inversely proportional to defect density. The possible reason may be defect coalescence at high temperatures reduces defect density. While other factors such as irradiation temperature, neutron fluence, and burnup level can influence defect sizes, no straightforward correlation was found among the five analyzed particles.

Interestingly, Ag-110m retention and void size has an inverse relation, as shown in Fig. 7. This indicates that the void size has a possible effect on enhancing Ag release through SiC layer.

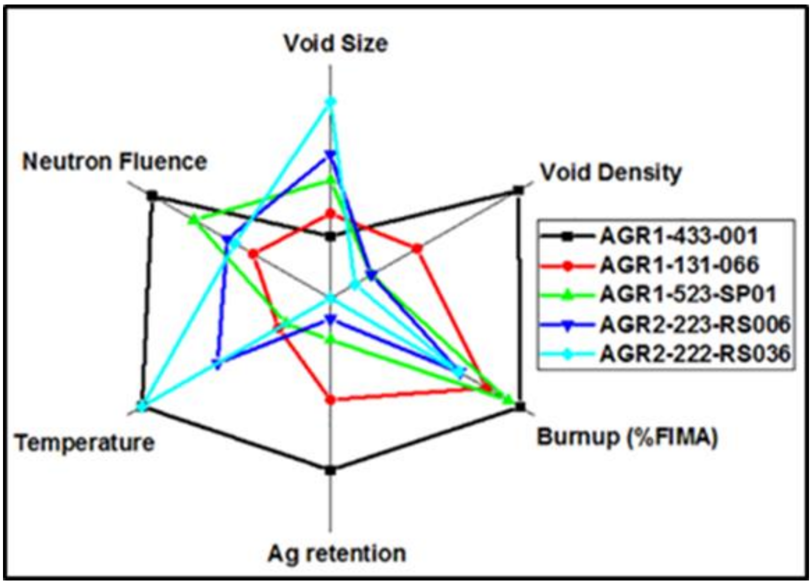

Fig. 6: A radar diagram that shows the relation of various parameters on void size and density.

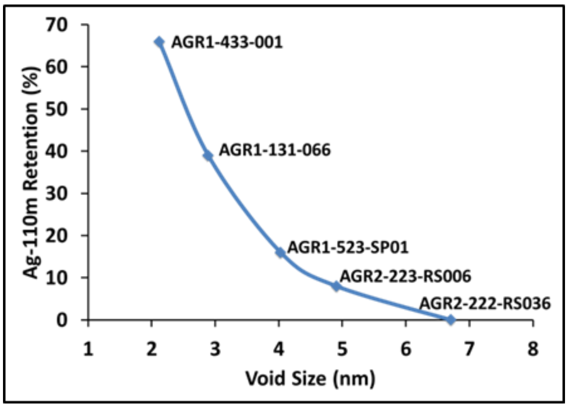

Fig. 7: A plot of Ag-110m retention in percent against the void sizes in various AGR-1 and AGR-2 experiments.

III.E. Defect density and irradiation temperature

Although the time- and volume-averaged, and time-average peak temperature of the compacts containing the TRISO-coated fuel particles to be studied is known (Table 1), the actual irradiation history of individual TRISO particles can deviate significantly from the average compact values. However, the void characteristics can give indication of irradiation-temperature experience of the individual particle. Analysis of two AGR-2 particles in Fig. 6, where the Ag retention, burnup, and neutron fluence are quite similar, shows the defect size is higher while defect density is lower in case of particles subjected to higher temperature (AGR2-222-RS036).

III.F. Comparative relationships between AGR-1 and AGR-2 particles

Excluding the particle subjected to safety tested conditions (AGR1-433-001 and AGR2-222-RS036), the time-average peak temperature for AGR-2 experiment was determined to be higher by nearly 200°C. From the Fig. 6, it can be deduced that the AGR-2 particle has slightly higher void size when compared with that in other two AGR-1 experiments.

IV. CONCLUSIONS

The present study of neutron-irradiation induced defects that spans AGR-1 and AGR-2 experiments, reports the following conclusions:

1) Neutron irradiation can produce black spots, polygonal voids, and Frank loops. Frank loops have been observed to act as a nucleation site for intragranular α-SiC and Pd silicide precipitates.
2) Void distribution has been found to be non-uniform. A high concentration of smaller voids are found at stacking faults compared to those in the β-SiC matrix.
3) Voids and dislocation loops in one safety-tested AGR-2 particle, appear to segregate to intragranular α-SiC and Pd silicide precipitates.
4) An inverse relation of void size and density was unambiguously found for all particles examined.
5) The Ag-110m retention in SiC layer appears to have an inverse relation with void sizes. But isolated influence of neutron fluence, burnup level on void size or density was not observed.

This work will continue to analyze further the Frank loop presence and distributions, to potentially provide an indication of actual irradiation level and temperatures.

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