

METHODS FOR JOINING SILICON CARBIDE COMPOSITES FOR HIGH TEMPERATURE STRUCTURAL APPLICATIONS - C. A. Lewinsohn, R. H. Jones (Pacific Northwest National Laboratory)*, M. Singh (NASA Lewis Research Center), T. Shibayama (Center for Advanced Research of Energy Technology, Hokkaido University), T. Hinoki, M. Ando, Y. Kato, and A. Kohyama (Institute of Advanced Energy, Kyoto University)

OBJECTIVE

Reliable and practical joining techniques are required to enable the use of silicon carbide composites in fusion energy systems. Based on criteria relevant to fusion applications, silicon carbide has been selected as a promising joint material. The objective of this work is to evaluate the thermal stability and mechanical properties of silicon carbide joints fabricated via reaction-forming.

SUMMARY

Joining methods are required to allow affordable fabrication of large or complex SiC/SiC components for fusion energy systems. Previous analysis of the criteria for successful and functional joints indicates that reaction-formed and polymer-derived silicon carbide should be considered as candidate joint materials. This report summarizes preliminary mechanical properties of silicon carbide joints formed by a reaction-based approach. Both the test methods and materials are preliminary in design and require further optimization. The values of the room temperature strength of the joints, tested in flexure, are one-third to one-quarter the expected value. It is believed that the material evaluated was not fully reacted during fabrication. Further annealing, in vacuum, also decreased the strength of the joints.

PROGRESS AND STATUS

Introduction

Silicon carbide has many desirable properties for use as a "first-wall" in a fusion energy system [1-4]. Composites consisting of continuous silicon carbide fibers in a matrix of silicon carbide offer mechanical reliability that is not available in unreinforced silicon carbide. A limitation of these composite materials, however, is that they can only be produced in limited sizes and shapes. Therefore, to fabricate a complete fusion energy system a method of joining SiC/SiC components, without compromising the properties that are needed, is required. In fusion energy systems utilizing silicon carbide first-wall materials it would be undesirable to use a joining technique that introduces dissimilar materials at the inner face of the first wall. In addition, to avoid poisoning the plasma the first wall must be hermetic. Therefore, two attractive methods of joining silicon carbide with other forms of silicon carbide have been developed: reaction bonding [5-8], and preceramic polymer adhesives [9-15]. In this paper, preliminary results obtained from joints formed by reaction based forming will be presented. Although other investigators have demonstrated that joints with required values of strengths can be formed [5-8], the long term stability of the joint, and consequent effects on the mechanical properties of the joint, require further attention. In this study joints were

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annealed under long term static and cyclic, high temperature conditions and the resulting mechanical properties and microstructures were examined.

Experimental Technique

To evaluate the suitability of joints formed by the reaction-based forming approach, plates of monolithic silicon carbide (Hexoloy SA, Carborundum Co., Niagara, NY) were joined using the ARCjoinT technique [6-8]. Two plates of monolithic silicon carbide were cut into 25 mm-long by 4 mm-thick pieces. A carbonaceous mixture was applied to the ends of the plates that were to be joined and this was cured at 110-120°C for 10 to 20 minutes. Subsequently, a slurry of pure silicon powder was applied to the surface of the joint region and heated up to 1425°C for 5-10 minutes. Capillary forces drew the molten silicon into the joint region where it reacted with the carbon to form silicon carbide. The resulting joint material consisted of silicon carbide with controllable amounts of silicon and other phases as determined by the composition of the raw materials and infiltrant.

A limited number of joints between pieces of silicon carbide composite material were also fabricated. This composite was reinforced with Hi-Nicalon fibers (Nippon Carbon Co., Yokohama, Japan) that had been coated with a 1 μm -thick layer of carbon prior to matrix infiltration via chemical vapor infiltration. In addition, an approximately 2 μm -thick layer of silicon carbide was deposited on the outside of the composite to inhibit oxidation at high-temperatures.

The plates that were joined using the method described above were cut into bars that were 44 x 4 x 4 mm. The bars were cut so that the joint was at the middle of the bar and the plane of joining was aligned so that it was parallel to the applied load. Several of the bars were annealed in a resistively-heated, quartz-image, furnace under vacuum. A series of specimens was annealed at 1100°C for ten consecutive 10 h long cycles. The microstructure of untreated and annealed 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).

Mechanical tests were performed by applying flexural loading to the bars described above. Two types of loading configurations were used. The first was standard 1/4, four-point bending (Figure 1a). This configuration subjects the specimens to a constant tensile stress in the region between the two inner load points. This test, therefore, is a measure of the tensile strength of the joint. The other test uses asymmetrical four-point loading (Figure 1b). This test, as described by Unal [16], subjects the specimens to a constant, through-thickness shear stress in the middle of the specimen. This test, therefore, is a measure of the shear properties of the joint material. Flexural loading was obtained by applying a compressive force on the fixtures using a rigid, mechanical test frame.

Results

As reported earlier [17], (Figure 2), joining pieces directly or between the outer coating of silicon carbide was equally successful and no evidence of a deleterious reaction between the joint material and the composite was observed, despite the high-temperature used to melt the pure silicon infiltrant. Apparently, these composite materials are not affected by exposure to the liquid silicon during the short infiltration time.

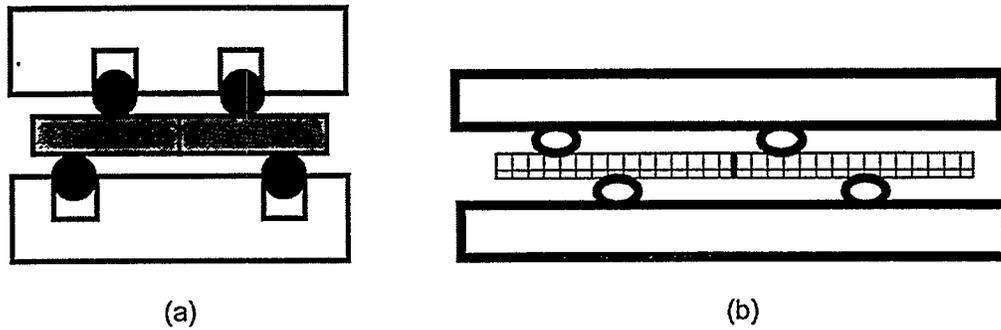


Figure 1. Schematic drawings of mechanical test configurations: (a) 1/4, four-point bending, and (b) asymmetric, four-point bending.

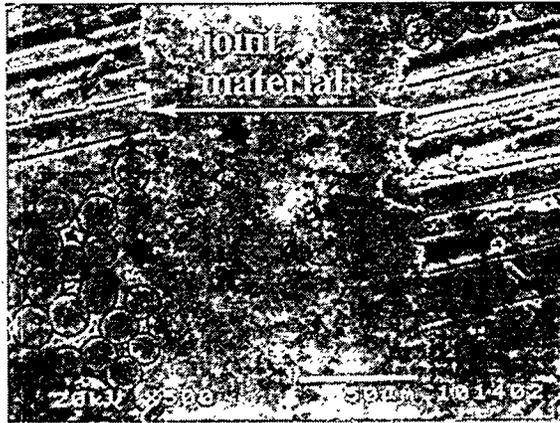


Figure 2. Micrograph of a cross section of two pieces of silicon carbide joined with reaction-bonded silicon carbide.

Experiments were conducted to investigate the long term stability of the reaction-formed joint material at elevated temperatures. Several specimens were annealed in a treatment that involved heating the specimens from 25°C to 1100°C in 55 min, holding the temperature at 1100°C for ten hours, rapidly cooling the specimens to 25°C (about 30 min), and repeating this cycle until the specimen had been subject to ten hold periods. Initial SEM microscopy and EDX results, Figure 3, indicate that after this treatment the silicon signal across a typical cross section of a joint is relatively constant. In addition, the appearance of the joint material near the interface with the monolithic silicon carbide is different from that in the middle of the joint. It is possible that further reactions between unreacted silicon and the extremely fine-grained carbon in the untreated joints occurred during annealing. Further microscopy will be conducted to investigate this hypothesis.

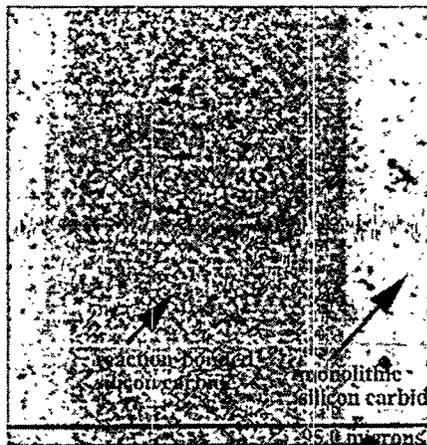


Figure 3. Scanning electron micrograph, with EDX spectra from silicon superimposed, of a cross section of two pieces of monolithic silicon carbide joined by reaction-bonded silicon carbide after annealing for ten cycles to 1100°C.

At room temperature, the maximum tensile stress obtained in flexural loading of joined composites and unreinforced SiC (monolithic) were similar (Figure 4). Unfortunately, the measured values of strength were lower than the first matrix-cracking stress, roughly 80-100 MPa [18], for the composites used. As mentioned earlier, the materials were not optimized for mechanical properties. The room temperature strength of reaction-formed silicon carbide joints has been reported as 255 ± 3.2 MPa [6]. The reasons for the lower than anticipated tensile stress values are not fully understood. The initial microstructural investigation [17], however, revealed that the untreated joints contained excess carbon and silicon that is indicative of an incomplete reaction. Therefore, it is likely that the joints were weaker than expected because the reaction to silicon carbide had not been completed.

The through-thickness shear properties of the joints were also tested. As described above, asymmetric, four-point bend tests were used to measure this property. As expected, the maximum through-thickness shear strength is lower than the maximum tensile stress in flexure (Figure 5). Despite the less than optimum strength properties of these joints, the through-thickness shear strengths were approximately one-third to one-quarter of a typical value for a reliable joint. Of course, the actual strength value required is highly dependent on the joint geometry and service stresses. After annealing in vacuum at 1000°C for either 100 h or ten 10 h cycles, the value of the through-thickness shear strength decreased significantly (Figure 6). Initial microscopy [17] revealed that these annealing conditions promoted the formation of additional SiC at the interface between the joint material and the substrate. Although the formation of additional silicon carbide at the interface between the joint and the substrate would seem to be desirable, diffusion of the required reactants from the joint material may have created porosity in the joint and made it weaker (Figure 3). This hypothesis must be examined further. These results emphasize the importance of carefully controlling the processing conditions during fabrication of the joints. Finally, the strength of composite material joined between approximately 2 μm -thick outer layers of unreinforced CVD SiC oxidation-protection coating were almost identical to the strength of those joined

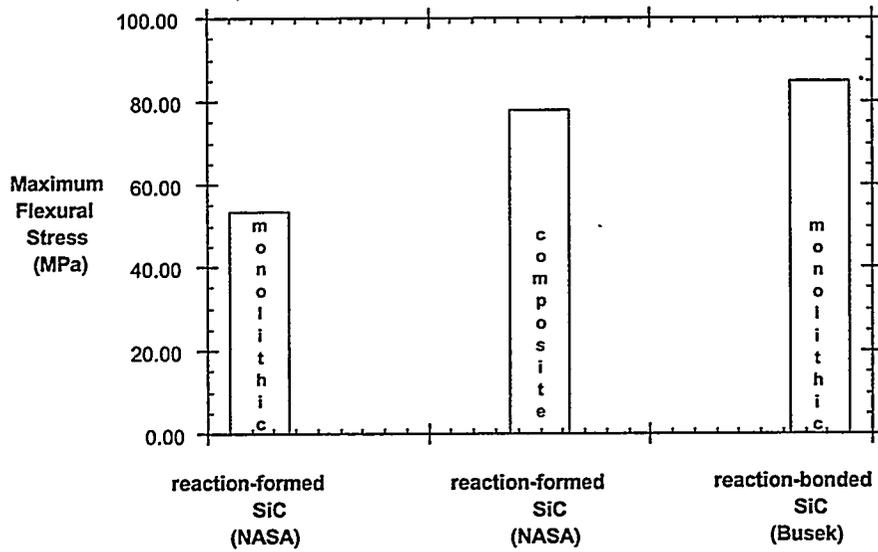


Figure 4. Comparison of room-temperature flexural strength.

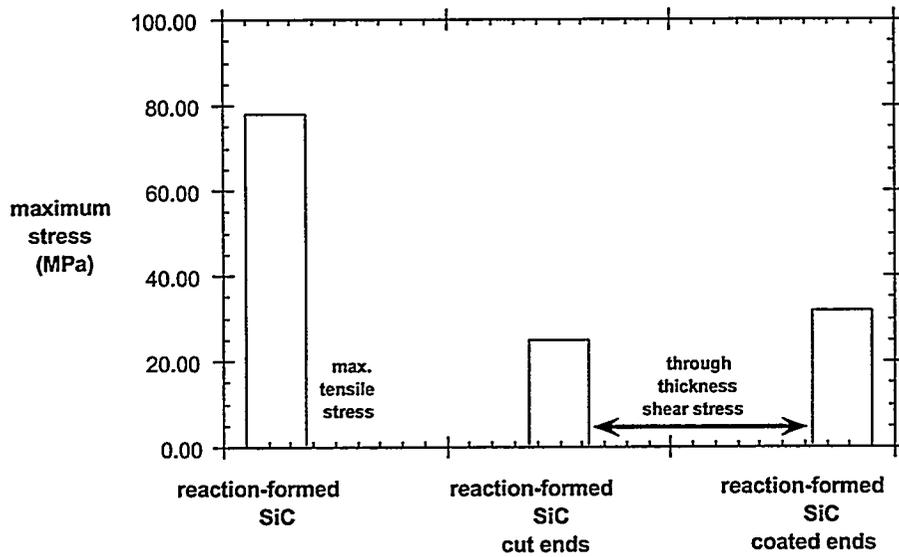


Figure 5. A comparison of the room temperature tensile stress and the through-thickness shear strength.

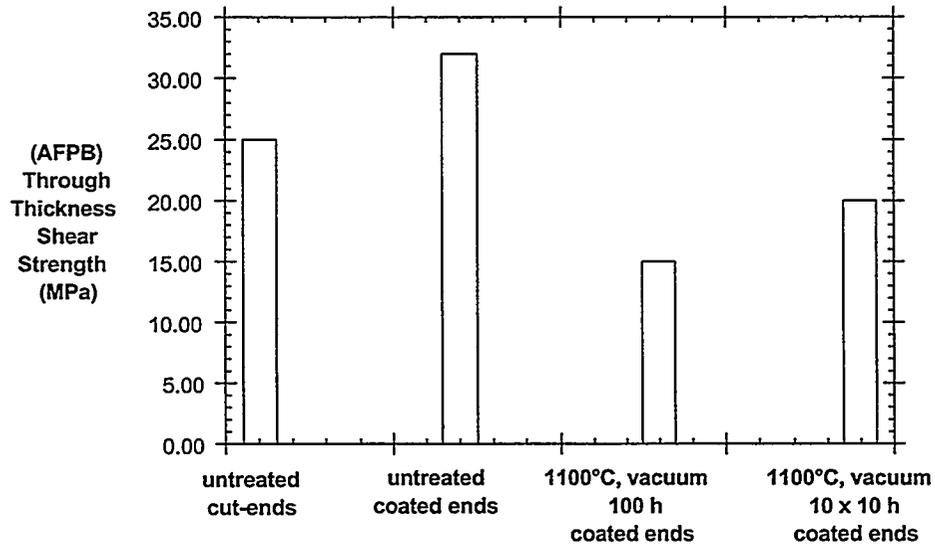


Figure 6. A comparison of the through-thickness shear strength after thermal annealing.

between cut surfaces. These results indicate that there need not be any special fabrication requirements prohibiting the cutting of materials with a protective layer supplied by a manufacturer.

CONCLUSIONS

Preliminary studies have shown that joints between silicon carbide and silicon carbide composites can be fabricated by a reliable, low-cost reaction-forming technique. Initial mechanical test data indicates that the joints have lower than anticipated strength values. In addition, annealing further decreases the measured values of strength. Additional work is required to characterize the microstructure and mechanical properties of untreated and annealed joints.

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