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Brazing of Si₃N₄ to Metals with Al Filler (Report II)†
—Si₃N₄/Fe, Ni, Cu, Al or SUS304 Joint—

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

Abstract

The joining of silicon nitride Si₃N₄ to metal was conducted using aluminum filler metal in a vacuum, where metal was Fe, Ni, Cu, Al or SUS 304.

Si₃N₄ 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 α·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 Si₃N₄/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 metals1- 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 ceramics3-4). Naka et al.5) have reported that aluminum among aluminum, copper and silver shows the lowest equilibrium contact angle with the highest negative temperature dependence 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 thermal expansion between ceramic and metal.

This work in the successive report from the previous one6) tries to join silicon nitride Si₃N₄ to metals using aluminum filler and clarify the dominating factors in mechanical properties of the ceramic/metal joint.

2. Experimental Procedure

The pressureless sintered Si₃N₄ containing a few percent of Al₂O₃ and Y₂O₃ was used. The purity of aluminum was 99.99 mass %. The metals were Fe, Ni, Cu, Al and SUS304 in high purity. Si₃N₄ 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 Si₃N₄ 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
$10^{-2}$ mms$^{-1}$ as shown in Fig. 2. The microstructure of Si$_3$N$_4$/metal was investigated by means of scanning electron microscope and EDX microanalyser.

3. Results and Discussion

3.1 Joining strength of Si$_3$N$_4$/metal joint

Although the sound brazing of Si$_3$N$_4$ to metal was done using Al filler, the detrimental effects of mismatches between the thermal expansion coefficient of Si$_3$N$_4$ and that of the metal components are shown in Figs. 3 and 4, where $\alpha$ 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. The larger mismatch in thermal expansion coefficient between Si$_3$N$_4$ and metal reduces definitely the strength of the joint. The thermal stress in the joint is arisen from the difference in thermal shrinkage between ceramic 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 $\sigma_t$ in ceramic/metal joint is represented by the following equation.

$$\sigma_t = A_t \Delta \alpha \Delta T$$  \hspace{1cm} (1)

where $A_t$ is related to the elastic modulus $E$ and Poisson ratio $\nu$ of materials, and

$$\Delta \alpha = \alpha_{\text{metal}} - \alpha_{\text{ceramic}}$$ \hspace{1cm} (2)

$$\Delta T = T_{\text{joining}} - T_{\text{room}}$$ \hspace{1cm} (3)

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

![Fig. 3](image1.png)

**Fig. 3** Joining strength of Si$_3$N$_4$/metal joint plotted as a function of $\alpha$; metal thermal expansion coefficient.

![Fig. 4](image2.png)

**Fig. 4** Joining strength of Si$_3$N$_4$/metal joint plotted as a function of $\alpha \cdot E$, where $\alpha$ and $E$ are thermal expansion coefficient and elastic modulus of metal, respectively.

![Fig. 5](image3.png)

**Fig. 5** Microstructure and line analyses for Ni and Al in Ni/Al/Si$_3$N$_4$ joint.

![Fig. 6](image4.png)

**Fig. 6** Microstructure and line analyses for Si in Ni/Al/Si$_3$N$_4$ joint.
Eq. (1) suggests that the smaller factor of $\Delta \alpha$ provides the decrease in thermal stress $\sigma_1$ 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$_3$N$_4$ to metal.

3.2 Joining microstructure of Si$_3$N$_4$/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$_3$N$_4$/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.

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$_2$Al$_3$ 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$_2$Al$_3$ and NiAl$_3$ are formed.

The joining Si$_3$N$_4$ 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$_3$N$_4$/Al joint fractures at the brittle 4004 filler.

4. Conclusion

The silicon nitride Si$_3$N$_4$ was brazed to metals using aluminum filler in a vacuum. The silicon nitride Si$_3$N$_4$ 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 $\text{Si}_3\text{N}_4$/metal joint is related to $\alpha$ or $\alpha \cdot E$, where $\alpha$ 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, definitely reduce the strength of the $\text{Si}_3\text{N}_4$/metal joint.

The intermediate phases of $\text{Ni}_2\text{Al}_3$ and NiAl$_x$ are formed at the joining layer of $\text{Si}_3\text{N}_4$/Ni joint with Al filler.

References