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Fabrication and characterization of joined silicon carbide cylindrical components for nuclear applications

H.E. Khalifa [*], C.P. Deck, O. Gutierrez, G.M. Jacobsen, C.A. Back

General Atomics, San Diego, CA, USA

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ABSTRACT

The use of silicon carbide (SiC) composites as structural materials in nuclear applications necessitates the development of a viable joining method. One critical application for nuclear-grade joining is the sealing of fuel within a cylindrical cladding. This paper demonstrates cylindrical joint feasibility using a low activation nuclear-grade joint material comprised entirely of β-SiC. While many papers have considered joining material, this paper takes into consideration the joint geometry and component form factor, as well as the material performance. Work focused specifically on characterizing the strength and permeability performance of joints between cylindrical SiC–SiC composites and monolithic SiC endplugs. The effects of environment and neutron irradiation were not evaluated in this study. Joint test specimens of different geometries were evaluated in their as-fabricated state, as well as after being subjected to thermal cycling and partial mechanical loading. A butted scarf geometry supplied the best combination of high strength and low permeability. A leak rate performance of 2×10^{-9} mbar l s^{-1} was maintained after thermal cycling and partial mechanical loading and sustained applied force of 3.4 kN, or an apparent strength of 77 MPa. This work shows that a cylindrical SiC–SiC composite tube sealed with a butted scarf endplug provides out-of-pile strength and permeability performance that meets light water reactor design requirements.

1. Introduction

Silicon carbide (SiC) and silicon carbide fiber-reinforced, silicon carbide matrix (SiC–SiC) composites exhibit desirable properties for use as structural components in accident tolerant and advanced nuclear reactor concepts. Among these properties are the retention of strength at high temperature, extremely low reaction rate in high temperature steam associated with a loss of coolant accident (LOCA), and excellent stability under neutron irradiation [1–6]. However, the deployment of silicon carbide and SiC–SiC composites in advanced reactor designs necessitates the development of a joining method and joint material to form the complex functional sections of the reactor core. The measurement of joint strength in ceramic systems has received significant attention due to the dearth of established techniques for characterizing these ceramic–ceramic joint systems. Ongoing efforts to develop a suitable joint described in the literature have focused primarily on development and evaluation of the joint material [7–11]. In those studies, the torsion test or lap shear test was used to characterize shear strength due to the pure shear strain maintained during the test [12–14].

Shear testing is critical for assessing the performance of the joint material, however the shear strength of the joint material alone is not enough to identify a suitable joining technique. For nuclear applications, additional required properties of the joint assembly include: resistance to irradiation damage, chemical compatibility with the fuel and coolant systems, sufficient mechanical strength, low permeability, and minimal differential thermal and irradiation-induced dimensional change between the joint material and joint components. When measuring these properties, the performance of the entire assembly comprised of the joint material, the components being joined, and the configuration of the joint interface, must be characterized. Taking the Energy Multiplier Module (EM^2) gas-cooled fast reactor as an example, its 30-year lifetime requires both fully ceramic and compositionally-pure SiC–SiC composites and SiC materials for the cladding and joint materials to satisfy irradiation resistance and chemical compatibility requirements [15]. To survive handling, retain fission gases and operate under the high temperatures for EM^2 or any other reactor, it is necessary to meet the mechanical strength, permeability, and dimensional change requirements through engineering and process control.

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[*] Corresponding author. Tel.: +1 858 455 2027.
E-mail address: hesham.khalifa@ga.com (H.E. Khalifa).

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The work reported in this paper characterizes joined tubular geometries, the typical geometry of nuclear fuel cladding. Joints were formed by hybrid preceramic polymer/chemical vapor infiltration to bond a cylindrical SiC–SiC cladding tube with an endplug. Different candidate geometries were fabricated and the mechanical and permeability performance of the joined assemblies was evaluated. To obtain measurements in a cylindrical geometry, customized test rigs were designed and fabricated. In particular, an endplug pushout test was developed and implemented. The paper describes the role played by different geometric joint features and provides a summary as to which of these are most likely to succeed in providing the requisite strength and permeability performance needed for nuclear applications.

2. Experimental

In this study, different joint specimen types were made and characterized iteratively to determine critical factors affecting joint performance. Since tubular geometry test methods are not established in the literature, planar joint test specimens were first examined to establish relationships between different joint geometries via planar flexural testing on monolithic SiC materials using a well-understood test method. Then, candidate tubular joint specimens were fabricated with monolithic SiC tubes, and ultimately with SiC–SiC composite tubes. Mechanical and permeability testing was utilized to evaluate the tubular joint specimens. Thus, the paper discusses the types of specimens and the testing through planar specimens, monolithic tube joint specimens, and composite tube joint test specimens in sequence, with the goal of demonstrating robust performance of joined cylindrical SiC–SiC specimens.

2.1. Joint test specimens

Current nuclear fuel cladding typically has a 9.5 mm outer diameter with a 0.6 mm wall thickness. Any alternate claddings will need to remain thin-walled to allow heat to be efficiently transferred. This constraint is the dominant factor affecting joint design for strength and ease of manufacturing. Based on existing literature on fiber-reinforced polymer composites, planar scarf and double-butted lap (DBL) joint geometries were expected to perform well because they both deliver improved tensile and shear reinforcement, as well as increased surface area compared to a pure lap or butt joint [16,17]. When implemented in cylindrical geometries, the DBL joint entails a simpler setup for joint surface preparation because grinding is parallel or perpendicular to the component surface while the scarf joint adds the complexity of an angle tolerance. For these reasons, both double butted lap and scarf joints were examined in planar studies and refined for the cylindrical studies.

All joint test specimens were comprised of monolithic SiC and/or SiC–SiC composite components bonded with a high purity β-SiC joint material. A detailed description of the joint material and processing can be found in [18]. The joint was fabricated using a hybrid preceramic polymer, chemical vapor infiltration (CVI) joining approach. The polycarbosilane-based preceramic polymer[1] slurry is goes through a 3 step process that includes a low temperature cure, followed by pyrolysis and subsequent heat treatment at temperatures in excess of 1300 °C to refine and homogenize the grain structure. The specimen is subsequently infiltrated via CVI to densify the joint layer. The joint material was developed with a priority on maintaining compositional and microstructural homogeneity between the joint and surrounding components. Testing shows the joint material achieves uniform grain size and purity comparable to that of the constituent SiC composite fibers. The composition of the joint material and compilation of existing data on SiC material suggest that this joining approach should maintain performance to temperatures in excess of 1000 °C. Full results together with irradiation results will be described in a subsequent report, however, most importantly for this study, the out-of-pile strength was measured by a well-established torsion test method at Oak Ridge National Laboratory [8,12]. Using CVD β-SiC substrates,[2] the torsion testing measured the characteristic shear strength of this joint material to be 78 ± 14 MPa. Test specimens were the standard 6 mm × 6 mm × 3 mm square torsion hourglass type with a .4 mm notch radius. All 15 of the test specimens failed by apparent shear within the joint layer region. This hybrid joining approach was used in all joint test specimens.

A critical factor affecting joint strength is the interfacial surface area. To estimate the area needed for a robust nuclear fuel cladding joint, calculations were based on the nominal internal pressure of 15.5 MPa for a PWR fuel rod at the end of life and the experimentally measured shear strength of the joint material used in this study. This case is used because the fuel rod must retain fission gases, and thus is more demanding than other fast reactor designs where fuel may be vented. For the 10.5 mm OD, 7.5 mm ID SiC tubing used in this study and the addition of a relatively conservative safety margin of 5, a minimum endplug joint length of 5 mm is calculated for the pure lap geometry. For an alternate testing length, 10 mm was also selected for added safety margin. These linear joint lengths provided the starting point for the planar flexural testing.

All of the planar joint test specimens were prepared with monolithic direct-sintered α-SiC[3] with joints containing double butted lap or scarf features and subjected to flexural testing. The bond lengths were 5 and 10 mm long. Specimens were cut with a high speed diamond saw. The joining surfaces were prepared via polishing or lapping assisted by diamond suspension such that a RMS surface roughness less than 1 μm was achieved. For the two geometries, DBL and scarf, this resulted in an evaluation of four joining conditions.

The joint for all cylindrical specimens was made between the tube and a monolithic SiC endplug. The endplug sealing approach, where a subcomponent plug inserts into the mating tube, is preferred over an endcap that slides over the outside of the tube because it allows existing LWR fuel rod dimensions to remain unchanged. The SiC endplugs were fabricated via typical transient eutectic phase (TEP) processing in a hot press. The precursor compositions and processing parameters used are described elsewhere in [19–21]. The TEP precursor consisted of SiC, Al_2O_3, Y_2O_3, and SiO_2 nanopowders[4] that were ball-milled to ensure homogeneity. TEP consolidation takes advantage of the increased reactivity resulting from the high specific surface area of nanopowders to achieve rapid densification at >1500 °C temperatures under applied pressure. TEP has the added benefit of near-net to shape processing such that different endplug geometries can be fabricated directly with appropriate hot press tooling. The primary joint geometries evaluated were the butt, butted lap, scarf, and butted scarf. Renderings of the different endplug geometries are shown in Fig. 1.

The cylindrical joint test specimens used either a monolithic SiC tube[5] or a SiC–SiC composite tube. Monolithic cylindrical joint test

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[1] Starfire Systems, 2165 Technology Dr, Schenectady, NY 12308, SMP-10 polycarbosilane, batch # 20867, 2013.

[2] Morgan Technical Ceramics, 4 Park Avenue, Hudson, NH 03051, CVD SiC plate, vintage 2013.

[3] Ortech, Incorporated, 6720 Folsom Blvd., Suite 219, Sacramento, CA 95819, SSiC sintered silicon carbide plate, vintage 2012.

[4] US Research Nano Materials, 3302 Twig Leaf Lane, Houston, TX, 77084, β-SiC 45–65 nm, US2028, α-Al_2O_3, 80 nm US3008, Y_2O_3 30–45 nm, US3551, SiO_2 20–30 nm US3438.

[5] Saint Gobain Ceramic Materials, 23 Acheson Drive, Niagara Falls, NY 14303, Hexoloy SE tube, vintage 2012.