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Ultrasonic Brazing of Alumina to Copper Using Zn-Al-Cu Fillers†

Masaaki NAKA* and Michimasa MAEDA**

Abstract

Ultasonic Brazing was applied to join to coopper using Zn-Al-Cu filler metals containing Al plus Cu content up to 30 mass% where the ratio of Al/Cu is fixed as 3/2. The intensity of ultrasound was 1 kW and 18 kHz. The joining mechanism was investigated by measuring the joining strength and analyzing the microstructure at interface of the joint.

First, Al_2O_3 was metallized by applying ultrasound in Zn-Al-Cu filler bath. Then, the metallized Al_2O_3 was brazed with Cu using the same fillers. The increase in application time of ultrasound and metallizing temperature raises the joinig strength of Al_2O_3/Zn-Al-Cu/Cu joint. The joining strength of Al_2O_3/Cu joint exhibits the maximum at Al+Cu content of 20 mass% in Zn-Al filler, and loweres with further addition of Al+Cu content.

The application of ultrasound during brazing accelerates the removal of bubbles and the reaction at the interface between alumina and filler metal. This gives rise to wetting of Zn-Al-Cu filler against alumina, and increasing the joining strength of Al_2O_3/Cu joint.

KEY WORDS: (Ceramic-Metal Joining) (Ceramics) (Alumina) (Copper) (Ultrasonic Brazing) (Joining) (Brazing) (Zinc) (Filler Metal)

1. Introduction

Ceramics are becoming increasing important in engineering application, but their fabrication into useful components frequently requires techniques of joining ceramics to metals. The brazing methods of ceramics to metals are divided into the heat-resistant meals method[1], active filler metals method[2], molten aluminum method[3~5], oxide utilizing method[6,7]. The thermal stress arisen from the difference of thermal expansion between ceramic and metal affords detrimental effects in ceramic/metal joint after joining. Soft metals which are inserted between ceramics and metals are often used to relax the thermal stress in joints. Although the decrease in joining temperature also reduces the thermal stress in ceramic/metal joints, the wettability of brazing fillers is simultaneously impared. We have tried hitherto to improve the wettability of fillers by applying the ultrasonic wave to cermics[8,9].

This work reports, in successive from the previous one[8,9], to improve the brazeability of Zn-Al-Cu filler metals during joining of alumina to copper by applying the ultrasound.

2. Experimental

Materials used were 99.62 mass% Al_2O_3 containing 0.1 mass% SiO_2 and others and tough pitch copper containing 0.03 mass% O. The size of materials was 6 mm in diameter and 4 mm in thickness as shown in **Fig. 1. Table 1** gives a series of composition for Zn-Al-Cu filler metals containing the sum of Al plus Cu content up to 30 mass% where the ratio of Al/Cu is fixed as 3/2. **Figure 2** shows the process of ultrasonic brazing of ceramics to metal in Zn-Al-Cu filler bath. Alumina were first metallized by applying the ultrasound in Zn-Al-Cu filler bath. The intensity of ultrasound was 1 kW and 18 kHz. The brazing temperature and applying time of ultrasound were 673 to 773 K and 0 to 90 s, respectively. After metallizing alumina, alumina was lapped with copper which was coated with the same filler metal by applying the ultrasound in 3 s.

The joining strength of Al_2O_3/Cu joints brazed with

Table 1 Chemical composition of Zn–Al–Cu alloys used.

Zn-Al-Cu	Al(mass%)	Cu(mass%)	Zn(mass%)	Liquidus temperature (K)
Zn-6Al-4Cu	6	4	bal.	650
Zn-12Al-8Cu	12	8	bal.	663
Zn-18Al-12Cu	18	12	bal.	675

† Received on May 7, 1991

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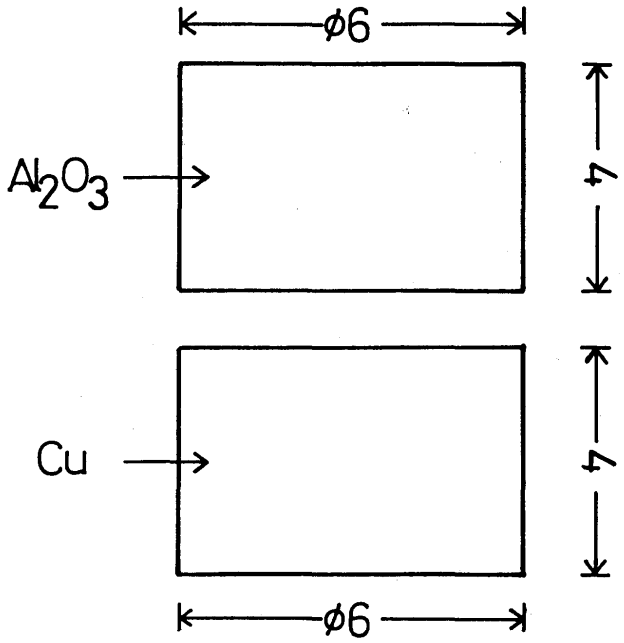

Fig. 1 Specimen size.

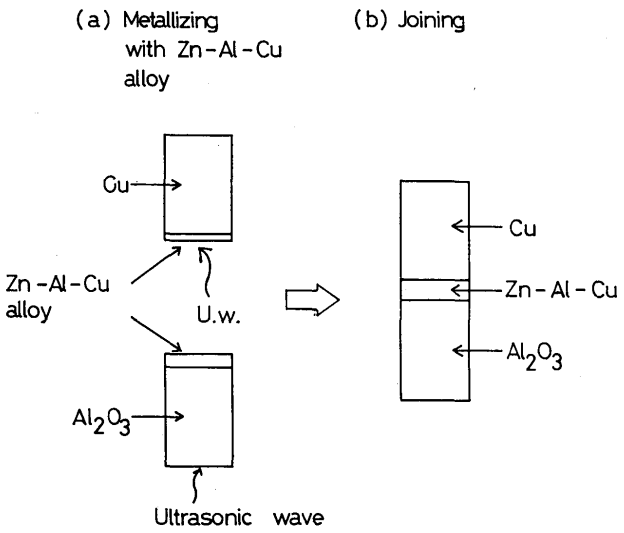

Fig. 2 Process of ultrasonic brazing of ceramics to meal.

Zn-Al filler was evaluated by fracture shear loading using a cross head speed of 1.67×10^{-2} mm/s, and the microstructure was analysed by scanning electron microscopy and EDX microanalysis.

3. Results and Discussion

3.1 Joining strength of Al_2O_3/Cu joint formed by ultrasonic brazing

Figure 3 shows the change in joining strength of Al_2O_3/Zn-6Al-4Cu/Cu joint with application time of ultrasound

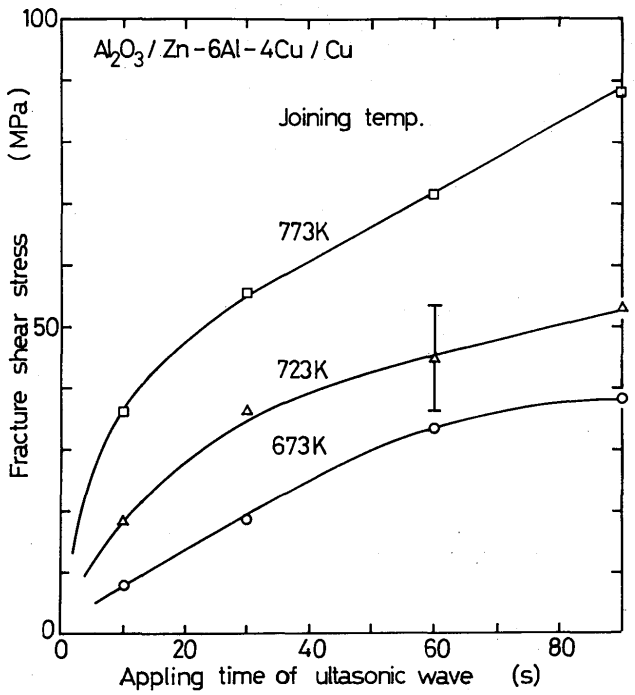

Fig. 3 Change in joining strength of Al_2O_3/Cu joint using Zn–6Al–4Cu filler with application time of ultrasonund.

at joining temperatures from 673 to 773 K. The application of ultrasound during brazing improves the strength of Al_2O_3/Cu joint at all joining temperatures. For instance, the strength of the joint increases up to 88 MPa at 90 s with increasing the application time of ultrasound at joining temperature of 773 K, although the joint without applying ultrasound doesn't be joined soundly. The ultasonic wave accelarates the motion of molten alloys in the filler bath and removes bubbles at the alumina-filler interface. Thus, the ultrasound accelerates the wetting of alloys against alumina. This results in the improvement of joining strength of Al_2O_3/Cu joints.

The similar effects of ultrasound are seen in Al_2O_3/Cu joints using other Zn-12Al-8Cu and Zn-18Al-12Cu filler metals as shown in **Figs. 4** and **5**. The longer time duration of applying ultrasound over 60 sec represents the higher strength of joint with Zn-12Al-8Cu filler than that of joint with Zn-6Al-4Cu filler. The Zn-18Al-12Cu filler with large amounts of alloying elements of Al and Cu reaches the lower strength of Al_2O_3/Cu joint, compared with other fillers.

The increase in joining temperature raises the joinig strength at a constant appying time of ultrasound. **Figure 6** represents the change in joining strength of Al_2O_3/Cu joint using Zn-6Al-4Cu filler with joining temperature at the constant applying time of 60 s. The strength of joint changes from 34 MPa at 673 K to 71 MPa at 773 K at brazing condition of 60 s. The increase in bath temperature makes the spread of ultrasound on ceramics easy.

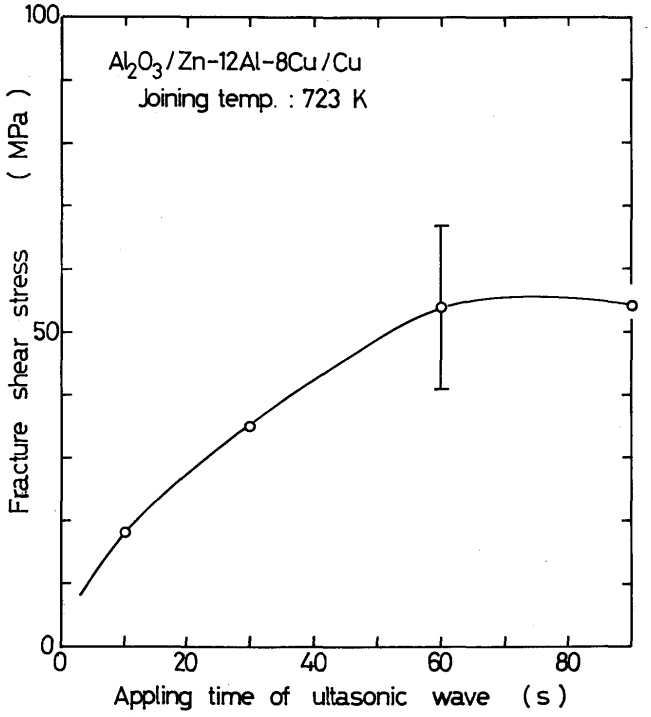

Fig. 4 Change in joining strength of Al_2O_3/Cu joint using Zn–12Al–8Cu filler at joining temperture of 723 K with application time of ultrasound.

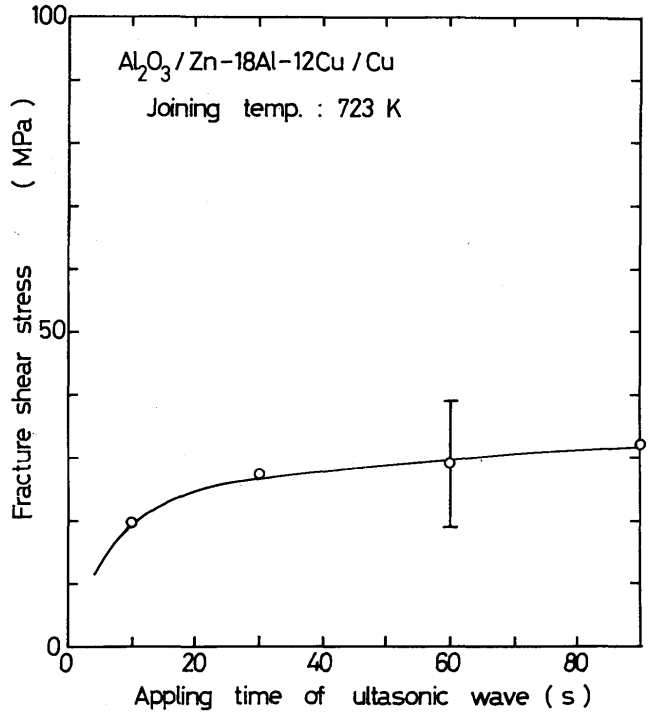

Fig. 5 Change in joining strength of Al_2O_3/Cu joint using Zn–18Al–12Cu filler with application time of ultrasound at joining temperture of 723 K.

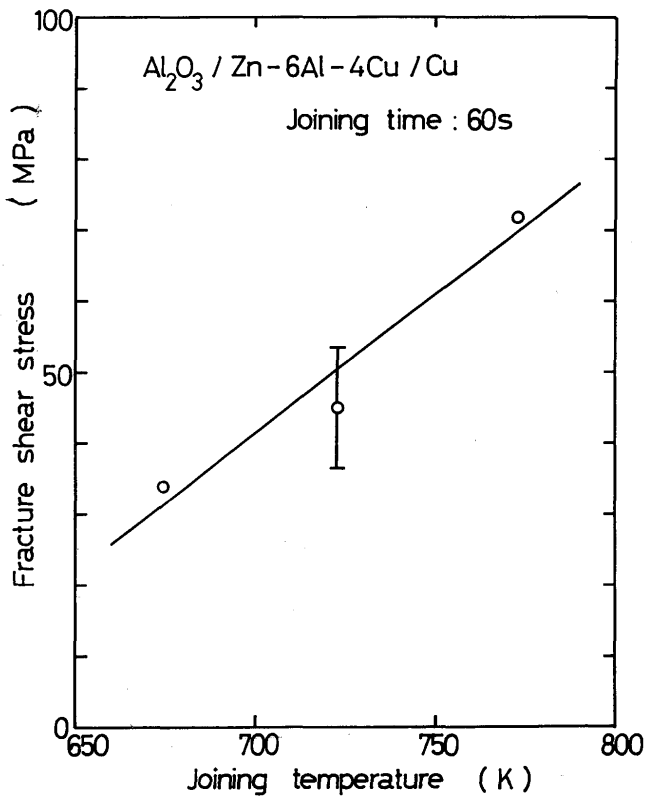

Fig. 6 Change in strength of Al_2O_3/Cu joint using Zn–6Al–4Cu filler with joining temperture at application time of 60 s.

The change in joining strength of Al_2O_3/Cu joint with Al plus Cu content in Zn-Al-Cu filler at joining condition of 723 K and 60 s is shown in **Fig. 7** where the ratio of Al/Cu is fixed as 6/4. The joining strength of Al_2O_3/Cu joint increases with increasing Zn plus Cu content in the filler from 12 MPa at 0 mass% Al, and exhibits the maximum value of 54.5 MPa at 20 mass% Al+Cu. With further adding Al+Cu content, the strength lowers to 29.5 MPa at 30 mass% Al + Cu through the maximum value. Included in the figure for comparison are the values of Al_2O_3/Cu joints with Zn and Zn-Al fillers[9]. The mixing of Al and Cu provides the higher strength, compared with that of joint containing Al as alloying element.

The increase in joining strength of Al_2O_3/Cu joint is arisen from two factors. First, the wetting of molten Zn-Al-Cu filler against Al_2O_3 is improved by an addition of aluminum in the initial stage up to Al + Cu content of 20 mass%. MacDonald[3] and Naka et al[5] have indicated that the molten aluminum definitely wets Al_2O_3. The aluminum in Zn-Al filler dissolves or reacts with Al_2O_3. This leads to accelarate the wetting of the filler against Al_2O_3. Secondly, the mechanical properties of Zn-Al-Cu fillers are improved by mixing Al plus Cu into Zn. This also raises the joining strength of Al_2O_3/Cu joint.

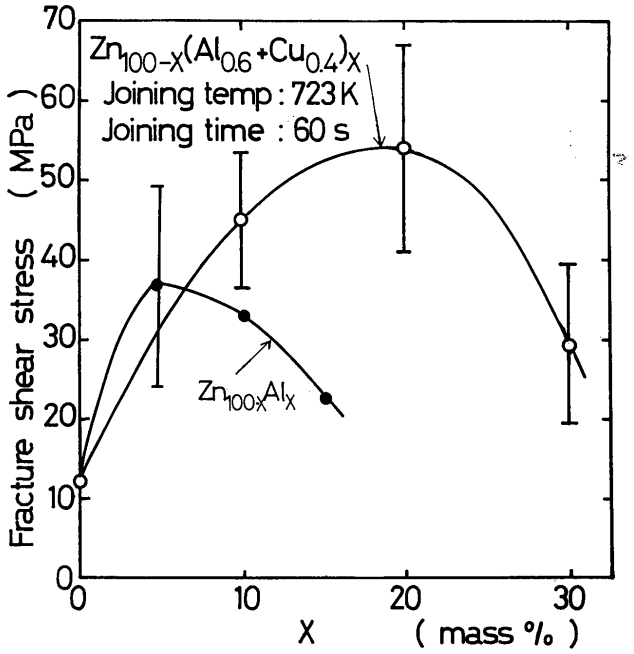

Fig. 7 Change in strength of Al_2O_3/Cu joint brazed at 723 K for 60 s with allying content X in Zn–Al–Cu or Zn–Al filler.

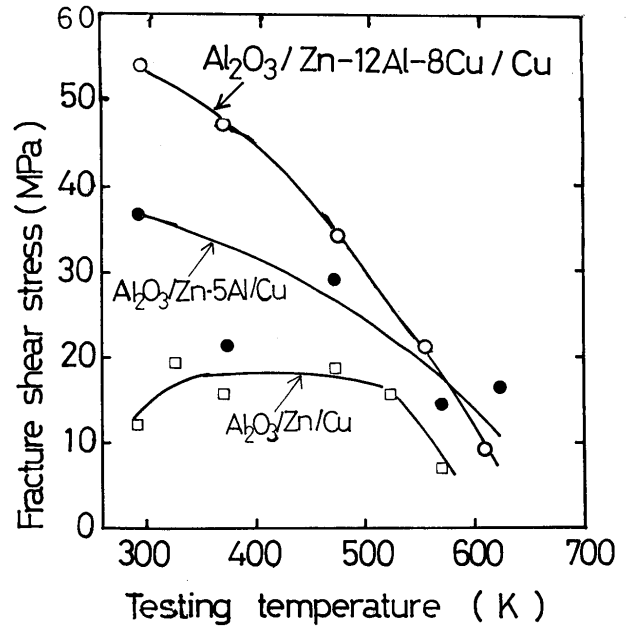

Fig. 8 Change in strength of Al_2O_3/Cu joint Zn–12Al–8Cu filler brazed at 723 K for 60 s with testing temperature.

However, the mixing of excess amounts of Al plus Cu raises the viscosity of molten Zn-Al fillers. The ultrasonioc wave can't be effectively applied to Al_2O_3 in such viscous molten fillers. Then, the high viscosity of the filler results in lowering the joining strength of Al_2O_3/Cu joint.

Figure 8 shows the joining strength of Al_2O_3/Cu joint using Zn-12Zn-8Cu filler brazed at 723 K for 60 s at testing tempertures from room temperature to 623 K. Al_2O_3/Cu joint with Zn filler maintains the joining strength up to 373 K, though the joining strength rises a litte with increasing testing temperature from room temperature. With further increasing testing temperature the joining strength of the joint lowers monotonously. Included in the figure for comparison are values of joints with Zn and Zn-5Al fillers. The joint with Zn-Al-Cu filler represents the higher strength at elevated tempertures up to 550 K, compared with the joint with Zn or Zn-5Al filler. The mixing effect of Al and Cu operates at the higher tempertures.

The fracture of Al_2O_3/Cu joint with Zn-12Al-8Cu filler takes place in the intreface between filler and alumina at room temperture in **Fig. 9**. The alumina fracture surface of joint with Zn-12Al-8Cu filler show partly the adhesion of

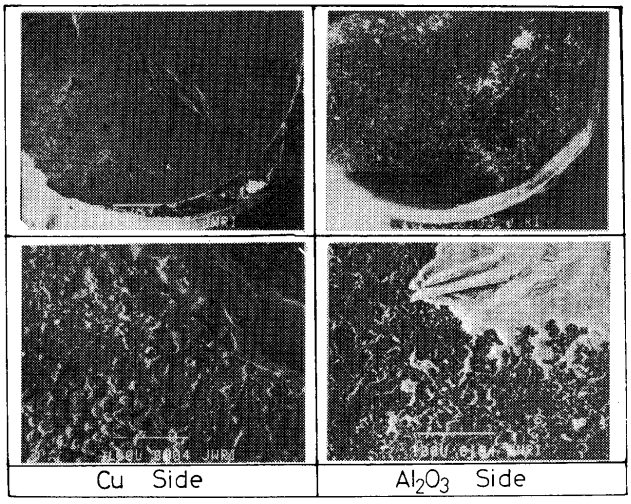

Fig. 9 Fracture surface of Al_2O_3/Cu joint using Zn–12Al–8Cu tractured at room temeprature.

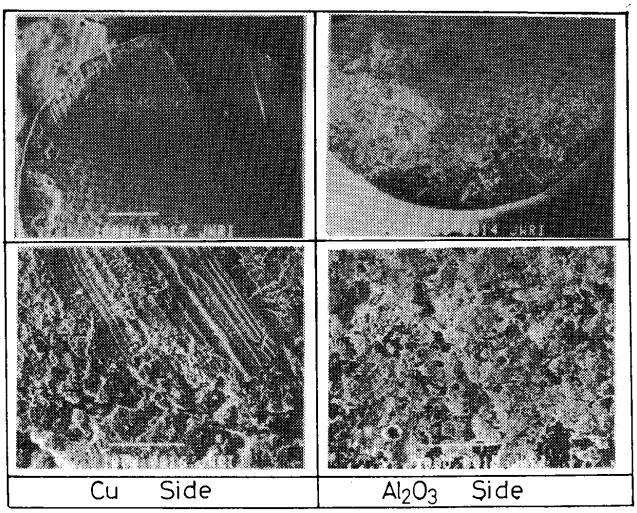

Fig. 10 Fracture surface of Al_2O_3/Cu joint using Zn–12Al–8Cu fractured at 543 K.

the filler on Al_2O_3. **Fig. 10** represents the fracture surface of Al_2O_3/Cu joint with Zn-12Al-8Cu filler at testing temperature of 543 K. The ductile shearing takes place at the filler. These observations of fracture surface of joint demonstrate that the decrease in strength of Zn-12Al-8Cu fillers at high testing temperature is attributable to the decrease in the joining strength of Al_2O_3/Cu joints.

4. Joining microstructure at interface of Al_2O_3/Cu joint

Figure 11 represents the microstructure (a), interfaces of Al_2O_3/filler(b) and filler/Cu(c) of Al_2O_3/Cu joint with Zn-6Al-4Cu filler brazed at 673 K for 60 s. The filler, which is accelerated by applying the ultrasound, well joins with Al_2O_3 without defects as shown in Fig. 11(b). The two intermediate phases are formed at the interface between the filler and copper material in Fig.11 (c). The chemical compositions of intermediate phases at filler/Cu interface of the joint with Zn-6Al-4Cu filler brazed at 673 K for 60 s are shown in **Table 2**. The phase I adjacent to the filler in Fig. 11(c) is identified as ε phase which is the same in Cu-Zn system[10]. The phase II adjacent to copper material is also identified as γ phase in Cu-Zn system. The aluminum stabilizes the Zn rich intermediates by dissolving into the phases in the table.

Figure 12 shows the microstructure(a) and interfaces of Al_2O_3/filler(b) and filler/Cu(c) of Al_2O_3/Cu joint with Zn-12Al-8Cu filler brazed at 723 K for 60 s. The filler also well wets the Al_2O_3. At the interface between the filler and Cu the two intermediate phases are formed in Fig. 12(c), and the chemical composition of the two phases are represented in **Table 3**. The phase I adjacent to the filler and phase II adjacent to copper are identified as γ phase in Cu-Zn system and τ' in Al-Cu-Zn system[12]. The intermediate phases in the joint with Zn-12Al-8Cu filler containing less amounts of zinc are formed, compared with the phases in the joint with Zn-6Al-4Cu filler.

Table 2 Chemical composition of intermediate phases in Al_2O_3/Cu joint using Zn–6Al–4Cu filler brazed at 673 K for 60 s.

	Zn	Cu	Al
		(at%)	
Phase I	75.94	20.94	3.12
Phase II	50.68	41.44	7.88

Table 3 Chemical composition of intermediate phases in Al_2O_3/Cu joint using Zn–12Al–8Cu filler brazed at 723 K for 60 s.

	Zn	Cu	Al
		(at%)	
Phase I	67.45	24.05	8.50
Phase II	41.97	31.20	26.83

5. Conclusions

The joinnig on Al_2O_3 to Cu with Zn-Al-Cu filler metals containing the sum of Al plus Cu content up to 30 mass% where the ratio of Al/Cu is fixed as 3/2 was conducted by ultrasonic brazing at brazing condition of 673–773 K and 0-90 s. The intensity of ultrsound used was 1 kW and 18 kHz. The joinig mechanism of the ultrasonic brazing was investigated by measuring the joint strength and observing the microstructure of the interface of the Al_2O_3/Cu joint.

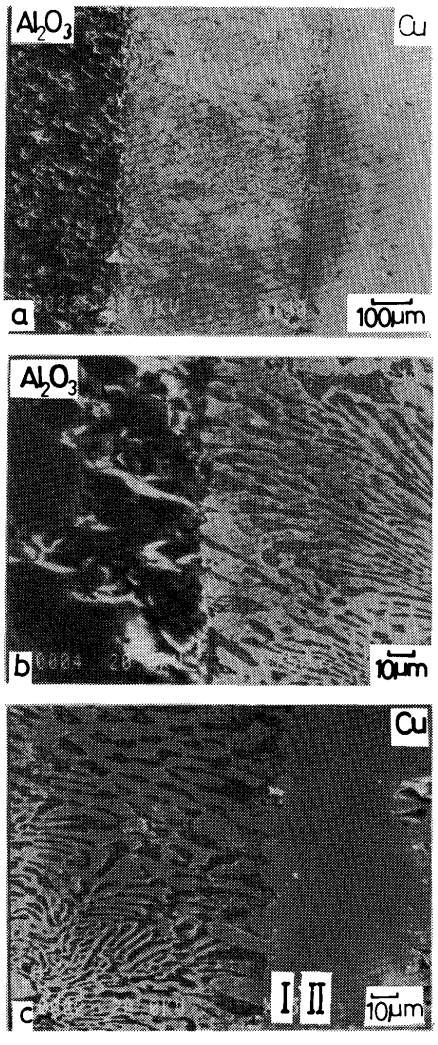

Fig. 11 Microstructure(a), interfaces of Al_2O_3/filler(b) and filler/Cu (c) in Al_2O_3/Cu joint with Zn–6Al–4Cu filler brazed at 673 K for 60 s.

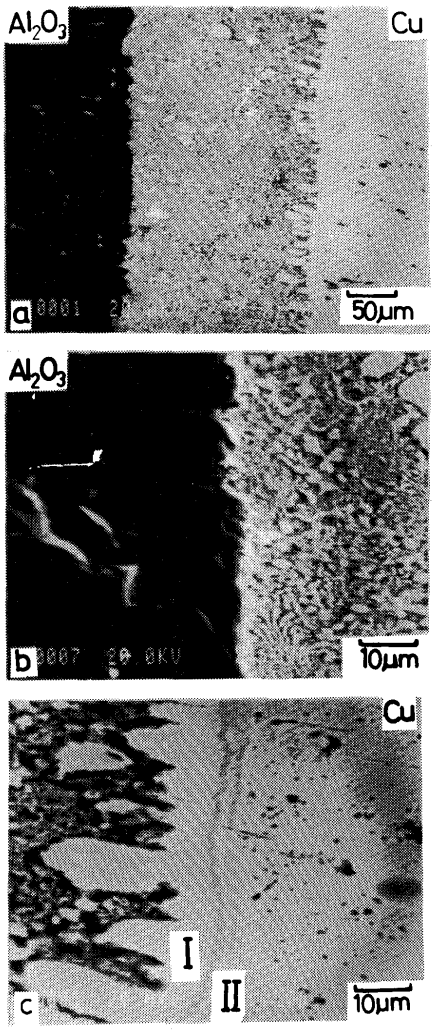

Fig. 12 Microstructure(a), interfaces of Al_2O_3/filler(b) and filler/Cu (c) in Al_2O_3/Cu joint with Zn–12Al–8Cu filler brazed at 723 K for 60 s.

The increase in applying time and brazing temperature raises the joining strength of Al_2O_3/Cu joints with Zn-Al-Cu filler. The ultrasonic wave activates the motion of molten alloys in the Zn-Al fillers, and accelarates the wetting of alloys of alloys against Al_2O_3. This leads to the increase in joining strength of the joint.

The addition of Al plus Cu to Zn filler up to 20 mass% improves the wettability and mechanical properties of the filler against Al_2O_3. This results in the superior strength of Al_2O_3/Cu joints brazed with Zn-Al-Cu filler under applying the ultrasound.

While the Zn-Al-Cu fillers directly join with Al_2O_3, the fillers form the intermediate phases between the fillers and copper.

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