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Joining of C_f/SiC Composite and TC4 Using Ag-Al-Ti Active Brazing Alloy

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Carbon fiber reinforced SiC (C_f/SiC) composite was successfully joined to TC4 with Ag-Al-Ti alloy powder by brazing. Microstructures of the brazed joints were investigated by scanning electron microscope, energy dispersive spectrometer, and x-ray diffraction. The mechanical properties of the brazed joints were measured by mechanical testing machine. The results showed that the brazed joint mainly consists of TiC, Ti_3SiC_2, Ti_5Si_3, Ag, TiAl, and Ti_3Al reaction products. TiC + Ti_3SiC_2/Ti_5Si_3 + TiAl reaction layers are formed near C_f/SiC composite while TiAl/Ti_3Al/Ti + Ti_3Al reaction layers are formed near TC4. The thickness of reaction layers of the brazed joint increases with the increased brazing temperature or holding time. The maximum room temperature and 500 °C shear strengths of the joints brazed at brazing temperature 930 °C for holding time 20 min are 84 and 40 MPa, respectively.

Keywords Ag-Al-Ti, brazing, C_f/SiC, TC4

1. Introduction

Carbon fiber reinforced SiC (C_f/SiC) ceramic matrix composites are lightweight, hard, and wear-resistant and stable in oxidizing environment up to the high temperature. Owing to the embedded carbon fibers, it has an excellent combination of mechanical properties. Therefore, C_f/SiC composite is a promising new structural material for a variety of high-temperature burner environments, including in hypersonic aircraft thermal structure, advanced rocket propulsion thrust chambers, cooled panels for nozzle ramps, turbo pump blisks/shaft attachments, and brake disks (Ref 1).

Like most ceramics, however, C_f/SiC composite is brittle and difficult to manufacture into workpieces with large dimensions and complex shapes, which substantially increases preparation cost. Therefore, the development of joining techniques is very important for joining of C_f/SiC composite to itself or to metals, especially Ti alloy. Fabrication of complex large-scale structural components requires robust integration technologies capable of assembling smaller, geometrically simple parts. Bolting and riveting are not advisable to join C_f/SiC composite due to its brittle nature and machine holes of C_f/SiC composite. Srivastava (Ref 2) used adhesives for joining C_f/SiC composite. The results showed that the strengths of the joint were very low. Meanwhile, the high service temperature of the joint impedes the use of bonding adhesives. Li et al. (Ref 3) joined C_f/SiC composite and Ni-based superalloy used Cu/W multiple interlayers. The results showed that the strengths of the joint are 102 MPa. Xiong et al. (Ref 4) joined C_f/SiC composite and Ni-based superalloy used Ti-Cu bi-foil interlayer. The results showed that the strengths of the joint are 34 MPa. Although the diffusion bonding can join the C_f/SiC composite and metal, the high bonding temperature (up to 1200 °C) limits the application of the method of diffusion bonding. But for brazing, due to its simplicity, lower cost investment and potential as a mass production process, is used extensively. Thus, brazing is becoming an effective method to join C_f/SiC composite with itself or metal. Asthana and Singh (Ref 5) and Singh et al. (Ref 6) brazed C_f/SiC composite and Al_2O_3 or Ti used Ag-Cu-Ti active metal brazes, but it is a preliminary study.

The eutectic composition of Ag and Al in weight percent is 94Ag-6Al, and it exhibits a low melting point (778 °C) as well as excellent fluidity upon melting. Eutectic Ag-Al alloys with a few percent of Ti are the most frequently considered active brazing fillers. Ag-Al-Ti active brazing filler metal possesses not only good wettability on ceramics but also an appropriate brazing temperature for TC4, which does not degrade the TC4. Decreasing the brazing temperature and time are always recommended with the advantages of decreased interfacial reactions, decreased erosion of substrates, and minimum loss of base-metal properties. However, the brazing mechanism and the possibly related interfacial reactions between C_f/SiC composite and Ag-Al-Ti active filler have been less often reported. The microstructure evolution and strength evaluation of the brazed joints using Ag-Al-Ti filler metal need further study.

In this study, the microstructures of C_f/SiC composite and TC4 brazed joints are investigated, the interface evolution mechanism of the C_f/SiC composite/Ag-Al-Ti/TC4 joint is analyzed and the mechanical properties of the joints are also discussed with the Ag-Al-Ti alloy brazing filler metal.

2. Experimental

Three-dimensional carbon fiber reinforced SiC matrix (3D C_f/SiC) composite and TC4 were used as components to be

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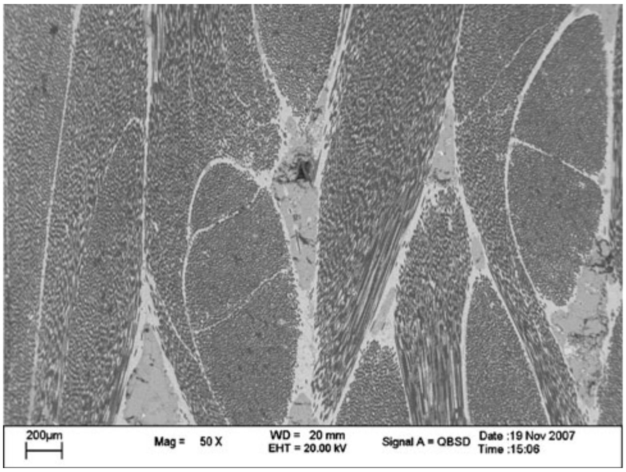

Fig. 1 The microstructure of C_f/SiC composite

joined in this study. Density of the C_f/SiC composite was 1.8 g/cm^3, its porosity was 10-15 vol.%, its three-point flexural strength is 300-400 MPa at room temperature. The carbon fibers distributed in the C_f/SiC composite were in the form of bundles and each bundle consisted of 12×10^3 pieces of carbon fibers. The carbon fiber volume fraction was 45-50%. The microstructure of C_f/SiC composite is shown in Fig. 1. The chemical composition of TC4 was Ti-6Al-4V (wt.%). The C_f/SiC composite and 15 mm diameter rod TC4 were cut into blocks measuring $5 \times 5 \times 5$ mm and cylinders of $Ø15 \times 4$ mm, respectively. The surfaces to be joined were ground by 400 grit silicon carbide papers for the C_f/SiC composite and 60 grit for the TC4. They were then cleaned in ethanol and dried at about 50 °C.

The constituents of the brazing material were 98(94Ag-6Al)-2Ti (wt.%) alloy powder with particle size of 320 mesh. The brazing materials were mixed with ethanol into paste, which was applied between the C_f/SiC composite and the TC4 by coating on the 5×5 mm surface of the C_f/SiC composite. The coaxial assemblies of the C_f/SiC composite and TC4 were brazed at 910-950 °C for 10-30 min in a vacuum furnace with vacuum levels better than 6×10^{-3} Pa to suppress undesirable reactions with oxygen. The heating rate was set at 15 °C/min. After brazing, all the specimens were furnace-cooled to room temperature.

To characterize microstructures of the joints, specimen was cut and subsequently processed by standard metallographic procedure prior to inspection. The cross-sections of the brazed joints were examined by means of a LEO-1450 scanning electron microscope (SEM) equipped with a KEVEX Sigma energy dispersive spectrometer (EDS). The joints were analyzed by x-ray diffraction (XRD) with Cu Ka radiation. As shown in Fig. 2, room temperature and 500 °C shear tests were conducted by means of a specially designed fixture at a crosshead rate of 0.5 mm/min in air and the average strength of three joints brazed under the same conditions was used.

3. Results and Discussion

3.1 Microstructures of the Brazed Joints

Figure 3 is the backscattered electron images of the joint brazed at 930 °C for 10 min. Table 1 gives the average

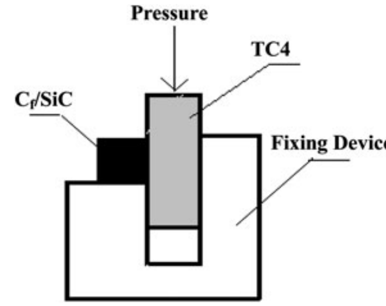

Fig. 2 The schematics of the shear test of the joined sample

chemical compositions of brazed joint at 930 °C for 10 min by EDS. As shown in Fig. 3(a), upper side is C_f/SiC composite and lower side is TC4, between the C_f/SiC composite and the TC4 is interlayer. The joint interfaces are microstructurally sound, well-bonded, and devoid of imperfections such as cracks and voids. It is expected that the molten braze tends to separate into two liquids during brazing. One is rich in Ag, and the other liquid is rich in both Al and Ti. Therefore, as shown in Fig. 3(b), during the following cooling process, Ag phase (white) and the Ti-Al phase (gray) distribute in interlayer. The Ti-Al phases were of two kinds, one is unshaped and mainly constituted of TiAl, the other is of short strip-shape and mainly constituted of Ti_3Al. The XRD diffraction presented in Fig. 4 indicates Ag and Ti-Al phases were reserved in the interlayer.

During brazing process, Ti element in the alloy powder reacts with the C_f/SiC composite surface, resulting in protrusion of carbon fibers on the interface between the C_f/SiC composite and the interlayer as shown in Fig. 3(c). This appearance should be beneficial to increasing interfacial bonding strength and also indicates that the reaction of Ti with SiC matrix is more violent than with carbon fibers. According to the further analysis of EDS, as shown in Table 1, Ti_5Si_3 compound of the reaction products for the SiC matrix of the C_f/SiC composite is evidenced as dark gray dots near the interface between the C_f/SiC composite and the interlayer. Another product appears in the form of Ti_3SiC_2 compound distributed on the surface of the SiC ceramic. The carbon fibers react with Ti element in the alloy powder, forming surface reaction layer TiC.

These compounds were produced by a series of reactions between the Ag-Al-Ti liquid phase and the SiC matrix. As Ag-Al-Ti liquid phase contacted with the SiC matrix of C_f/SiC composite, liquid partially infiltrated into the C_f/SiC composite during brazing, but there was still a layer of residual Ag-Al-Ti liquid phase in the joint. The Ti in the residual Ag-Al-Ti liquid phase first reacted with SiC and produced TiC and Si according to Reaction (1):

$$SiC + Ti \rightarrow TiC + Si \qquad \text{(Eq 1)}$$

The Gibbs free energy (about −100 kJ/mol at 900 °C) of this reaction is negative (Ref 7). The TiC crystal then nucleated at the surface of SiC matrix and grew into the liquid. Secondly, Ti continued to react with Si and TiC, the products of Reaction (1), and produce Ti_3SiC_2 compound according to Reaction (2) (Ref 8):

$$Ti + Si + TiC \rightarrow Ti_3SiC_2 \qquad \text{(Eq 2)}$$

Consequently, a mixture of TiC and Ti_3SiC_2 gradually covered the SiC matrix and finally formed the reaction layer as

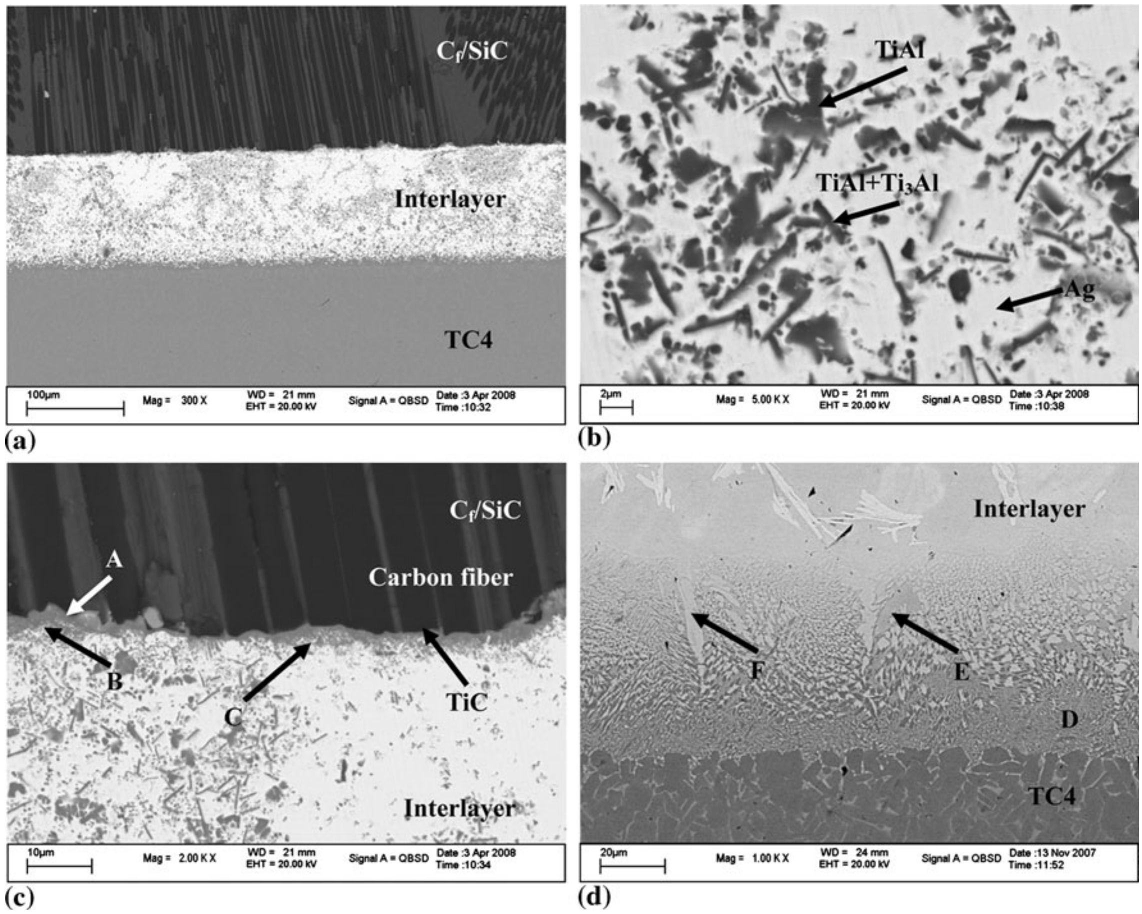

Fig. 3 Backscattered electron micrographs of joint brazed by Ag-Al-Ti alloy powder at 930 °C for 10 min. (a) Micrograph of the joint; (b) high-magnification image of interlayer; (c) interface between C_f/SiC composite and interlayer; (d) interface between interlayer and TC4

Table 1 Average chemical compositions of brazed joint by EDS (%)

Elements	C	Ti	Al	Si	Possible phase
A	30.44	56.10	0.12	12.09	Ti_3SiC_2
B	1.02	53.07	3.95	41.96	Ti_5Si_3
C	2.51	52.20	42.8	2.49	TiAl + Ti_3Al
D	5.35	70.72	15.67	8.26	Ti + Ti_3Al
E	3.02	61.31	27.01	8.66	Ti_3Al
F	6.23	43.54	46.47	3.76	TiAl

shown in Fig. 3(c). Thirdly, except for those reacting with Ti and TiC, the Si atoms produced by Reaction (1), driven by their own concentration gradient, diffused from the reaction layer into the liquid, and then reacted with the Ti of the liquid to form Ti_5Si_3 according to Reaction (3):

$$Si + Ti \rightarrow Ti_5Si_3 \quad \text{(Eq 3)}$$

The Gibbs free energy (about −580 kJ/mol at 900 °C) of this reaction is negative (Ref 7). As a result, Ti_5Si_3 particles were dispersedly embedded in residual TiAl (Ti_3Al) phase after cooling down.

During brazing process, the applied Ag-Al-Ti alloy melts in terms of Ag-Al-Ti and Al-Ti phase diagrams and forms liquid phase. In the liquid phase, Ti atom from TC4 is formed into intermetallics preferentially with Al, not with Ag from the Ag-Al-Ti filler metal, TC4 constantly dissolves and Al diffuses into the TC4, forming the diffusion reaction layers between interlayer and TC4 on the lower side of Fig. 3(d). The consumption of Al from the molten filler metal results in the depletion of Al from the Ag-Al eutectic phase during the brazing. Accordingly, the primary phase is Ag-rich and Al is

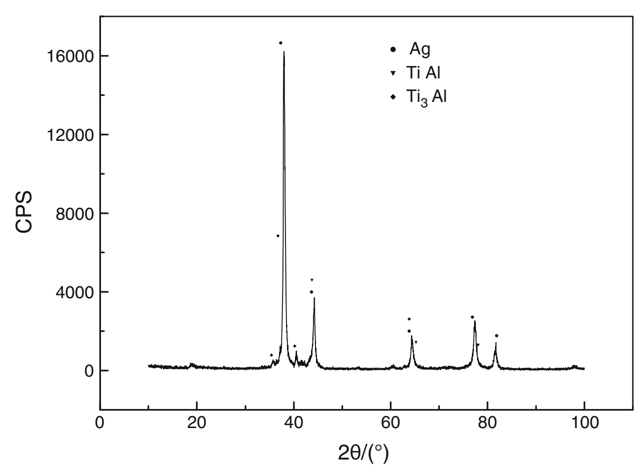

Fig. 4 XRD pattern of interlayer (at 930 °C for 10 min)

absent. Based on EDS analysis, as shown in Table 1, these phases were identified and are marked in Fig. 3(d), which array in sequence from TiAl down to the layer composed of coarse Ti_3Al phase and eutectic microstructure of Ti + Ti_3Al, which can be clearly seen with high magnification.

3.2 The Interface Evolution Mechanism of the C_f/SiC Composite/Ag-Al-Ti/TC4 Joint

A conceptual interface evolution model for the C_f/SiC composite/Ag-Al-Ti/TC4 brazed joint is displayed in Fig. 5. The whole reaction process can be divided into four stages:

(1) *Melting of Ag-Al-Ti alloy powder* from the room temperature to the melting point of Ag-Al-Ti alloy powder, the Ag-Al-Ti alloy powder gradually contacts with the base materials. Following the increased brazing temperature, Ag-Al-Ti alloy powder begins to melt.

(2) *Diffusion of atoms* when the brazing temperature is up to the melting point of the Ag-Al-Ti alloy powder, it melts and becomes liquid, some Ti and Al atoms in the alloy powder diffuse to the C_f/SiC composite/Ag-Al-Ti interface and Ag-Al-Ti/TC4 interface, respectively, some Si atoms in the C_f/SiC composite diffuse to the C_f/SiC composite/Ag-Al-Ti interface and some Ti atoms in TC4 diffuse into the alloy powder, as shown in Fig. 5(a).

(3) *Forming of the reaction layers* the Ti_3SiC_2 + TiC/TiAl + Ti_5Si_3 and TiAl/Ti_3Al/Ti + Ti_3Al reaction layers are formed at the C_f/SiC composite/Ag-Al-Ti and Ag-Al-Ti/TC4 interfaces, respectively. TiAl and Ti_3Al phases are formed in the interlayer with an increasing brazing temperature, as shown in Fig. 5(b).

(4) *Thickening and freezing of the reaction layers* the thicknesses of Ti_3SiC_2 + TiC/TiAl + Ti_5Si_3 and TiAl/Ti_3Al/Ti + Ti_3Al reaction layers increase with increasing brazing temperature and holding time. When the brazing temperature begins to decline, Ti_3SiC_2 + TiC/TiAl + Ti_5Si_3 reaction layers and TiAl/Ti_3Al/Ti + Ti_3Al reaction layers begin to freeze. Ag is also formed when the brazing temperature declines to the freezing point of Ag, as shown in Fig. 5(c).

Finally, the microstructure of the joint is C_f/SiC composite/Ti_3SiC_2 + TiC/TiAl + Ti_5Si_3/Ag + TiAl + Ti_3Al/TiAl/Ti_3Al/Ti + Ti_3Al/TC4.

3.3 Mechanical Properties of Joints

Using Ag-Al-Ti active brazing alloy powder, the joint brazed at 930 °C for 20 min shows maximum shear strength, being 84 MPa at room temperature and 40 MPa at 500 °C.

As shown in Fig. 6, it can be seen that with the increased brazing temperature or holding time, the joint strength increases at first then decreases as brazing temperature or holding time increase. When the brazing temperature is too lower or holding time is too shorter, the shear strength is low, e.g., the shear strength of the joint brazed at 910 °C for 10 min is only 38 MPa. This is because the amount of atomic diffusion is low and the reaction is insufficient between the filler materials and the base materials. The active element Ti cannot sufficiently react with the C_f/SiC composite to form a strong interfacial bonding between the interlayer and the C_f/SiC composite. This insufficient reaction results in lack of bonding between C_f/SiC composite and interlayer, as shown in Fig. 7. The fracture

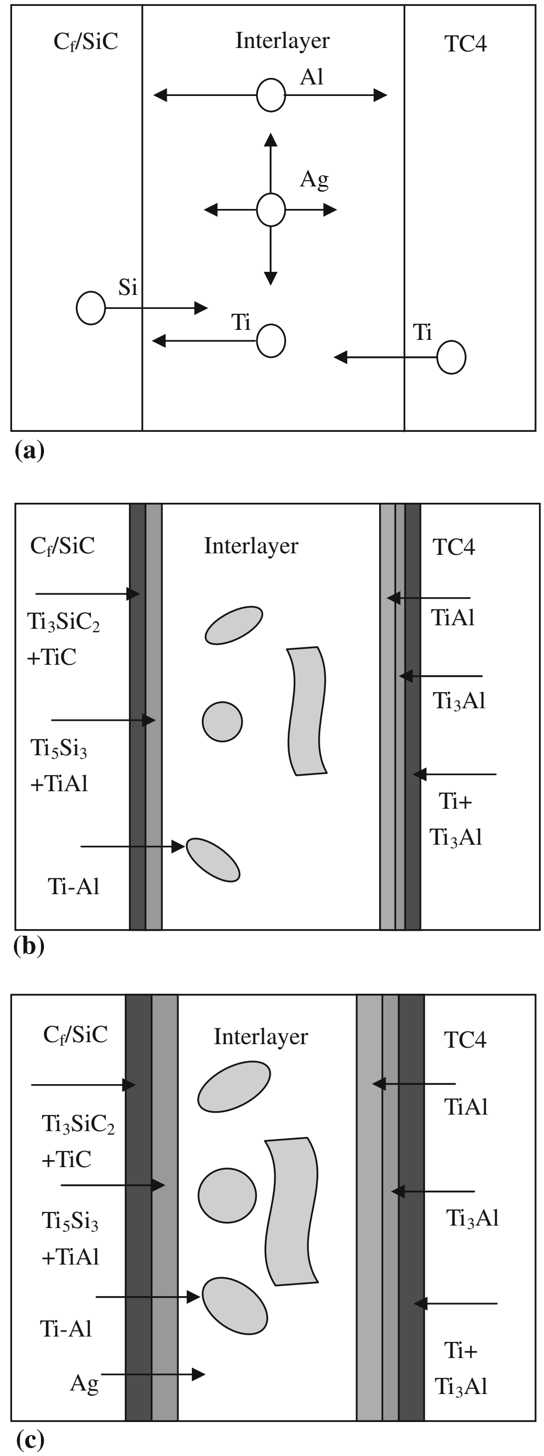

Fig. 5 Interface evolution model: (a) diffusion; (b) forming of the reaction layers; (c) thickening and freezing of reaction layers

usually occurred between the interfacial reaction layer and the C_f/SiC composite when shear testing.

With further increasing the brazing temperature or the holding time, the shear strength decreases, e.g., the shear strengths of the joints brazed at 950 °C for 10 min and 930 °C for 30 min are only 51 and 42 MPa, respectively. This is because the overreaction between C_f/SiC composite and interlayer, the accumulation of the brittle compounds would impair the joining properties of the joint, this overreaction

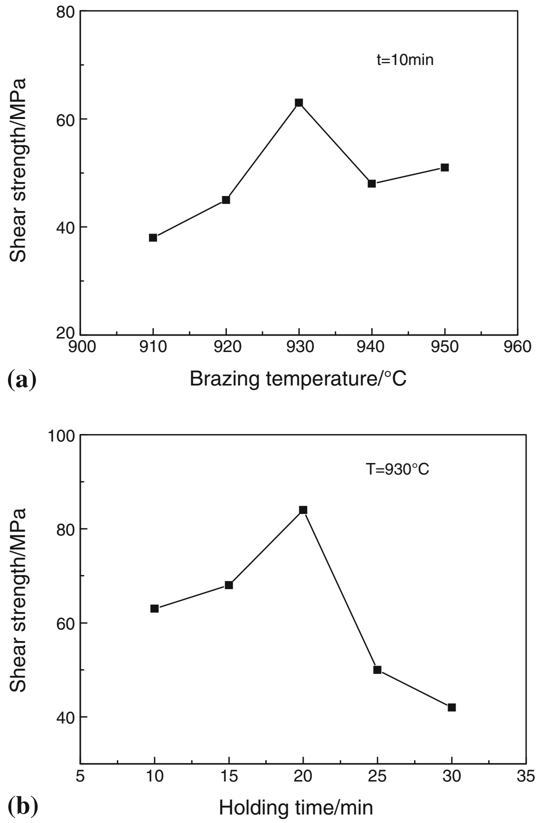

Fig. 6 Mechanical properties of the brazed joints (room temperature). (a) Effect of brazing temperature on shear strengths of joints; (b) effect of holding time on shear strengths of joints

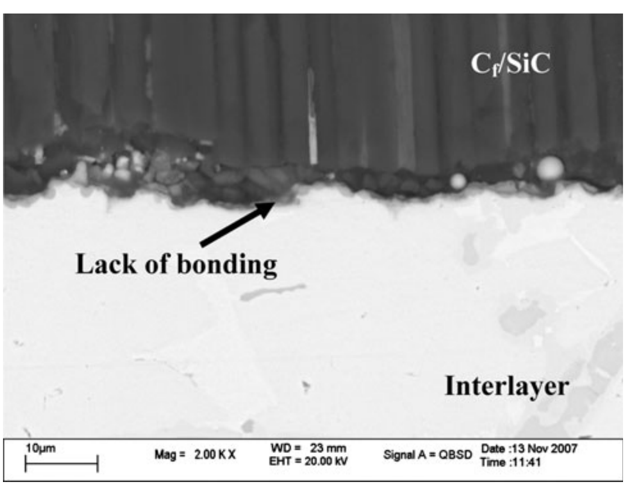

Fig. 7 Backscattered electron micrograph of joint brazed by Ag-Al-Ti at 910 °C for 10 min (interface between C_f/SiC composite and interlayer)

results in crack between C_f/SiC composite and interlayer, as shown in Fig. 8.

4. Conclusions

In conclusion, C_f/SiC composite was successfully joined to TC4 with Ag-Al-Ti alloy powder by brazing. Based on the

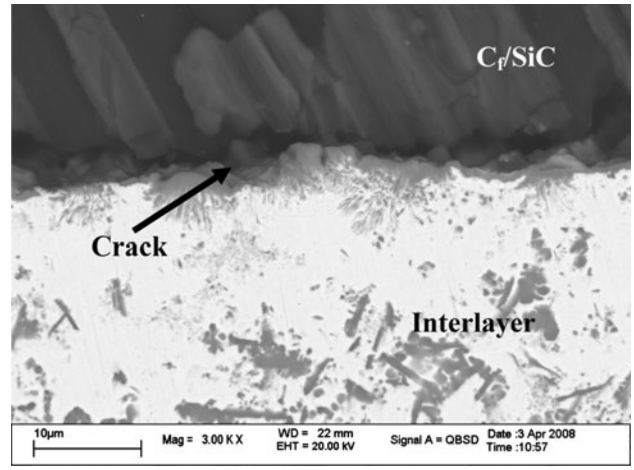

Fig. 8 Backscattered electron micrograph of joint brazed by Ag-Al-Ti at 930 °C for 30 min (interface between C_f/SiC composite and interlayer)

results obtained in this study, the following conclusions can be drawn.

(1) Ti element in the alloy powder can react with the C_f/SiC composite, a mixture of Ti_3SiC_2, TiC, and Ti_5Si_3 composites finally formed the reaction layers between C_f/SiC composite and interlayer. TC4 constantly dissolves and Al diffuses into the TC4, forming the diffusion reaction layers between interlayer and TC4. The microstructure of the joint is C_f/SiC composite/Ti_3SiC_2 + TiC/TiAl + Ti_5Si_3/Ag + Ti-Al/TiAl/Ti_3Al/Ti_3Al + Ti/TC4.

(2) The interface evolution process can be divided into four stages: 1. Melting of Ag-Al-Ti alloy powder; 2. diffusion of atoms; 3. forming of the reaction layers; and 4. thickening and freezing of the reaction layers.

(3) The maximum room temperature and 500 °C shear strengths of the joints brazed at brazing temperature 930 °C for holding time 20 min are 84 and 40 MPa, respectively.

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