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Brazing of Si_3N_4 to Metals with Al-Cu Filler Metals†

Masaaki NAKA*, Masao KUBO and Ikuo OKAMOTO*****

Abstract

The sessile drop technique has been used to measure the contact angle of Al-Cu alloys on Si_3N_4 in a vacuum, and evaluate the work of adhesion of the alloys as a measure of adhesion intensity.

The work of adhesion for Al-Cu alloys on Si_3N_4 at 1373 K departs to the plus side from the ideal mixing of aluminum and copper at the copper content below 50 at%, and exhibits the maximum of 1.42 Jm^{-2} for 4 mass% Cu. The wettability of aluminum on Si_3N_4 is improved by mixing of aluminum and copper at aluminum rich alloys.

The brazing of Si_3N_4 to metal was performed using Al-4 mass% Cu filler where metal is Ti, Nb, Ni or SUS304. The superior work of adhesion and mechanical property of the Al-Cu filler provide the superior strength of Si_3N_4/metal joint.

KEY WORDS : (Ceramic Joining) (Ceramic Metal Joining) (Joining) (Brazing) (Ceramics) (Brazing) (Silicon Nitride) (Aluminum) (Copper)

1. Intorduction

Joining of ceraimcs to metals expands the practical application of ceramics. Since the filler metals have to possess the good wettability against ceramics, the reactive elements such as titanium are used for the filler metals.

On the other hand, aluminum also wets various kinds of ceramics of Al_2O_3[1], ZrO_2[2], Si_3N_4[3] and SiC[4]. Aluminum possesses the high work of adhesion as a measure of wettability on ceramics with high temperature dependence among aluminum, copper and silver[2,3,4].

The stress arisen from the difference in thermal expansion between ceramics and metals affects the ceramic/metal joint. Aluminum is a soft metal which relaxes the stress in the joint. These facts indicates that aluminum is used as the brazing filler. The studies, however, are so far focused on the behavior of pure aluminum.

The purpose of present work is to clarify the effect of copper as alloying element on the wettability of aluminum on Si_3N_4, and join to metals with Al-Cu filler metals.

2. Expeimental Procedure

The ceramics used was pressureless sintered Si_3N_4 with a few percent of Al_2O_3 and Y_2O_3, and the metals used were Nb, Ti, Ni and SUS 304. Al-Cu alloys containing 0, 4, 10, 30, 60, 75 and 100 mass% Cu were prepared by arc-melting the high pure aluminum and copper in argon gas. **Table 1** presents the chemical composition and liquidus temperature of filler metals.

The wettability of molten alloy was evaluated by measuring the contact angle between the peripheral surface of a small drop of molten metal and horizontal surface of the ceramic substrate in a vacuum below 1.33 mPa.

Si_3N_4 of 15 mm in diameter and 3 mm in thickness, were lapped to metal of 6 mm in diameter and 3 mm thickness with Al-Cu fillers under a loading of 10 g. First, the metallizing of Si_3N_4 with Al-4 mass% Cu filler was conducted at 1373 K for 3.6 ks in 1.33 mPa, and then the lap joint with the filler thinned down to 0.1 mm thickness was made in the joining codition of 1273 K ~ 973 K and 300 s where SUS304 was Ni-plated. The thickness of the filler metal after joining was about 25 μm.

The joinig strength of the lap joint was determined by fracture loading using a crosshead speed of 1.7×10^{-2}

Table1 Chemical composition and liquidus temperture of Al-Cu alloys used.

Filler metal	Copper content (mass%)	Liquidus temperture (K)
Al	0	933
Al-4Cu	4	918
Al-10Cu	10	903
Al-30Cu	30	838
Al-60Cu	60	928
Al-75Cu	75	1321
Cu	100	1356

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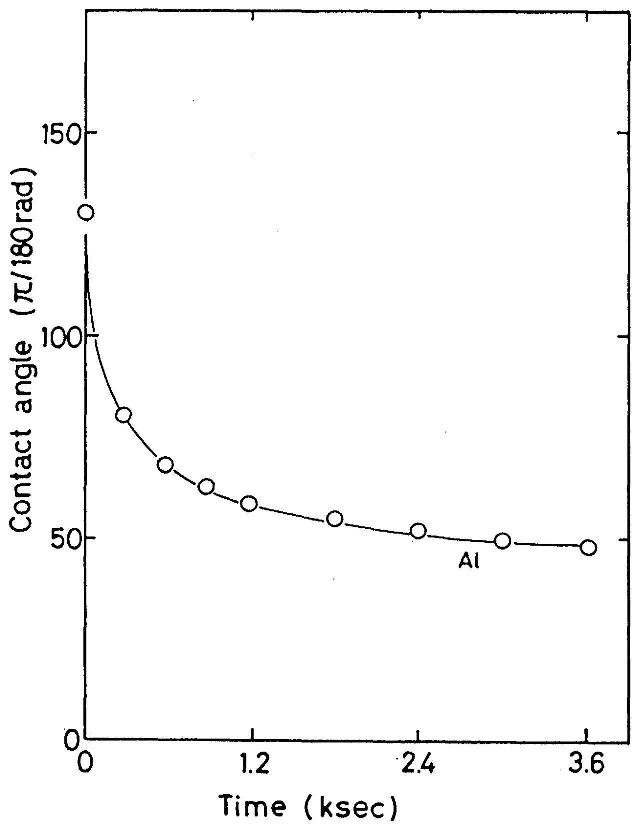

Fig. 1 Change in contact angle of molten aluminum on Si_3N_4 with time at 1373 K.

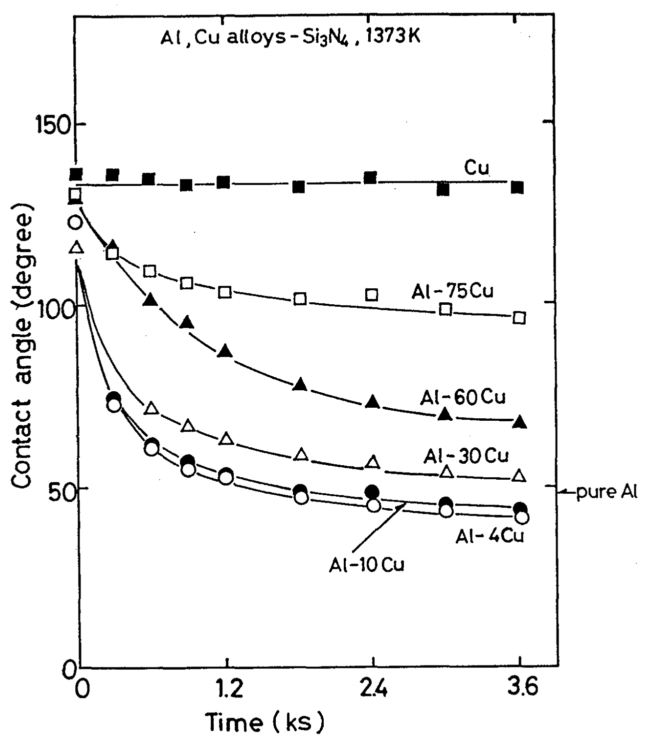

Fig. 2 Change in contact angle of Al-Cu alloys on Si_3N_4 with time at 1373 K.

mm·sec^{-1}. The microstructures and element distribution of joints were determined by means of a scanning elecron microscope and an energy dispersive microanalyser, respecticvely.

3. Results and Discussion

3.1 Wetting behavior of Al-Cu alloys

Figure. 1 shows the change in contact angle of molten aluminum on Si_3N_4 with time at 1373 K. The contact angle of molten aluminum lowers definitely with increasing time, and reaches the equilibrium value of 49 degree as shown in the figure. **Fig. 2** represents the change in contact angle of Al-Cu alloys on Si_3N_4 with time at 1373 K. Al-Cu alloys exhibits the time dependence of contact anagle. The addition of copper content up to 10 at% represents the lower contact angles than that of aluminum, and the more addition of copper raises the contact angle. The contact angle of molten copper is 135 degree with no time dependence. **Fig. 3** represents the equilibrium contact angle of Al-Cu alloys

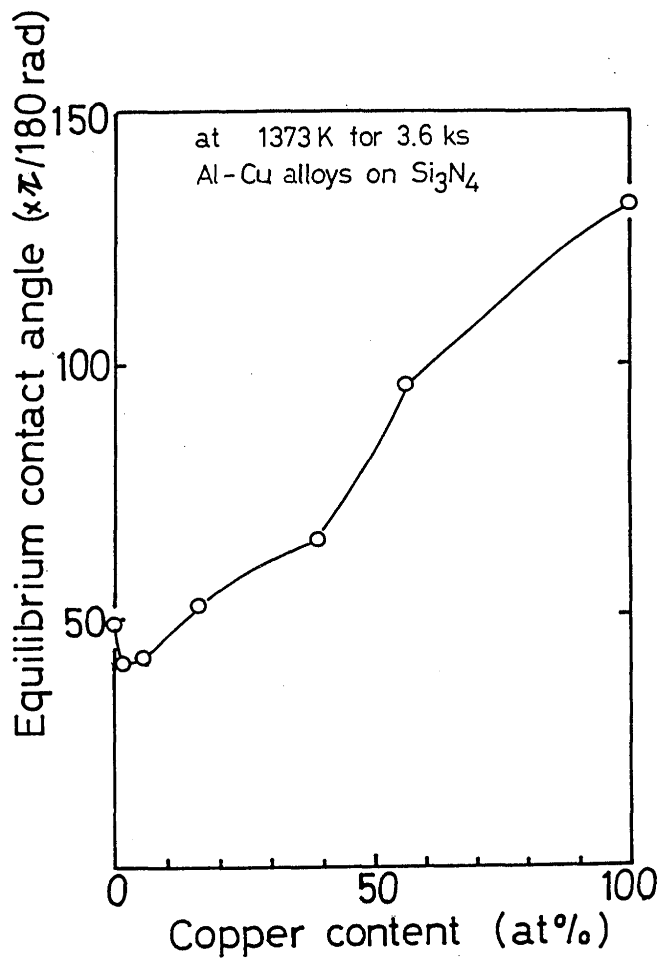

Fig. 3 Copper content dependence of equilibrium contact angle for Al-Cu alloys on on Si_3N_4 with time at 1373 K.

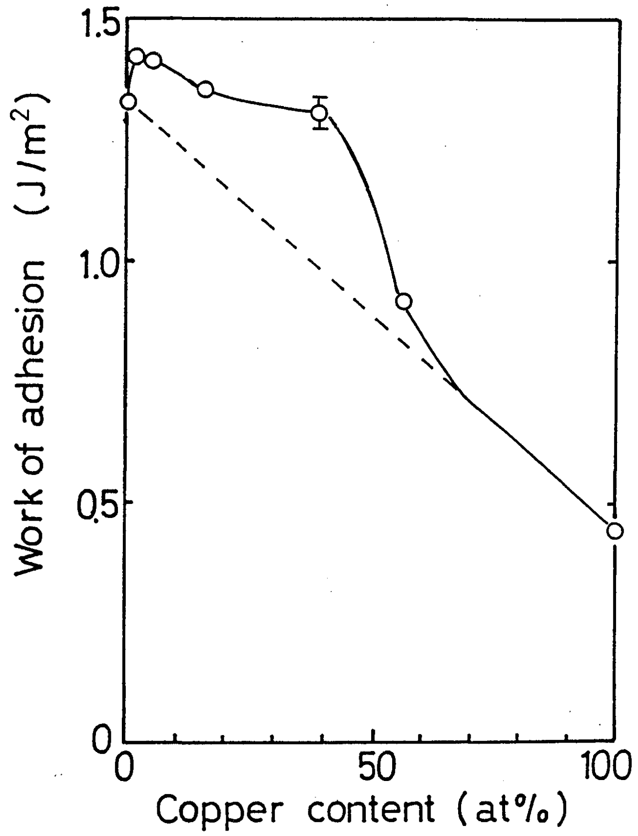

Fig. 4 Copper content dependence of work of adhesion for Al-Cu alloys on on Si_3N_4 with time at 1373 K.

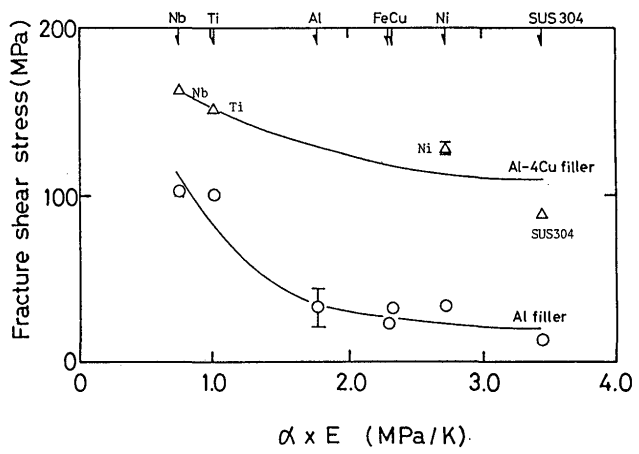

Fig. 5 Strength of Si_3N_4/metal joint plotted as a function of α ·E, where and E are thermal expansion coefficient and elastic modulus of metal, respectively.

as a function of copper contact at 1373 K. The equilibrium contact angle exhibits a minumun value of 40 degree at a copper contact of 4 mass%, and the angle increases with higher copper contact. The melt at the contact angle between 0 and 90 degree is said to be the wetting state. The wetting of Al-Cu alloys on Si_3N_4 is said to occur at the copper contact below 50 at%.

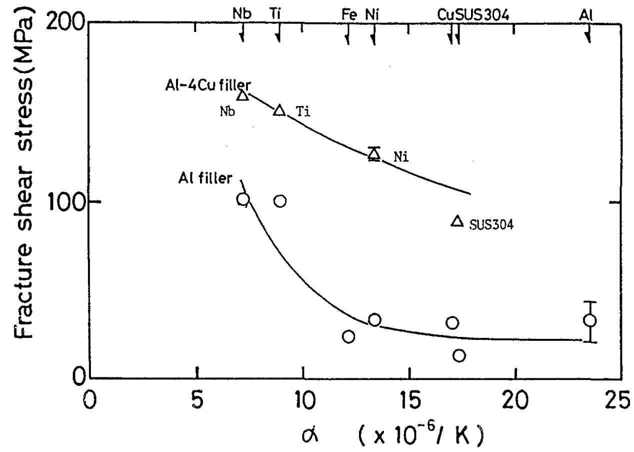

Fig. 6 Strength of Si_3N_4/metal joint plotted as a function of α, where α is thermal expansion coefficient elastic modulus of metal.

The wettability of ceramics by metals is often described by the work of adhesion, W_{ad}, for the metal. Wad is defined by the Young-Duprè equation as follows,

$$W_{ad} = \gamma_{LG} (1 + \cos \theta_{\infty}) \quad (1)$$

where γ_{LG} is the liquid energy, and the present values of θ_{∞} and γ_{LG} for Al-Cu alloy at 1373 K[5] are used. As shown in **Fig. 4** in which the copper content is plotted as at% the W_{ad} exhibits a maximum of 1.42 Jm^{-2} for 1.7 at% (4 mass%) Cu alloy from 1.33 Jm^{-2} for aluminum and the addition of copper content up to 20 at% increases the W_{ad} of the alloys. The alloys containing 40 at% copper content gives almost the same value as that of aluminum, and then the W_{ad} decreases markedly with increasing copper content to 0.45 Jm^{-2} for copper. The W_{ad} of Al-Cu alloys containing a copper up to about 65 at% departs to the plus side from the ideal mixing, indicationg by the dotted line in Fig. 4. This results that the wettability of alloys is improved by the addition of copper to aluminum.

3.2 Joining strength of Si_3N_4/Metal joints using Al-Cu filler metals

The joining of Si_3N_4 to a variety of metals was performed by Al-4 mass% Cu filler metal which provides the highest wettability against Si_3N_4. **Fig. 5** represents the relationships between the strength of Si_3N_4/metal joint with Al-4 mass% Cu filler and the factor of α ·E, where α and E are the thermal expansion coefficient and the elastic modulus of metal, respectively. In this figure included the strength of Si_3N_4/metal joint with Al filler metal. The factor of α ·E is a measure of stress in the Si_3N_4/metal joint. Although the sound joining of Si_3N_4 to metal is made using Al base filler metal, the stress arisen from the thermal expansion between Si_3N_4 to metal affects the strength of joint. The metals of Nb and Ti which possess the lower $\alpha \cdot E$ factor provide the higher strength of Si_3N_4/metal joint. The strength of Si_3N_4/metal joint is also plotted against factor of α , where α is the thermal expansion coefficient in **Fig. 6.** The factor represents the same measure as the factor of $\alpha \cdot E$.

The strength of Si_3N_4/metal joint using Al-4 mass% Cu filler metal provides the higher strength than that of Si_3N_4/metal joint using Al filler metal. The mixing of copper to aluminum improves the work of adhesion of aluminum on Si_3N_4. Furthermore, the addition of copper to aluminum privides the superior mechanical property of alloys. In other words, the superior work of adhesion and mechanical property of the Al-4 mass% Cu filler metal provide the superior strength of Si_3N_4/metal joint brazed with the filler metal.

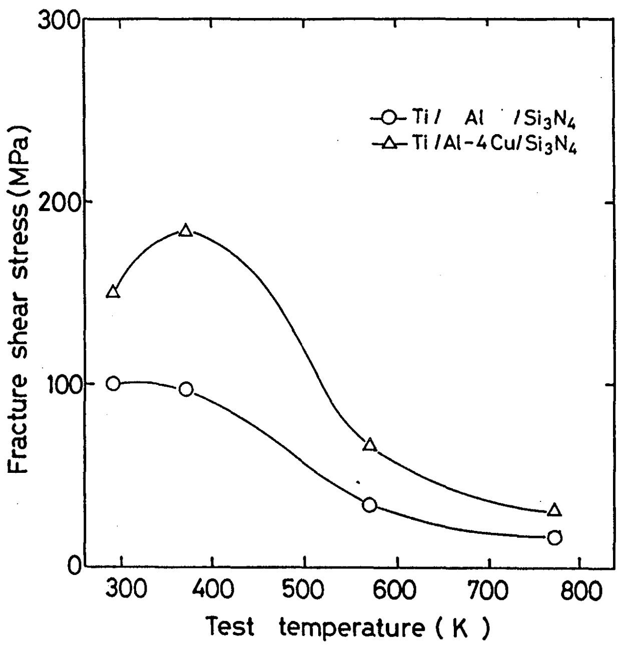

Fig. 7 Strength of Si_3N_4/Ti joint at elevated tempertures.

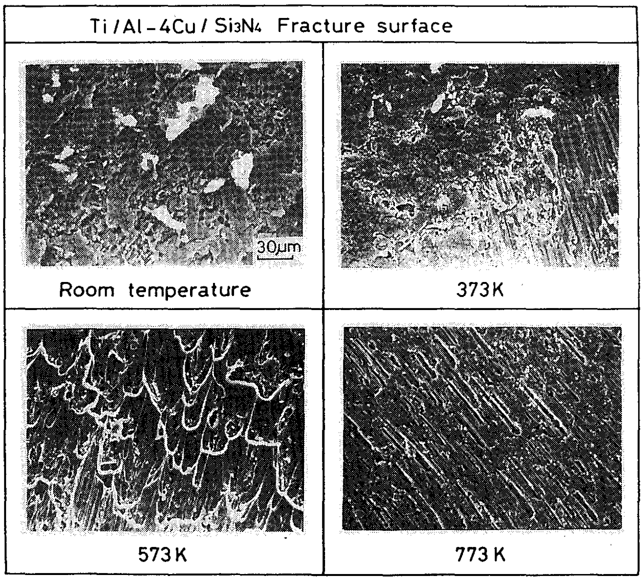

Fig. 8 Fracture structure of Si_3N_4/Ti joint at elevated tempertures.

The testing tempature dependence of the strength of Si_3N_4/Nb joints in **Fig. 9** using Al and Al-4 mass% Cu maximum at 373 K in **Fig. 7** where the strength of Si_3N_4/Ti joint with Al filler is included. The strength of joint with Al-4 mass% Cu filler lowers with higher testing temperature. The general ductile fracture of the joint at temperatures above 573 K in **Fig. 8** demonstrates that the decrease in strength of joint with Al-4 mass% Cu filler is atributable to the decrease in strength of Si_3N_4/Ti joint. The strength of joint with Al-4 mass% Cu filler is higher than that of joint with Al filer at every temperature.

The testing tepature dependence of the strength of Si_3N_4/Nb joints in **Fig. 9** using Al and Al-4 mass% Cu filllers exhibits the maximum values of 110 and 220 MPa, respectively. The release of stress in the joints accounts for the increase in strenght at the jopints at testing temperature of 373 K. Furthermore, the strength of Si_3N_4/Nb joints falls with increasing testing temperatures. The superior strength of Si_3N_4/Nb joint with Al-4 mass% Cu filler is mainted at testing temperatures up to 773 K.

The change in fracture structure of Si_3N_4/Nb joint with Al-4 mass% Cu filler with testing temperture is shown in **Fig. 10.** The fracture structure mixed with the ductile fracture structure of the filler and the brittle fracture of Si_3N_4 in the Si_3N_4/Nb joint is seen at testing temperatures

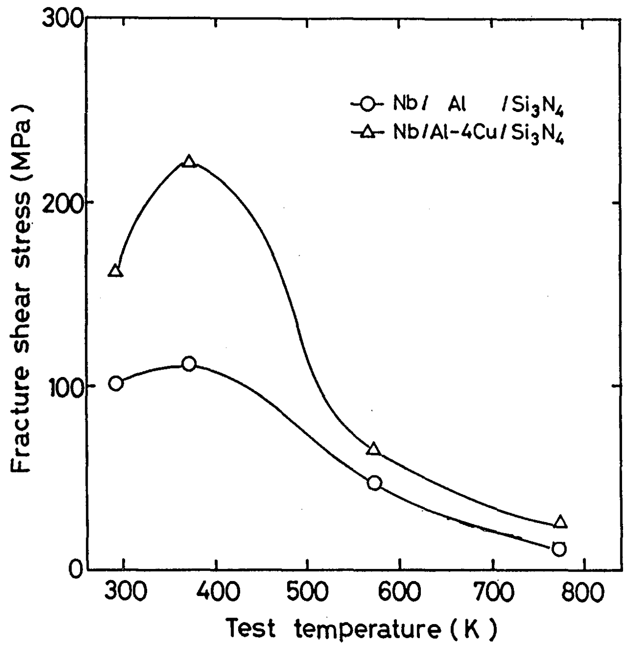

Fig. 9 Strength of Si_3N_4/Nb joint at elevated tempertures.

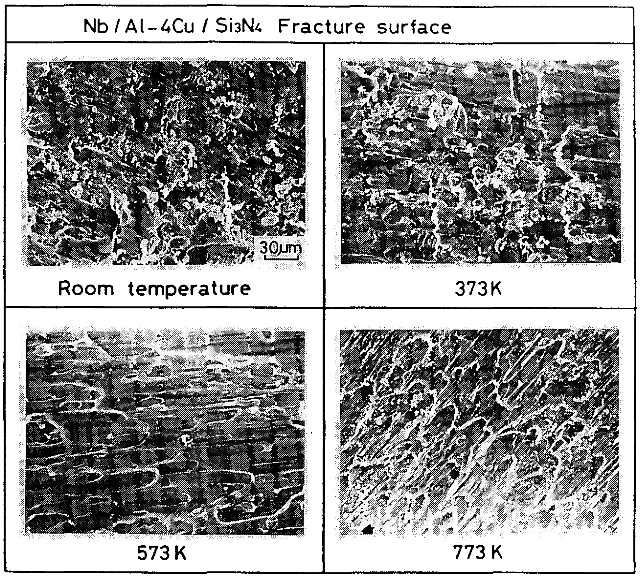

Fig. 10 Fracture structure of Si_3N_4/Nb joint at elevated tempertures.

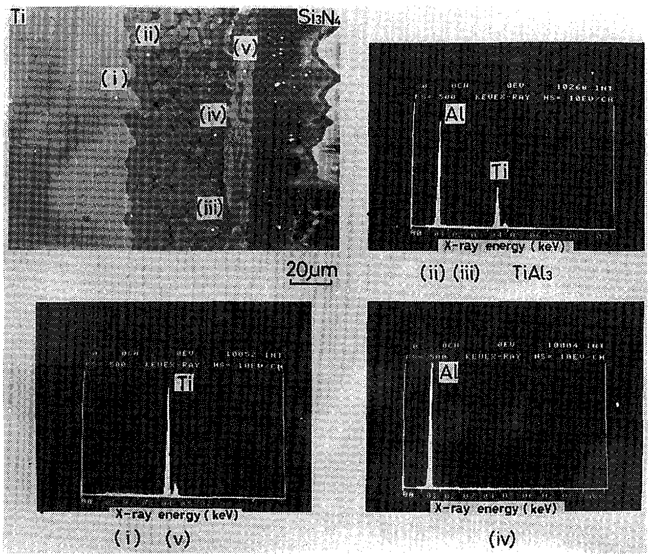

Fig. 11 Microstructure and spot analyses for Ti and Al in Si_3N_4/Ti joint.

below 373 K, and the general ductile fracture structure of the filler is seen at tempertures above 573 K. The decrease in strength of Al-4 mass% Cu filler accounts for the decrease in strength of Si_3N_4/Nb joint.

3.3 Joining microstructure of Si_3N_4/Ti or Si_3N_4/Nb joint

The microstructure and spot analyses for Ti and Al in Si_3N_4/Ti joint with Al-4 mass% Cu Filler are shown in **Fig. 11.** The granular $TiAl_3$ phases in (ii and iii) are found at the joining layer beside Ti, and the titanium erroded is seen at the interface beside Si_3N_4 in (iv) of the figure.

The microstructure and spot analyses for Nb, Al and

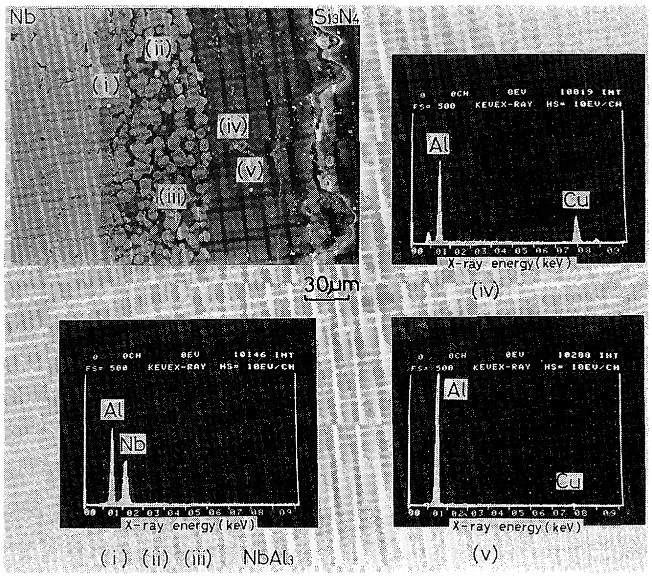

Fig. 12 Microstructure and spot analyses for Nb, Al and Cu in Si_3N_4/Nb joint.

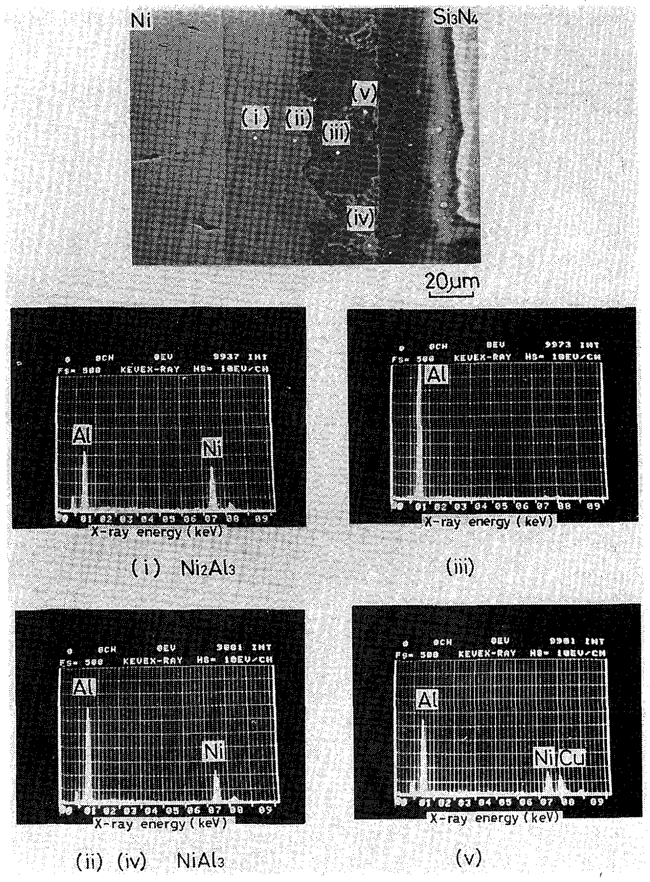

Fig. 13 Microstructure and spot analyses for Ni, Al and Cu in Si_3N_4/Ni joint.

Cu in Si_3N_4/Nb joint with Al-4 mass% Cu Filler are seen in **Fig. 12.** The granular $NbAl_3$ phases in Fig. 12 (i)-(iii) are found in the joining layer beside Nb. The Al-4 mass% Cu Filler remains in the filler. The microhardness of H_V

= 600 for $NbAl_3$ in the joint with Al-4 mass% Cu Filler is higher than that of H_V = 500 for $NbAl_3$ in the joint with Al Filler. The inprovement of mechanical properties of $NbAl_3$ phase and Al-4 mass% Cu filler increase the strength of Si_3N_4/Nb joint using Al-4 mass% Cu filler.

Figure 13 shows the microstructure and spot analyses of elements for Si_3N_4/Ni joint using Al-4 mass% Cu filler. The layer phases of Ni_2Al_3 in (i) and $NiAl_3$ in (ii) and (iv) of the figure are found in the joining layer beside Ni, and the Al-4 mass% Cu filler remained in the joining layer.

4. Conclusions

The contact angle of Al–xCu alloys (x = 0–100 mass%) on Si_3N_4 were measured by a sessile drop technique in a vacuum. The equilibrium contact angle of alloys at 1373 K exhibits a minimum of 40 degree at 4 mass% Cu content, and furthermore the equilibrium contact angle rises with increasing copper content in the alloys. In contrary to the trend of contact angle, the work of adhesion for alloys on Si_3N_4 at 1373 K changes from 1.33 Jm^{-2} for Al through the maximum of 1.42 Jm^{2-2} for Al-4 mass% Cu alloy to 0.45 Jm^{-2} for Cu. The wettability of aluminum on Si_3N_4 is improved by mixing the copper up to 20 at%.

The joining of Si_3N_4 to metal was performed using Al-4 mass% Cu which possesses the highest wettability on the ceramics where metal is Ti, Nb, Ni or SUS304. The superior work of adhesion and mechanical property of the Al-4 mass% Cu filler provide the higher strength of Si_3N_4/metal joint with the Al- Cu filler, compared with the strength of Si_3N_4/metal joinat with Al filler.

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