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Joining of carbon/carbon composites for nuclear applications

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Abstract Using hot pressing, carbon/carbon composites were joined using a Ti_3SiC_2/SiC interlayer which was in situ synthesized by the reaction of TiC and Si. Phase composition of the interlayer was characterized by X-ray diffraction. Morphologies of the joints before and after shear test were determined by scanning electron microscope and energy dispersive spectroscopy. The mechanical strength of the joints was assessed by shear strength test. Phase analysis reveals that the interlayer was mainly composed of ternary Ti_3SiC_2, SiC, and little TiC. The microstructure observation results show that the dense and uniform interlayer adheres firmly to the C/C composites. A composition gradient reaction layer was formed at the joining interface between C/C substrates and interlayer. The room temperature average shear strength of the joints is about 38.9 ± 3.6 MPa. The joining mechanism and failure behavior of the joints were also discussed.

Introduction

Carbon/carbon (C/C) composites are attractive structural materials for high-temperature applications due to their low density, low coefficient of thermal expansion (CTE), excellent high temperature strength and good ablation resistance [1]. Besides, they possess excellent thermo-mechanical properties, such as high thermal conductivity, high thermal shock, and thermal fatigue resistance. Therefore, C/C composites have been successfully employed in many fields such as engines in the aerospace industry and in the international thermonuclear fusion industry are proposed as plasma facing components [2–4].

The engineering design often requires large-volume and complex-shape components of C/C composites. Unfortunately, direct fabrication of these components was restricted by long preparation period and limited fiber-weaving technique. Alternatively, one promising way of achieving this is to build up complex-shape components by joining geometrically simple ones [5].

Several joining techniques for C/C composites are under development, which include solid-state reaction bonding, active metal brazing, pre-ceramic polymers joining, and joining by amorphous glass or glass-ceramic [6–10]. However, limited by the mechanical property of interlayer, the joints prepared by these techniques have a weak joining strength and cannot satisfy service requirements for nuclear applications.

In order to obtain the high strength and high thermal conductivity joints, an appropriate choice of interlayer material is necessary. Ti_3SiC_2 reinforced with SiC (Ti_3SiC_2/SiC) is an ideal candidate material which can be used as interlayer, owing to its unique combination of excellent properties, such as high elastic modulus and strength, high fracture toughness and thermal conductivity, as well as good thermal shock resistance [11–13]. The aim of this article is to propose a simple and reliable way to join C/C composites using Ti_3SiC_2/SiC interlayer, in which the interlayer was synthesized by in situ reaction between TiC and Si.

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Experimental

The C/C composites were manufactured in our laboratory by isothermal chemical vapor infiltration with a density of 1.75 g/cm^3 and with no impurity involved besides carbon. The C/C composites were cut by a diamond saw and the machining direction was vertical to the orientation of the carbon fibers. Rectangular samples (30 × 25 × 3 mm^3) were polished with 500 grit SiC paper, cleaned ultrasonically in acetone for 30 min and dried at 373 K for 10 min.

Commercially available powders of TiC (purity ≥99.99%, $d \leq 43$ μm) and Si (purity ≥99.99%, $d \leq 43$ μm) were employed in this study. The powders were mixed in the TiC/Si molar ratio of 3:2 and ball-milled for 12 h along with dispersant to form homogeneous and suitable viscosity coefficient slurry (about 500–600 Pa s).

One piece of C/C composites was sprayed with the as-prepared slurry, and another piece of C/C composites was put on them to form a "sandwich-like" structure. Then this structure was fastened using graphite clamp and put into vacuum hot-press furnace. Followed by heating at 1593 K for 1 h to form the interlayer by reaction of TiC and Si and dwelling at 1723–1823 K for 2–3 h for interlayer densification, the C/C composite joint was prepared. During the whole process, rapid heating rate (about 20 K/min) was preferred to avoid the volatilization of Si. A constant pressure of 30 MPa was applied on the joints to accelerate progress of the in situ reaction and densification.

The crystal-phase of the interlayer was determined by X-ray diffraction (XRD, X'Pert Pro MPD) with Ni filtered Cu Kα radiation (wavelength = 0.1542 nm). Microstructure and morphology of the joints were characterized by scanning electron microscope (SEM, Model JSM-6460) and energy dispersive spectroscopy (EDS). The room temperature shear strength tests of pure C/C composites and the joined samples (joint size: 10 × 10 mm^2) were performed with universal testing machine (Model Instron 1185) with a crosshead of 0.5 mm/min. The schematic figure of the shearing set-up was shown in Fig. 1. Five samples were measured for each group and arithmetical average value was regarded as the average strength of joints.

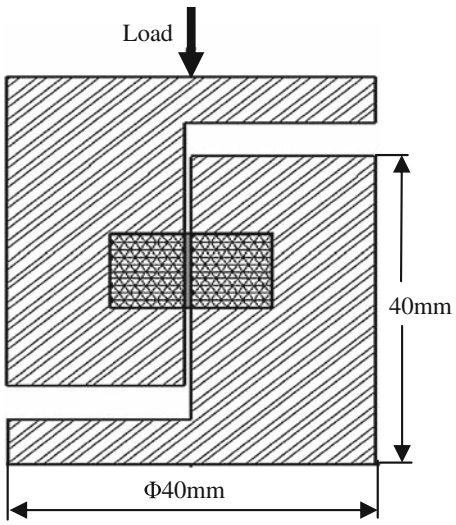

Fig. 1 Sample and clamp configuration for the shear strength test

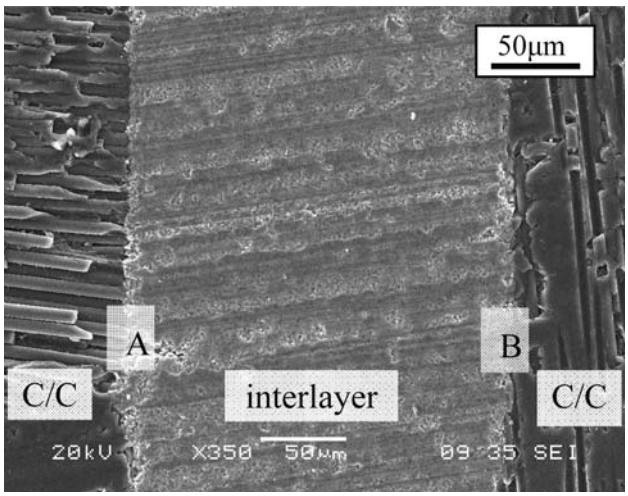

Fig. 2 The cross-section SEM image of the joints prepared at 1773 K for 3 h

Results and discussion

Figure 2 shows the cross-section SEM image of as-prepared joints prepared by the hot-press process. As shown in Fig. 1, the interlayer is dense and uniform with a thickness of about 200 μm. Visible discontinuities and micro-cracks are absent from the bonding interface between the interlayer and C/C composites. In particular, the interlayer exhibited excellent contact with carbon fiber and matrix, which can be seen clearly from the interface A and B, respectively. Few vertical cracks can be found at the interlayer, which should be attributed to great tensile stress was induced in the interlayer during cooling.

Figure 3 shows XRD patterns of the fracture surface of joints after shear test. XRD analysis identifies the presence of SiC and ternary Ti_3SiC_2 phase in the interlayer. As expected, solid-state displacement reaction occurred in the interlayer as follows:

$$3TiC(s) + 2Si(s) \rightarrow Ti_3SiC_2(s) + SiC(s) \quad (1)$$

However, residual TiC is still detected in the interface zone, which implies that TiC is relatively in excess and Si is insufficient. The mass loss of Si may result from two aspects: (1) the gradual evaporation effect during

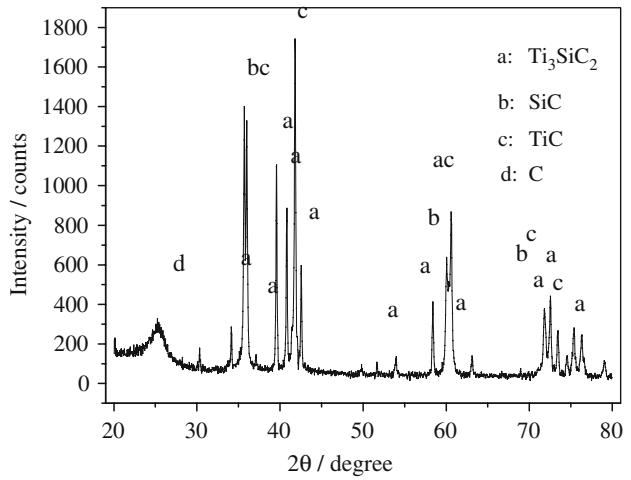

Fig. 3 XRD patterns of the fractured surface of joints after shear strength testing

hot-pressing preparation of joints above 1683 K (the melting point of silicon); (2) partial Si diffusion into C/C composites and reaction with carbon matrix to form the interface zone. Consequently, the structure and mechanical property of the interlayer must be weaken to some extent. It is suggested to eliminate this negative effect by adding some more Si into the slurry.

Figure 4 shows an EDS concentration profile of the joining interface between Ti_3SiC_2/SiC and C/C composites. Element line-scanning analysis reveals the diffusion trace of element C, Si, and Ti. From Fig. 3, it can be seen that the concentration of element Ti decreases, but that of element C increases gradually at the interface zone. In addition, Si concentration presents a peak, indicating that

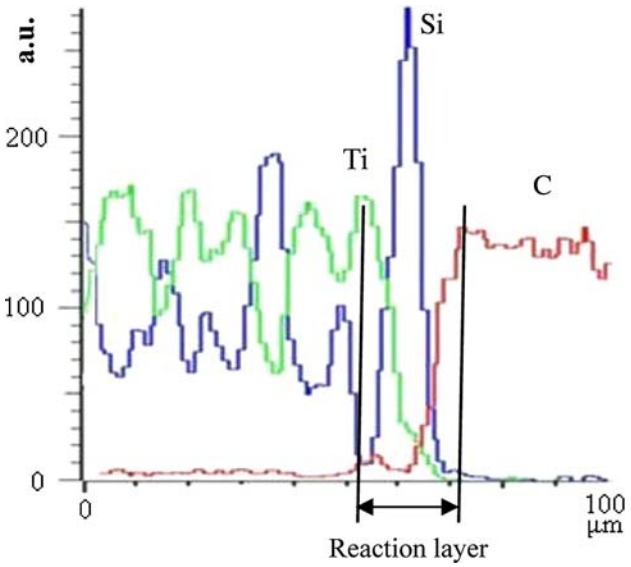

Fig. 4 The EDS concentration profile of the joining interface of the as-prepared joints

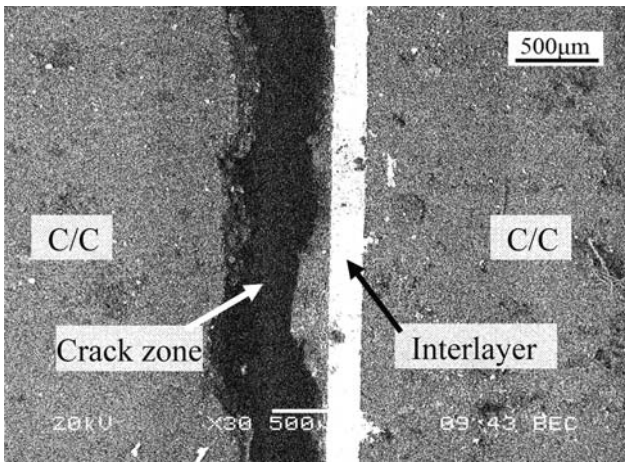

Fig. 5 Typical back-scattering electron image of the joint couple after shear test

Si diffused to the joining interface during hot-pressing. The results of element diffusion enable the formation of a composition gradient reaction layer at the interface and make interface zone actually compose of composites/transition layer/interlayer. The composition gradient reaction layer has a gradient CTE and can effectively alleviate the residual thermal stress at the interface zone. Therefore, the reaction layer helps to improve the shear strength of joints.

Figure 5 shows the typical fracture morphology of as-prepared joints after shear test. The average room temperature shear strength of the joints was about 38.9 ± 3.6 MPa and the maximum value can be up to 42.5 MPa, which was just lower than that of pure C/C composites (about 45.1 ± 2.7 MPa). As can be seen in Fig. 5, the interlayer is nearly intact and slightly damaged and the joints are destroyed invariably along the joining interface. Occasionally, the fracture extends into C/C composites. The fact was also demonstrated by XRD analysis, shown in Fig. 2, in which the crystalline diffraction peak of carbon was detected after shear strength testing. So, it can be concluded that shear strength of joints is not determined by the strength of interlayer, but by the C/C composites. Due to the great mismatch of CTE between C/C substrates and interlayer, residual stresses was induced at the joining zone during cooling process. The residual stresses damage the joining interface and weaken the shear strength of joints actually.

Conclusion

A reliable and novel joining technique for C/C composites was developed by means of a one-shot process. Using Ti_3SiC_2/SiC as interlayer, the joints of C/C composites were successfully produced via vacuum hot-press process,

where the interlayer was simultaneously synthesized by the reaction of TiC and Si. The remarkable average shear strength of the as-prepared joints is about 38.9 ± 3.6 MPa, which approaches shear strength of C/C composites. The reactive layer formed at the interface zone contributes a lot to shear strength of the joints. Meanwhile, residual thermal stress induced at interface zone results in the failure of the joints always occurred along the joining interface.

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