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Joining of silicon carbide composites for fusion energy applications

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Abstract

Joining of silicon carbide based materials has been recognized as one of the enabling technologies for the successful utilization of ceramic components in fusion energy systems. Sintered silicon carbide (Hexoloy SA) and silicon carbide (Hi-Nicalon™) fiber reinforced silicon carbide matrix composites have been joined using reaction forming/bonding based joining technologies. The room- and high-temperature mechanical properties and fractography of ceramic joints have been reported.

1. Introduction

Silicon carbide fiber reinforced silicon carbide matrix composites (SiC_f/SiC_m) have been selected as candidate materials for fusion energy systems. These materials can withstand high temperatures and neutron fluxes and provide a safe and environmentally benign life cycle [1]. A current limitation is that SiC_f/SiC_m can only be produced in restricted sizes and shapes. A method of joining SiC_f/SiC_m components that satisfies the requirements of radiation resistance, mechanical integrity, desirable thermal properties, safety during operation and maintenance or accident, and acceptable waste management characteristics is required [2,3]. Since thermomechanical stresses should be minimized by a material with properties similar to that of the material to be joined, silicon carbide was selected as the joining material [4].

The joining technique must be compatible with the materials and processes used during the assembly of fusion energy systems. Since designs for fusion energy systems incorporate large SiC_f/SiC_m components, the use of applied pressure is not feasible. It is preferable that joining be performed in an ambient environment. The joining temperature must be below 1200–1400°C to avoid degradation of composites.

Fragomeni and El-Rahaiby [5] presented a review of various techniques used for the joining of silicon carbide based materials. Silicon carbide has been joined to itself via direct diffusion bonding, brazing, co-densification of green bodies and binders, reactive metal bonding, in-situ displacement reactions, preceramic polymer adhesives, glassy interlayers and reaction bonding. In this study, silicon carbide joints fabricated by the reaction forming method [6,7] were investigated. In addition, one set of samples was fabricated by a reaction bonding approach similar to the technique described by Rabin and Moore [8,9].

2. Experimental methods

Due to the high cost of SiC_f/SiC_m composites, monolithic (unreinforced) silicon carbide [1] was used for

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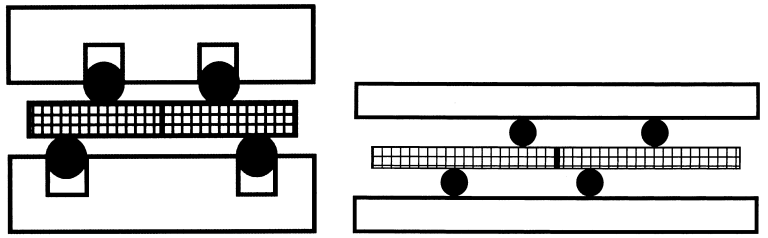

Fig. 1. Schematic drawings of mechanical test configurations: (a) 1/4, four-point bending and (b) asymmetric four-point bending.

evaluation. Plates of silicon carbide were cut into 25 mm × 4 mm thick pieces. A detailed description of the joining process has been presented elsewhere [6,7]. A carbonaceous mixture was applied to the surfaces to be joined and cured at 110–120°C for 10–20 min. Subsequently, a slurry of silicon powder was applied to the surface of the joint region and heated to 1425°C for 5–10 min. Capillary forces drew the molten silicon into the joint, where it reacted with the carbon to form silicon carbide. Some specimens [2] were prepared using a tape-cast carbonaceous material. A limited number of joints between SiC_f/SiC_m [3] were also fabricated. The SiC_f/SiC_m was coated with an approximately 2 μm thick layer of silicon carbide to inhibit oxidation at high temperatures. The resulting joint thickness, in all cases, was approximately 115 μm.

The plates were cut into bars that were $44 \times 4 \times 4$ mm^3 with the joint at the middle of the bar (Fig. 1). The microstructure of the specimens was investigated via scanning electron microscopy (SEM), energy-dispersive X-ray analysis (EDX), transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM).

Two methods for measuring mechanical properties were used. In the first technique, specimens were subject to flexural loading in 1/4, four-point bending (Fig. 1(a)). Specimens tested in this manner failed due to the tensile stresses. In the second technique, a flexural load was applied asymmetrically (Fig. 1(b)), giving rise to through-thickness shear at the midpoint [10].

3. Results

There was no evidence of a deleterious reaction in, or near, the joint (Fig. 2). A TEM image indicated that the joint consisted of carbon, silicon carbide and pores (Fig. 3). The pores may have resulted from ion thinning.

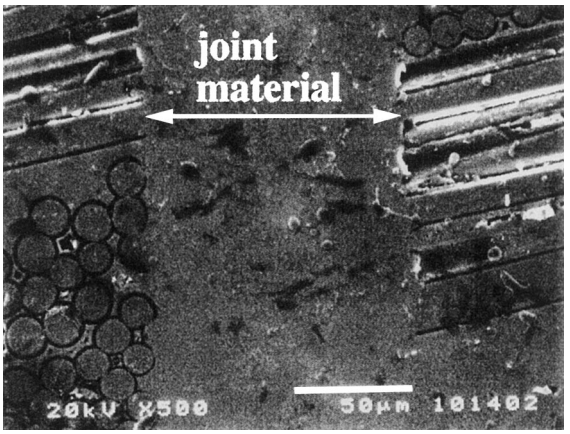

Fig. 2. Micrograph of joined SiC_f/SiC_m composites.

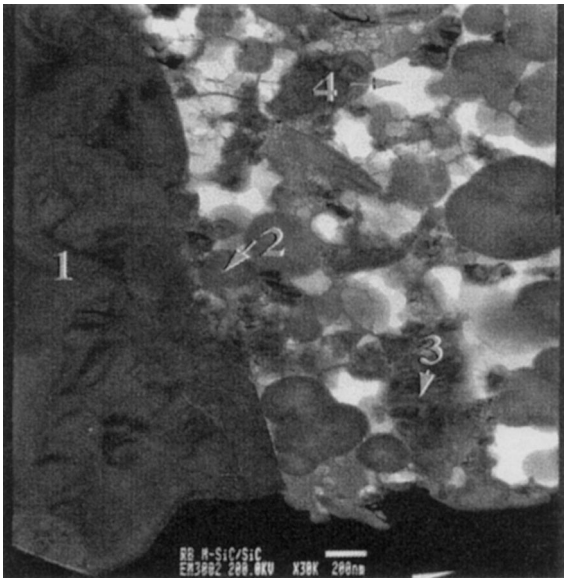

Fig. 3. A low magnification TEM micrograph indicating: (1) monolithic silicon carbide, (2) epitaxial carbon, (3) reaction formed silicon carbide and (4) a pore.

[2] Specimens provided by Busek, Natick, MA, USA.

[3] Fabricated by Honeywell Advanced Composites, Wilmington, DE, USA.

Table 1
Strength of joined specimens

Substrate	Joint material	Test method	Test temp. (K)	Joint thickness (μm)	Strength (MPa)
RBSC[a]	RFSC[b]	4PBS[c]	298	10	210 ± 6
Hexoloy SA	RFSC	4PBS	298	45–50	255 ± 3.2
Hexoloy SA	RFSC	4PBS	298	52	53 ± 6
Hexoloy SA	RBSC	4PBS	298	130	85 ± 10
SiC_f/SiC_m	RFSC	A4PB[d]	298	115	28 ± 7
SiC_f/SiC_m	RFSC	4PBS	298	115	78 ± 8
SiC_f/SiC_m	RFSC	4PBS	298	125	65 ± 5
SiC_f/SiC_m	RFSC	4PBS	1073	125	66 ± 9
SiC_f/SiC_m	RFSC	4PBS	1473	125	59 ± 7

[a] RBSC = reaction-bonded silicon carbide.
[b] RFSC = reaction-formed silicon carbide.
[c] 4PBS = 1/4, four-point bend strength.
[d] A4PB = asymmetrical, four-point bend strength.

The typical particle size of the carbon and the reaction-formed silicon carbide was a few hundred nanometers.

At room temperature, the bending strength of joined composites was 78 ± 8 MPa, similar to that in previous reports [11]. The through-thickness shear strength of joined composites was 28 ± 7 MPa. On the other hand, the strength of joined monolithic substrates was significantly lower than that in previous reports [6,7]. Joints between monolithic substrates fabricated using a carbonaceous slurry exhibited a bending strength of 53 ± 6 MPa and those using tape cast methods, 85 ± 10 MPa. The results are summarized in Table 1.

4. Discussion

Composite degradation, due to the joining procedure, was not observed. In an earlier study [7], 45–50 μm thick reaction-formed joints in sintered silicon carbide ceramics exhibited a flexural strength of 255 ± 3.2 MPa, which is significantly higher than that measured in this study. The flexural strengths of joints between composite substrates, however, were similar to those for 125 μm thick joints reported previously [11]. The measured values of flexural strength were lower than the first matrix-cracking stress, approximately 80–100 MPa for the composites. This implies that the joints will fail before the matrix.

The joints examined were thicker than those investigated previously, perhaps leading to larger residual stresses. Since the composite materials are more compliant than the monolithic substrates, these residual stresses may not be as deleterious. Thinner joints are known to exhibit better mechanical properties [13]. It is also possible that processing variations caused the contradictory results. Excess silicon and carbon that suggest an incomplete reaction were observed by TEM [12]. Insufficient specimens were examined to determine whether this was characteristic of all joints and whether these areas were related to the mechanical properties. It is likely that the mechanical properties of all the joints can be improved by process optimization and control.

The bending strength of SiC_f/SiC_m joined directly between the outer oxidation-protection coatings was similar to that of SiC_f/SiC_m joined between cut surfaces. These results suggest that surface preparation may be unnecessary. The actual strength value required is highly dependent on the joint geometry and stresses in a fusion energy system.

5. Summary and conclusions

Reaction-formed joints in silicon carbide fiber reinforced silicon carbide matrix composites were fabricated without evidence of fiber degradation. The results indicated that improvements in joint properties require process control and optimization. The strength of joints between cut surfaces and surfaces coated by a protective layer were similar.

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