Brazing of Si_3N_4 to Metals with Al Filler (Report II)†

— Si_3N_4/Fe, Ni, Cu, Al or SUS304 Joint —

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Abstract

The joining of silicon nitride Si_3N_4 to metal was conducted using aluminum filler metal in a vacuum, where metal was Fe, Ni, Cu, Al or SUS 304.

Si_3N_4, first, was metallized at 1373K for 3.6 ks and then brazed to metal by remelting the metallized aluminum layer. The strength of silicon nitride-metal joints are related to factor α or $\alpha \cdot E$ of metal, where α and E are the thermal expansion coefficient and elastic modulus of metal, respectively. Cu and SUS 304 which possess the high thermal expansion coefficient reduce the strength of Si_3N_4/Metal joint.

KEY WORDS : (Ceramic-Metal Joining) (Joining) (Brazing) (Ceramics) (Silicon Nitride) (Aluminum) (Filler Metal) (Iron) (Nikel)

1. Introduction

One of the more difficult problems in the application of structural ceramics has been the joining of ceramics to metals[1, 2]. Ceramic/metal joining expands the application of ceramics for the structural components. The filler metals for brazing ceramics have to the superior wettability against ceramics. The wettability of metals is often measured by observing the contact angle of molten metals on ceramics[3,4]. Naka et al.[5] have reported that aluminum among aluminum, copper and silver shows the lowest equlibrium contact angle with the highest negative temperature dependencence against silicon nitride, indicating the best wettability among the metals.

Aluminum also have the superior mechanical properties to relax the stress in the ceramic/metal joint, which is arisen from the difference of themal expansion between ceramic and metal.

This work in the successive report from the previous one[6] tries to join silicon nitride Si_3N_4 to metals using aluminum filler and clarify the dominating factors in mechanical properties of the ceramic/metal joint.

2. Experimental Procedure

The pressureless sintered Si_3N_4 containing a few percent of Al_2O_3 and Y_2O_3 was used. The purity of aluminum was 99.99 mass %. The metals were Fe, Ni, Cu, Al and SUS304 in high purity. Si_3N_4 of 15 mm in diameter and 3 mm in thickness, and metal of 6 mm in diameter and 3 mm in thickness were used for a lap joint as shown in **Fig 1.** Cu and SUS304 were nickel-electroplated to prevent the reaction Al filler metal with the metals.

First, the metallizing of Si_3N_4 with aluminum was conducted at 1373 K for 3.6 ks in 1.3 mPa, and then the lap joint with aluminum thinned down to 0.1 mm thickness was made in the joining condition of joining temperature of 973K and brazing time of 300s under a load of 10 g. The joining condition for Al using 4004 (Al-Si-Mg filler) was 973 K for 180s.

The joining strength of the lap joint was determined under shear fracture loading at a cross head speed of 1.7

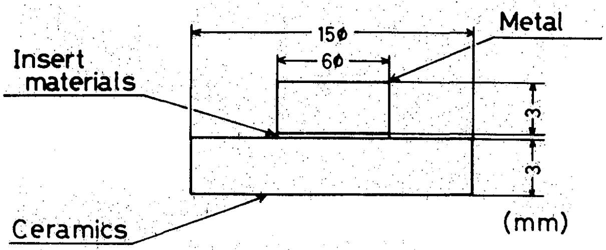

Fig. 1 Si_3N_4/metal joint.

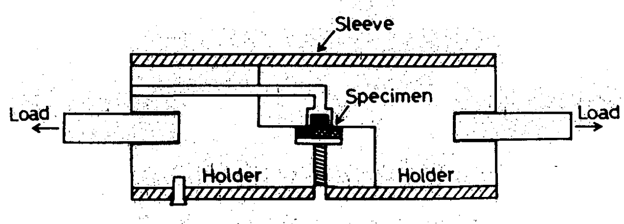

Fig. 2 Jig for fracture shear testing

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$\times 10^{-2}$ mms^{-1} as shown in **Fig. 2.** The microstructure of Si_3N_4/metal was investigated by means of scanning electron microscope and EDX microanalyser.

3. Results and Discussion

3.1 Joining strength of Si_3N_4/metal joint

Although the sound brazing of Si_3N_4 to metal was done using Al filler, the detrimental effects of mismatches between the thermal expansion coefficient of Si_3N_4 and that of the metal components are shown in **Figs. 3** and **4**, where α and E are the metal thermal expansion coefficient and metal elastic modulus, respectively. The data of joints with Nb and Ti is also included in the figures.[6]. The larger mismatch in thermal expansion coefficient between Si_3N_4 and metal reduces definitely the strength of the joint. The thermal stress in the joint is arisen from the difference in thermal shrinkage between cermic and metal during cooling after brazing. Niobium and titanium which possess the lower thermal expansion coefficients provide the higher joining strength, and Cu, Al and SUS304 that have the higher thermal expansion coefficients represent the lower joining strength.

The tensile thermal stress σ_i in ceramic of ceramic/metal joint is represented by the following equation.

$$\sigma_i = A_i \Delta \alpha \Delta T \qquad (1)$$

where A_i is related to the elastic modulus E and poisen ratio ν of materials, and

$$\Delta \alpha = \alpha_{metal} - \alpha_{ceramic} \qquad (2)$$

$$\Delta T = T_{joining} - T_{room} \qquad (3)$$

where $\Delta \alpha$ and ΔT are the difference in thermal expansion coefficient between ceramic and metal, and the difference between joining temperature and room temperature, respectively.

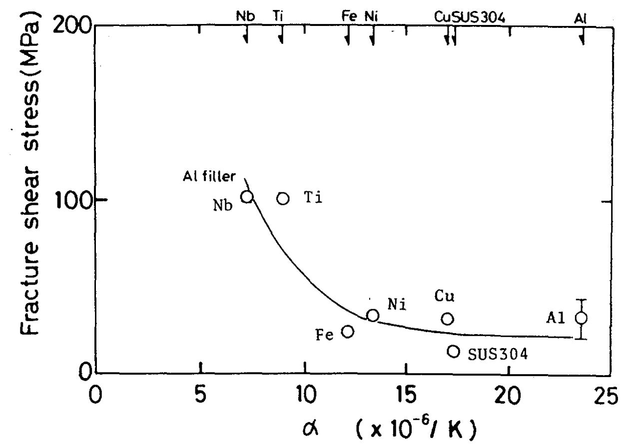

Fig. 3 Joining strength of Si_3N_4/metal joint plotted as a function of α ; metal thermal expansion coefficient.

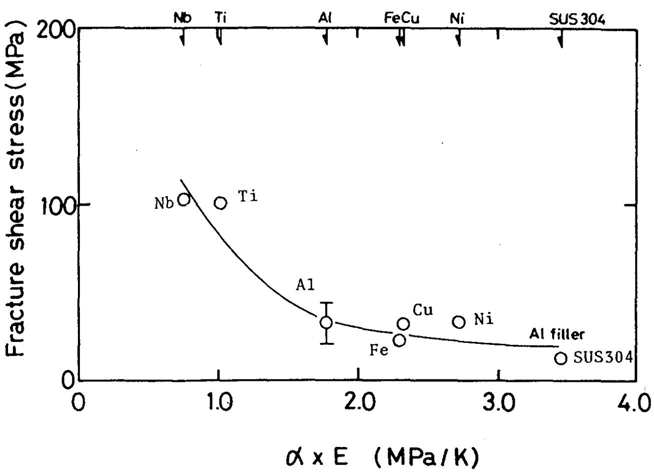

Fig. 4 Joining strength of Si_3N_4/metal joint plotted as a function of $\alpha \cdot$E, where α and E are thermal expansion coefficient and elastic modulus of metal, respectively.

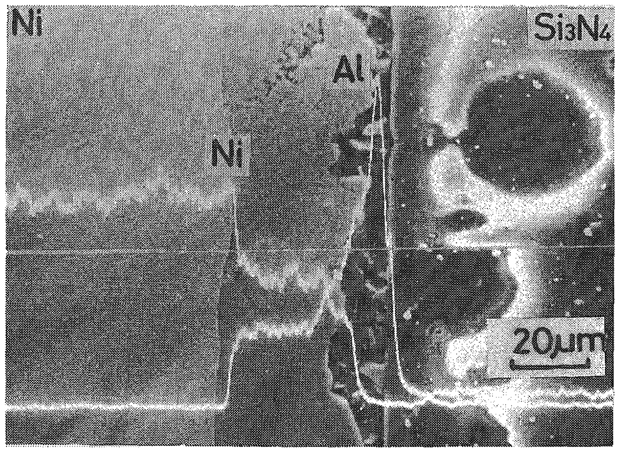

Fig. 5 Microstructure and line analyses for Ni and Al in Ni/Al/Si_3N_4 joint.

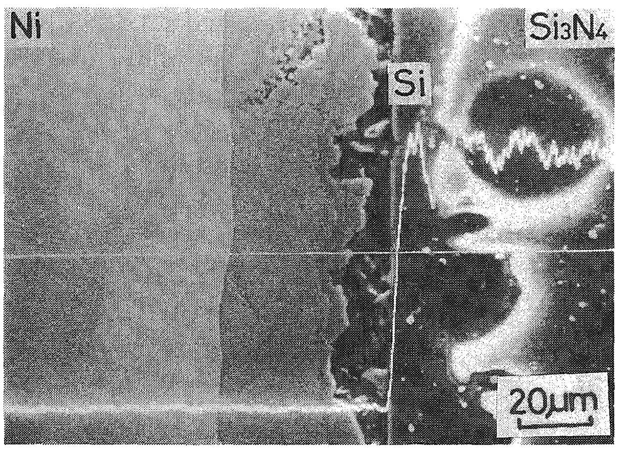

Fig. 6 Microstructure and line analyses for Si in Ni/Al/Si_3N_4 joint.

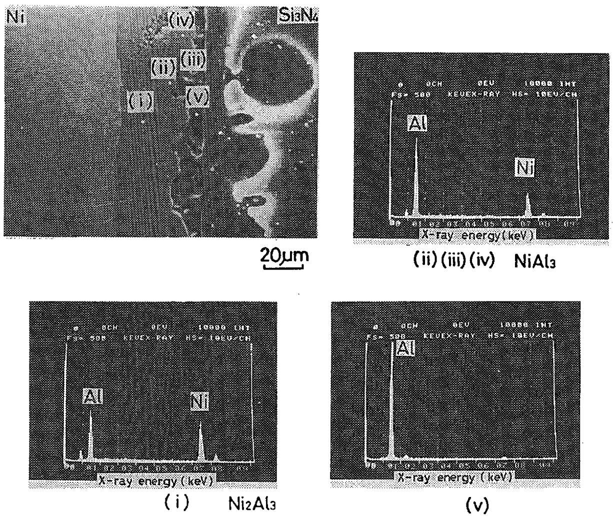

Fig. 7 Microstructure and spot analyses in $Ni/Al/Si_3N_4$ joint.

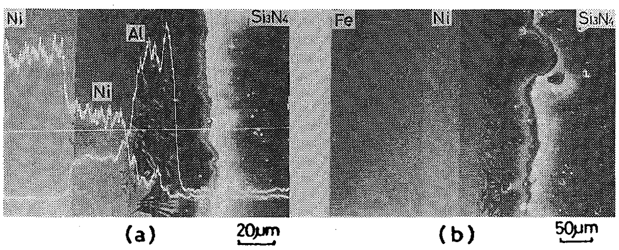

Fig. 8 Microstructure and line analyses for Ni and Al in $Fe/Ni/Al/Si_3N_4$ joint.

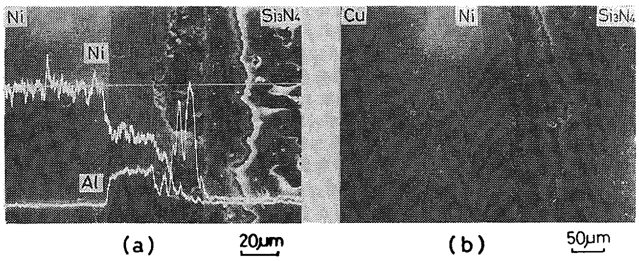

Fig. 9 Microstructure and line analyses for Ni and Al in $Cu/Ni/Al/Si_3N_4$ joint.

Eq.(1) suggests that the smaller factor of $\Delta\alpha$ provides the decrease in thermal stress σ_i in ceramic/metal joint. Thus, the usage of Nb and Ti which possess the lower thermal expansion coefficient gives the higher joining strength of Si_3N_4 to metal.

3.2 Joining microstructure of Si_3N_4/metal joint

The microstructural analyses were performed using scanning electron microscopy and X-ray (EDX) microanalysis.

The microstructure and line analyses for Ni, Al and Si in Si_3N_4/Al/Ni joint brazed with Al filler at 973 K for 300 s are shown in **Figs. 5 and 6.** In the joining layer, two intermediate phases are formed between Ni and Al filler.

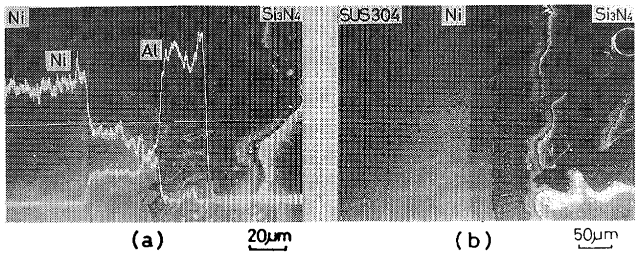

Fig. 10 Microstructure and line analyses for Ni and Al in SUS $304/Ni/Al/Si_3N_4$ joint.

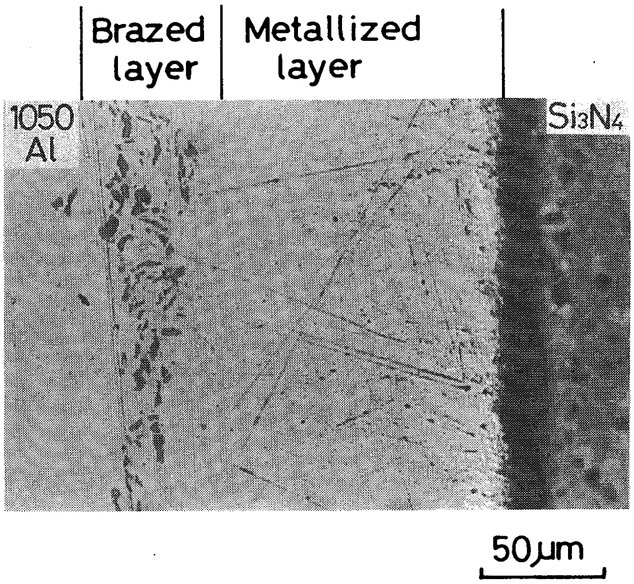

Fig. 11 Microstructure of Al/Si_3N_4 joint with 4004 filler metal.

The intermediate phases are analysed by using EDX spot analyses as shown in **Fig. 7.** The first thick phase beside Ni, and the second phase are identified as Ni_2Al_3 and $NiAl_3$ using EDX spot analyses as shown in Fig. 7.

In order to prevent the formation of the brittle intermediate phases between Fe, Cu and SUS 304, and Al filler the metals and alloys are Ni electroplated as shown in **Figs. 8, 9, 10.** At the joining layer between these metals and alloy the same intermediate phases of Ni_2Al_3 and $NiAl_3$ are formed.

The joining Si_3N_4 to Al was done using 4004 filler by remelting the metallizing Al layer on the ceramics as shown in **Fig. 11.** The 4004 filler was remained between aluminum and the metallizing layer. Since the remained 4004 filler contains the large amounts of Si, the Si_3N_4/Al joint fractures at the brittle 4004 filler.

4. Conclusion

The silicon nitride Si_3N_4 was brazed to metals using aluminum filler in a vacuum. The silicon nitride Si_3N_4 was first metallized using Al filler, and then brazed to metals, Fe, Ni, Cu, Al and SUS 304 by remelting the metallizing aluminum layer.

The joining strength of Su_3N_4/metal joint is relatd to α or $\alpha \cdot E$, where α and E are the thermal expansion coefficient and the elastic modulus of metal, respectively. The metal which possesses the higher thermal expansion coefficient gives the higher thermal stress in the ceramic/metal joint. The high thermal expansion coefficient of metals such as Al, Ni and SUS 304, difinitely reduce the strength of the Si_3N_4/metal joint.

The intermediate phases of Ni_2Al_3 and $NiAl_3$ are formed at the joinnig layer of Si_3N_4/Ni joint with Al filler.

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