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Microstructure and formation mechanism of SHS joining between C_f/Al composites and TiAl intermetallic with Al–Ni–CuO interlayer

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Abstract In this study, it was reported a novel approach for joining C_f/Al composites and TiAl intermetallic by self-propagating high-temperature synthesis (SHS). Mixed powders of 14Al–2Ni–3CuO were used as the SHS interlayer, and differential thermal analysis test of Al–Ni–CuO interlayer was conducted to analyze the exothermic characteristic. Sound joint was got by SHS joining under the conditions of 600 °C, 30 min, and 5 MPa. The joint was characterized by scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and X-ray diffraction (XRD). $TiAl_3$ and $NiAl_3$ are, respectively, formed in the TiAl/interlayer and C_f/Al/interlayer interfaces. Reaction products of Ni_2Al_3, $NiAl_3$, Al_2O_3, and Cu were observed in the interlayer. And the formation mechanism of SHS joining was investigated.

Keywords SHS; Joining; Microstructure; Formation mechanism; Al–Ni–CuO interlayer

1 Introduction

Carbon fiber-reinforced aluminum matrix composites (C_f/Al composites) have low density and thermal expansivity, high specific strength and stiffness, and excellent electrical and thermal conductivity. Owing to their excellent physical and mechanical properties, C_f/Al composites have successfully demonstrated their potential in aerospace and automotive applications [1, 2]. In these fields, reliable joining, especially with other materials, is required, and which is also an essential way to widen the application of the composites. Titanium aluminide is characterized by low density and high specific strength at elevated temperatures compared with other materials, and is regarded as the potential alternative for titanium alloys in aircraft industry [3–6]. The development of joining technique between C_f/Al composites and TiAl intermetallic can significantly expand their applications, which has great meanings. However, joint of C_f/Al composites and TiAl intermetallic is not easy to be obtained due to their great differences in properties, and conventional welding methods cannot achieve satisfied joints with lower cost. Clearly, innovative method should be developed.

Self-propagating high-temperature synthesis (SHS) is a promising technique, and can be used to synthetize ceramics, intermetallic compound and composites, join dissimilar materials [7–14], especially materials with high melting points. During the joining process, heat is released by interlayer's exothermal reaction and localized to the joint's interface, which can be used to join temperature sensitive materials without thermal damage [15].

In this work, Al–Ni–CuO interlayer was used to join C_f/Al composites and TiAl intermetallic by self-propagating high-temperature synthesis. Microstructure and formation mechanism of the joining process were investigated.

2 Experimental

All the experiments were carried out with the mixture of high purity Al (99.9 %, 45 μm), Ni (99.9 %, 58 μm), CuO (99.9 %, 50 μm) powders. Appropriate amounts of Al, Ni, and CuO powders were weighed out according to the composition of 14Al–2Ni–3CuO (at%). Milling was performed at room temperature in a hardened steel bowl using

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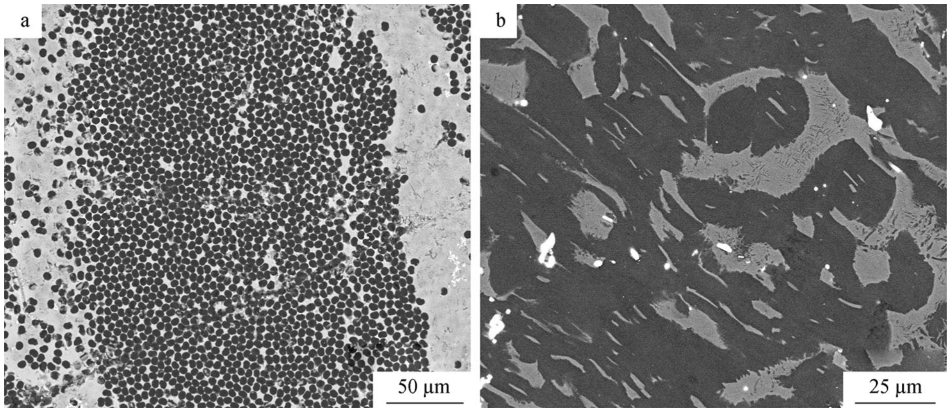

Fig. 1 SEM images of C_f/Al composites and TiAl intermetallic: **a** TiAl intermetallic and **b** C_f/Al composites

Al_2O_3 balls for 2 h, with the rotation speed of 300 r·min^{-1}. The ball-to-powder weight ratio was 10:1. Then milled powders were cold pressed into cylindrical shape of 10 mm in diameter and 0.5 mm in height under the pressure varying from 10 to 20 MPa. The green density of the specimen ranged from 55 % to 65 % of theoretical density.

TiAl intermetallic applied in this study has the nominal composition of Ti–48Al–7V–0.3Y. The microstructure of TiAl mainly consisted of γ-TiAl and α_2-Ti_3Al. The maximum tensile strength of TiAl substrate at room temperature reaches 451.2 MPa.

C_f/Al composites applied in this study has the density of 2.189 g·cm^{-3}, and were achieved through extrusion casting method, with fiber volume percent of 50 % and metal matrix of 6,061 aluminum alloy. Maximum tensile strength of C_f/Al composites at room temperature reaches 558 MPa. Microstructures of TiAl intermetallic and C_f/Al composites are shown in Fig. 1. Specimens were cut to 5 mm × 3 mm × 3 mm for metallographic analysis. All joined surfaces were polished by SiC papers up to 1200 grit and ultrasonically cleaned by acetone prior to the SHS joining.

Green compacts were sandwiched between C_f/Al composites and TiAl intermetallic for direct SHS joining. The joining system was heated to 600 °C under the pressure of 5 MPa with the heating rate of 10 °C·min^{-1}. Then the system was hold in 600 °C for 30 min. Schematic of SHS joining apparatus is shown in Fig. 2.

Differential thermal analysis (DTA) test of Al–Ni–CuO interlayer was conducted to analyze the exothermic characteristic prior to the SHS joining. SHS joining was conducted in the radiation heating vacuum diffusion welding machine, whose studio vacuum degree was able to reach 1.33 × 10^{-4} Pa. After joining process, microstructure and phase composition of reaction products were characterized employing scanning electron microscopy (SEM, S-4700) and X-ray diffraction (XRD).

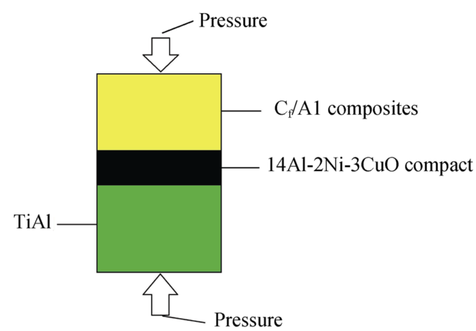

Fig. 2 Schematic of SHS joining apparatus

3 Results and discussion

3.1 DTA analysis of interlayer

DTA test of Al–Ni–CuO interlayer was conducted prior to the SHS joining. DTA result was obtained using a heating rate of 10 °C·min^{-1} under argon flow on the initial powders. Figure 3 represents the DTA curve of the interlayer. Two exothermic peaks and an endothermic peak are detected in the curve, which, respectively, correspond to

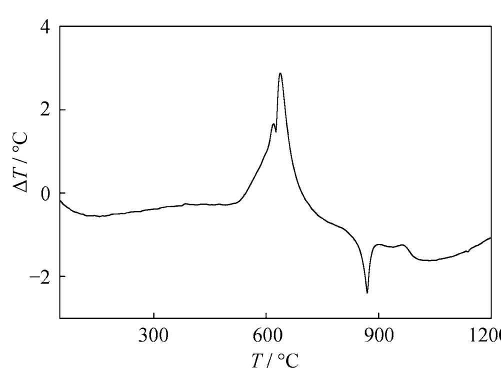

Fig. 3 DTA curve of Al–Ni–CuO interlayer

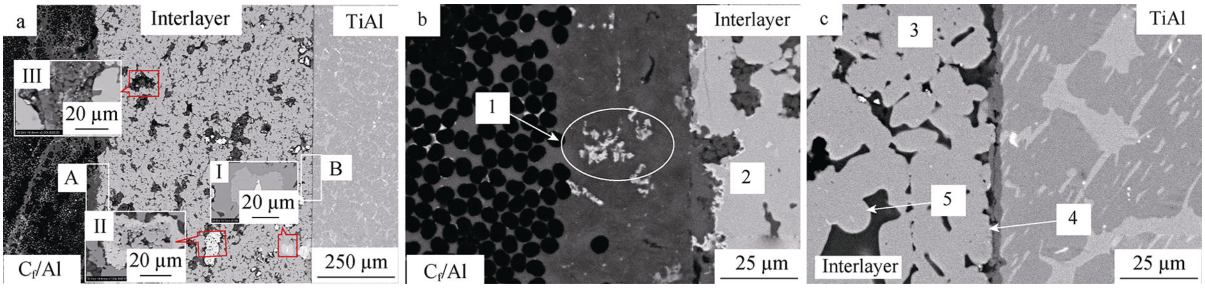

Fig. 4 SEM images of joint microstructure ($T = 600$ °C, $t = 30$ min): **a** overall SHS joint interface, **b** joint region at the interface (Location A), and **c** joint region at interface (Location B)

Table 1 EDS results of spots in Fig. 4 (at%)

Spots	Al	Ni	Ti	Si	Possible phases
1	75.61	24.39	0	0	$NiAl_3$
2	77.17	22.83	0	0	$NiAl_3$
3	76.40	23.60	0	0	$NiAl_3$
4	74.28	0	19.12	6.61	$TiAl_3$, Si
5	100	0	0	0	Al

Table 2 EDS results of spots in Fig. 5 (at%)

Spots	Al	Ni	O	Cu	Mg	Possible phases
1	–	100	–	–	–	Ni
2	64.38	35.62	–	–	–	Ni_2Al_3
3	76.40	23.60	–	–	–	$NiAl_3$
4	–	–	44.99	55.01	–	CuO
5	–	–	–	100	–	Cu
6	19.01	–	42.53	–	38.46	MgO, Al_2O_3
7	40.42	–	35.14	–	24.44	Mg–Al–O

619, 637, and 870 °C. First exothermic peak starts when the temperature reaches 500 °C. It can be seen from the curve that, the second exothermic peak releases the largest quantity of heat, which supplies the most quantity of heat needed in the SHS joining process.

3.2 Interfacial microstructure of SHS joint

SEM image of the SHS joint is shown in Fig. 4a. It clearly indicates that the joining between interlayer and base metals is reliable without obvious pores or flaws. Obvious continuous layer generates in the interface between TiAl intermetallic and interlayer. New white phase is observed in the interlayer and the interface in C_f/Al composites side. In order to further investigate the interfacial microstructure, more details are presented in a larger visual field in back scattered electron (BSE) imaging mode, as shown in Fig. 4b, c and the EDS results taken from different phases in Table 1.

As seen from Table 1, compositions of Spots 1, 2, and 3 are almost the same with each other. According to related binary phase diagrams, the possible phase of Spots 1, 2, and 3 is $NiAl_3$, and the possible phases of Spots 4 and 5 are, respectively, $TiAl_3$–Si solid solution and residual Al.

Three obvious areas are observed in the interlayer. They are gray region I which has bright white phase inside, faveolate white region II, and erose black region III. In order to observe them conveniently, SEM images of Regions I, II, and III taken at higher magnification are shown in Fig. 5.

Phases in three regions are identified by EDS, and analysis results are shown in Table 2. According to Ni–Al phase diagrams, it could be confirmed that three phases in Region I, which correspond to Spots 1, 2, and 3 in Fig. 5, are residual Ni, Ni_2Al_3, and $NiAl_3$. Molar contents of nickel in Ni, Ni_2Al_3, and $NiAl_3$ are, respectively, 100 %, 40 %, and 25 %, which show a decreasing trend. This is

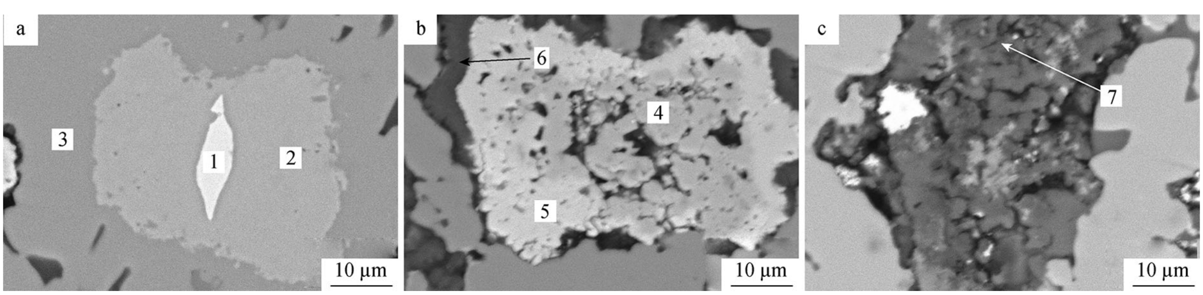

Fig. 5 SEM images of Regions I, II, and III taken at higher magnification: **a** Region I, **b** Region II, and **c** Region III

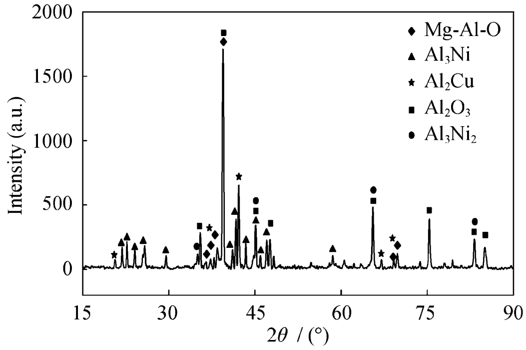

Fig. 6 XRD pattern of joint fracture surface

possibly caused by the interdiffusion of Ni and Al. Molten Al first reacts with Ni particles and generates $NiAl_3$ layer. As the proceeding of the reaction, $NiAl_3$ layer becomes thicker. Only a few Ni atoms can diffuse through the layer and react with Al, which causes the decrease of nickel's molar contents and generation of Ni_2Al_3.

Phase 4 centered in Region II is residual CuO. Cu surrounds CuO, which demonstrates the reaction between Al and CuO. As can be seen from the faveolate structure in the interlayer, the reaction between Al and CuO is nearly complete, Al_2O_3 generates and only a little CuO remains. The metal matrix of C_f/Al composites is 6061 aluminum alloy, whose alloying elements are Mg and Si. A small quality of impurity such as MgO, Mg, etc, also exist in the CuO powders. Mg in the metal matrix and the impurity of CuO powders reacts with CuO and generates MgO, which exists as the black phase 6 and surrounds Cu. In Region III, there is a mixture of MgO, residual Al, etc. Shear test of the joint is conducted, and joint is fractured in the interlayer. XRD pattern of the joint fracture surface is conducted. The result is shown in Fig. 6, which proves the correctness of the above inference.

3.3 Formation mechanism of SHS joint

As can be seen from the above analysis, C_f/Al composites and TiAl intermetallic are joined by the heat released from reactions in the interlayer and the quantity of heat influences, and determines joint's quality. $NiAl_3$, $TiAl_3$, Ni_2Al_3, Cu, and Al_2O_3 generate in the interlayer, and form the joint. From the macroscopic perspective, enough quantity of generated heat, higher heating temperature, and longer holding time make aluminum melt completely in the interlayer, and the interlayer joins well with base metals in both sides. In order to analyze the heat's influence on joint's interface and properties, it is necessary to investigate the formation mechanism of SHS joining, and analyze interfaces' forming process in microscopic view. The reaction process of joint mainly consists of following parts.

(1) Physical contact between the interlayer and joining materials. The interlayer is sandwiched between C_f/Al composites and TiAl intermetallic for direct self-

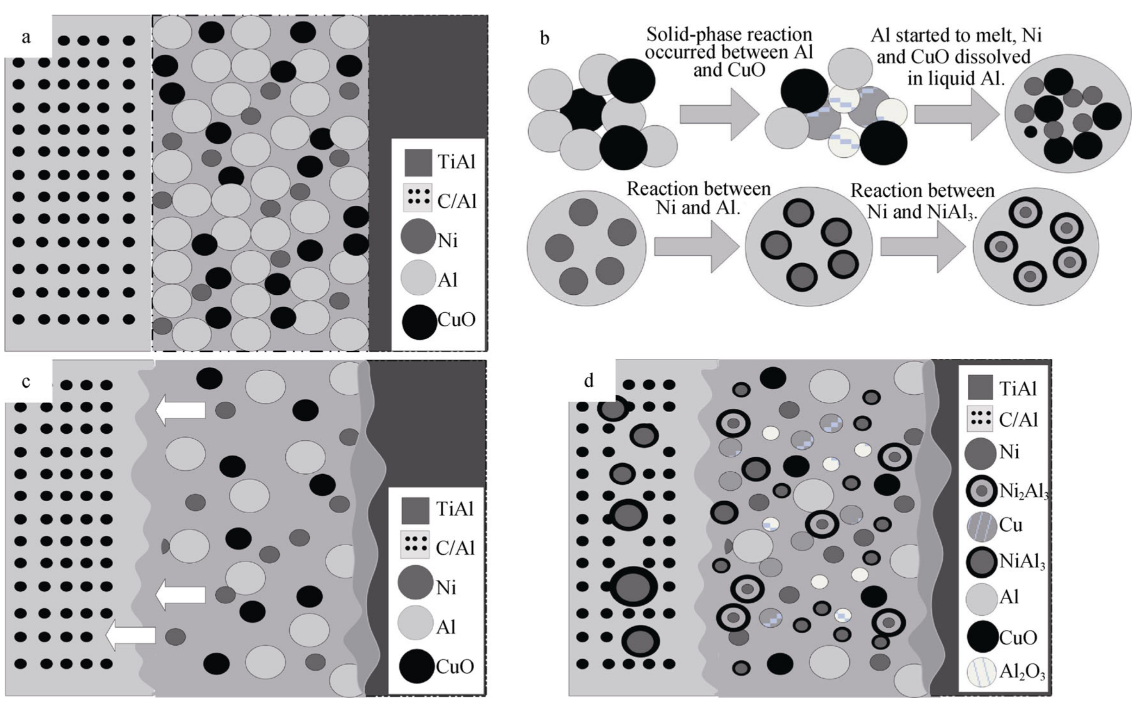

Fig. 7 Mechanism schematic of C_f/Al and TiAl SHS joining with Al–Ni–CuO interlayer: **a** physical contact process, **b** SHS reaction of Al–Ni–CuO in the interlayer, **c** reaction between interlayer and C_f/Al, TiAl, and **d** formation of final joint

propagating joining under certain pressure, which is shown in Fig. 7a. When temperature is low, interlayer and base metal in both sides contact physically under small pressure. But the pressure is too small to make them contact completely. Exothermic reactions occur as temperature gradually increases, liquid aluminum emerges. Then interlayer and base metals in both sides gradually get completely contacted.

(2) SHS of Al–Ni–CuO interlayer. When temperature reaches 500 °C, solid-phase reactions occur between Al and CuO, and releases small quantity of heat. Al_2O_3 and Cu generate in the reaction. First exothermic peak of the interlayer's DTA curve starts as the quantity of heat releases by the reaction between Al and CuO. Temperature in interlayer's local area reaches the melt point of Al, and liquid aluminum emerges in the interlayer and surrounds Ni and CuO particles. Ni particles react with liquid Al and generate $NiAl_3$, release large quantity of heat. Owing to the heat released from the reaction, the whole interlayer's temperature almost reaches adiabatic temperature of the interlayer, which is calculated to be 1,767 °C. The aluminum matrix of C_f/Al composites, which contacts with interlayer, melts.

(3) Formation of the final joint. Interlayer's temperature increases rapidly as the proceeding of Al–Ni–CuO SHS reaction. On the TiAl side, melted aluminum reacts with TiAl and forms $TiAl_3$ layer, which is shown in Fig. 7b. $TiAl_3$ layer becomes continuous and thicker gradually as the increase of holding time. On the C_f/Al side, Al particles in the interlayer and aluminum matrix of C_f/Al melt and dissolve. Interlayer's Ni particles react with molten aluminum matrix of C_f/Al and forms $NiAl_3$. In the interlayer, Al reacts with CuO, forming Al_2O_3. Liquid Al surrounds Ni particles. Al reacts with Ni and forms $NiAl_3$ layer, which in turn surrounds Ni particles. With the reaction proceeding, $NiAl_3$ layer becomes thicker and thicker, separating Ni particles from liquid Al. As the further reaction occurs between Al and Ni particles, $NiAl_3$ layer gets cut off. As temperature continues to increase, internal Ni reacts with ambient $NiAl_3$ and forms Ni_2Al_3. Consequently, Ni_2Al_3 layer surrounds Ni particles, outmost layer is $NiAl_3$ layer.

4 Conclusion

In this study, joining of C_f/Al and TiAl by SHS was successfully performed, and sound joint was got. Typical microstructure of SHS joint was investigated. Adjacent to the TiAl substrate, $TiAl_3$ reaction layer is detected in the interface. Reaction occuring in the interlayer is not complete. Unreacted Ni, Al, and CuO are observed in the interlayer. Reaction products Ni_2Al_3, $NiAl_3$, Al_2O_3, and Cu are distributed in the interlayer. Adjacent to the C_f/Al substrate, Ni reacts with Al in the matrix of C_f/Al and forms $NiAl_3$. Reaction process of joint consists of three parts. First, interlayer and joining materials contact physically. Second, SHS occurs in the Al-Ni-CuO interlayer. Finally, SHS joint of C_f/Al composites and TiAl intermetallic forms.

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