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The microstructural observation and wettability study of brazing Ti-6Al-4V and 304 stainless steel using three braze alloys

C. C. LIU, C. L. OU, R. K. SHIUE*
Department of Materials Science and Engineering, National Dong Hwa University, Hualien 974, Taiwan, ROC
E-mail: rkshiue@mail.ndhu.edu.tw

Both Ti-6Al-4V and 304 stainless steels (304SS) are good engineering alloys and widely used in industry due to their excellent mechanical properties as well as corrosion resistance. Well-developed joining process can not only promote the application of these alloys, but also can provide designers versatile choices of alloys. Brazing is one of the most popular methods in joining dissimilar alloys. In this study, three-selected silver base filler alloys, including Braze 580, BAg-8 and Ticusil®, are used in vacuum brazing of 304SS and Ti-6Al-4V. Based upon dynamic sessile drop test, Braze 580 has the lowest brazing temperature of 840°C, in contrast to 870°C for BAg-8 and 900°C for Ticusil® braze alloy. No phase separation is observed for all brazes on 304SS substrate. However, phase separation is observed for all specimens brazed above 860°C on Ti-6Al-4V substrate. The continuous reaction layer between Braze 580 and 304SS is mainly comprised of Ti, Fe and Cu. The thickness of reaction layer at Braze 580/Ti-6Al-4V interface is much larger than that at Braze 580/304SS interface. Meanwhile, a continuous Cu-Sn-Ti ternary intermetallic compound is found at the Braze 580/Ti-6Al-4V interface. Both Ticusil® and BAg-8 brazed joint have similar interfacial microstructures. Different from the Braze 580 specimen, there is a thick Cu-Ti-Fe reaction layer in both BAg-8/304SS and Ticusil®/ 304SS interfaces. The formation of Cu-Ti-Fe interfacial layer can prohibit wetting of BAg-8 and Ticusil® molten brazes on 304SS substrate. Meanwhile, continuous Ti_2Cu and TiCu layers are observed in Ti-6Al-4V/BAg-8 and Ti-6Al-4V/Ticusil® interfaces. © *2002 Kluwer Academic Publishers*

1. Introduction

With ever increasing many special purpose alloys in the market place, joining processes, such as welding, brazing . . . etc., plays an important role in practical application of these alloys. Well-developed joining process can not only promote the application of these alloys, but also can provide designers versatile choices of materials. Brazing is one of the most popular methods in joining dissimilar alloys [1, 2]. Both Ti-6Al-4V and 304 stainless steels (304SS) are good engineering alloys and widely used in industry due to their excellent mechanical properties as well as corrosion resistance. However, there are two major problems in brazing dissimilar materials. First, residual thermal stresses are usually developed after brazing due to thermal expansion mismatch between the joined alloys. Second, brittle intermetallic compounds are formed, especially at the interface between braze alloy and substrate after brazing. Both the above problems will be encountered in brazing Ti-6Al-4V and 304SS.

Ti-6Al-4V is a type of α-β titanium alloy. With aid of β stabilizer addition in the alloy, the β phase can be stabilized at room temperature. Therefore, Ti-6Al-4V can be heat treated by solution and aging procedure in order to obtain proper mechanical properties [3–5]. The brazing of Ti alloys has been extensively studied [2, 6, 7]. Dececco *et al.* suggested using Ag and its alloys to braze many Ti alloys [8]. The joint demonstrates certain degree of toughness due to the formation of TiAg intermetallics during brazing. Fig. 1 shows the SEM metallograph of Ti-6Al-4V and 304SS joint vacuum brazed by pure Ag filler at 1000°C for 600 seconds. A huge crack is found at the interface between the pure Ag braze and 304 SS. According to the SEM observation, the bonding strength between 304SS and pure Ag braze is not strong enough to effectively overcome the residual thermal stress between Ti-6Al-4V and 304SS. On the other hand, it is also observed that many silver base braze alloys can effectively braze many Ti alloys [9–11]. The primary advantage in using the silver base braze alloy to bond Ti alloy is that the melting point of most silver base braze alloys is lower than β transus temperature of most Ti alloys [6]. Heating Ti alloys above (or close to) its β transus temperature will result

*Author to whom all correspondence should be addressed.

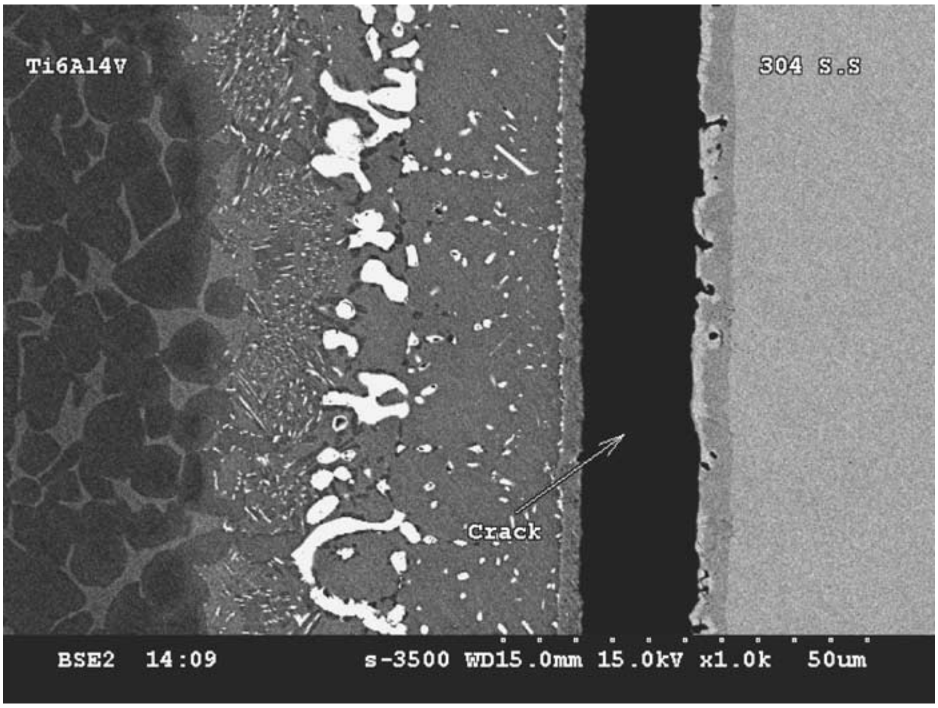


Figure 1 The SEM metallograph of Ti-6Al-4V and 304SS joint vacuum brazed by pure Ag filler at 1000°C for 600 seconds.

in excessive growth of the β phase in Ti alloy, and the mechanical properties of Ti alloys will be greatly deteriorated. Therefore, it is required that brazing of Ti alloys should not exceed its β transus temperature in order to avoid deterioration of its mechanical properties.

304SS is a typical type of austenitic stainless steels [12]. It is one of the most popular species among all stainless steels, and is characterized of both excellent corrosion resistance and mechanical property [12]. By addition of austenite stabilizing element Ni into the alloy, the austenite phase is stable at room temperatures [12]. It is important to note that sensitization of 304SS in heat affected zone in brazing or welding should be avoided [12, 13]. As the 304SS heated above 1000°C, huge chromium carbide precipitates will be formed along austenite grain boundaries. The consumption chromium content in 304SS may result in loss of corrosion resistance along grain boundary of the steel [13, 14]. The sensitized 304SS will be suffered from intergranular corrosion in service. Therefore, it is crucial for 304SS to avoid sensitization in brazing [13, 15]. Compared with the nickel base braze alloys, the low liquidus temperature of silver or copper base braze alloy makes it more suitable in brazing 304SS from the viewpoint of low brazing temperatures. The purpose of this investigation is focused on vacuum brazing of Ti-6Al-4V and 304SS. There are three braze alloys, including: Braze 580, BAg-8 (Ag-Cu eutectic braze alloy) and Ticusil®, evaluated in the experiment. Dynamic sessile drop measurements on the two substrates and formation of the intermetallics at the bonding interface will be extensively studied.

2. Experimental procedures

The substrates used in the experiments were 304SS and Ti-6Al-4V alloys. The 304SS rod was sliced into disks with 12.7 mm in diameter and 2.0 mm in thickness. The size of Ti-6Al-4V plate was 15 mm × 15 mm × 3 mm. Three commercially available braze alloys were used in the study for the purpose of comparison, and they are Braze 580 (57Ag-33Cu-7Sn-3Mn, wt%), BAg-8 (Ag-Cu eutectic alloy, 71.9Ag-28.1Cu, wt%) and Ticusil® (68.8Ag-26.7Cu-4.5Ti, wt%), respectively. Both solidus and liquidus temperatures of these braze alloys are listed in Table I [7]

Samples of total 2.5 g master alloys were prepared from high purity element pellets of 99.9 wt% by vacuum arc remelting with the operation voltage of 60 V and 150 A. At least remelting three times were necessary in order to avoid the inhomogeneity of the braze alloy, and the total accumulated weight loss of final

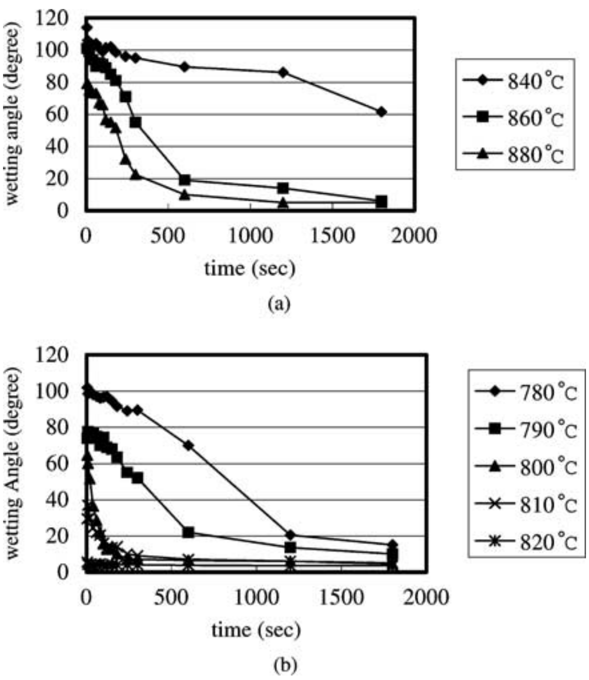


Figure 2 The dynamic wetting angle measurement of Braze 580 alloy on (a) Ti-6Al-4V and (b) 304SS substrate.

TABLE I Solidus and liquidus temperatures of three braze alloys used in the experiment [7]

Braze alloy	Solidus (°C)	Liquidus (°C)
Braze 580 (57Ag-33Cu-7Sn-3Mn, wt%).	605	730
BAg-8 (71.9Ag-28.1Cu, wt%)	780	780
Ticusil® (68.8Ag-26.7Cu-4.5Ti, wt%)	830	850

master alloy was less than 1 wt%. An ultrasonic bath using acetone as the solvent was used to clean samples prior to brazing. Furnace brazing was performed in a vacuum of 5×10^{-3} Pa at temperatures between 810 and 940°C for various time periods. All specimens were preheated at 700°C for 600 seconds prior to the brazing temperature. The heating rate of the furnace was set at 10°C/min throughout the experiment. Similarly, dynamic wetting angle measurements were made using the same furnace at the temperature ranges between 810 and 940°C for 0–1800 seconds. 0.15 g master alloy was put on the substrate, and the heating rate of the furnace was 10°C/min. The image of wetting angle was instaneously recorded by a camera.

The brazed specimens were cut by a low speed diamond saw, and followed by a standard metallographic procedure. An etching solution of 10 g $FeCl_3$ + 150 ml H_2O + 50 ml HCl + 300 ml C_2H_5OH was used prior to metallographic examination. The cross section of the brazed specimens was examined using a Hitachi 3500 H scanning electron microscope with the operation voltage of 15 KV. Quantitative chemical analysis was performed by using a JEOL JXA-8600SX electron probe microanalyzer (EPMA) equipped with a WDS (Wavelength Dispersive Spectroscopy), and its spot size was 1 μm throughout the analysis.

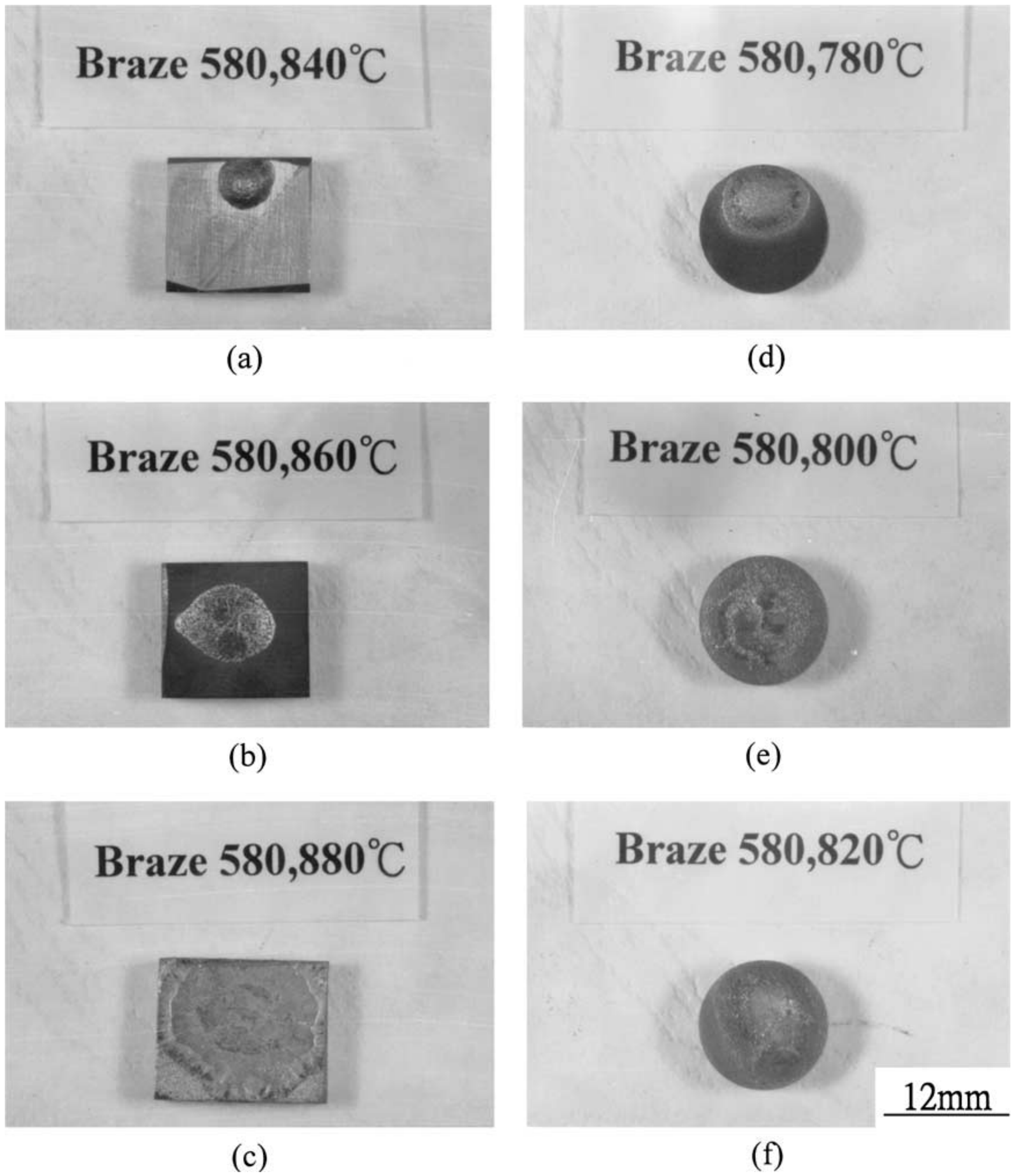
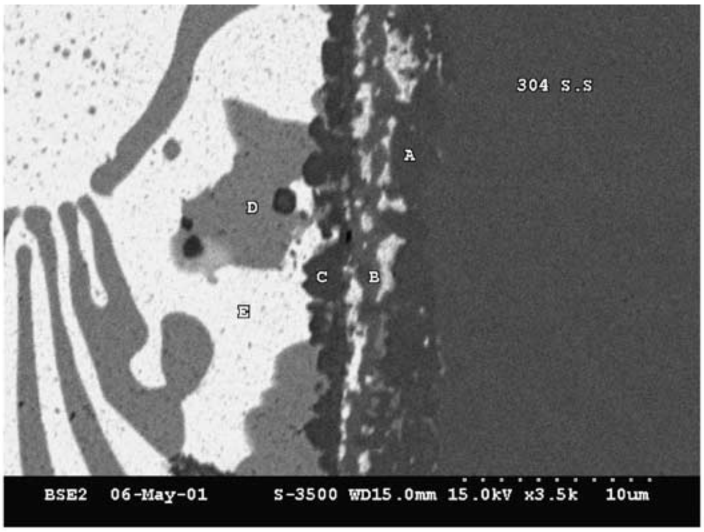


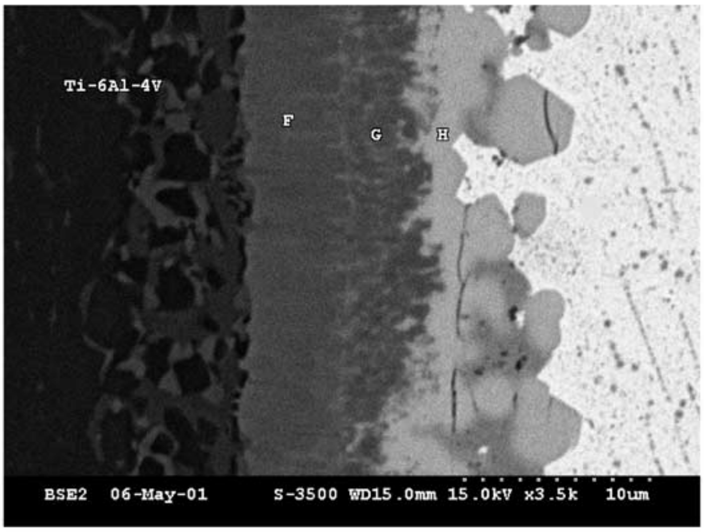
Figure 3 The spreading of Braze 580 alloy on (a)-(c) Ti-6Al-4V and (d)-(f) 304SS substrate.

3. Results and discussion

3.1. Braze 580 filler metal

Braze 580 has the lowest solidus and liquidus temperatures among three braze alloys [7]. However, there is a wide melting range between solidus and liquidus temperatures of the alloy. For a brazing filler alloy with different solidus and liquidus temperatures, the composition of the melt will gradually change as the temperature increases from the solidus to liquidus [16]. If the portion that melts first is allowed to flow out, the remaining solid may not melt and may retain behind as a residue, which is called liquation [16]. Filler alloys with narrow melting ranges do not tend to liquation, so they flow quite freely into joints with narrow clearance. On the other hand, a filler metal with a wide melting range needs rapid heating cycles to minimize separation





wt%	Ag	Fe	Si	Ni	Cr	Cu	Sn	Mn	Ti	Al	V
A	3.3	59.0	0.2	0.6	25.6	5.3	0.8	2.4	2.6	0.1	0.3
B	5.3	58.1	0.1	0.3	23.0	7.2	0.7	1.4	3.5	0.1	0.3
C	2.1	38.5	0.1	0.2	1.3	6.7	0.9	0.2	49.6	0.2	0.1
D	7.7	0.1	-	0.1	0.1	82.9	4.1	4.2	0.4	0.4	0.1
E	93.7	0.6	0.9	0.1	0.1	3.2	0.9	0.7	0.2	0.4	0.1
F	2.1	0.2	-	1.0	-	52.2	2.9	0.3	40.7	0.4	0.3
G	2.6	0.3	-	0.9	-	56.5	7.0	0.4	28.2	3.8	0.4
H	0.4	0.9	0.1	0.5	-	10.9	50.4	0.2	36.0	0.2	0.6

Figure 4 The SEM backscattered electron image and EPMA chemical analysis results of the Braze 580 specimen brazing at 850°C for 600 sec.

during brazing. Therefore, it is important to note that the liquation of Braze 580 should be avoided in vacuum brazing.

Fig. 2 displays dynamic wetting angle measurement of Braze 580 alloy on Ti-6Al-4V and 304SS substrates, respectively. Based on the Fig. 2a, Braze 580 can not effectively wet Ti-6Al-4V substrate below 840°C. As the brazing temperature increases above 860°C, comprehensive wetting can be obtained in 600 seconds. On the other hand, Braze 580 can wet 304SS substrate at much lower temperatures as compared with the Ti-6Al-4V substrate. For example, the wetting angle of Braze 580 on 304SS substrate is below 20° for the specimen brazed at 780°C for 1200 seconds. Completely spreading of the molten braze can be achieved for 304SS substrate brazed above 800°C. As described earlier, the liquidus of Braze 580 alloy is 730°C, and the heating rate of vacuum furnace is set at 10°C/min. The molten braze will have enough time in spreading if the brazing temperature is above 800°C. Based on the wetting angle measurement results, it is preferred that the brazing of Ti-6Al-4V and 304SS exceeds 840°C. Fig. 3 shows the spreading of Braze 580 alloy on both Ti-6Al-4V and 304SS substrate at various brazing conditions. There is no phase separation except for Ti-6Al-4V substrate brazed at 880°C for 1800 seconds.

There are at least two types of images in SEM observations, including: secondary electron image (SEI) and backscattered electron image (BEI) [17]. The low-energy secondary electrons escaped from a relatively shallow depth (1–10 nm) in the specimen. Secondary emission, therefore, is very sensitive to the topography of the sample surface. The more irregular the topography of the specimen surface, the more electrons escape from that area of the specimen. On the other hand, a backscattered electron is defined as an electron escaping the surface of the specimen with energy of more than 50 eV [17]. The number of backscattered electrons produced per electron beam is continuously increased with the atomic number of the specimen. Thus, a specimen area containing high-atomic number elements appears light in the cathode ray tube, whereas an area with low-atomic number elements appears dark. The backscattered image does not provide topographic contrast but primarily shows the element distribution in the joint. Thus, a specimen area containing high-atomic number elements appears light, while an area with low-atomic number elements appears dark [17].

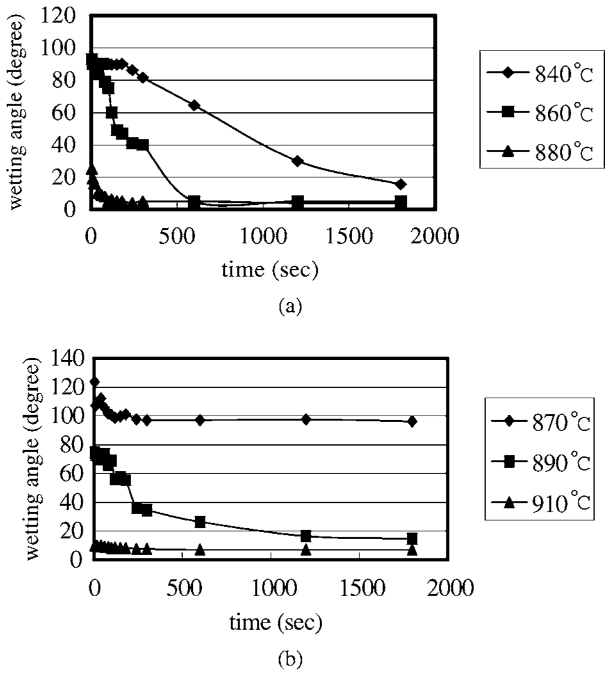


Figure 6 The dynamic wetting angle measurement of BAg-8 braze alloy on (a) Ti-6Al-4V and (b) 304SS substrate.

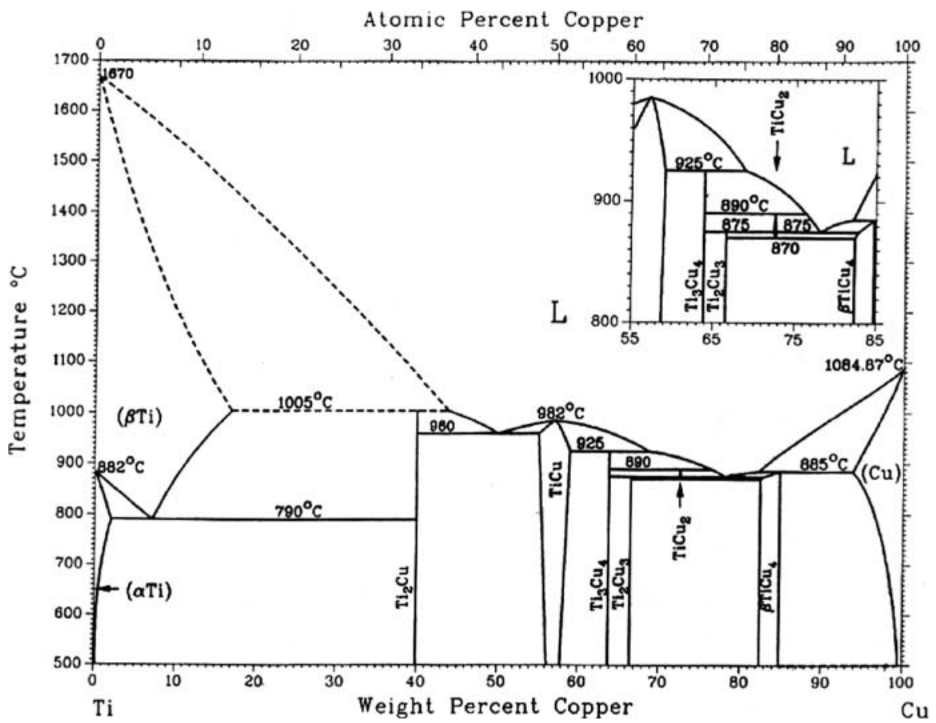


Figure 5 The Cu-Ti binary alloy phase diagram [18].

Fig. 4 shows the SEM backscattered electron image and EPMA chemical analysis results of the Braze 580 specimen brazing at 850°C for 600 sec. Several reaction layers exist in both Braze 580/304SS and Braze 580/Ti-6Al-4V interfaces. Points A and B are basically Fe-Cr alloy. Point C is the reaction layer mainly comprised of Ti, Fe and Cu. Since there is no Ti content in Braze 580, it is expected that Ti atoms are transported from Ti-6Al-4V substrate via Ti diffusion in the molten braze. The area of braze alloy is primarily comprised of Ag-rich and Cu-rich phases alloyed with a few other elements as displayed in point D and E (Fig. 4). There are three reaction layers observed at the interface between Braze 580 and Ti-6Al-4V as shown in Fig. 4. The thickness of reaction layer at Braze 580/Ti-6Al-4V interface is much larger than that at Braze 580/304SS interface. The chemical composition at point F and G in Fig. 4 mainly consists of Cu and Ti. Fig. 5 is Cu-Ti binary alloy phase diagram [18]. According to the phase diagram, the stoichiometry of Cu/Ti ratio at point F is very close to TiCu intermetallics. Similarly, the stoichiometry of Cu/Ti ratio at point G is close to Ti_2Cu_3 intermetallic compound. It is also noted that high Sn content at point G is observed due to its location close to the braze alloy. A continuous layer of Cu-Sn-Ti ternary intermetallic compound is found. The Cu-Sn-Ti intermetallic compound has been observed in copper base active braze alloys in previous study [19]. In this research, Sn is strongly associated with Ti, and forms a Cu-Sn-Ti intermetallic compound at the interface between Braze 580 and Ti-6Al-4V. Therefore, most of Sn content in Braze 580 will be depleted due to the interfacial reaction. Since there are some cracks in Cu-Sn-Ti intermetallics as shown in the figure, the ternary

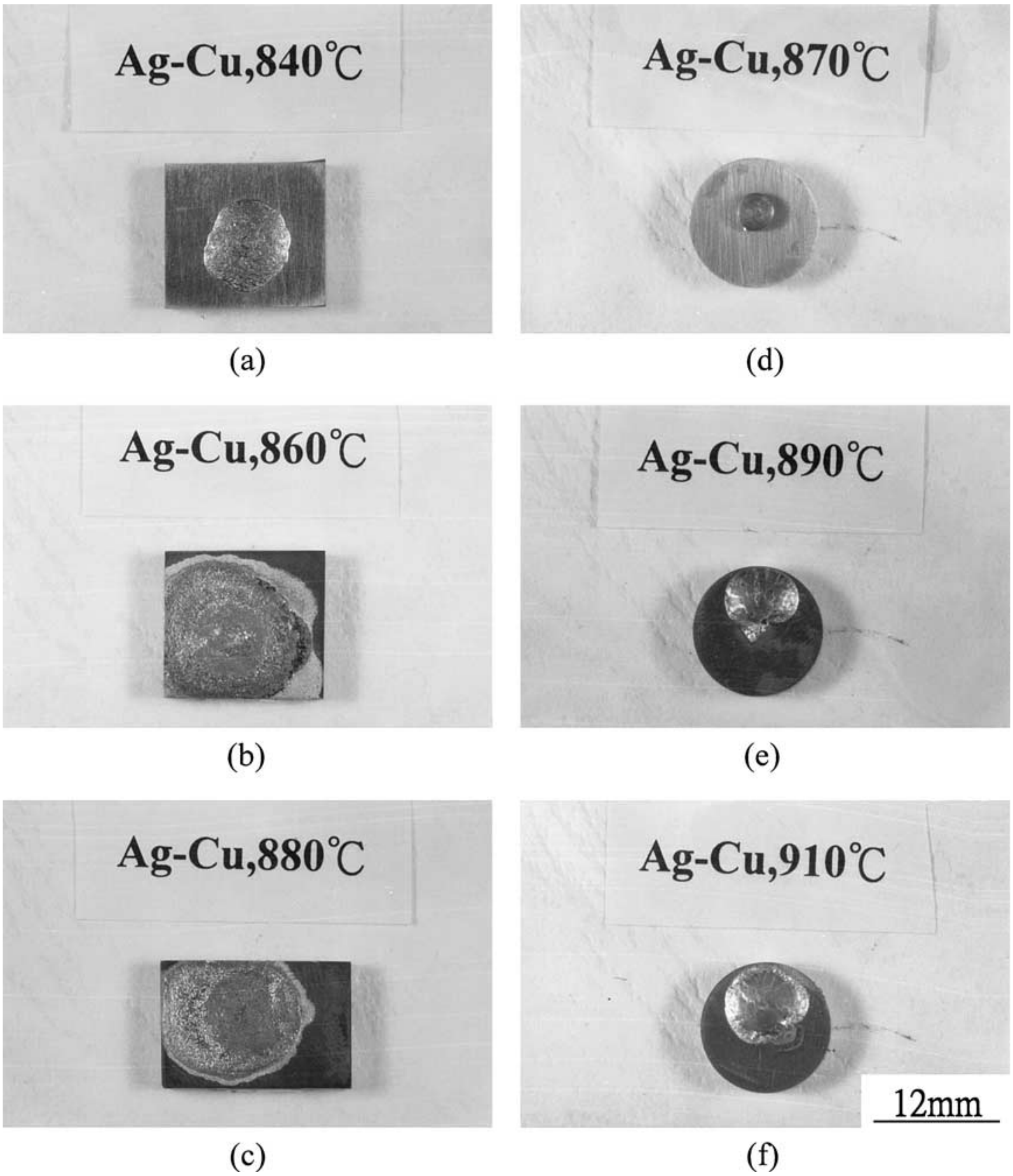
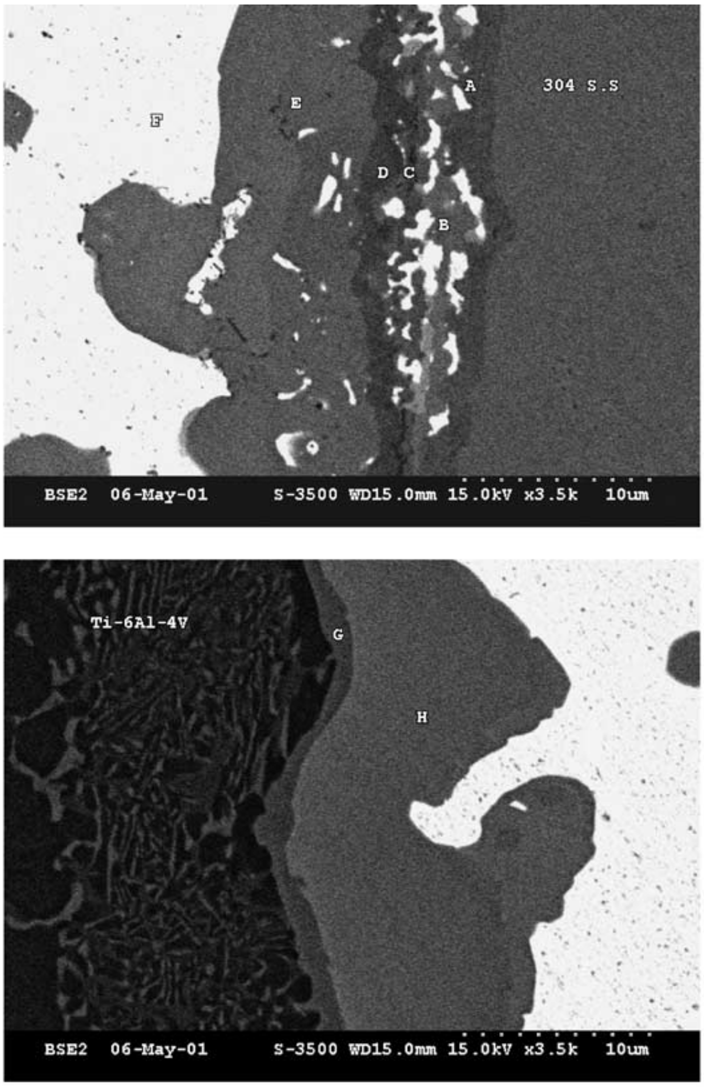


Figure 7 The spreading of BAg-8 braze alloy on (a)–(c) Ti-6Al-4V and (d)-(f) 304SS substrate.

intermetallics is brittle. It is expected that existence of the continuous Cu-Sn-Ti interfacial intermetallics is detrimental to its bonding strength.

3.2. BAg-8 filler metal

BAg-8 has a eutectic composition of Cu and Ag with a melting point of 780°C. Fig. 6 displays dynamic wetting angle measurement of BAg-8 braze alloy on both Ti-6Al-4V and 304SS substrates. Similar to Braze 580, BAg-8 can not effectively wet Ti-6Al-4V substrate until 840°C. As the brazing temperature further increases to 860°C, comprehensive wetting can be obtained in 600 seconds. Meanwhile, BAg-8 can not effectively wet 304SS substrate below 870°C, and its wetting behavior is very different from Braze 580. According to experimental results, it is preferred that the brazing of



wt%	Ag	Fe	Si	Ni	Cr	Cu	Mn	Ti	Al	V
A	1.0	57.2	0.1	1.1	26.5	9.7	1.7	2.5	0.1	0.1
B	0.8	57.5	0.1	1.9	24.0	9.0	0.4	5.6	0.3	0.2
C	2.4	48.9	0.3	4.0	6.2	0.7	0.2	36.6	0.3	0.1
D	0.6	32.6	0.1	7.4	1.6	13.8	0.2	41.3	0.8	1.6
E	2.5	10.9	0.1	3.9	1.1	46.0	0.1	33.5	0.6	1.3
F	92.1	1.2	0.7	0.6	0.1	3.3	0.1	0.4	0.6	0.4
G	3.6	0.1	-	0.1	0.1	34.2	0.1	58.9	2.2	0.8
H	5.8	-	-	0.1	0.1	51.0	0.1	42.1	0.5	0.4

Figure 8 The SEM backscattered electron image and EPMA chemical analysis results of BAg-8 brazed specimen at 890°C for 600 sec.

Ti-6Al-4V and 304SS exceeds 870°C. Fig. 7 shows the spreading of BAg-8 braze alloy on both Ti-6Al-4V and 304SS substrate at various brazing conditions. There is phase separation for Ti-6Al-4V substrate brazed above 860°C. It is resulted from the interfacial reaction between BAg-8 and Ti-6Al-4V substrate as demonstrated in following microstructural observations.

Fig. 8 shows the SEM backscattered electron image and EPMA chemical analysis results of BAg-8 brazed specimen at 890°C for 600 sec. Similar to previous case,

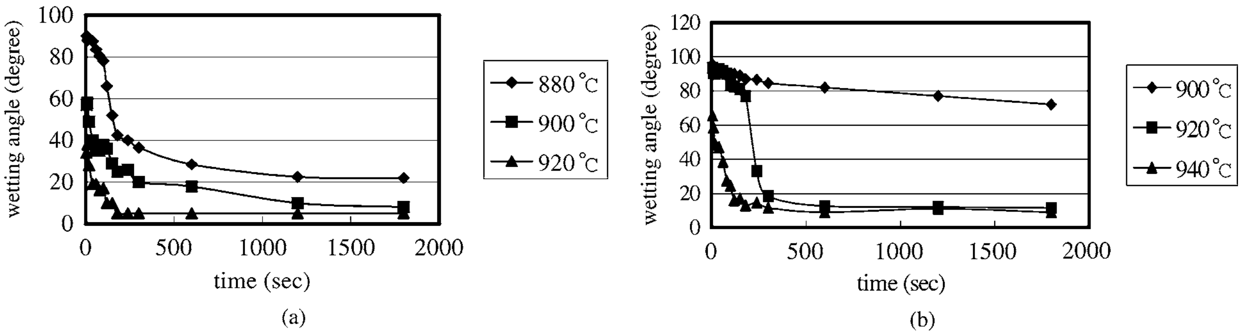


Figure 9 The dynamic wetting angle measurement of Ticusil® braze alloy on (a) Ti-6Al-4V and (b) 304SS substrate.

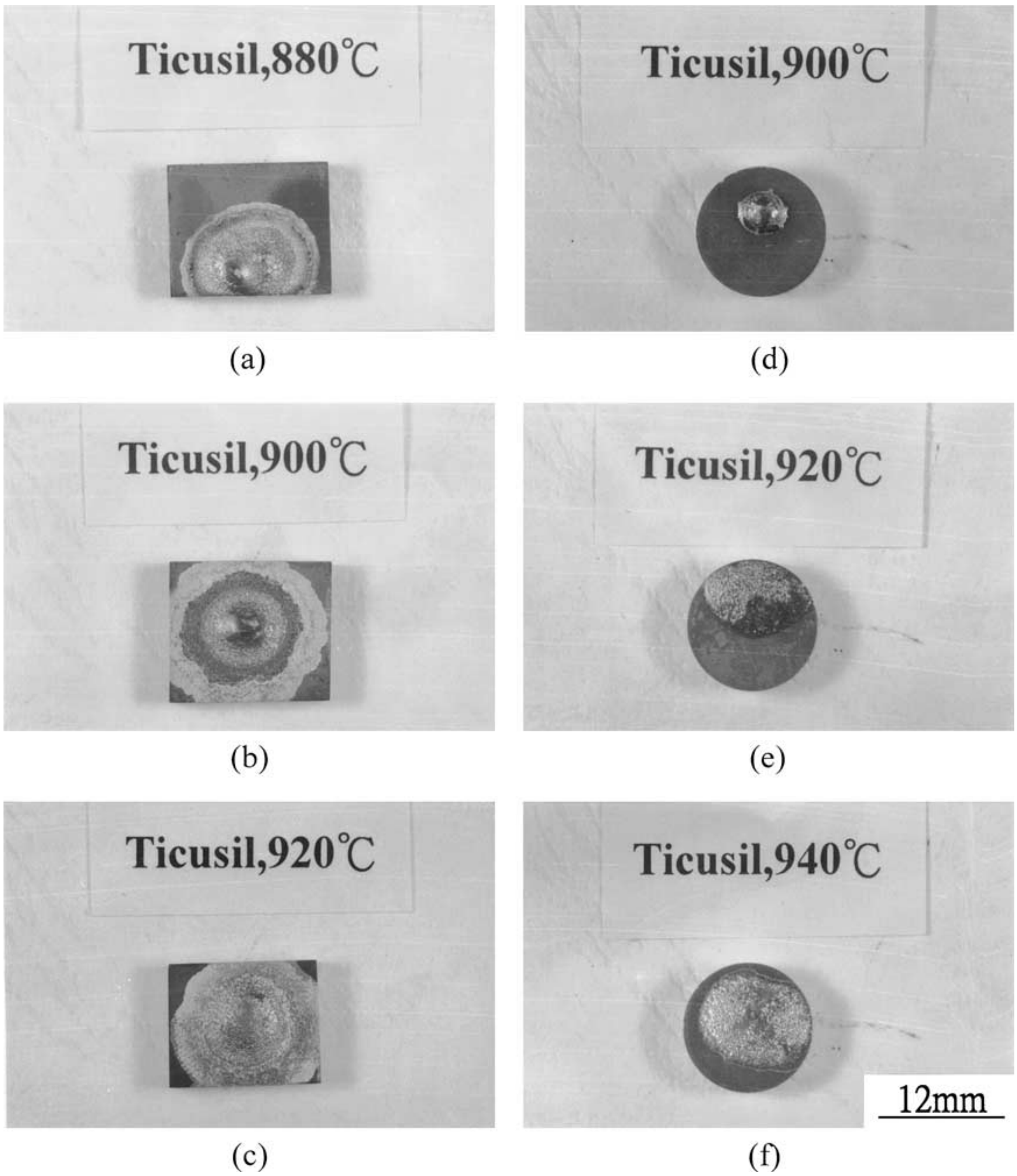
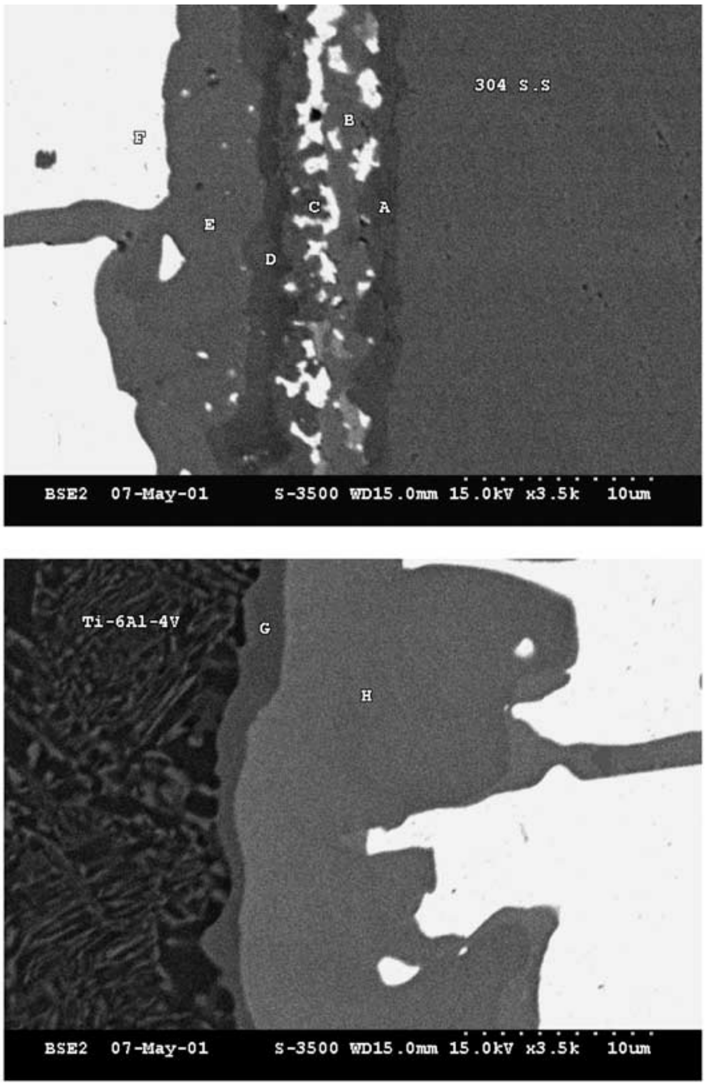


Figure 10 The spreading of Ticusil® braze alloy on (a)-(c) Ti-6Al-4V and (d)-(f) 304SS substrate.

many reaction layers exist in both Braze 580/304SS and Braze 580/Ti-6Al-4V interfaces. Points A and B are basically Fe-Cr alloy. Point C is the reaction layer mainly comprised of Ti, Fe and Cr, and point D is the reaction layer mainly comprised of Ti, Fe and Cu. Different from the interface of Braze 580/304SS, there is a thick Cu-Ti-Fe reaction layer (point E) at the interface between BAg-8 and 304SS. The formation of Cu-Ti-Fe interfacial layer may prohibit wetting of BAg-8 molten braze on 304SS substrate. Huge difference in wetting behavior of 304SS is found in using Braze 580 and BAg-8 braze alloys. Similarly, there is no Ti content in BAg-8, so it is expected that Ti atoms are transported from Ti-6Al-4V substrate via Ti diffusion in the molten braze. The brazing temperature of BAg-8 is higher than that of Braze 580, so it is expected that



wt%	Ag	Fe	Si	Ni	Cr	Cu	Mn	Ti	Al	V
A	1.6	58.6	0.1	1.3	27.5	1.3	1.3	8.1	0.1	0.1
B	2.8	57.2	0.2	2.2	23.7	3.8	1.5	8.4	0.1	0.1
C	3.6	48.8	0.2	1.8	4.9	4.9	0.6	33.1	1.4	0.7
D	0.9	19.3	0.1	8.4	0.8	26.3	0.2	41.9	1.0	1.1
E	2.4	4.9	0.1	2.6	0.8	56.6	0.1	29.6	2.4	0.5
F	93.3	0.8	0.8	0.1	0.2	3.4	0.1	0.8	0.3	0.2
G	3.2	0.1	-	0.1	-	31.6	-	61.1	3.0	0.9
H	5.7	0.1	-	0.1	-	50.7	-	42.4	0.4	0.6

Figure 11 The SEM backscattered electron image and EPMA chemical analysis results of Ticusil® brazed specimen at 910°C for 600 sec.

more Ti atoms dissolute from Ti-6Al-4V substrate into the molten braze. Moreover, Ti is strongly associated with Cu and Fe, and forms a thick Cu-Ti-Fe reaction layer at the interface between BAg-8 and 304SS.

There are only two reaction layers observed at the interface between Braze 580 and Ti-6Al-4V as shown in Fig. 8. The chemical compositions of point G and H in Fig. 8 mainly consist of Cu and Ti. According to the phase diagram shown in Fig. 5, the stoichiometry of Cu/Ti ratio at point G is close to Ti_2Cu intermetallics [18]. Similarly, the stoichiometry of Cu/Ti ratio at point H is close to TiCu intermetallic compound. The growth rate of TiCu is much faster than that of Ti_2Cu at 890°C. Both dissolution of Ti-6Al-4V and strong reaction between Ti and Cu are primarily responsible for the phase separation of BAg-8 on Ti-6Al-4V substrate brazed above 860°C.

3.3. Ticusil® filler metal

Ticusil® is a reactive braze alloy with the chemical composition of Ag-Cu eutectic plus 4.5 wt% Ti. It has the highest melting range between 830 and 850°C among all three tested braze alloys. Fig. 9 shows the dynamic wetting angle measurement of Ticusil® braze alloy on both Ti-6Al-4V and 304SS substrates. It can effectively wet Ti-6Al-4V substrate above 880°C. Unlike previous two braze alloys, Ticusil® can not effectively wet 304SS substrate below 900°C. Extensive wetting of the Ticusil® braze on 304SS can be acquired if the brazing temperature exceeds 900°C. Consequently, it is preferred that the used of Ticusil® in brazing of Ti-6Al-4V and 304SS exceeds 900°C. Fig. 10 shows the spreading of Ticusil® braze on both Ti-6Al-4V and 304SS substrate at various brazing conditions. Similar to previous observations, there is phase separation for Ti-6Al-4V substrate brazed above 880°C, and it is resulted from the interfacial reaction between Ticusil® and Ti-6Al-4V substrate. Meanwhile, there is no phase separation observed in 304SS substrate.

Fig. 11 shows the SEM backscattered electron image and EPMA chemical analysis results of Ticusil® brazed specimen at 910°C for 600 sec. Since the Ti can be transported from Ti-6Al-4V substrate via Ti diffusion in the molten braze. It is expected that both Ticusil® and BAg-8 brazed joint have similar interfacial microstructures. However, the brazing temperature of Ticusil® brazed specimen is higher than that of BAg-8 brazed specimen. The higher the brazing temperature, the more interfacial reaction available during brazing. In Fig. 11, Points A and B are basically Fe-Cr alloy. Point C is a reaction layer mainly comprised of Ti and Fe. Point D is the reaction layer mainly comprised of Ti, Cu and Fe. Similar to BAg-8 braze alloy, there is a thick Cu-Ti-Fe reaction layer at the interface between Ticusil® and 304SS.

There are only two reaction layers observed at the interface between Ticusil® and Ti-6Al-4V substrate as shown in Fig. 11. The chemical compositions of point G and H in Fig. 11 are very similar to those of points G and H in Fig. 8. Basically, both continuous Ti_2Cu and TiCu layers are observed in the SEM examination. Moreover, the thickness of TiCu in Fig. 11 is larger than that in Fig. 8 due to the higher brazing temperature of Ticusil® braze alloy.

4. Conclusion

1. Based on the sessile drop tests, Braze 580 has the lowest brazing temperatures above 840°C, in contrast to 870°C for BAg-8 and 900°C for Ticusil® braze alloy. No phase separation is observed for all brazes on 304SS substrate. However, phase separation is observed for all specimens brazed above 860°C on Ti-6Al-4V substrate. Both dissolution of Ti-6Al-4V substrate and strongly interfacial reactions between Ti and Cu are primarily responsible for the phase separation of braze alloys on Ti-6Al-4V brazed above 860°C.

2. The reaction layer between Braze 580 and 304SS is mainly comprised of Ti, Fe and Cu. The thickness of reaction layer at Braze 580/Ti-6Al-4V interface is much larger than that at Braze 580/304SS interface. A continuous Cu-Sn-Ti interfacial intermetallic compound is found. Since Sn is strongly associated with Ti, it forms a ternary Cu-Sn-Ti intermetallic compound at the interface between Braze 580 and Ti-6Al-4V. Most of Sn content in Braze 580 is depleted due to the interfacial reaction.

3. Both Ticusil® and BAg-8 brazed joint have similar interfacial microstructures. Different from the Braze 580 specimen, there is a thick Cu-Ti-Fe reaction layer in both BAg-8/304SS and Ticusil®/304SS interfaces. The formation of Cu-Ti-Fe interfacial layer can prohibit wetting of BAg-8 and Ticusil® molten braze on 304SS substrate. Moreover, continuous Ti_2Cu and TiCu layers are also observed in both Ti-6Al-4V/BAg-8 and Ti-6Al-4V/Ticusil® interfaces.

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