

A NEW APPROACH TO JOINING SiC/SiC COMPOSITES—C. H. Henager, Jr. (Pacific Northwest National Laboratory)¹

OBJECTIVE

The objective of this task is to develop a suitable joining method for SiC/SiC composites that retains the strength and toughness of the composite material, is low-activation, is easily applied, and can be used in the field for repairs, if required.

SUMMARY

A new approach to joining SiC-based ceramics is described and evaluated for Fusion Energy systems. The joining method is based on pre-ceramic polymers filled with reactive and inert filler powders and is similar to other approaches that use such materials. This approach differs in the particular polymer system and in the details of the processing. A principal advantage of this approach relative to other, similar approaches is that the polymer system is easily handled in ambient air and can be processed in air. This makes the joining process simple and field repairable. The joining compound is a liquid that can be painted, sprayed, or applied by dip coating.

PROGRESS AND STATUS

Introduction

Prior to this new approach, ceramic joining of SiC composites at PNNL either made use of solid state displacement reactions [1,2] or pre-ceramic polymers based mainly on polycarbosilane [3-7]. Both of these approaches were used to make strong joints between monolithic SiC, with joining of SiC/SiC composites being more problematic due to surface morphology considerations. However, both of these approaches suffer from requiring inert environments for handling and/or processing, which is less desirable compared to air handling. Thus, a new polymer system was found so that joining could be performed in air. Others have also attempted this, with varying success [8,9].

A polysiloxane polymer, polyhydridosiloxane (PHMS), from Gelest, Corp. is used as the pre-ceramic polymer for this work [10]. It converts to SiO₂ starting at 600 C and is completely converted at 1200 C in air. The backbone Si is bonded to hydrogen and a methyl group, in addition to the Si-O bonds that form the polymer chain. The hydrogen is readily removed and can be substituted with OH or with any number of organic groups, such as an ethoxy group or larger hydrocarbon groups. Doing so adds carbon to the resulting pyrolysis product so that a Si-O-C glass forms. The oxygen-carbon ratio is also sensitive to the processing environment and pyrolysis in nitrogen or argon results in higher carbon retention. At higher temperatures, SiO₂ and SiC are formed as crystalline phases and the SiO₂:SiC ratio can also be manipulated. Thus, this polymer is an attractive joining candidate for SiC-composites.

However, approximately 5% mass loss accompanies the pyrolysis and glass phase formation. This results in several percent linear shrinkage of the polymer on pyrolysis, which will either cause high residual tensile stresses in the joint or, more likely, will fracture the joint during pyrolysis and cool-down. To offset these shrinkage stresses, Greil [11] understood that reactive filler powders could expand enough on conversion in air or nitrogen to accommodate these shrinkages and, thus, form near-net shape ceramics using pre-ceramic polymers. We follow this route, partly, and make use of Al, alumina, and silicon carbide powders to make ceramic joints. The use of Al is not as desirable as SiC due to activation concerns but Al promotes the formation of SiC in the joint, which can help adhesion.

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Experimental Procedure

Powder slurries for joining were prepared by mixing powders, cyclohexane, and PHMS polymer in a standard roller mill. Approximately 50 v/o of the various powders were used to prepare the slurries. The Al powders are flake aluminum powders from AAE, the alumina powders are 2-micron size from Cerac, and the SiC powders are 0.7-micron size from Pred Materials.

The slurries were applied to monolithic Hexaloy SiC blocks that were 20 mm x 6 mm x 3 mm in size. Two blocks were joined using the slurry such that a 2 mm overhang at either end was present so that a simple shear test specimen was fabricated. The specimen and joint are shown schematically in Fig. 1. Enough slurry compound was applied so that as the blocks were pressed together it formed a uniform layer of joining compound.

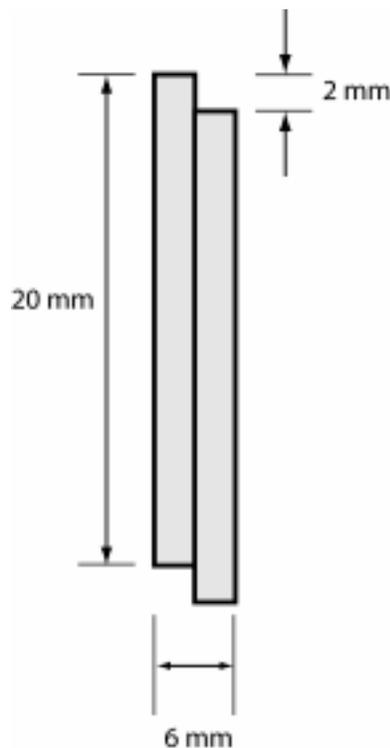


Fig. 1. Schematic of simple shear-lap joint using Hexaloy SiC blocks. The joining compound is applied all along the shared surface of the blocks.

The joints were processed by drying and curing the polymer-powder slurry at 150 C in moist air, which serves to crosslink the polymer. The joined blocks were then pressed normal to the joint at approximately 1 MPa pressure and heated to 1200 C in air for 1 hour. The joints were examined with an SEM. They were then tested at room temperature for shear strength as an indication of joint strength.

Results

Figure 2 shows the joint strength as a function of joint composition and processing conditions. Joints made with SiC powders and processed at 1200 C had the highest strength, 22 MPa in shear. The Al/SiC-filled joint also exhibited good strength at 14.5 MPa. The Al/Al₂O₃-filled joint had a lower strength and the joint processed without applied pressure during pyrolysis was very weak.

The SEM photos of this joint show in Fig. 3 that densification was good and that only minimal pressure is required to achieve a relatively dense joint. Figure 4 shows that without applied pressure that the joint density is low. Note the porosity of the Al/SiC joint processed without applied pressure.

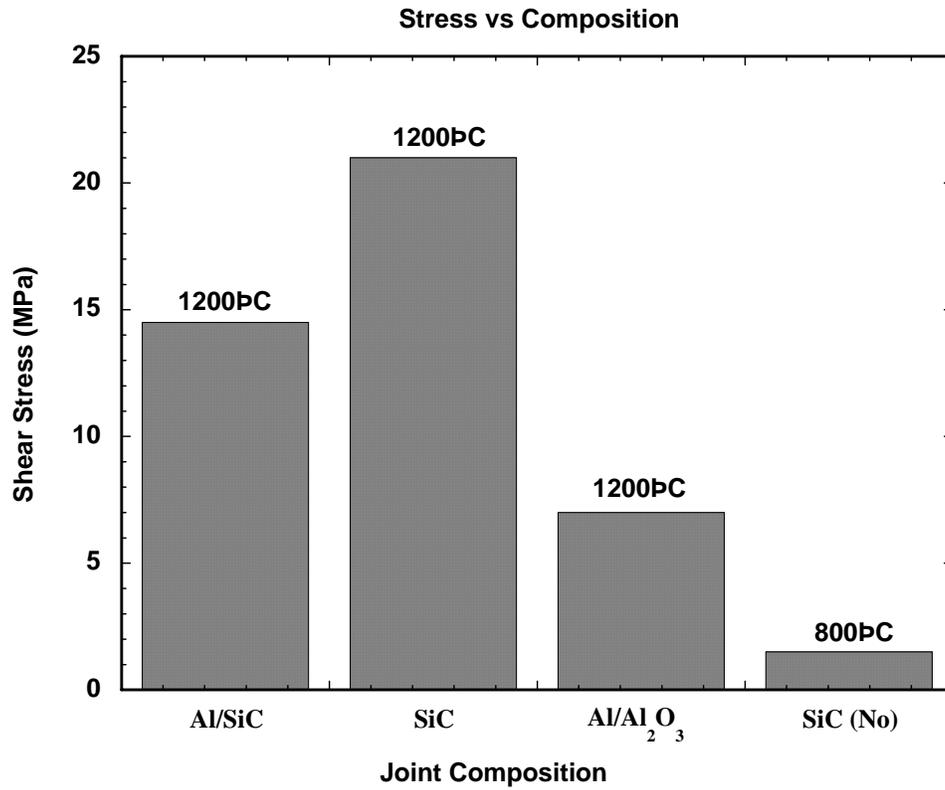
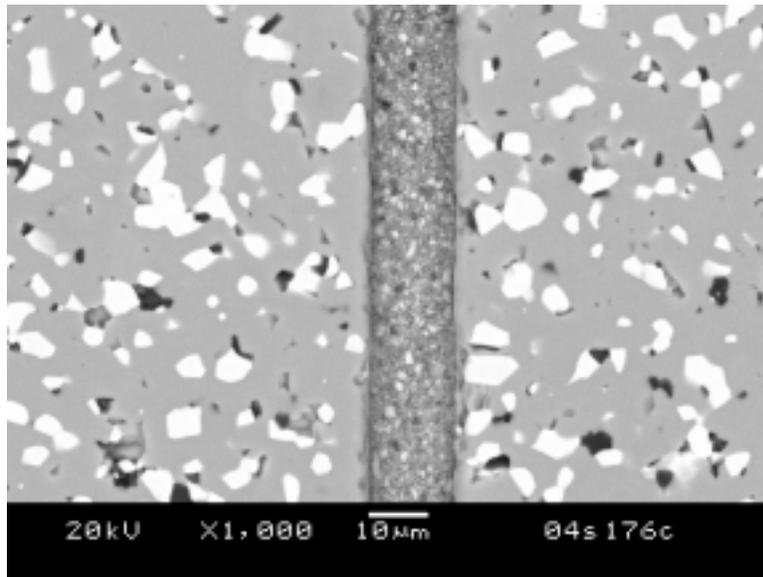
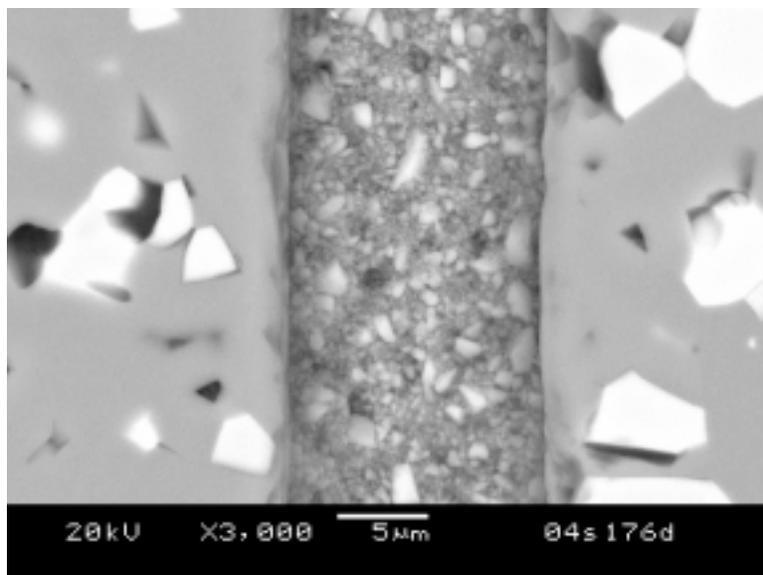


Fig. 2. Shear strength results for joints prepared at 1200 C with minimal pressure and one joint prepared at 800 C with no applied pressure.



(a)



(b)

Fig. 3. SEM photos of SiC-filled preceramic polymer joint at two different magnifications.

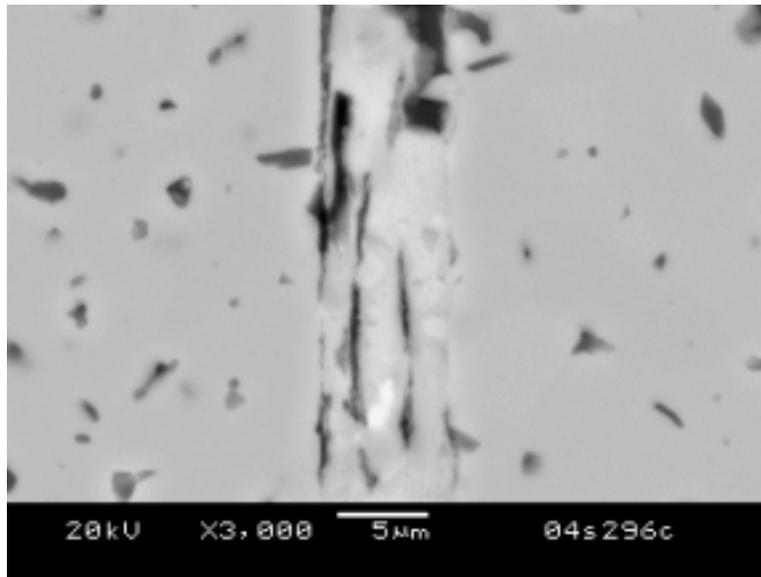


Fig. 4. SEM photo of Al/SiC-filled preceramic polymer joint.

Discussion

This work demonstrates a new approach to joining SiC-based materials for Fusion by emphasizing low-cost processing in air. While others have joined SiC using similar polymers, this is the first use of PHMS and the first to use processing in air. The advantages of PHMS are that we can control the Si-O-C ratios in the final pyrolysis product and that SiC is produced during pyrolysis. The use of Al powders assists in the production of SiC by reducing the Si-O during pyrolysis. However, the formation of Al_2O_3 is not helpful in joining SiC since the two materials are not very reactive. The best results in the present, limited study were obtained using SiC powders in the PHMS polymer. However, strengths were low compared to what have been reported by others. Apparently, the use of applied stress during processing is important and may be required for obtaining strong joints. However, Colombo et al. [12] report that reasonable joints can be obtained with unfilled siloxane polymers at 1200 C. The use of filler powders, which is required to provide shrinkage control, may make the application of pressure necessary.

Future Work

Since advanced ceramic fibers can withstand temperatures higher than 1200 C we plan to explore higher temperature joint processing to take full advantage of the filler powders and interface bonding characteristics of the polymer. Other combinations of filler powders will also be explored.

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