Available online at www.sciencedirect.com
ScienceDirect
Trans. Nonferrous Met. Soc. China 22(2012) 324−329
Transactions of Nonferrous Metals Society of China
www.tnmsc.cn

Microstructure and shear strength of reactive brazing joints of TiAl/Ni-based alloy

LI Hai-xin[1], HE Peng[1], LIN Tie-song[1], PAN Feng[1], FENG Ji-cai[1], HUANG Yu-dong[2]

1. State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Harbin 150001, China;
2. School of Chemical Engineering and Technology, Harbin Institute of Technology, Harbin 150001, China

Received 9 March 2011; accepted 11 May 2011

Abstract: Reactive brazing of TiAl-based intermetallics and Ni-based alloy with Ti foil as interlayer was investigated. The interfacial microstructure and shear strength of the joints were studied. According to the experimental observations, the molten interlayer reacts vigorously with base metals, forming several continuous reaction layers. The typical interfacial microstructure of the joint can be expressed as GH99/ $(Ni, Cr)_{ss}$ (γ)/TiNi (β_2)+$TiNi_2Al$ (τ_4)+Ti_2Ni (δ)/δ+Ti_3Al (α_2)+Al_3NiTi_2 (τ_3)/α_2 +τ_3/TiAl. The maximum shear strength is 258 MPa for the specimen brazed at 1000 °C for 10 min. Higher brazing temperature or longer brazing time causes coarsening of the phases in the brazing seam and formation of brittle intermetallic layer, which greatly depresses the shear strength of the joints.

Key words: TiAl; Ni-based alloy; reactive brazing; interfacial microstructure; shear strength

1 Introduction

TiAl-based alloys are regarded as excellent materials for high temperature applications, because of their low density, high melting temperature and high temperature strength [1, 2]. Studies of the practical application of TiAl for components in aircraft and automobile have been carried out [2, 3]. In order to further extend their scopes of application and elevate the operating temperature of thermal structures, the joining of TiAl and Ni-based superalloys is a promising approach [4, 5]. As a result, various joining processes have been utilized to join these two alloys [5−7]. Since TiAl-based intermetallics suffer from severe low-temperature brittleness and high crack sensitivity, it is difficult to obtain satisfactory joints using conventional fusion welding techniques [8]. It has been reported that sound joints could be achieved by solid-state diffusion bonding, such as superplastic forming/diffusion bonding (SPF/DB) [5] and reaction-assisted diffusion bonding [6, 9]; however, the complex process, long processing time and high pressure may limit this method to practical applications [10]. Vacuum brazing is considered to be a good choice to solve these problems [8, 10]. However, a few studies have been reported on brazing TiAl-based intermetallics to Ni-based alloys. TETSUI [11] used silver brazing to join TiAl and Ni-based alloy. However, the joint obtained by this method cannot satisfy high-temperature applications (above 500 °C). Because TiAl-based intermetallics and Ni-based alloys are often used in high-temperature environment, brazing fillers expected to have certain high-temperature strength should be selected for brazing. Researchers have demonstrated that Ti-based braze alloys can be used for brazing TiAl-based intermetallics [12, 13]. Compared with Ag-based braze alloys, Ti-based braze alloys can be suitable for higher temperature applications, especially Ti−Ni braze alloys (a lowest melting temperature of 942 °C). Furthermore, Ti−Ni braze alloy has been used as insert metals to join TiAl and Inconel 718 by employing transient liquid phase bonding technology [7]. However, there is little information on the interfacial microstructure and mechanical properties of the joints.

Considering that a Ti−Ni eutectic reaction occurs

Foundation item: Projects (50975062, 51105107, 51021002) supported by the National Natural Science Foundation of China; Projects (QC2011C044) supported by the Natural Science Foundation of Heilongjiang Province, China; Project (20112302130005) supported by the Specialized Research Fund for the Doctoral Program of Higher Education, China; Project (CUGA4130902510) supported by the China Postdoctoral Science Foundation Funded

Corresponding author: LIN Tie-song; Tel/Fax: +86-451-86403422; E-mail: hitjoining@hit.edu.cn

DOI: 10.1016/S1003-6326(11)61178-3

between Ti foil and Ni-based alloy, a Ti foil was chosen in this study as the interlayer for reactive brazing the TiAl-based intermetallics to the Ni-based alloy. The interfacial microstructure and effects of brazing parameters on the microstructure along with the shear strength of the brazed joints were also investigated.

2 Experimental

The nominal composition of the TiAl-based intermetallics and the Ni-based alloy (type GH99) used in this study is shown in Table 1. The TiAl-based intermetallics and the GH99 alloy were cut into the dimensions of 7.0 mm×5.0 mm×2.0 mm and 14.0 mm×5.0 mm×1.2 mm, respectively. Titanium foil, which was cut into slices of the same size as TiAl-based intermetallics, was used as the brazing interlayer. The thickness of titanium foil was 80 μm. The schematic diagram of assembling the brazing parts is shown in Fig. 1. The materials of the three brazing parts, TiAl-based intermetallics, Ni-based alloy and Ti foil should be taken through grinding and cleaning steps carefully before assembling.

Table 1 Chemical composition of materials

Material	x(Al)/%	x(Ni)/%	x(Cr)/%	x(Ti)/%	x(V)/%
TiAl	52.82	−	1.20	44.88	1.10
GH99	5.44	57.29	20.63	1.96	0.42

Material	x(Co)/%	x(W)/%	x(Mo)/%	x(Si)/%
TiAl	−	−	−	−
GH99	7.46	3.03	2.98	0.80

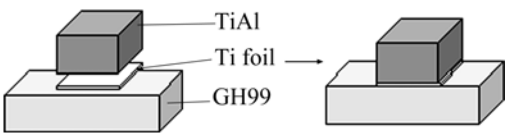

Fig. 1 Schematic diagram of assembling brazing parts

The specimens were brazed at temperature ranging from 960 °C to 1040 °C for 1 to 30 min in a vacuum furnace (1 MPa). Heating and cooling rates of 10 °C/min were used. After brazing, the joint plate was cut perpendicular to the joint interface, and the cross-sections of the joints were prepared by using standard metallographic techniques. The microstructures of the joints were examined by scanning electron microscopy (SEM, S−570). The chemical analysis of the reaction products was performed by electron probe X-ray microanalysis (EPMA, JXA−8600). Shear tests were performed on an Instron−1186 universal testing machine to evaluate the shear strength of the brazed joints. The brazed specimens were compressed with a cross-head speed of 1 mm/min.

3 Results and discussion

3.1 Microstructure of TiAl/Ti/GH99 joints

The interfacial microstructures of the TiAl/Ti/GH99 joints brazed at 1000 °C for 5 min are shown in Fig. 2. It can be clearly seen from Fig. 2(a) that four kinds of reaction layers occurred between the TiAl-based intermetallics and the GH99 alloy, marked by layers I, II, III and IV. Layer I is a diffusive layer of the GH99 alloy, which contains white dispersed phase. Layer II is composed of coarse grayish black phase, a small quantity of grayish phase and grayish white blocky phase. Layer III consists of three kinds of phases: coarse grayish phase is the matrix in which coarse grayish black phase and grayish white blocks disperse. Layer IV contains lamellar grayish phases which disperse in a grayish black reaction layer adjacent to the TiAl-based intermetallics substrate.

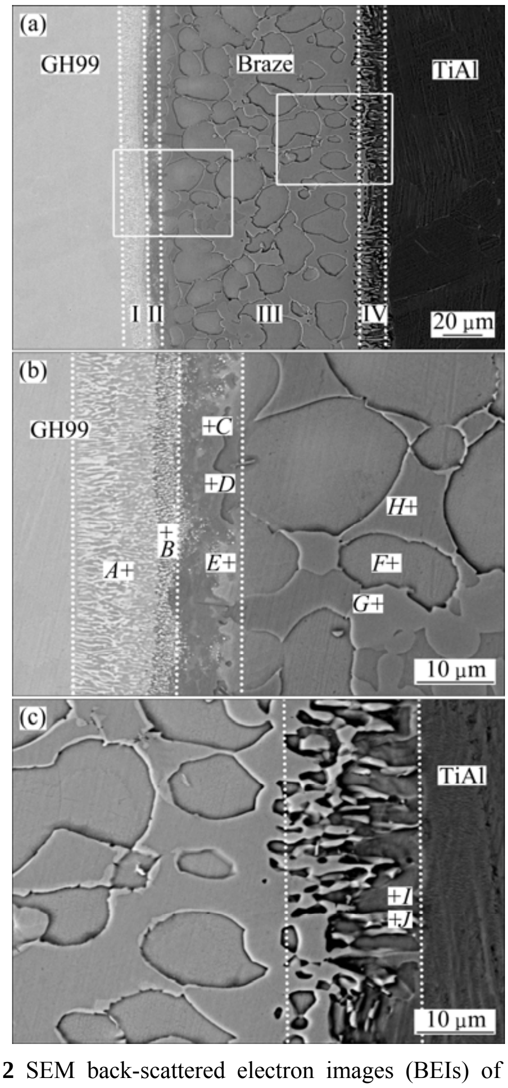

Fig. 2 SEM back-scattered electron images (BEIs) of joints brazed at 1000 °C for 5 min: (a) Whole joint interface; (b) Brazing layer close to GH99 alloy; (c) Brazing layer close to TiAl-based intermetallics

Table 2 Average contents of major elements of TiAl/Ti/GH99 joint brazed at 1000 °C for 5 min

Point	x/%							Possible phase
	Al	W	Mo	Co	Ti	Cr	Ni	
A	8.06	3.93	3.46	5.66	5.73	25.12	48.04	γ
B	9.55	4.46	5.08	4.80	17.64	28.90	29.33	γ
C	9.65	4.03	0.20	9.04	36.76	2.28	38.03	β_2
D	25.46	0.58	0.29	0.39	26.98	1.97	44.33	τ_4
E	11.05	4.75	0.34	2.51	45.89	7.80	27.66	δ
F	19.87	3.24	0.29	3.21	54.39	8.22	11.07	α_2
G	11.05	0.71	0.38	0.32	53.88	3.41	27.95	δ
H	22.55	0.48	−	0.34	45.74	6.22	24.68	τ_3
I	36.50	0.39	−	−	58.40	3.19	1.43	α_2
J	41.69	0.19	0.46	0.35	36.78	2.67	18.86	τ_3

The EPMA results of chemical compositions of the spots marked by letters *A*−*J* in Figs. 2(b) and (c) are shown in Table 2. The EPMA results of spots *A* and *B* show that the chemical composition of these regions is close to that of the GH99 alloy, just the quantity of Cr, Co, W, Mo, Al and Ti is higher than that in the GH99 alloy due to the interdiffusion and reaction between the interlayer and the GH99 alloy. The EPMA result of grayish phase designated by spot *C* suggests that the grayish phase is composed of Ti and Ni in 1:1 mole ratio, according to the Ti−Ni binary phase diagram. This phase can be attributed to the TiNi (β_2) phase. The EPMA results of spots *E* and *G* show that these regions in the reaction layer are mainly composed of Ti and Ni, and the molar ratio of Ti to Ni is 2:1, so they can be inferred to be the Ti_2Ni (δ) phase. The EPMA result of coarse grayish black phase designated by spot *D* suggests that the grayish black phase is composed of Ti, Ni and Al in 1:2:1 molar ratio, according to the Ti−Ni−Al ternary phase diagram [14]. This phase can be attributed to the $TiNi_2Al$ (τ_4) phase. The EPMA results of spots *F* and *I* show that these regions in the reaction layer are mainly composed of Ti and Al, and the molar ratio of Ti to Al are about 2.7:1 and 1.6:1, respectively. From the Ti−Al binary phase diagram [15], it is clear that the molar ratios of Ti to Al in the Ti_3Al (α_2) phase range between 1.5:1 and 3.2:1. Therefore, the regions marked by spots *F* and *I* can be attributed to the α_2 phase. The EPMA results of spots *H* and *J* show that these regions in the reaction layer are mainly composed of Ti, Ni and Al, and the molar ratios of Ti, Ni and Al are about 2:1:1 and 2:1:2.2. From the Ti−Ni−Al ternary phase diagram, the location of these two phases is in or near the region of the Al_3NiTi_2 (τ_3) phase. Furthermore, combining with the studies of JULIUS et al [14], the regions marked by spots *H* and *J* can be inferred to be the τ_3 phase.

Thus, the whole brazing process can be simply divided into two steps. Firstly, when the brazing temperature is up to the eutectic temperature of Ti−Ni (942 °C), the liquid forms at the interface of the GH99 alloy and the Ti interlayer by a binary eutectic reaction (1).

$$\beta\text{-Ti}+Ti_2Ni \rightarrow L \tag{1}$$

Due to the diffusion and consumption of Ni of the GH99 alloy, a diffusive layer $(Ni, Cr)_{ss}$ (γ) (here, ss means solid solution), which is rich in alloying elements (namely Cr, Co, W and Mo), forms on the GH99 alloy side of the joints. Similarly, studies of EIJK et al [16] showed that Ni seemed to easily diffuse into the brazing seam, while other alloying elements of superalloy were much more reluctant to diffuse. A diffusive layer which was rich in alloying elements formed. When the Ti interlayer melts completely, the molten interlayer reacts with the TiAl-based intermetallics, and the Al enters into the molten interlayer. Meanwhile, the Ti also diffuses from the molten interlayer to the GH99 alloy and the TiAl-based intermetallics. The primary elements of the brazing seam are Ti, Ni and Al.

Next, since more Ni atoms diffuse from the GH99 alloy into the brazing seam, the isothermal solidification occurs at the interface of GH99 alloy side, and a TiNi layer forms. At 987 °C, a peritectic reaction (2) occurs, which results in the τ_4 and δ phases formed in layer II [14].

$$L+TiNi \rightarrow Ti_2Ni+\tau_4 \tag{2}$$

The α_2 phase of layer Ⅲ forms by reaction (3) from the liquid phase during the isothermal solidification process at the brazing temperature, while the α_2 phase of layer IV forms by reaction (4) between the TiAl-based intermetallics and molten interlayer [17].

$$Al_L+Ti_L \rightarrow Ti_3Al_{L,\ RZ} \tag{3}$$

$$TiAl_S+Ti_L \rightarrow Ti_3Al_{RZ} \tag{4}$$

Studies of JULIUS et al [14] showed that the τ_3 phase coexisted with liquid phase of Ti−Ni−Al above 969 °C. Therefore, it can be considered the τ_3 phase precipitates during the temperature-fall period, and the δ phase precipitates from the residual liquid which is rich in Ti. So, the reaction products of the joints brazed with Ti interlayer are respectively γ, β_2+τ_4+δ, δ+α_2+τ_3 and α_2+τ_3 from the GH99 alloy to the TiAl-based intermetallics side.

3.2 Effects of brazing parameters on microstructures and shear strength of brazed joints

The interfacial microstructures of the joints brazed at 960, 1000 and 1040 °C for 10 min are shown in Fig. 3. It can be seen that there are four reaction layers, and their thickness increases with increasing the brazing temperature rate. When the brazing temperature is lower (960 °C, 10 min), no Ti−Ni−Al alloys are detected in the reaction layers (see Fig. 3(a)). The reason is that the quantity of atomic diffusion is not enough and the dissolution reaction is insufficient between the molten interlayer and the master alloys. When the brazing parameters are 1000 °C and 10 min, α_2, τ_3 and δ phases form and occupy majority of the brazing seam, and these phases of every reaction layer distribute uniformly (see Fig. 3(b)). With a further increase in the brazing temperature (1040 °C, 10 min), the τ_3 and α_2 phases become coarse, and the proportion of the δ phase is reduced because more Al atoms diffuse into the molten interlayer and react with Ti and Ni to the τ_3 and α_2 phases (see Fig. 3(c)).

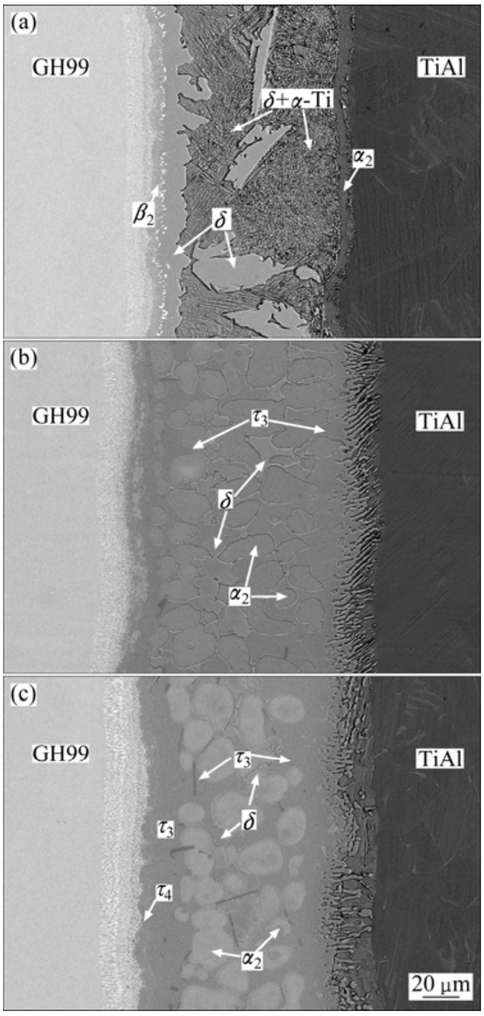

Fig. 3 SEM BEIs of joints brazed at different brazing temperatures for 10 min: (a) 960 °C; (b) 1000 °C; (c) 1040 °C

The interfacial microstructures of the joints brazed at 1000 °C for 1 min and 30 min are shown in Fig. 4. It can be seen that the interfacial microstructures of the joints, which was brazed at 1000 °C for 1 min (see Fig. 4(a)), are basically similar to those of the joint brazed at 1000 °C for 5 min (see Fig. 2(a)), just the thickness of reaction layers is thinner. It can be inferred that the interlayer melts completely and reacts with the master alloys at a high rate when the brazing temperature is 1000 °C. When the brazing time increases, the influence on the change of the interfacial microstructures of the joints is similar to that of brazing temperature increment (see Fig. 4(b)).

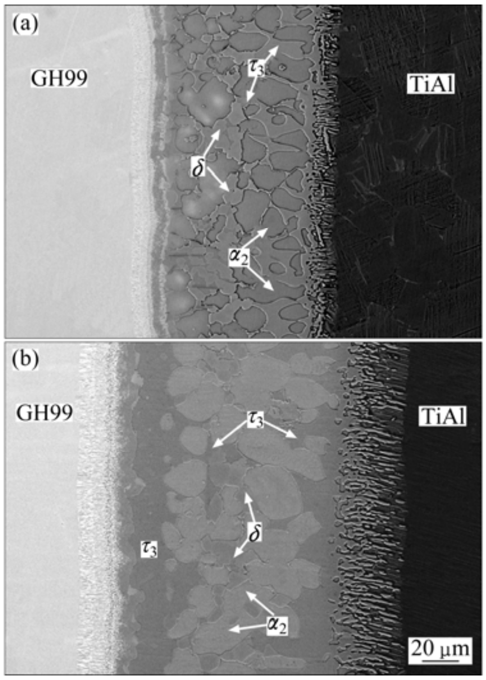

Fig. 4 SEM BEIs of joints brazed at 1000 °C for different brazing time: (a) 1 min; (b) 30 min

The effect of brazing parameters on shear strength of the joints is shown in Fig. 5. The shear strength increases with increasing the brazing temperature or time, and attains a maximum (258 MPa) at 1000 °C for 10 min, and thereafter falls. In a microstructural point of view, the shear strength is largely dependent on the microstructures of joints. When the brazing temperature is lower (960 °C, 10 min), the interlayer melts inadequately and the α_2+τ_3 layer does not appear at the TiAl-based intermetallics boundary (see Fig. 3(a)), which reduces the shear strength remarkably. When the brazing time is short (1000 °C, 1 min), the molten interlayer reacts with the master alloys, but the microstructure of the joints is nonuniform (see Fig. 4(a)). When the brazing parameters are 1000 °C and 10 min, the reaction layers become thick, and the phases of every reaction layer are mixed together and distribute uniformly in the brazing seam (see Fig. 3(b)), which results in the highest shear strength. With a further increase in the brazing temperature or time, the brazing seam is mainly composed of the τ_3 and α_2 phases. In addition, the phases in the brazing seam become coarse and a brittle intermetallic layer forms at the GH99 alloy side, which results in the reduction of the shear strength.

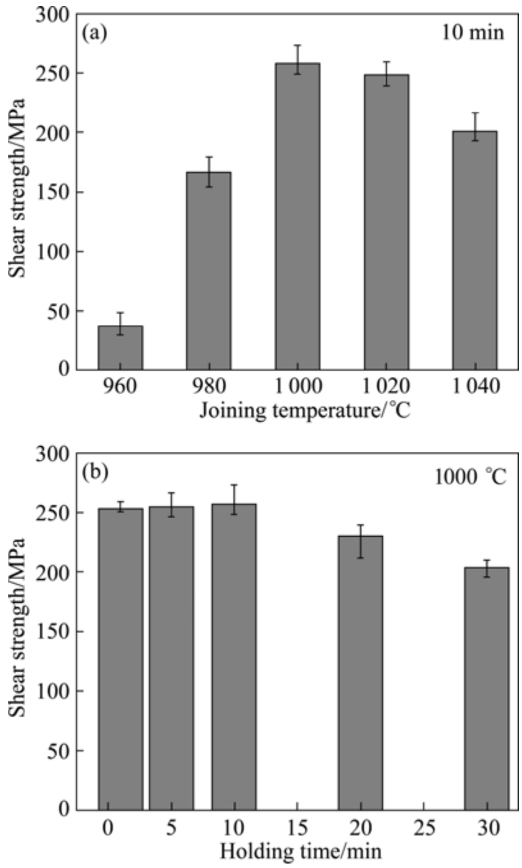

Fig. 5 Effect of brazing parameters on shear strength of joint: (a) Brazed at different temperatures for 10 min; (b) Brazed at 1000 °C for different brazing time

4 Conclusions

1) TiAl-based intermetallics and Ni-based alloy can be joined successfully by reactive brazing with Ti interlayer. The interfacial microstructures of the joints are remarkably affected by the brazing parameters. When the brazing parameters are 960 °C and 10 min, the dissolution reaction is insufficient and no Ti−Ni−Al alloy forms at the reaction layer. When the brazing temperature is 1000 °C, the interlayer melts completely and reacts with the master alloys at a high rate. And four kinds of reaction layers form, namely, γ, $\beta_2+\tau_4+\delta$, $\delta+\alpha_2+\tau_3$ and $\alpha_2+\tau_3$, from the Ni-based alloy to TiAl-based intermetallics side.

2) The relationship among brazing parameters, microstructure and shear strength of the joints was discussed, and the optimum brazing parameters were obtained. The sample brazed at 1000 °C for 10 min has the highest shear strength (258 MPa). Further increasing the brazing temperature or time causes coarsening of brittle phases and formation of brittle intermetallic layer, which greatly depresses the shear strength of the joints.

References

[1] LI Yu-long, FENG Ji-cai, HE Peng. Vacuum brazing of TiAl to 42CrMo with Ag−Cu−Ti filler metal [J]. Transactions of Nonferrous Metals Society of China, 2005, 15(s3): s331−s334.

[2] NODA T. Application of cast gamma TiAl for automobiles [J]. Intermetallics, 1998, 6: 709−713.

[3] LI Yu-long, FENG Ji-cai, HE Peng, CAO Jian. Research progress in brazing technology for TiAl-based alloy [J]. Welding & Joining, 2007, 12: 13−18.

[4] LUO Zhi-chun, WANG Xiu-feng, LIU Xue-bin, LIN Jian-guo. Diffusion bonding of γ-TiAl based alloy to different alloys [J]. Titanium Industry Progress, 2006, 23(6): 20−23.

[5] LUO G X, WU G Q, HUANG Z, RUAN Z J. Diffusion bonding of laser-surface-modified gamma titanium aluminide alloy to nickel-base casting alloy [J]. Script Materialia, 2007, 57: 521−524.

[6] RAMOS A S, VIEIRA M T, SIMOES S, VIANA F, VIEIRA M F. Reaction–assisted diffusion bonding of advanced materials [J]. Defect and Diffusion Forum, 2010, 297−301: 972−977.

[7] DUAN Hui-ping, LUO Jun, ZHANG Tao. Transient liquid phase bonding of TiAl and Inconel 718 [J]. Journal of Beijing University of Aeronautics and Astronautics, 2004, 30(10): 984−988. (in Chinese)

[8] HE P, FENG J C, XU W. Mechanical property of induction brazing TiAl-based intermetallics to steel 35CrMo using AgCuTi filler metal [J]. Materials Science and Engineering A, 2006, 418: 45−52.

[9] RAMOS A S, VIEIRAL M T, SIMOES S, VIANA F, VIEIRA M F. Joining of superalloys to intermetallics using nanolayers [J]. Advanced Materials Research, 2009, 59: 225−229.

[10] SHIUE R K, WU S K, CHEN S Y. Infrared brazing of TiAl intermetallic using BAg-8 braze alloy [J]. Acta Materialia, 2003, 51: 1991−2004.

[11] TETSUI T. Gamma Ti aluminides for non-aerospace applications [J]. Current Opinion Solid state & Materials Science, 1999, 4: 243−248.

[12] FAN Kun, KANG Hui, ZHU Ying, QU Ping. Present research ststus of brazing technology of TiAl-based alloy [J]. Welding Technology, 2004, 33(1): 3−5.

[13] HE P, LIU D, SHANG E J, WANG M. Effect of mechanical milling on Ni-TiH_2 powder alloy filler metal for brazing TiAl intermetallic alloy: The microstructure and joint's properties [J]. Materials Characterization, 2009, 60: 30−35.

[14] JULIUS C S, ZHU P, LIU S H, WEITZER F, DU Y. On the constitution of the ternary system Al−Ni−Ti [J]. Intermetallics, 2007, 15: 1257−1267.

[15] JUNG I S, KIM M C, LEE J H, OH M H, WEE D M. High temperature phase equilibria near Ti-50 at% Al composition in Ti−Al system studied by directional solidification [J]. Intermetallics, 1999, 7(11): 1247−1253.

[16] EIJK C V D, SALLOM Z K, AKSELSEN O M. Microwave brazing of NiTi shape memory alloy with Ag−Ti and Ag−Cu−Ti alloys [J]. Script Materialia, 2008, 58: 779−781.

[17] WALLIS I C, UBHI H S, BACOS M P, JOSSO P, LINDQVIST J, LUNDSTROM D, WISBEY A. Brazed joints in γ TiAl sheet: Microstructure and properties [J]. Intermetallics, 2004, 12: 303−316.

TiAl/Ni 基合金反应钎焊接头的微观组织及剪切强度

李海新[1]，何 鹏[1]，林铁松[1]，潘 峰[1]，冯吉才[1]，黄玉东[2]

1. 哈尔滨工业大学 先进焊接与连接国家重点实验室，哈尔滨 150001；
2. 哈尔滨工业大学 化工学院，哈尔滨 150001

摘 要：以 Ti 为中间层，对 TiAl 基金属间化合物与 Ni 基高温合金进行反应钎焊连接，研究反应钎焊接头的界面微观结构及剪切强度。通过实验发现，熔融中间层与两侧母材反应剧烈，生成连续的界面反应层。典型的界面微观结构为 GH99/(Ni, Cr)$_{ss}$ (γ)/TiNi (β_2)+$TiNi_2Al$ (τ_4)+Ti_2Ni (δ)/δ+Ti_3Al (α_2)+Al_3NiTi_2 (τ_3)/α_2+τ_3/TiAl。当钎焊温度为 1000 °C，保温时间 10 min 时，所得接头的剪切强度最高为 258 MPa。进一步升高钎焊温度或延长保温时间，会引起钎缝组织中组成相粗化和脆性金属间化合物层的生成，从而导致接头剪切强度的降低。

关键词：TiAl；Ni 基合金；反应钎焊；界面微观结构；剪切强度

(Edited by YANG Hua)