LOW ACTIVATION JOINING OF SiC/SiC COMPOSITES FOR FUSION APPLICATIONS[1]—
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OBJECTIVE

This work discusses the latest developments in TiC + Si displacement reaction joining at PNNL based on new scanning electron microscopy and electron backscatter diffraction results.

SUMMARY

The use of SiC composites in fusion environments may require joining of plates using reactive joining or brazing. One promising reactive joining method is the use of solid-state displacement reactions between Si and TiC to produce Ti_3SiC_2 + SiC. We continue to explore the processing envelope for this type of joint for the Titan collaboration to produce the best possible joints to undergo irradiation studies in HFIR. Joining pressure appears to require almost 30 MPa at 1673K in order to produce strong and dense joints. Recently pressures of 40 MPa and temperatures of 1698K have been used to produce excellent joints with apparent optimal density and microstructure. This increased temperature and pressure appears to provide for better bonding between the SiC and joining compound due to interfacial SiC formation and growth during the joint processing. Electron backscatter diffraction was used to show that local epitaxy exists between the SiC and interfacial SiC grown in the joint region.

PROGRESS AND STATUS

Introduction

SiC is an excellent material for fusion reactor environments, including first wall plasma facing materials and breeder-blanket modules. It is low-activation, temperature-resistant, and radiation damage tolerant compared to most materials. In the form of woven or braided composites with high-strength SiC fibers it has the requisite mechanical, thermal, and electrical properties to be a useful and versatile material system for fusion applications, especially since microstructural tailoring during processing allows control over the physical properties of interest [1-7]. However, it is difficult to mechanically join large sections of such materials using conventional fasteners so the analog of welding is being pursued for these ceramic materials [2, 6, 8-15]. Such methods include metallic brazes [9, 16], glass ceramics [8, 17], preceramic polymers [15], and displacement reactions [2]. This paper reports on the current status of SiC and SiC-composite joining for fusion applications based on displacement reactions between Si and TiC. This has been used to produce bulk composite material consisting of SiC-Ti_3SiC_2, with small amounts of TiC determined by the phase equilibria conditions [18].

Experimental Procedure

Joints are made using a tape calendering process using organic binders and plasticizers together with a mixture of TiC and Si powders, with 99.99% purity, average diameters less than 45 μm, and a TiC:Si ratio of 3:2. The flexible calendered tapes were 200 μm thick and were cut to shape and applied between two Hexoloy SiC or SiC/SiC composite coupons cut to a rectangular parallelepiped shape 20mm x 4mm x 2mm in size. Joints were formed by heating the coupon sandwiches in argon to 1698K at 10K/min and holding for 2 hours at 30 to 40 MPa applied pressure. The resulting joints were dense and approximately 12 to 30 μm thick. They were prepared for optical and scanning electron microscopy using standard ceramographic cutting, grinding, and polishing methods. A final polish using colloidal silica followed by a light argon ion etch was preferred for joints taken into the SEM and characterized using electron backscatter diffraction (EBSD) methods.

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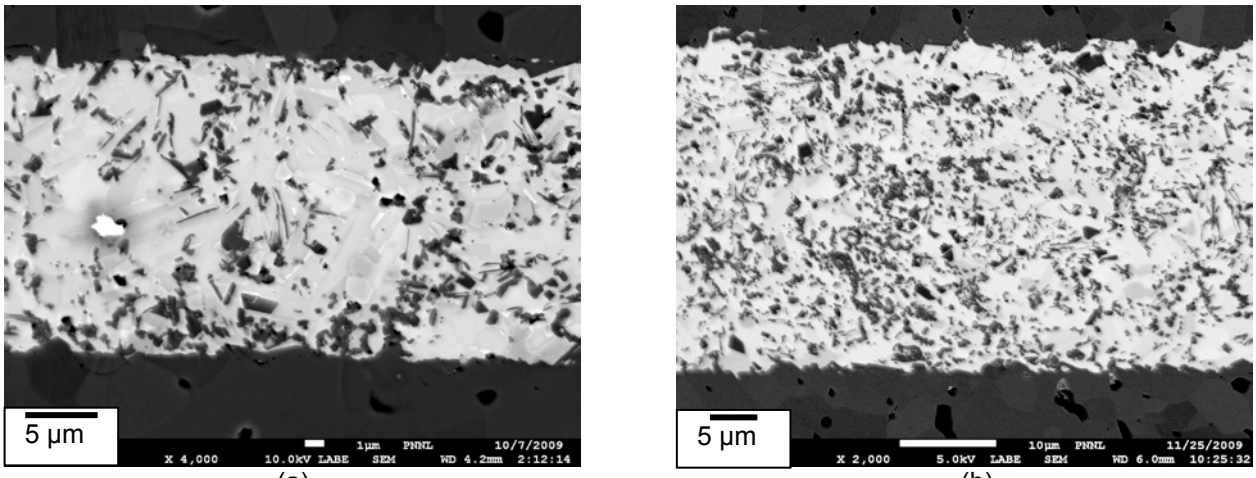
Results

Figure 1 shows scanning electron micrographs of a joint processed at 1673K for 2 hours at 30 MPa and one processed at 1698K at 30 MPa. Both joints appear dense in these images but the joint shown in Figure 1a has regions of lower density as shown in Figure 2. Figure 2 shows a less dense a region of the joint in Fig. 1(a) and processed at 1673K, 30 MPa. The joints processed at 1698K, 30 MPa or 40 MPa, did not exhibit these less dense regions, although each joint does have some porosity that may be unavoidable due to gas release during tape binder burnout. Based on these more extensive SEM examinations, the PNNL joint processing has been modified to 1698K, 40 MPa for 2 hours in argon.



(a) (b)

Figure 1. SEM micrographs of Ti_3SiC_2+SiC joint between Hexoloy SiC coupons. Shown in (a) is a joint processed at 1673K and (b) processed at 1698K, both at 30 MPa applied pressure for 2h during joining in purified argon gas. Both joints (taken at different magnifications; note micron markers) show mixture of SiC particles in Ti_3SiC_2 with significant in-growth of SiC at the joint/Hexoloy interfaces.

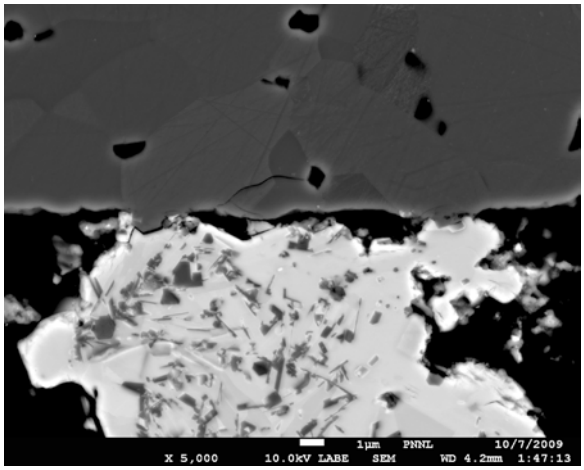


Figure 2. SEM micrograph of a region of a Ti_3SiC_2+SiC joint between Hexoloy SiC coupons showing less dense region after 1673K processing for 2h at 30 MPa. Based on this result, processing temperature was increased to 1698K for 2h.

A newly installed JEOL 7600 SEM was used for more detailed studies of the joint line interface and joint microstructure. One key parameter that appears to be controlling the joint strength is the excellent

bonding observed between the displacement reaction formed joint and the original SiC materials. For this study the original Hexoloy interfaces were polished flat and smooth using submicron diamond paste such that interface growth features formed during joining could be observed and studied. Figure 3 shows a SEM micrograph of the joint interface and clearly shows small voids at the original Hexoloy interface along with SiC that has grown in at the interface during the joining process.

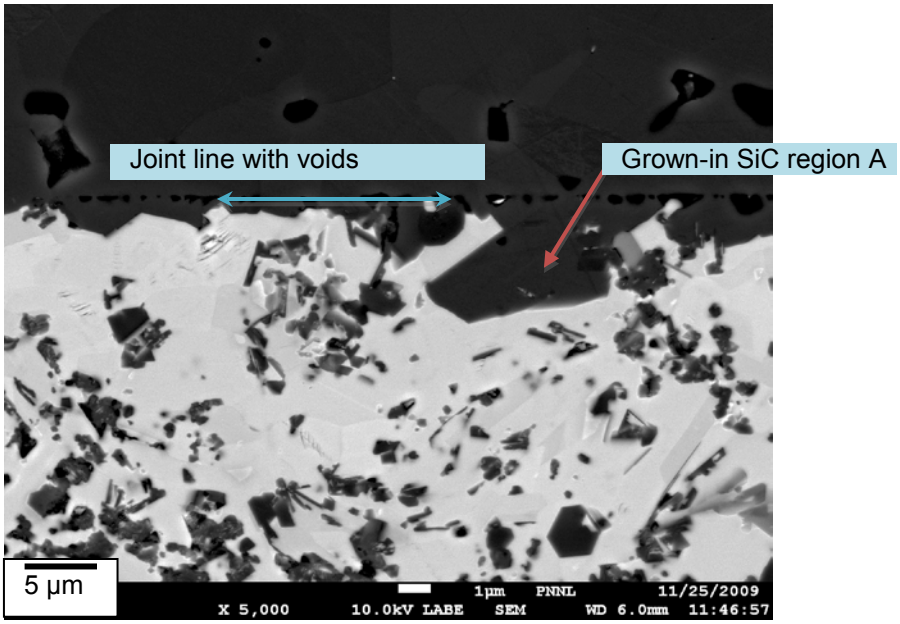


Figure 3. SEM micrograph showing joint line voids and large grown-in SiC regions (marked on micrograph). EBSD data indicates that the grown-in SiC regions have the same crystallographic orientation (epitaxy) as the grain they are attached to at the original SiC interface.

These regions, in particular, and the interior joint regions were studied using electron backscatter diffraction (EBSD) in the JEOL 7600 SEM. EBSD provides detailed crystallographic orientation information from near surface Kikuchi diffraction patterns that can be used to determine orientation relations between phases or grains present in the sample. For this work, EBSD data was acquired in the Hexoloy and joint regions with an emphasis on the joint line. This was more successful than EBSD data obtained from the joint interior because of the small grain sizes within the joint region compared to the larger SiC grains at the interface and within the Hexoloy. Briefly, this work supports our earliest findings from TEM studies that there is local epitaxy between the interface SiC grains that have grown into the joint region at the interface and the existing Hexoloy SiC grains [2]. Three interface regions were studied using EBSD and all of them indicated that grown-in SiC was epitaxial to existing SiC grains at the interface. This is shown in Figure 4 for one such region, where local epitaxy is found across the SiC/joint interface. Sample charging, thermal drifting, sub-micron grain sizes, and the complex crystallography of a-SiC all contributed to making it a difficult task to obtain high-quality EBSD patterns from these joints.

The EBSD data confirm that SiC formed during the displacement reaction preferentially nucleates at the pre-existing SiC interface and adopts the crystallography of the local grains. This is an important result, and one that was already suspected from our earlier TEM work but in this work we observe this phenomenon over a much larger region compared to the previous TEM study. Every region that was examined via EBSD showed local epitaxy across the joint line interface. The importance of this lies in understanding the strength of the joints thus formed. Early observations showed that joint shear fractures also produced fracture paths within the joined SiC pieces. Thus, local epitaxy produces a strong bond across the interface and a bond that can cause fracture to shift to the joined materials, which can be used to help design even stronger joints in the future.

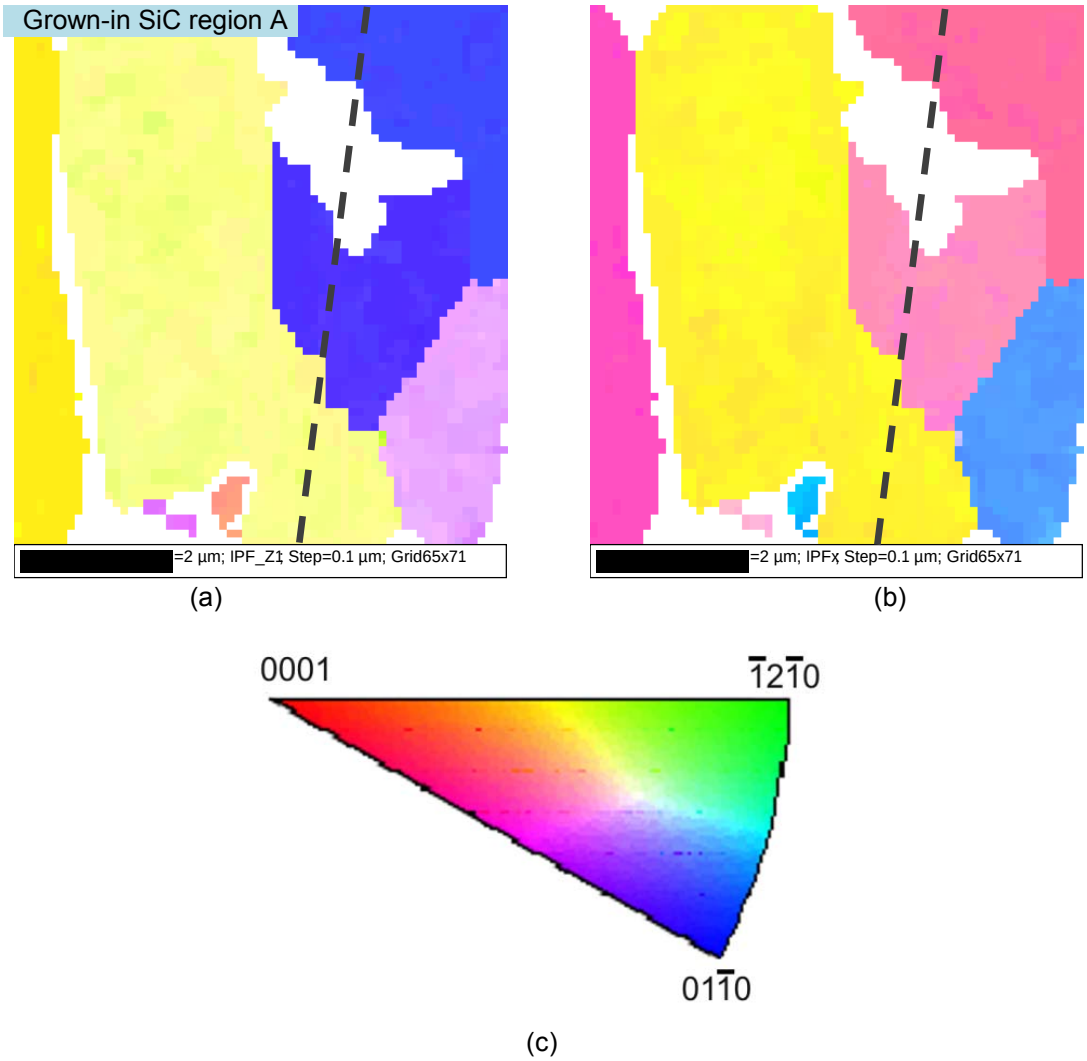


Figure 4. EBSD orientation maps showing crystallographic orientation for grown-in SiC region A shown in Fig. 3 SEM image. The location of the original SiC interface is indicated in the figure with the dashed line. The data show a continuity of crystallographic orientation, or epitaxy, across the SiC interface grains. The colors indicate the orientation relative to the standard stereographic triangle shown in Fig. 4(c) for the hexagonal SiC-6H phase. Fig. 4(a) shows the orientation map for z-axis normal to the plane of the image, while (b) shows the orientation map in the plane of the figure normal to the interface line.

CONCLUSIONS

Solid-state reaction joining was further optimized by processing tape cast TiC + Si joints in argon at 1698K and 30 to 40 MPa bonding pressure using Hexoloy SiC coupons and SiC/SiC coupons. Joints with desirable microstructures and thicknesses have been obtained. Local SiC-phase epitaxy was observed at the original joint interface indicating that SiC nucleation is facilitated by the pre-existing surface. Further work is underway to characterize the joints using transmission electron microscopy for improved phase identification and compositional mapping.

References

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