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Radiation stable, hybrid, chemical vapor infiltration/preceramic polymer joining of silicon carbide components☆

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ABSTRACT

This paper reports on a nuclear-grade joining material for bonding of silicon carbide-based components. The joint material is fabricated via a hybrid preceramic polymer, chemical vapor infiltration process. The joint is comprised entirely of β-SiC and results in excellent mechanical and permeability performance. The joint strength, composition, and microstructure have been characterized before and after irradiation to 4.5 dpa at 730 °C in the High Flux Isotope Reactor. The hybrid preceramic polymer-chemical vapor infiltrated joint exhibited complete retention of shear strength and no evidence of microstructural evolution or damage was detected following irradiation.

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 nuclear reactors [1,2]. The Department of Energy's Accident Tolerant Fuels (ATF) and Advanced Reactor Technologies (ART) programs aim to utilize materials like SiC–SiC composite to improve reactor safety, performance and fuel utilization. Enhanced reactor performance and fuel utilization can be achieved by increasing burnup and fuel cycle length. However, achieving these goals with SiC–SiC necessitates development of suitable joining materials that can withstand the higher fluences anticipated in advanced light water reactors (ATF) and high temperature fast reactors like the Energy Multiplier Module (EM^2) [3]. Furthermore, SiC joining technology is expected to enable the fabrication of more complex internal core components such as fuel rod supports structures [4].

Previous work in the area of radiation tolerant joining has generated several joining materials with promising unirradiated performance, including transient eutectic phase (TEP) sintered SiC, glass ceramic, Ti–Si–C MAX phase, and metal diffusion bonding [4–8]. While these approaches are promising for lower fluence applications such as LWR fuel cycles, strength degradation or increased data scatter were observed in some of these joint materials following irradiation to 5 dpa at 800 °C [9]. For instance, Ti-diffusion bonding and Ti–Si–C MAX phase materials are comprised largely of titanium which has a high neutron cross section and coefficient of thermal expansion (CTE) nearly 3 times that of SiC. TEP processed joints deliver extremely high strength and are predominately SiC, but moderate strength degradation of monolithic TEP SiC reported at 830 °C to 5.9 dpa was attributed to differential neutron-induced swelling of oxide phases [10]. While the neutron induced defects in stoichiometric SiC undergo an annealing process at elevated temperatures [11], any compositional inhomogeneities or dissimilar materials within the joint may lead

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to premature failure in longer fuel cycle advanced reactor applications. For this reason a high purity SiC joining material is desirable.

This paper describes the irradiated performance of a nuclear-grade joint material for SiC and SiC–SiC composites. It is the first nuclear-grade joint comprised entirely of β-SiC and designed specifically to withstand higher temperature and fuel burnup applications such as EM^2. The β-SiC joint is created in a hybrid process involving preceramic polymer and chemical vapor infiltration-derived SiC [12]. Previous out-of-pile work has shown that this joining approach can successfully be applied to cylindrical joint geometries. In addition, testing shows the joint material is impermeable and provides sufficient mechanical strength for anticipated ATF and advanced reactor needs [13]. Features of the joining approach will be described and the effect of irradiation at 730 °C to 4.5 dpa on microstructure and torsional shear strength will be presented.

2. Experimental

2.1. Joint specimen preparation

The shear strength of the joint material was evaluated using hourglass-type torsion specimens [14]. Torsion joint specimens were prepared with chemical vapor deposited β-SiC substrates (Morgan Advanced Materials, Hudson, New Hampshire). Specimens have approximate dimensions of 6 mm × 6 mm × 3 mm, with a 0.4 mm notch radius cut to a depth of 0.5 mm such that the joint surface area is a circle with 2.5 mm radius at the mid plane of the specimen. Substrates were cut with a high speed diamond saw. The joining surfaces were prepared via polishing or lapping assisted by 1 μm diamond suspension. All parts were sonicated in isopropanol and dried prior to joining. An example hourglass torsion joint specimen is shown in Fig. 1.

The hybrid preceramic polymer, chemical vapor infiltrated (GA-HSiC) joints were made via application of a SMP-10 allylhydridopolycarbosilane (Starfire Systems Inc, Glenville, New York) preceramic polymer slurry loaded with SiC whiskers (Advanced Composite Materials, Greer, South Carolina) in a 1:1 mass ratio. The slurry was mixed in a Thinky ARV 310 orbital centrifuge. Polymer slurry was applied between two torsion substrates and then cured, pyrolized, and subsequently heat treated at a maximum temperature greater than 1500 °C for 120 min in Argon atmosphere to convert to β-SiC. A ramp rate of 2 °C/min is used below 400 °C. Above 400 °C, a ramp rate of 10 °C/min is used. Following preceramic polymer conversion to SiC, the specimens were further densified via the decomposition reaction of methyltrichlorosilane and hydrogen with an argon purge in a chemical vapor infiltration (CVI) process. Additional details of the joining process are described in Ref. [12].

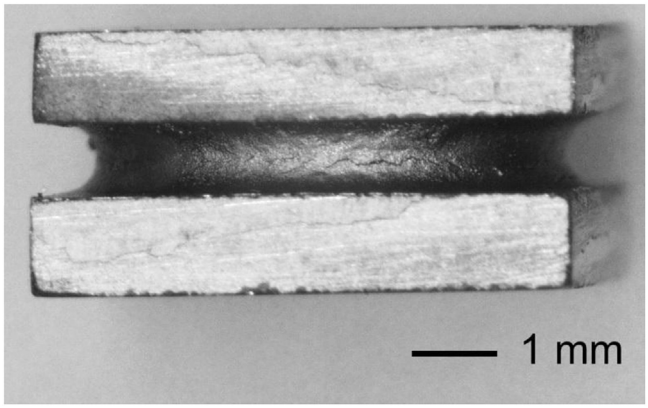

Fig. 1. Appearance of typical hourglass torsion joint specimen.

2.2. Neutron irradiation

Irradiation was carried out in the target irradiation facility of the High Flux Isotope Reactor (HFIR) at Oak Ridge National Lab (ORNL) using the rabbit irradiation vehicles. Sixteen solid, hourglass, torsion joint specimens, were placed in a single rabbit capsule. Irradiation was carried out to approximately 4.5 × 10^{25} n/m^2 at 730 °C. An equivalence of 1 × 10^{25} n/m^2 (E > 0.1 MeV) with 1 dpa is assumed for SiC [9]. In this temperature range, which is within the transient swelling regime for SiC, this fluence exceeds the saturation fluence for SiC such that no progressive dimensional change is anticipated with further irradiation in this high purity SiC joint material [11]. Irradiation of other SiC joining candidates to similar temperature and fluence levels has effectively revealed dimensional change and microstructural phenomena arising at the joint interface that effect joint performance [9].

2.3. Pre- and post-irradiation examination

Post-irradiation examination was carried out in the Low Activation Materials Development and Analysis (LAMDA) laboratory at ORNL. Detailed examination in LAMDA included torsional shear strength, fracture surface examination, crystallographic phase examination and cross-sectional microstructures on a limited number of specimens. The shear strength test procedure is described elsewhere [14]. Scanning electron microscopy (SEM) examination and analyses were performed using SEM Hitachi Model S4700. X-ray diffraction (XRD) analysis was performed using Thermo ARL Model Scinatag Pad V. Porosity was determined from X-ray computed tomography (Nikon XT H LC 450 XCT) volumes of the joint using the void detection analyses in VolumeGraphics software package.

3. Results and discussion

3.1. Joint microstructure

An example of typical joint morphology is shown in Fig. 2. The SiC joint material design objective focused on maintaining compositional and microstructural homogeneity between the joint and surrounding components. Typical polycarbosilane pyrolysis temperatures range from 850 to 1300 °C, and often result in amorphous or only partially nanocrystalline structures [15]. However by post pyrolysis heat treatment above 1500 °C, the crystal structure was further refined such that the joint material had an average grain size similar to that of the nuclear grade fibers used in SiC–SiC composites, which have demonstrated retention of strength to 70 dpa at 800 °C [16]. It should be noted that heat treatment at higher temperatures for longer times will result in more complete crystallization and additional coarsening. However, the heat treatment temperature must also be selected to minimize damage to the fibers. Among the nuclear grade fibers, Hi-Nicalon Type S (Nippon) and Tyranno SA3 (Ube, Inc.) strength degradation occurs following exposure to temperatures of 1400–1900 °C [17,18]. Peak broadening analysis via Scherrer's method shows average grain size for the pyrolized and heat treated preceramic polymer of 90 nm. This more complete crystallization relative to previous data on preceramic polymer-derived SiC [15] is expected to stabilize the material against neutron induced shrinkage, and resultant degradation.

The CVI SiC application subsequent to the preceramic polymer