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High-temperature property of carbon/carbon composite joints bonded with ternary Ti–Si–C compound

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ABSTRACT

Carbon/carbon composites were joined using Ti–Si–C compound as interlayer and high-temperature shear strength and thermal shock resistance of these joints were investigated in this paper. In order to improve thermal shock resistance of the joints, a SiC transition layer was introduced between carbon/carbon substrates and interlayer. Microstructures and phase composition of the as-received joints were characterized by electron backscattered diffraction and X-ray diffraction. Mechanical strength and thermal shock resistance of the joints were measured by high-temperature shear test and thermal cycle test, respectively. The shear test results show that, with test temperature increasing from 273 K to 1673 K, shear strength of joints gradually increases to a maximum value of 46 MPa at 1473 K, then drops suddenly at higher temperature. Joints with SiC transition layer exhibit better thermal shock resistance than the ones without SiC transition layer and strength retention can be kept up to 85%, even after 30 thermal cycles from 1673 K to room temperature.

1. Introduction

Carbon/carbon (C/C) composites were regarded as one of attractive candidate materials for thermostructural application, due to their low density, low coefficient of thermal expansion (CTE), high thermal conductivity, excellent mechanical properties and thermal shock resistance at high temperature [1]. Presently, C/C composites are widely used in aviation and space field, such as high thrust–weight ratio turbine engines and leading edges of wing for space shuttle [2].

Owing to inherent brittleness and poor machinability of C/C composites, however, it is difficult to design and manufacture large-scale or complex-shape C/C component by conventional method, which inhibits some indispensable applications of these material. In order to achieve large and complex components, some researchers proposed to solve this problem by joining simple-shape C/C composites [3].

There are several methods developed to join C/C composites, such as glass material bonding [4,5], homogeneous pre-ceramic polymers joining [6] and reactive metal brazing [5,7]. Nevertheless, joints produced with above-mentioned interlayer material only can be used for low temperature application, which is limited to the high-temperature mechanical properties of interlayer material. Using boron, refractory borides or carbides as interlayer, Dadras et al. [8] and Dadras and Mehrotra [9] investigated the feasibility to join C/C composites by solid diffusion bonding. Those joints can be used at high temperature; however, high joining temperature more than 2173 K was needed during joining process, which made it hard to join large-scale C/C components. White et al. [10] bonded C/C composites by combustion joining with exothermic mixture of Ti–C and Ti–B system, but the tensile strength of the joints was lower than 5.5 MPa.

The choice of suitable interlayer was rather important before joining. Ti_3SiC_2 can be easily synthesized [11,12] and was regarded as potential "plastic" ceramics [13], because that it combined excellent characteristics of metal and ceramic, such as high stiffness, chemical resistance, high fracture toughness, excellent thermal and electric conductivity. Furthermore, its high-temperature stability (stable in inert atmosphere to temperature above 2473 K [14]), relatively low CTE ($8–10 \times 10^{-6}\,K^{-1}$ [15]) and good thermal shock resistance [16] made it possible to be used at high temperature.

Few literatures related to the C/C joints for high-temperature application and especially paid attention to thermal shock resistance of C/C joints. The purpose of this work is to join C/C composites using Ti_3SiC_2 as interlayer material by an easy and cost-effective way. The high-temperature shear strength and thermal shock resistance of as-prepared joints were investigated.

2. Experimental

Rectangular samples (30 mm × 25 mm × 3 mm) used in this experiment were cut from bulk 2D C/C composites with a density of 1.75 g/cm^3 and carbon fiber is more than 35 vol.%. The machining direction was vertical to in-plane orientation of carbon

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fiber. These samples were polished with 500 grits SiC paper, cleaned ultrasonically in acetone for 30 min and dried at 373 K for 10 min.

SiC transition layer for alleviating thermal stress was prepared by pack-cementation. Primary compositions for pack-cementation were composed of 60–70 wt.% Si, 20–25 wt.% graphite and 1–5 wt.% Al_2O_3. Samples and the pack mixtures were put into a graphite crucible and heat-treated at 2073–2273 K under inert atmosphere to form SiC transition layer.

Interlayer material was in situ synthesized by reaction of TiC and Si. Commercially available TiC (purity ≥ 99.99%, $d \leq 43\,\mu m$) and Si (purity ≥ 99.99%, $d \leq 43\,\mu m$) were employed in this study. Powders were mixed in the TiC/Si molar ratio of 6:5 and ball-milled for 12 h along with carboxymethyl cellulose to form homogeneous slurry. The slurry was evenly sprayed onto the surface of pre-treated C/C samples and samples were assembled in sequence of substrate/slurry/substrate to form a "sandwich-like" structure. This structure was fastened using graphite clamp and put into vacuum hot-press furnace. By heating at 1593 K for 1 h to form interlayer via in situ reaction of TiC and Si and dwelling at 1823 K for further densification, the C/C composite joints were prepared in a one-shot way. During whole process, rapid heating rate was preferred to avoid the volatilization of silicon. A constant pressure of 30 MPa was applied on the structure to accelerate progress of the in situ reaction and densification.

The crystalline phase of interlayer was identified with X-ray diffraction (XRD). Microstructure and morphology of the joints were characterized by electron backscattered diffraction technique (BES) and scanning electron microscope (SEM). The shear strength of the joints (joints area: 10 mm × 10 mm) were performed on Instron universal testing machine (Model 1185) ranging from room temperature (RT) to 1573 K with crosshead rate of 0.5 mm/min.

The procedure of one cycle of thermal shock experiment: the joints were put into a quartz glass tube with constant flow of argon, dwelled at 1673 K for 10 min in an electrical furnace and then took them out to cool in the air for 10 min. Thermal shock resistance of C/C composite joints was evaluated by strength retention after thermal cycles from 1693 K to RT. The shear strength retention (R_S) was calculated by the following equation:

$$R_S = \frac{\sigma_a}{\sigma_b} 100\% \quad (1)$$

where σ_a is RT shear strength of joints after thermal cycles, σ_b is RT shear strength of joints before thermal cycles.

3. Results and discussion

Fig. 1 shows the cross-section SEM image and phase composition of C/C composite joints. It is apparent from Fig. 1A that the thickness of interlayer is about 100 μm and the interlayer is dense and uniform. The interface zone between C/C substrates and interlayer is free of defects. Some cracks vertical to the interface, with regular interval (~0.5 mm), are observed in interlayer, which should be attributed to the great mismatch of CTE between C/C substrates ($\sim 2 \times 10^{-6}\,K^{-1}$) and interlayer ($\sim 9 \times 10^{-6}\,K^{-1}$). As shown in Fig. 1B, the phase analysis reveals that the interlayer is mainly composed of ternary compound Ti_3SiC_2, SiC and residual TiC. No presence of Si peak proved the Si element was fully consumed. The cracking in interlayer may relax the thermal stress of interface zone to some extent, thus alleviates the damage effect of thermal stress and improves the joining strength of C/C composite joints. It is worthwhile to mention that the cracks vertical to interface have a less damage on joining strength than parallel crack does.

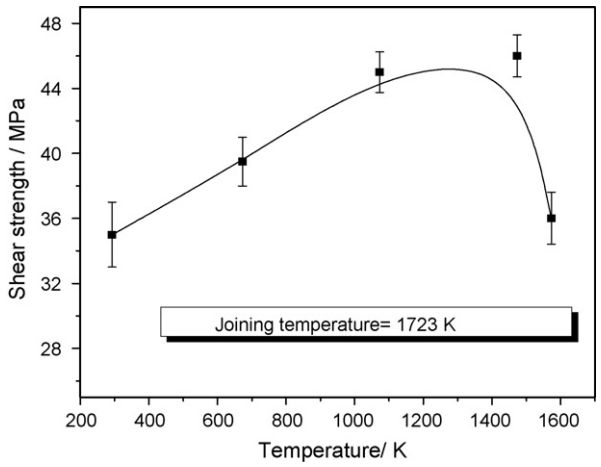

Fig. 2. Shear strength of joints measured at different temperatures from RT to 1573 K.

Fig. 2 shows the shear strength of as-prepared joints measured at 293 K, 673 K, 1073 K, 1473 K and 1573 K. It is apparent that shear strength of C/C joints increased from 35 MPa to a maximum value of 46 MPa with the test temperature increased from 293 K to 1473 K and then a sudden drop in shear strength was measured at higher test temperature. When the test temperature is lower than 1473 K, the increase in shear strength can be attributed to the relaxation of residual thermal stress at the interface zone. It is noted that the shear strength of joints exhibits a rapid drop from 46 MPa to 37 MPa when test temperature above 1473 K. The reason for this phenomenon mainly related to the fact that Ti_3SiC_2 exhibits a brittle-to-ductile transition at about 1473 K, above which the shear strength of Ti_3SiC_2 samples monotonically decreased rapidly with testing temperature [17]. All of the C/C joints fractured from interface zone between C/C substrates and Ti_3SiC_2 interlayer.

To alleviate the residual thermal stress at the interface zone, a SiC transition layer was introduced between C/C substrates and interlayer in this experiment. Fig. 3 shows the cross-section BES image of C/C composite joints with SiC transition layer. The selection of SiC based on the fact that SiC ceramic has an excellent mechanical strength at elevated temperature, a moderate CTE about $5.1 \times 10^{-6}\,K^{-1}$ and chemically compatible with both C/C substrates and Ti–Si–C interlayer. It is clear to see that the dense SiC transition layer and interlayer have a thickness about 50 μm and 130 μm, respectively. Absent of parallel crack and discontinuity along the interface zone indicated that the good bonding was formed between them. As expected, no detective vertical crack can

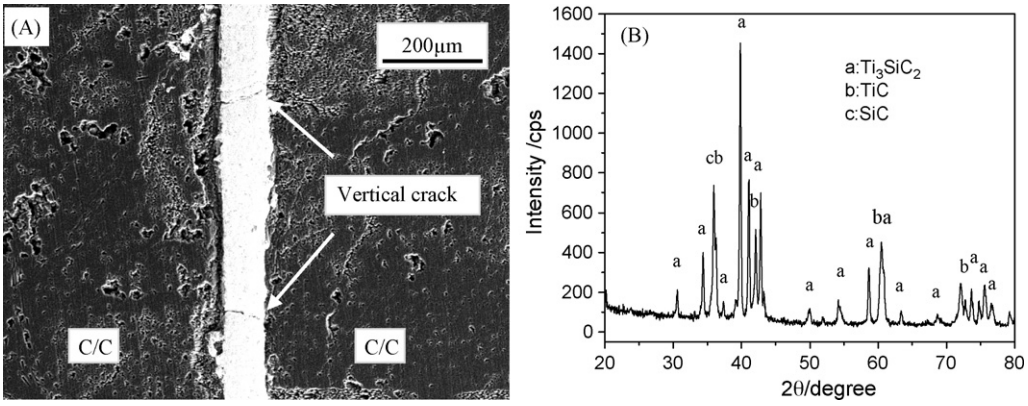

Fig. 1. The cross-section SEM image and XRD patterns of C/C composite joints.

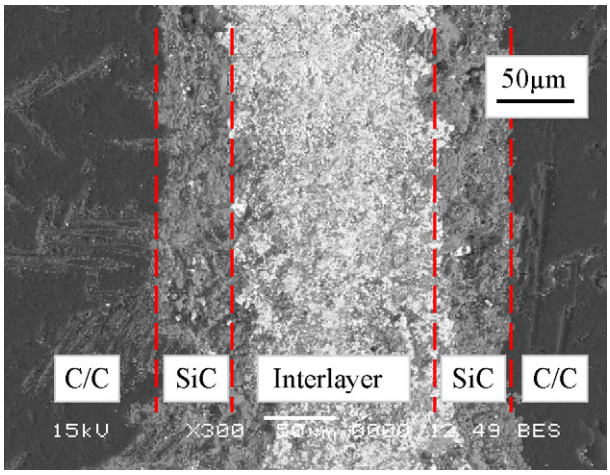

Fig. 3. The cross-section BES image of C/C composite joints with SiC transition layer.

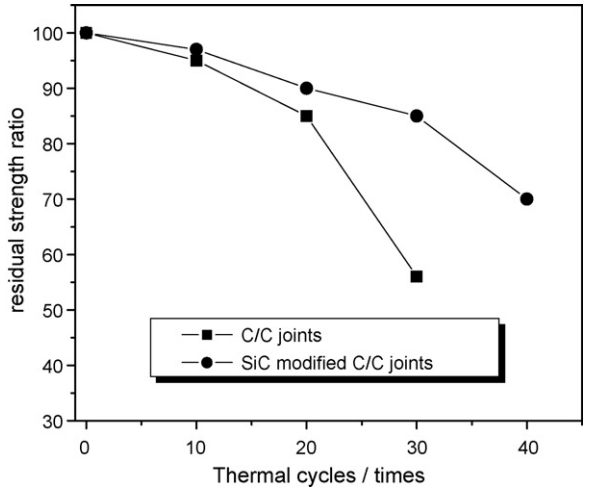

Fig. 4. Shear strength retention of both joints after thermal shock from 1673 K to RT.

be found in whole interlayer, which proved that SiC transition layer can effectively relax the mismatch between C/C substrate and interlayer.

Fig. 4 illustrates shear strength retention of joints after thermal cycles from 1673 K to room temperature. It is reported that Ti_3SiC_2 has excellent thermal shock resistance; nearly no loss of mechanical strength can be measured after thermal cycles from 1673 K to RT. When suffered from 10 thermal cycles, shear strength of the both kinds of joints exhibits good strength retention, even can be kept to 95% of initial shear strength. But after thermal cycles more than 20 times, joints with SiC transition layer show better shock resistance than the ones without SiC transition layer. Furthermore, the latter reveals an obviously accelerating decrease in shear strength. The decrease in shear strength primarily related to the generation and propagation of thermal crack at the joining zone. It can be postulated that some micro-cracks induced in interlayer firstly during cooling step; after several times thermal cycles, more and more cracks generated in interlayer and these micro-cracks spread gradually to form a network crack structure. At the same time, some cracks penetrated into C/C substrates, which resulted in the peeling of carbon matrix and breakage of carbon fiber around the cracking zone. As a result, the joints fractured under a lower load level with the increasing thermal cycles.

4. Conclusions

C/C composites were successfully joined using Ti–Si–C compound as interlayer by hot pressing. The Ti–Si–C interlayer was dense and uniform and the interface between C/C substrates and interlayer was defect-free. The shear strength of joints increased to a maximum value of 46 MPa at 1473 K and followed by a sudden drop at higher test temperature. A SiC transition layer prepared by pack-cementation can effectively improve the thermal shock resistance of the joints. Suffered from 30 thermal cycles in thermal shock experiment from 1673 K to RT, the C/C joints with SiC transition layer still have strength retention more than 85%.

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