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# Microstructure and mechanical properties of C/C composite joint brazed with Ni-based filler

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**Abstract:** The novel Ni-based brazing filler was used to join C/C composites. When brazing temperature increased from 1080 to 1100 °C, the wetting angle decreased from 23° to 14°, and the brazing filler had good wettability on the surface of C/C composites. The brazing seam of the brazed joint consisted of Ni(s,s) and $Cr_3C_2$ phases. As brazing temperature increased, lots of $Cr_3C_2$ phases were generated at the bonding interface, and the thick reaction layer was formed. When brazing temperature was 1120 °C, the shear strength of C/C joint reached the maximum value of 31.5 MPa. The fracture path extended in the C/C matrix close to the bonding interface.
**Key words:** brazing; microstructure; shear strength; C/C composites

## 1 Introduction

Carbon/carbon (C/C) composites have been widely applied in transportation and aerospace industries due to low density, high elastic modulus, and excellent heat resistance [1−5]. To expand engineering application, C/C composites are often required to join with high-temperature structural components [6−10]. There are several methods used to join, such as brazing [11−13], adhesive bonding [14], diffusion bonding [15−17], and resistance spot welding [18].

The great progress on brazing technology of C/C composites has been made because of the huge demand in aerospace industry. The active brazing method has become the important choice to achieve C/C joining owning to its simplicity, high structure adaptability, and low cost [19]. Currently, there are two main challenges. (1) Most of brazing fillers have poor wettability on the C/C composites. The metallurgical compatibility between C/C composites and brazing seam is poor. (2) The difference of expansion coefficient between C/C composites and metal causes large residual stress, which weakens the quality of C/C joints. The methods to improve the wettability of C/C composites include surface metallization and active brazing technology. Some metals in the brazing filler (such as Al, Ti, Zr, Cr, Mo and W) react with C/C composites, and the surface of C/C composites is modified to form carbides. The interfacial reaction can promote the metallurgical combination of fillers and C/C composites. The brazing fillers include Ti-based solder, Ag-based solder, Ni-based active solder, and Cu-based active solder. Recently, FENG's group chose the active brazing method to join C/C composites with many metals [20−22]. HE et al [23] introduced a novel two-step brazing method to obtain reliable C/C joints through gradient structure

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design and strong interfacial bonding. FENG et al [24] reported a mixed nanotube interlayer for joining C/C composites, which enhanced interfacial interaction. In addition, the addition of $B_4C$ could also greatly improve the mechanical properties of the adhesive due to the formation of borosilicate glass [25].

C/C components are usually used in high-temperature environment, so the brazing seam is required to have a high melting point. DADRAS and MEHROTRA [26] used Si foil as the brazing filler to braze C/C composites, and the brazing temperature was 1400 °C. Because Si reacted with C and SiC was formed, the shear strength of joint was 22 MPa. Ti−SiC−Si−C filler could also be used to join C/C composites, and the joints with ~160 μm thick filler had the maximum shear strength (38 MPa) [27]. Although Ti, Si and their alloys could be used to join C/C composites, the joints have poor oxidation resistance and cracks are easily formed at the interface due to the mismatch of thermal expansion.

In this work, the novel Ni-based brazing filler is used to braze C/C composites. The interfacial microstructure and brazing seam of the C/C joint are discussed. The reaction mechanism between Ni-based brazing filler and C/C composites is analyzed, and the shear strength of the joints is investigated.

## 2 Experimental

C/C composites used in this study were purchased from Boyun New Material Co., Ltd., (China) and consisted of preform and precursor after repeated high-pressure impregnation and carbonization treatments. The preform is the needle punched carbon felt fiber with 3D structure, and the precursor is novolac resin. Figure 1 shows the optical image of C/C composites. C/C composites were cut into blocks with dimensions of 5 mm × 10 mm × 15 mm. The Ni-based brazing filler was the mixed BNi-1a and DF-4b, and the mass ratio of BNi-1a to DF-4b was 1:1. Table 1 gives the chemical composition of Ni-based brazing filler. The BNi-1a was provided by Taizhou Zhenghua Co., Ltd., (China) and DF-4b was provided by Sulzer Metco Co., Ltd. (China).

Before brazing, the surface of C/C composites was ground by 320−1200 grid SiC papers and cleaned for 15 min in alcohol. The brazing of C/C composites was performed in a vacuum furnace with pressure less than $5.0\times10^{-3}$ Pa. The brazing temperature was 1080−1140 °C, and the brazing time was 15 min.

Microstructure and composition of the brazed C/C joints were characterized by a field emission scanning electron microscopy (SEM) and an energy dispersive spectrometry (EDS). The shear strength of the brazed joints was tested by a material machine (Gleeble 1500) at room temperature, and the shear speed was 0.5 mm/min.

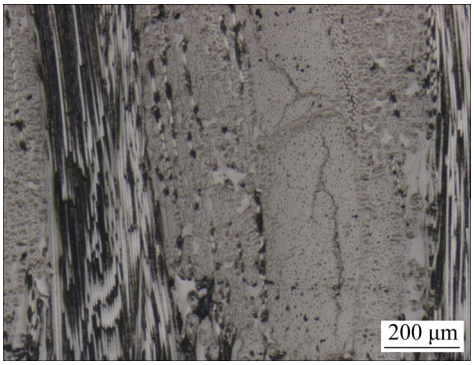


**Fig. 1** Optical image of C/C composites

**Table 1** Chemical compositions of Ni-based brazing filler (wt. %)

| Filler alloy | C | Cr | Co | B | Si | Al | Ta | Fe | Ni |
|---|---|---|---|---|---|---|---|---|---|
| BNi-1a | 0.06 | 14 | − | 3 | 4.5 | − | − | 4.5 | Bal. |
| DF-4b | − | 14 | 10 | 2.75 | − | 3.5 | 2.5 | − | Bal. |

## 3 Results and discussion

### 3.1 Wettability of brazing filler

Interfacial reaction was a common phenomenon in the process of wetting. There were two interfacial reaction mechanisms to determine wetting: the free energy of the interfacial reaction and the interfacial reaction product. According to thermodynamics, the spontaneous reaction in the system would inevitably reduce the free energy of the entire system. Therefore, the free energy change of interfacial reaction directly acted on the solid/liquid interface energy. Thus, the intenser the interfacial reaction was, the better the wettability was. The intense reaction was a necessary condition for obtaining good wettability.

Figure 2 shows the wetting angle of the mixed brazing filler on the surface of C/C composites at

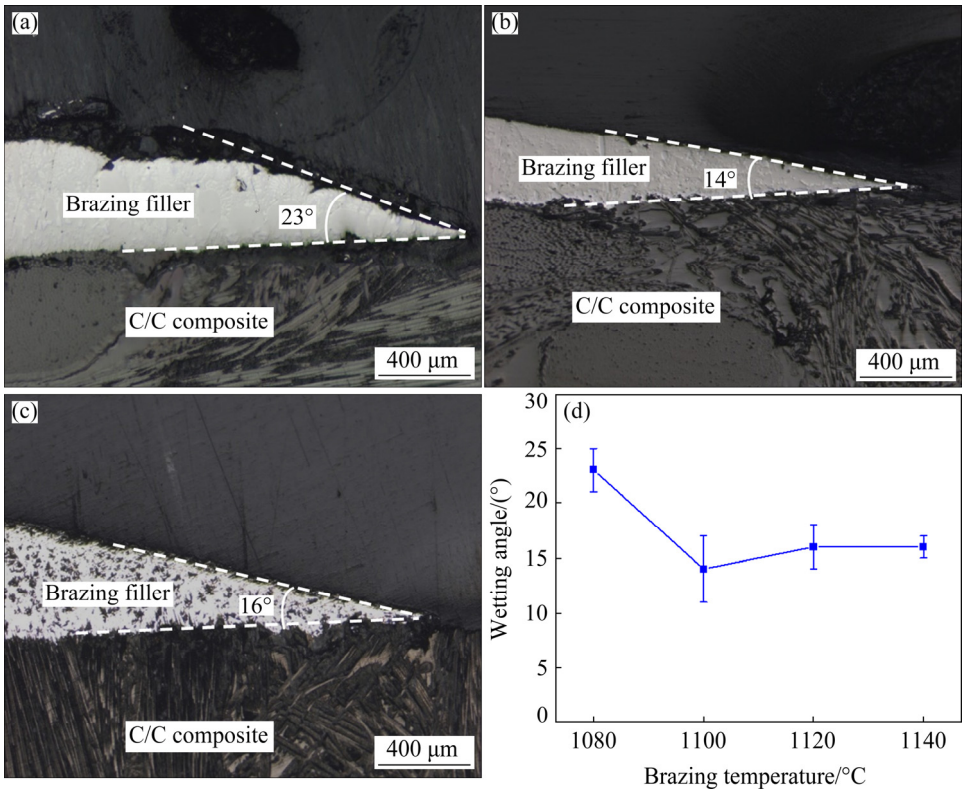


**Fig. 2** Wetting angles of mixed brazing filler on surface of C/C composites at different temperatures: (a) 1080 °C; (b) 1100 °C; (c) 1140 °C; (d) Relationship between wetting angle and brazing temperature

different temperatures. When brazing temperature increased from 1080 to 1100 °C, the wetting angle decreased from 23° to 14°. As the temperature continued to increase, the wetting angle was maintained at about 16°. There were fluctuations at 1100 °C, but the overall trend should be that the wettability was gradually improved as the temperature increased. Therefore, the mixed brazing filler had good wettability to the surface of C/C composites at high temperature.

### 3.2 Interfacial microstructure

Figure 3 shows optical images of the brazed C/C joints. When brazing temperature was 1080 °C, the brazing seam was dense, and its thickness was about 50 μm (Fig. 3(a)). When brazing temperature increased to 1100 °C, the brazing filler started to penetrate into the C/C matrix (Fig. 3(b)). When the temperature reached 1120 °C, the brazing seam became sparse, and lots of brazing filler penetrated into the C/C matrix (Fig. 3(c)). These results indicated that the wettability of filler was improved as brazing temperature increased, which was consistent with the wetting angle of Fig. 2(d). The brazing filler wetted well with the C/C matrix, and there were no defects in the brazing seam. When brazing temperature reached 1140 °C, more and more filler penetrated into the C/C matrix, indicating that the brazing filler metal had excellent wetting on the C/C matrix (Fig. 3(d)).

Figure 4 shows the SEM morphology of the brazed C/C joint. There were blocks of gray phases staggered along the bonding interface. When the temperature was 1080 °C, dark gray phases were distributed in the brazing seam (Fig. 4(a)). As temperature increased, much dark gray phases were generated at the bonding interface between brazing seam and C/C matrix, and a reaction layer was formed with 10 μm in thickness (Fig. 4(b)). In addition, part of bulk phases extended into the brazing seam. When brazing temperature was 1120 °C, much brazing filler penetrated into the C/C matrix, and the brazing seam became thin (Fig. 4(c)). Especially, when carbon fibers were perpendicular to the brazing seam, the degree of wetting became high, and the wetting distance could reach up to 150 μm (Fig. 4(d)). These gray phases were wedge-shaped and wedged into the C/C matrix, playing pinning role and strengthening the interfacial bonding force.

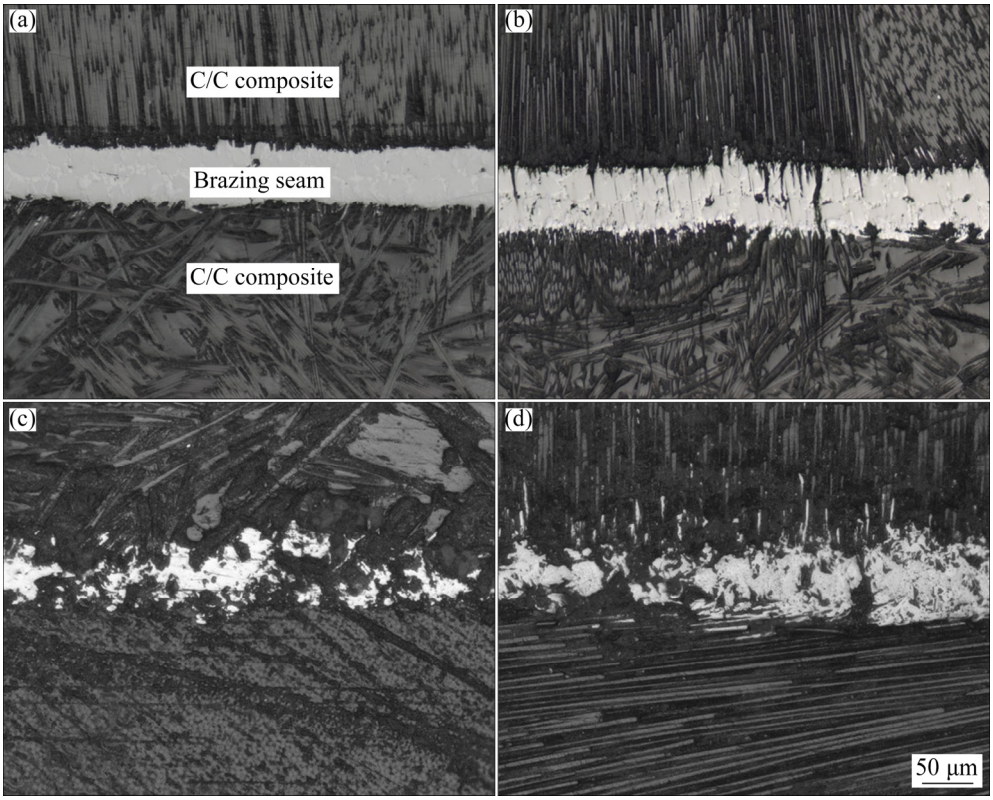


**Fig. 3** Optical images of C/C joints at different brazing temperatures: (a) 1080 °C; (b) 1100 °C; (c) 1120 °C; (d) 1140 °C

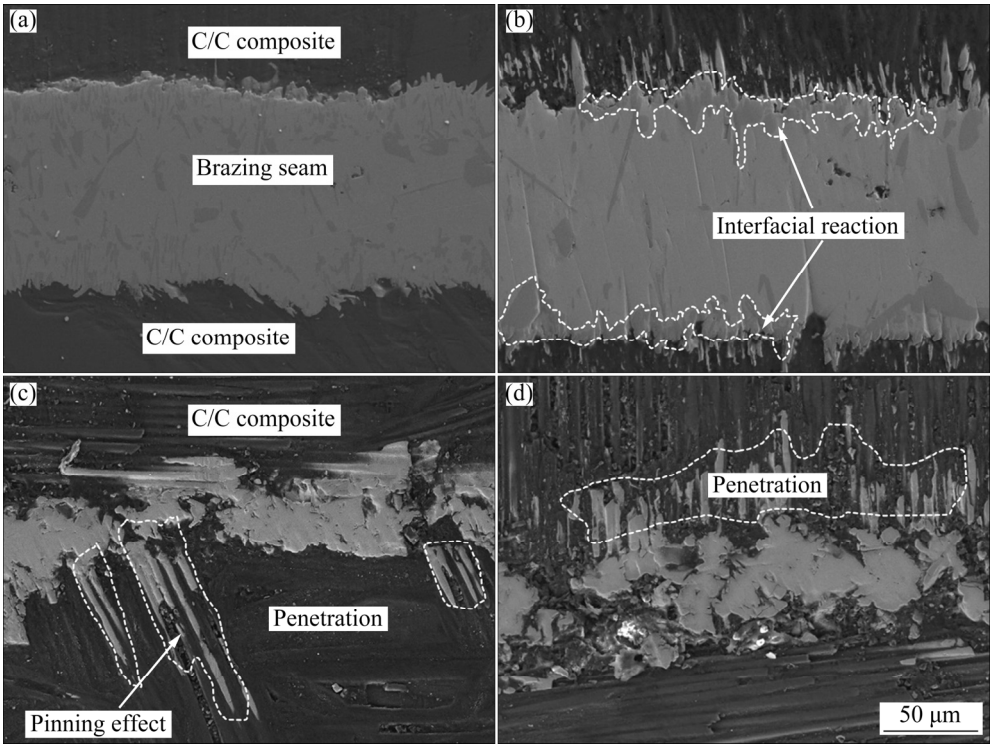


**Fig. 4** SEM images of C/C composite brazed joints at different temperatures: (a) 1080 °C; (b) 1100 °C; (c) 1120 °C; (d) 1140 °C

Figure 5 shows the magnified morphologies of the brazed C/C joint (brazing temperature: 1120 °C). Table 2 gives the EDS results of each area in Fig. 5. According to composition, the dark gray phase was $Cr_3C_2$, which was formed by the reaction of active element Cr with C at the interface. The reaction of

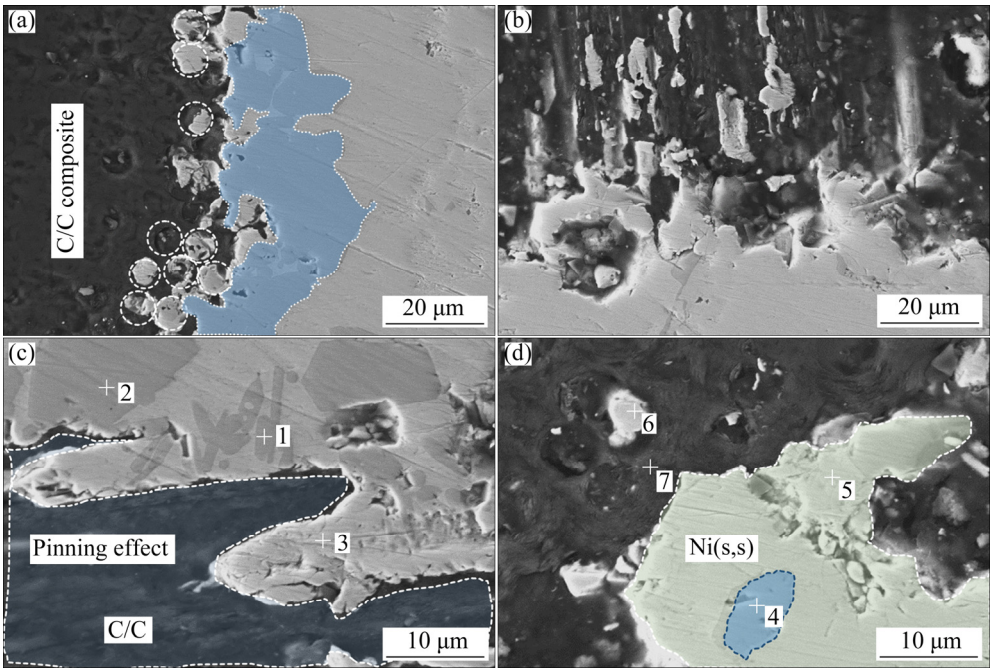


**Fig. 5** Interfacial microstructures of brazed joints (1120 °C): (a) Molten filler filling carbon fibers; (b) Filler penetration into matrix; (c, d) Interface of Ni(s,s) phase

**Table 2** Composition of C/C composite brazed joints (wt.%)

| Point | C | Cr | Ni | Co | Possible phase |
|---|---|---|---|---|---|
| 1 | 53.36 | 45.38 | 0.46 | 0.8 | $Cr_3C_2$ |
| 2 | 41.43 | 3.08 | 55.49 | − | Ni(s,s) |
| 3 | 44.51 | 55.49 | − | − | $Cr_3C_2$ |
| 4 | 51.36 | 46.47 | 1.36 | 0.81 | $Cr_3C_2$ |
| 5 | 35.18 | 3.02 | 61.8 | − | Ni(s,s) |
| 6 | 28.01 | 2.8 | 69.19 | − | Ni(s,s) |
| 7 | 100 | − | − | − | C |

Cr and C was as follows:

$$2C+3Cr\rightarrow Cr_3C_2 \quad (1)$$

$$\Delta G(Cr_3C_2)=-10500-0.3T \quad (2)$$

When the temperature was 1080 °C, the Gibbs free energy change of $Cr_3C_2$: $\Delta G<0$, so it was proven from the perspective of thermodynamics that this reaction could occur during brazing.

The interface-enriched $Cr_3C_2$ layer was thin when the brazing temperature was low, and the pinning effect on the C/C composites was not obvious. When the temperature reached 1100 °C, the diffusion ability of Cr in the brazing seam was enhanced, and the $Cr_3C_2$ layer at the interface was thickened. The thickened reaction layer caused the distribution of $Cr_3C_2$ to become continuous at the interface. Because brazing temperature increased, the wettability of filler was improved, and the molten filler filled the position of carbon fibers, as shown in Fig. 5(a). Compared with the parallel area, the wetting at the vertical area was intenser, and Ni-based brazing filler extended into the matrix along carbon fibers (Fig. 5(b)). At the interface close to the C/C composite, there was also light gray Ni solid solution (Ni(s,s)) besides $Cr_3C_2$ phases, and the interface was irregular to increase the joining area (Figs. 5(c, d)).

After brazing, a certain penetration of the brazing seam into the C/C matrix occurred, and a wrapping effect was formed on part of the carbon matrix along the C/C composites, indicating that the brazing filler was in full contact with the matrix, as shown in Fig. 6.

To get a better composition of the brazing seam, the area distribution of C, Ni, and Cr is obtained, as shown in Fig. 7. The brazing seam consisted of Ni(s,s) and $Cr_3C_2$. The size of Ni(s,s) was very large and some $Cr_3C_2$ phases connected into a large region.

### 3.3 Shear strength of brazed joints

Figure 8 shows the shear strength of the C/C joints brazed at different temperatures. When the

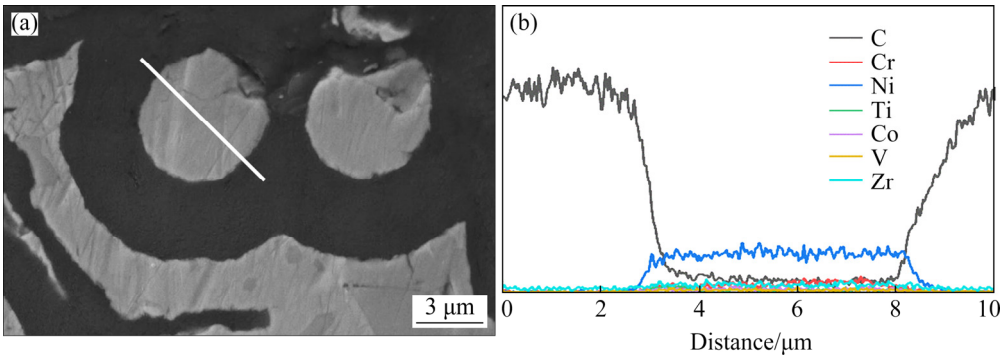


**Fig. 6** Line scanning at interface between C/C composite and brazing seam (1120 °C): (a) Scanning position; (b) Elemental distribution

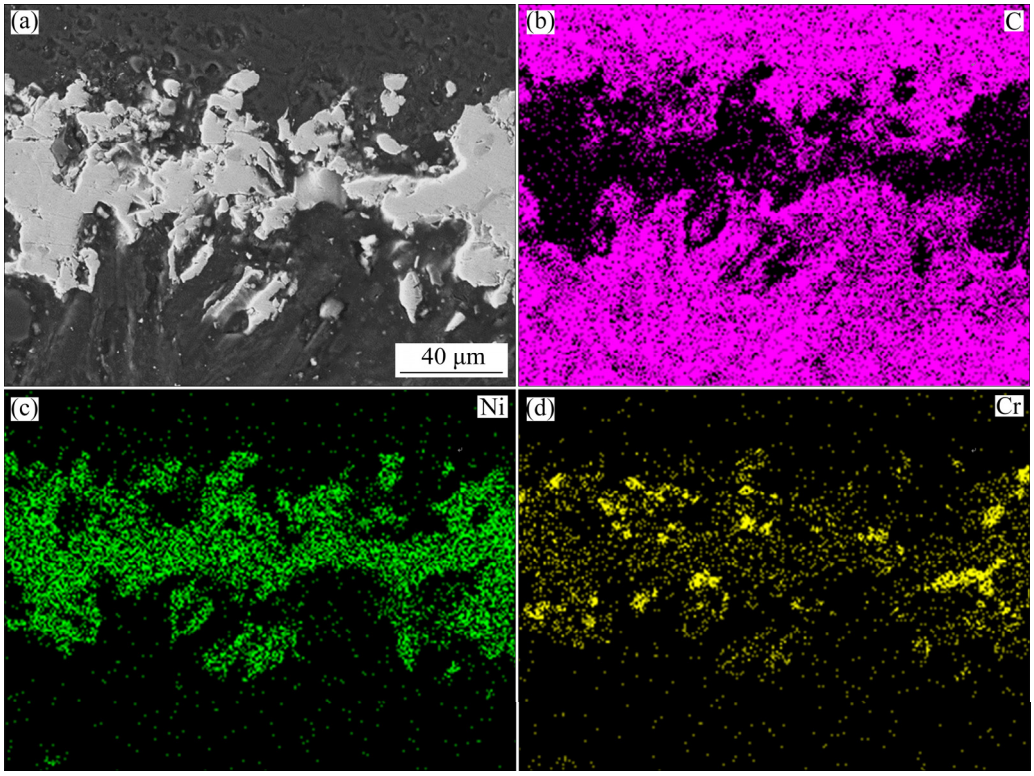


**Fig. 7** Elemental distributions of C/C joint brazed at 1120 °C

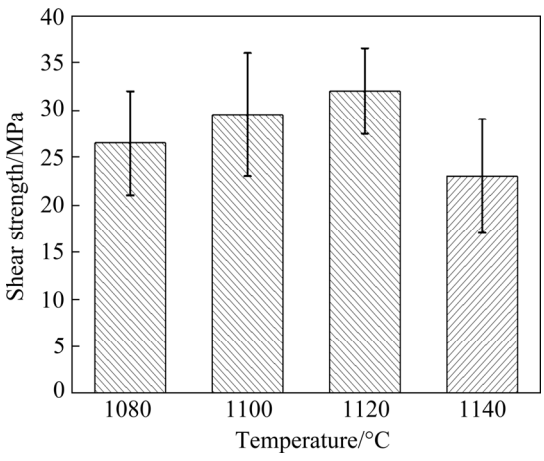


**Fig. 8** Shear strength of brazed C/C joints

temperature was 1080 °C, the average shear strength of C/C joints reached 26.8 MPa. As brazing temperature increased, the shear strength also increased. When the temperature was 1120 °C, the shear strength of joint reached the maximum value of 31.5 MPa. The reason should be related to the strong interface bonding and pinning effect. However, as brazing temperature increased further, the shear strength of joint began to decrease.

Figure 9 shows the fracture surface of C/C joint. From the SEM image of fracture surface, cracks initiated in the C/C composites close to the interface. The fracture path extended at the bonding interface, but did not enter the brazing seam.

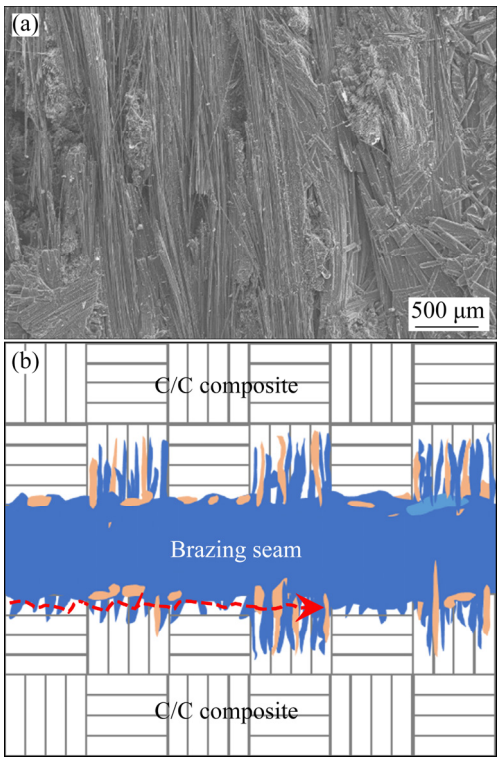


**Fig. 9** Morphology of fracture surface of C/C joint after shear test: (a) Fracture surface (1120 °C); (b) Crack initiation position

## 4 Conclusions

(1) The mixed Ni-based brazing filler had good wettability on the surface of C/C composites at high temperature. When brazing temperature increased from 1080 to 1100 °C, the wetting angle decreased from 23° to 14°.

(2) The brazing seam consisted of Ni(s,s) and $Cr_3C_2$ phases. As temperature increased, much $Cr_3C_2$ was generated at the bonding interface between brazing seam and C/C matrix, and a reaction layer was formed with thickness of about 10 μm.

(3) When brazing temperature was 1120 °C, the shear strength of C/C joint reached the maximum value of 31.5 MPa. Judged from fracture surface, the fracture path extended in the C/C matrix close to the bonding interface.

## Acknowledgments

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# 采用 Ni 基钎料连接 C/C 复合材料接头的显微组织和力学性能

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**摘 要**：采用新型 Ni 基钎料连接 C/C 复合材料。当钎焊温度从 1080 ℃升高至 1100 ℃时，润湿角从 23°降至 14°，该钎料在 C/C 复合材料表面具有良好的润湿性能。焊缝由 Ni 固溶体和 $Cr_3C_2$ 组成。随着钎焊温度的升高，大量的 $Cr_3C_2$ 在连接界面处产生，并形成一定厚度的反应层。当焊接温度达到 1120 ℃时，C/C 复合材料连接接头的剪切强度达到最大值 31.5MPa，其断裂路径为在靠近连接界面的 C/C 复合材料基体内延伸。

**关键词**：钎焊；显微组织；剪切强度；C/C 复合材料

**(Edited by Bing YANG)**