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Microstructure and phase constitution near the interface of Cu/Al vacuum brazing using Al–Si filler metal

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Abstract

Brazing of Cu to Al using Al–Si filler metal has been carried out by vacuum brazing technology. The microstructure and the phase constitution in Cu/Al joint were studied by means of metallography, electron probe microanalyser (EPMA) and X-ray diffraction (XRD). Experimental results obtained showed that two kinds of intermetallic compounds (IMCs) are formed near the interface of copper and brazing seam region and those are Cu_3Al_2 and $CuAl_2$ phases. Moreover, ε-$Cu_{15}Si_4$, Al–Si and $CuZn_2$ are formed on the α-Al solid solution in the brazing seam region. Technology parameters of vacuum brazing were: brazing temperature $T = 590$–$610\,^{\circ}C$, vacuum level 10^{-3} Pa, holding time $t = 5$–10 min.
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Keywords: Al/Cu; Al–Si filler metal; Vacuum brazing; Interface; Microstructure

1. Introduction

In recent years, considerable interest has been generated in welding nonferrous materials like copper and aluminium to form compound structure because Cu and Al exhibit excellent electrical and thermal properties, and also because of the light weight and high performance of industrial product [1,2]. For example, substitute Al for Cu in the area of electronic industries, can avoid many accidents induced by the high electrical resistivity of aluminium oxide film and its creep deformation.

The melting point, fusion heat and linear expansion of Cu and Al are different, and intermetallic compounds (IMCs) are formed easily between Cu and Al, therefore welding Cu and Al to make compound structure is difficult [3–5]. Generally, reports involved welding Cu and Al, for example friction welding [6], diffusion bonding [7,8] and ultrasonic welding [9], were based on solid diffusion principle to form a joint. However, all these methods need rather high pressure and the welded parent metal surface must be smooth and clean enough [10]. So, all these solid diffusion methods were not applicable to manufacturing components and parts with complicated shapes, or process in mass production. In this paper, sheet materials including Cu and Al were successfully brazed by vacuum brazing technology. Slag inclusion resulting from using brazing flux in common brazing methods can be avoided. This is favourable to the enhancement of the compactness and corrosion resistance of the Cu/Al joint.

In this study, Cu and Al (dissimilar materials) were brazed in a vacuum of 10^{-3} Pa using Al–Si filler metal and a Cu/Al joint with good combination was formed. Microstructure, microhardness distribution and phase constitution in the brazing seam region were analysed by means of metallography, electron probe microanalyser (EPMA) and X-ray diffraction (XRD). The results provide a favourable basis for further studies on joining Cu/Al and for the application of the compound structure.

2. Materials and experiments

The materials used in this study were copper (T2) and aluminium (1035). Aluminum matrix was mild and

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Table 1
Chemical composition and thermo-physical properties of Al (1035) and Cu (T2)

Materials	Chemical compositions (wt%)								
	Al	Cu	Zn	Si	Mg	Ti	Fe	Mn	S
Al (1035)	99.35	<0.10	<0.10	<0.35	<0.05	<0.03	<0.6	<0.05	V<0.05
Cu (T2)	–	99.0	–	–	–	Pb<0.005	O< 0.06	Bi<0.002	<0.005
Filler metal	Bal.	0.30	0.20	11.0–13.0	0.10	–	0.80	0.05	–

Materials	Thermo-physical properties								
	Melting point (°C)	Boiling point (°C)	Density ($g\,cm^{-3}$)	Specific heat volume ($J\,kg^{-1}\,K^{-1}$)	Heat conductivity ($W\,m^{-1}\,K^{-1}$)	Young's modulus (GPa)	Poisson ratio	Tensile strength (MPa)	Yield strength (MPa)	Brinell hardness (HB)
Al (1035)	660	2520	2.71	946	226	71	0.33	80	30	32
Cu (T2)	1083	2578	8.90	385	390	108	0.35	215	–	35–45

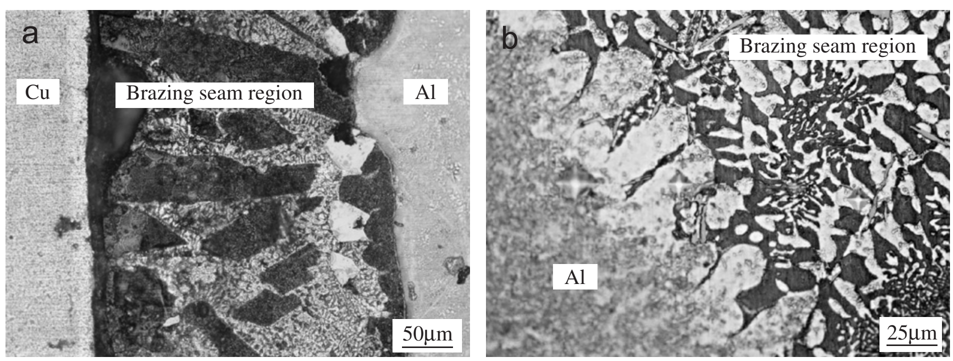

Fig. 1. Microstructure in interface zone of Cu/Al brazing joint: (a) microstructure feature; (b) Al side.

annealed. The chemical composition and thermo-physical properties of Cu and Al are shown in Table 1. Cu sample was machined into dimensions of 100 mm × 50 mm × 1 mm and Al sample into 100 mm × 50 mm × 6 mm. The filler metal used in this study was E4047 prepared into a flaky material, and the thickness was about 0.1 mm. The oxidation film and greasy dirt on the surface of the substrates and filler metal were eliminated by a series of mechanical and chemical methods before vacuum brazing. Then the test plates, to be brazed in vacuum brazing equipment, were assembled in the sequence Al, filler metal and Cu. The technology parameters during the vacuum brazing were: brazing temperature $T = 590–610\,°C$, vacuum level 10^{-3} Pa, holding time $t = 5–10$ min.

The samples were cut from the Cu/Al brazing joint by a line cut machine. Then they were ground by a series different type sand paper, polished and finally etched with solutions including 10 g pure NaOH and 60 ml H_2O. Metallographic examinations were carried out to analyse the microstructure feature near the Cu/Al interface. The microhardness distribution and phase constitution of the Cu/Al brazing joint were measured by means of Shimadzu microsclerometer, JXA-8800R EPMA and D/MAX-RCXRD, respectively.

3. Results and analysis

3.1. Microstructure feature of Cu/Al joint

Microstructure feature near the interface zone of the Cu/Al brazing joint was observed by means of optical metallographic microscope. The interface zone of Cu/Al brazing joint includes the transition region on Cu side, the middle brazing seam region and the transition region on Al side (Fig. 1). The boundary of the brazing seam region near the Cu substrate can be observed easily (Fig. 1a). It can be seen also (Fig. 1b), that bulky prismatic structure is formed on the side of Al substrate and perpendicular to Al surface. The growth of Al grain is the result of a eutectic reaction between Al substrate and Si element included in the filler metal. At this reaction Al fusing takes place at a temperature lower than the melting point.

According to Al–Cu phase diagram (Fig. 2), the generated prismatic structure is α-Al solid solution and because of the diffusion of Si atom in Al substrate [11], the concentration of Si element increases gradually from the root of the prismatic structure to the top. The proper growth of Al prismatic structure is favourable to improve mechanical property of the Cu/Al joint. However, if the

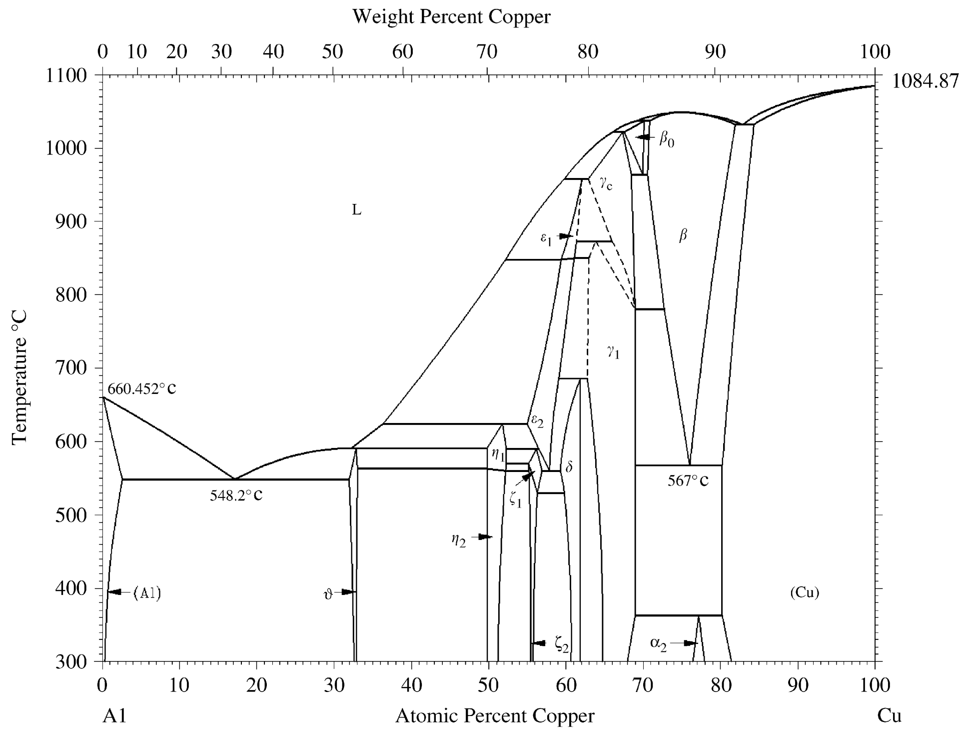

Fig. 2. Al–Cu phase diagram.

technology parameters are not proper, the growth control of prismatic structure is difficult. A heavy fusion of Al substrate could be induced and even the dimensional accuracy of the joint could be severely affected. Without a doubt, sheet materials would be fused breakthrough. So the eutectic reaction should be controlled appropriately.

Dendrite crystal is formed in the brazing seam region (Fig. 1b). The grey matrix is α-Al solid solution and the large volume white phase is eutectic $CuAl_2$ phase. Fine structure is α-Al + $CuAl_2$ eutectic. A bit of acicular compounds are formed at the boundary of grain. According to composition of the filler metal, acicular phase was primary crystal Si.

3.2. Microhardness near the joint

The microhardnesses of Cu substrate, brazing seam region and Al substrate were measured, and the results are shown in Fig. 3. The test instrument and parameters were: the Shimadze microsclerometer, 25 gf loading and a load time of 10 s. The results indicate that the microhardness of the prismatic structure is higher than that of the Al substrate (about 50 HM), and increases gradually from the root to the top. The problem arises in the diffusion of Si element in the eutectic reaction. The higher Si element concentration is, the more microhardness of the prismatic structure is. Three different microhardness zones are found, the microhardness of Cu substrate is low (about 100 HM), however the microhardness of the transition on Cu side is distinctly higher, 550 and 750 HM, respectively. At the case of Cu/Al vacuum diffusion bonding [3], the maximum of microhardness is 780 HM, on Cu side and the microhardness on Al side increases gradually, shown a similar diversification like the results obtained in this research. Under the environment of vacuum brazing, various reactions between Al and Cu occur, the elements concentration changes in a linear relationship with the distance, and two mainly IMCs come into being.

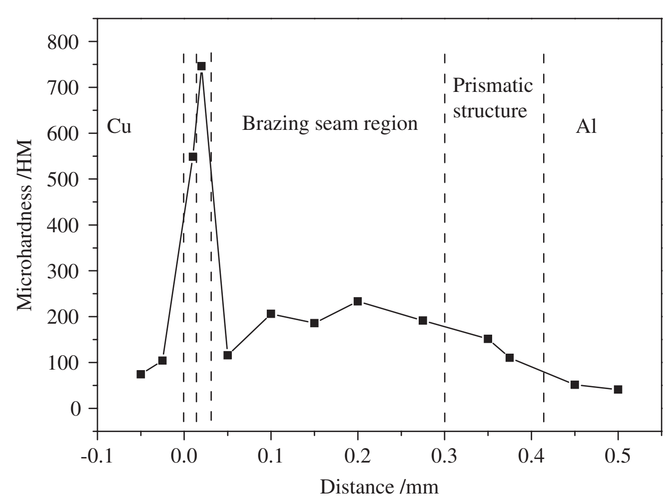

Fig. 3. Microhardness near the interface of Cu/Al brazing joint.

3.3. Element distribution near interface of Cu/Al vacuum brazing

The Al–Si filler metal is an excellent material for brazing aluminium alloy. Accordingly, quality of Cu/Al joint relied

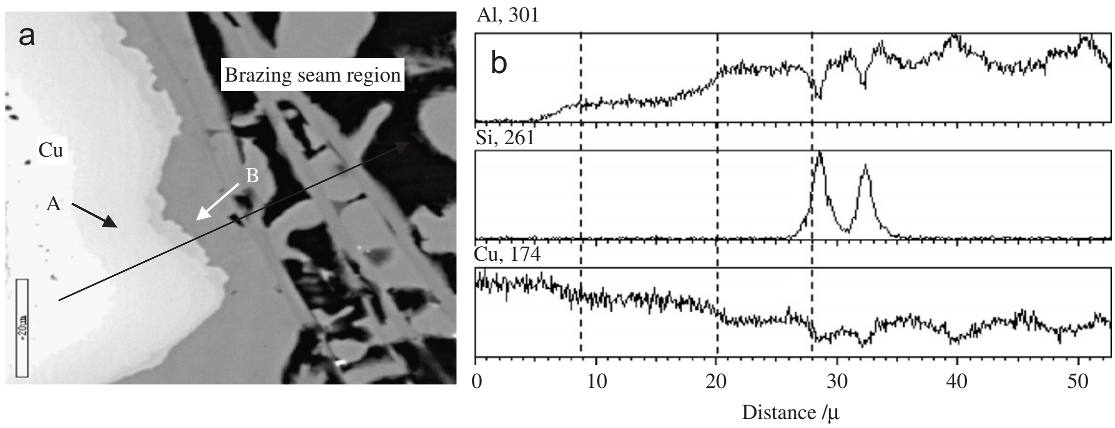

Fig. 4. Measured location and results of element distribution near the Cu side for brazing joint: (a) measured location; (b) element distribution.

heavily on reaction of Cu substrate and the filler metal. There exist various elements in the brazing seam region. The element distributions in different regions were measured by the electron probe microanalyser JXA-8800R EPMA. The test parameters were: working voltage 20 kV and working current 2.5×10^{-5} mA. The results including the Cu substrate and the brazing seam region are shown in Fig. 4. The line shown in Fig. 4a is the EPMA measured location.

According to Fig. 4, there are two different transition regions on Cu substrate side. For further studying elements percentage of the two different transition regions, constituents in two points A and B were measured (Fig. 4) and the results are shown in Table 2.

According to Al–Cu phase diagram (Fig. 2) and Table 2, the phase in point A is Cu_3Al_2 IMC and the phase in point B is $CuAl_2$ IMC. Therefore, two transition regions observed in Fig. 4 are Cu_3Al_2 and $CuAl_2$ IMCs, respectively. The average width of Cu_3Al_2 transition region is 12 μm, regularly distributed on the Cu substrate surface, while the width of $CuAl_2$ transition region is 8 μm. The $CuAl_2$ phase comes off the Cu substrate and is distributed outside the Cu_3Al_2 transition region, irregularly into the brazing seam region. In the brazing process, activity of Al element is higher than that of Cu, so Cu_3Al_2 is formed according to the following equation between Cu and Al:

$$2Al + 3Cu \rightarrow Cu_3Al_2. \quad (1)$$

Then concentration of Al element becomes higher and $CuAl_2$ is formed between Al and Cu_3Al_2 according to

$$4Al + Cu_3Al_2 \rightarrow 3CuAl_2. \quad (2)$$

In the brazing seam region, a eutectic reaction occurs between a part of $CuAl_2$ phase and Al matrix, and a binary eutectic phase α-(Al) + $CuAl_2$ is formed. The $CuAl_2$ phase plays an important role concerning mechanical property of the Cu/Al brazing joint. As shown in Ref. [12], most fractures on the Cu side take place inside the $CuAl_2$ phase. Therefore, shortening the treatment time at high temperature would reduce the amount of formed $CuAl_2$ phase and improve the performance of the Cu/Al joint.

Table 2
EPMA result of element composition in the brazing zone

Elements	Point A		Point B	
	wt%	at%	wt%	at%
Al	23.1	41.43	46.85	67.49
Cu	76.9	58.57	53.15	32.51

3.4. XRD analysis near the interface of Cu/Al vacuum brazing

To further clarify phase constitution near the interface of Cu/Al vacuum brazing, the XRD analysis was carried out with copper target under the following conditions: working voltage 60 kV and working current 40 mA. The XRD results are shown in Fig. 5. The comparison of the results obtained with the data from the Joint Committee on Power Diffraction Standards (JCPDS) is presented in Table 3.

According to the XRD analysis results, the brazing seam region is mainly consisted of α-Al solid solution, Cu_3Al_2 and $CuAl_2$ IMCs, what is in good agreement with the EPMA analysis data. Moreover, ε-$Cu_{15}Si_4$ phase, Al–Si phase and $CuZn_2$ phase are formed in the brazing seam region, and these phases appear in form of acicular compounds, as shown in Fig. 1. These acicular compounds are dispersed into the brazing seam region. No matter what brittle IMCs are formed, the microhardness of the brazing seam region would arise and the brittleness would increase. So, the technology parameters (holding time, brazing temperature and vacuum level) should be controlled properly to limit the Al diffusion into Cu substrate and the formation of any brittle IMC in the brazing seam region. Different IMCs Cu_9Al_4 and CuAl instead of Cu_3Al_2 are found by Ouyang et al. [6] in the friction stir welding of 6061 aluminum alloy to copper. The IMCs

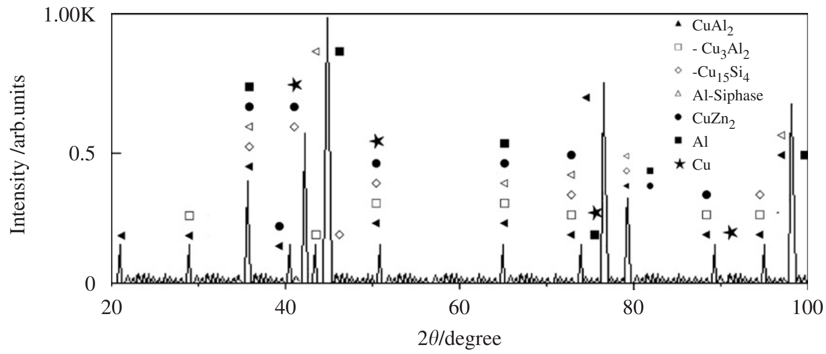

Fig. 5. XRD result of brazing zone of Cu/Al brazing joint.

Table 3
X-ray diffraction analysis results for the brazing zone of Cu/Al vacuum brazing joint

Measured data	Data from JCPDS														
	$CuAl_2$ (25-0012)			ε-Cu_3Al_2 (26-0015)			ε-$Cu_{15}Si_4$ (23-0222)			Al–Si phase (41-1222)			$CuZn_2$ (39-0400)		
d (nm)	*d* (nm)	*hkl*	I/I_0	*d* (nm)	*hkl*	I/I_0	*d* (nm)	*hkl*	I/I_0	*d* (nm)	*hkl*	I/I_0	*d* (nm)	*hkl*	I/I_0
0.4304	0.4304	110	100	–	–	–	–	–	–	–	–	–	–	–	–
0.3056	0.3037	200	35	0.2924	101	50	0.3040	310	40	0.3142	–	11	–	–	–
0.2340	0.2374	211	70	–	–	–	0.2410	400	40	0.2343	–	100	0.2355	311	50
0.2124	0.2121	112	90	–	–	–	–	–	–	–	–	–	0.2157	320	40
0.2089	–	–	–	–	–	–	0.2096	421	50	–	–	–	0.2080	321	100
0.2044	–	–	–	0.2068	102	100	0.2054	332	100	–	–	–	–	–	–
0.2026	–	–	–	–	–	–	–	–	–	0.2028		35	–	–	–
0.1810	0.1901	310	70	0.1794	200	20	0.1890	510	90	–	–	–	0.1892	410	10
0.1432	0.1408	411	6	0.1465	202	70	–	–	–	0.1433		18	0.1486	511	10
0.1279	0.1288	402	21	0.1266	004	30	0.1263	730	5	–	–	–	–	–	–
0.1234	0.1234	332	20	–	–	–	–	–	–	–	–	–	–	–	–
0.1221	0.1219	004	8	–	–	–	0.1229	650	90	0.1222	–	25	0.1226	620	30
0.1090	0.1091	413	3	0.1080	114	60	–	–	–	–	–	–	0.1096	710	10
0.1044	0.1040	530	2	0.1037	220	40	0.1045	760	60	0.1046	–	2	–	–	–
0.1013	0.1011	600	5	–	–	–	–	–	–	0.1013		3	–	–	–

formed are not only brittle but also rigid. This makes the process of friction stir welding difficult. However, with some IMCs formed, a good joint of Cu/Al vacuum brazing joint still can be obtained.

4. Conclusions

(1) The interface of Cu/Al brazing includes a transition region on Cu side, middle brazing seam region and a transition region on Al side. The proper technology parameters are: brazing temperature $T = 590$–$610\,^{\circ}C$, vacuum level $= 10^{-3}$ Pa, holding time $t = 5$–10 min.
(2) Layers Cu_3Al_2 and $CuAl_2$ are formed between the brazing seam and the Cu substrate. Average width of layered Cu_3Al_2 is about 12 μm, and $CuAl_2$ is about 8 μm. Microhardness of two layers is higher than that of the substrates. On the transition region of Al side, because of the diffusion of Si element, microhardness changes in a linear relationship with the distance.
(3) XRD results indicated that the brazing seam region is mainly consisted of α-Al solid solution, Cu_3Al_2 and $CuAl_2$ IMCs. Moreover, ε-$Cu_{15}Si_4$ phase, Al–Si phase and $CuZn_2$ phase are formed, and these phases appear in the form of acicular compounds. In the brazing seam region, α-(Al) + $CuAl_2$ binary eutectic phase is formed.

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