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Author(s)	Arata, Yoshiaki; Ohmori, Akira; Sano, Saburo
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Solid State Reaction Bonding of Non-Oxide Ceramics to Metals (Report I)[†]

— Bonding of Si₃N₄ to Al Alloys —

Yoshiaki ARATA*, Akira OHMORI** and Saburo SANO***

Abstract

The solid state reaction bonding of Si₃N₄ to Al alloys (A1050 and AC-8A) is studied. The bonding was done in vacuum (7×10^{-3} Pa) changing bonding temperature (627 – 877K), bonding time (0 – 180 min) and bonding pressure (0 – 350 MPa) respectively. The strength of bonded joint between Si₃N₄ and Al alloy increases with the increase of bonding temperature, bonding time and bonding pressure respectively.

The reaction between Si₃N₄ and aluminum is studied by using QMS (Quadrupole Mass Spectrometer) and XRD (X-ray Diffraction). Si₃N₄ reacted with aluminum at 1273K to give AlN and Si. The solid state reaction bonding of Si₃N₄ to Al alloy may be due to the formation of AlN and Si at the interface.

KEY WORDS: (Solid State) (Reaction Bonding) (Nitrogen Silicide) (Aluminum) (Al; Alloy)

1. Introduction

Recently, a remarkable attention have been paid to ceramics for their useful characteristics. To use ceramics widely, it is necessary to produce composite materials of ceramics with metals. So, many works have been done in this area of ceramics-metal bonding.

Many methods for bonding ceramics to metals have been undertaken as the application of ceramics.¹⁾ Most of them are the brazing or metallizing techniques. Some works have been done by using the fusion welding techniques like laser beam,²⁾ by using a electric field³⁾ or by applying a solid state reaction.⁴⁾

The solid state reaction bonding is a method to make directly a bonding of ceramics to metals at solid state. In previous paper, the solid state reaction bonding of stabilized ZrO₂ to Al-Mg alloy was reported,⁵⁾ and the mechanism of bonding was discussed.

In this work, the solid state reaction bonding was applied to the bonding of non-oxide ceramic, Si₃N₄, to Al alloys. As Al alloys, an industrial grade pure aluminum (A1050) and an Al-Si alloy for casting (AC-8A) were used.

In order to know the reaction between Si₃N₄ and

aluminum, the mixed powders were heated in a vacuum furnace. During the heating, gas phase reaction products in the furnace were monitored by Quadrupole Mass Spectrometer (QMS), and after heating the solid products were identified by X-ray diffraction (XRD).

2. Experimental Procedure

2.1 Method of solid state reaction bonding

As the ceramics, a pressureless sintered Si₃N₄ (SN220: Kyocera Co.) was used. Si₃N₄ for the bonding is 15 mm in diameter and 5 mm in thickness, and its four points bending strength is about 60 MPa. Each surface roughness of Si₃N₄ disk was < 3.2 μm as received.

Chemical compositions of Al alloys used in this study are shown in Table 1. A1050 is an industrial grade pure aluminum and AC-8A is an Al-Si alloy for casting use. From 10 mm diameter rods, Al alloys were machined into

Table 1 Chemical compositions of Al alloys used.

Materials	Chemical Compositions (wt%)							
	Cu	Si	Fe	Mn	Mg	Zn	Ti	Al
A1050	0.05	0.25	0.40	0.05	0.05	0.05	0.03	99.50
AC-8A	0.8~ 1.3	11.0~ 13.0	0.8	0.1	0.7~ 1.3	< 0.1	< 0.2	86.30

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* Professor

** Associate Professor

*** Graduate Student

6 mm diameter and 0.6 mm thick disks. Both surfaces of the disk were finished by polishing with #1500 emery paper. The surface roughness of metal disk was $< 1.6 \mu\text{m}$.

Figure 1 shows the apparatus for solid state reaction bonding used in this study and the detail of assembly for bonding. The bonding apparatus consists of a electric resistant heater, a high vacuum system and a oil pump for pressing. All experiments in this study were done in the vacuum of $7 \times 10^{-3} \text{Pa}$, and the bonding pressure was monitored both by measuring the oil pressure of the oil pump and by using the load cell as shown in Fig. 1 (a). Specimens for bonding were degreased in acetone with a ultrasonic cleaner. Then the specimens were assembled for bonding as shown in Fig. 1 (b), and it was heated to a required temperature at the rate of 50 deg/min. At the required temperature a required pressure was applied to the assembly for a bonding time. After the bonding, the assembly was cooled at the rate of 10 deg/min. The eval-

uation of mechanical strength of bonded joints was done by measuring the tensile strength.

2.2 Method of QMS analysis

Figure 2 shows the schematic diagram of the apparatus for Quadrupole Mass Spectroscopic (QMS) analysis. In order to detect easily the reaction of Si_3N_4 and aluminum, both powders were used for the analysis. An alumina boat containing about 1 g of the mixed powders was set in the vacuum furnace. During heating the powders in vacuum, the mass spectrum was monitored by QMS, and simultaneously temperature and chamber pressure were recorded. After the heating, products were investigated by X-ray diffraction (XRD).

3. Results and Discussions

3.1 Bonding of Si_3N_4 to A1050

Figure 3 shows the bonding temperature dependence

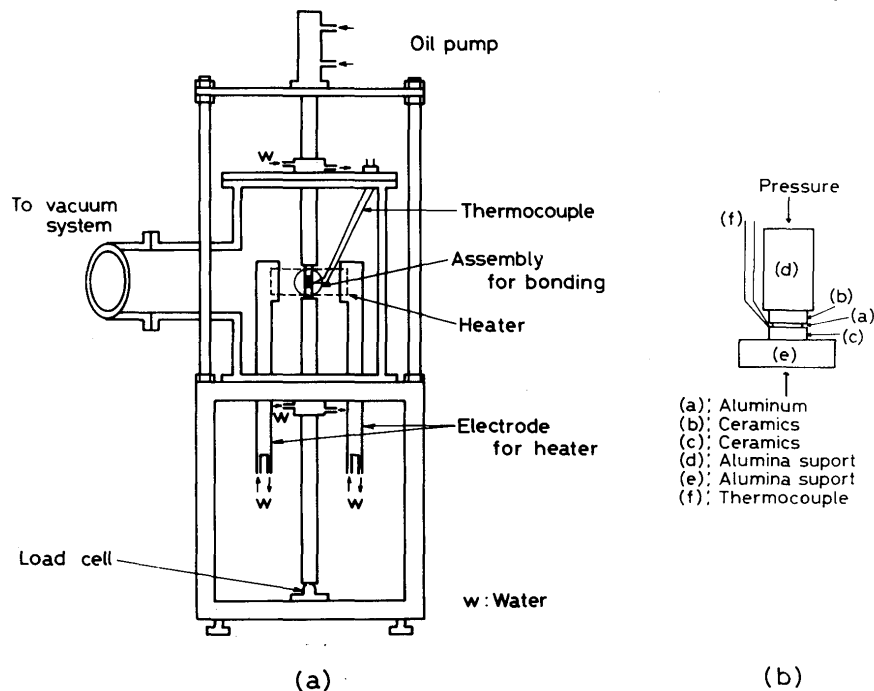


Fig. 1 Schematic diagram of apparatus and assembly for solid state reaction bonding.

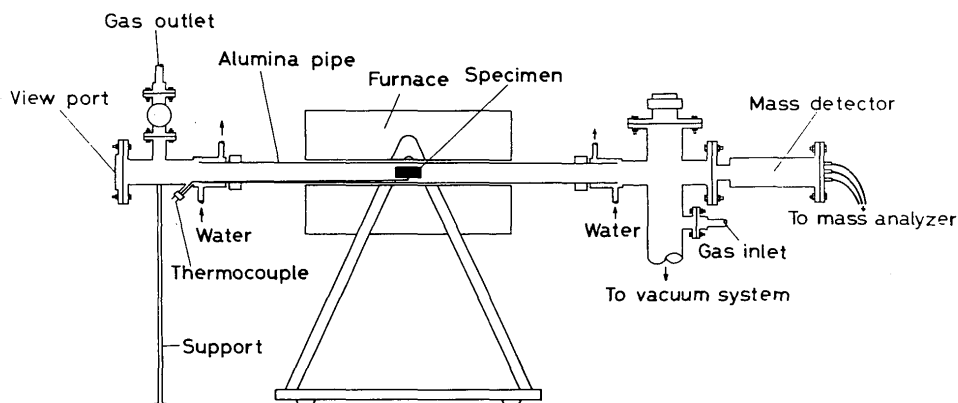


Fig. 2 Schematic diagram of apparatus for QMS analysis.

of the bonding strength. In this figure the tensile strength increases with the increase of bonding temperature, in the case of bonding over 823 K (550°C) the fracture occurs in Si₃N₄ as shown in Fig. 4. Figure 5 shows the bonding time dependence of the bonding strength. In this figure the tensile strength increases with the increase of bonding time. For bonding of the bonding time longer than 60 min, the fracture occurs in Si₃N₄ and the tensile strength becomes about 120 MPa. Figure 6 shows the bonding pressure dependence of the bonding strength of bonded joint at 773 K and 823 K for 10 min. For the bonding temperature of 773 K, the bonding strength increases with the bonding pressure and the fracture occurs at the interface of Si₃N₄ and A1050 even under the bonding pressure of 350 MPa. However, in the case of the bonding temperature of 823 K, the fracture occurs in Si₃N₄ under the

lower bonding pressure of 50 MPa. From Figs. 3 to 6 the bonding strength increases with the increase of the bonding temperature, the bonding time and the bonding pressure, respectively.

The tensile strength of Si₃N₄ used in this study is about 350 MPa. This value is about 3 times bigger than the tensile strength of the bonded joints fractured in Si₃N₄. It may be due to the difference of the thermal coefficient of Si₃N₄ and A1050.

Figure 7 shows the change in bonding interface part with the bonding temperature, when Si₃N₄ was bonded with A1050 for 10 min under 100 MPa bonding pressure. From this figure, it was seen that as the bonding temper-

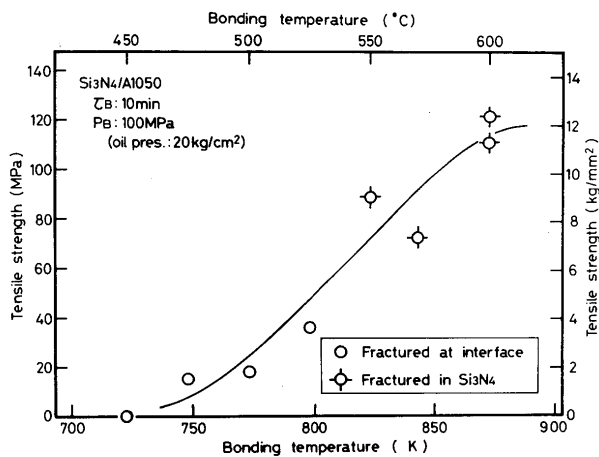


Fig. 3 Temperature dependence of bonding strength on bonding of Si₃N₄ to A1050.

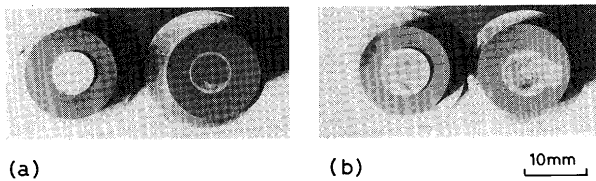


Fig. 4 Fractured surface of bonded joints, (a); fractured at interface and (b); fractured in Si₃N₄.

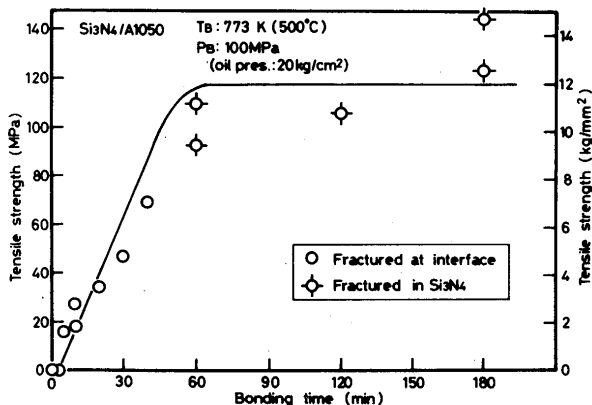


Fig. 5 Time dependence of bonding strength on bonding of Si₃N₄ to A1050.

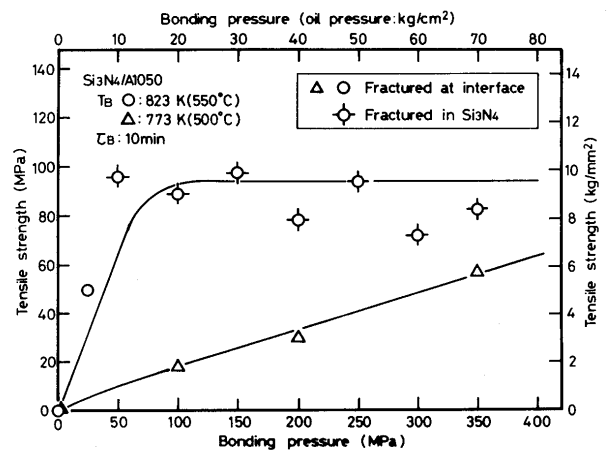


Fig. 6 Pressure dependence of bonding strength on bonding of Si₃N₄ to A1050.

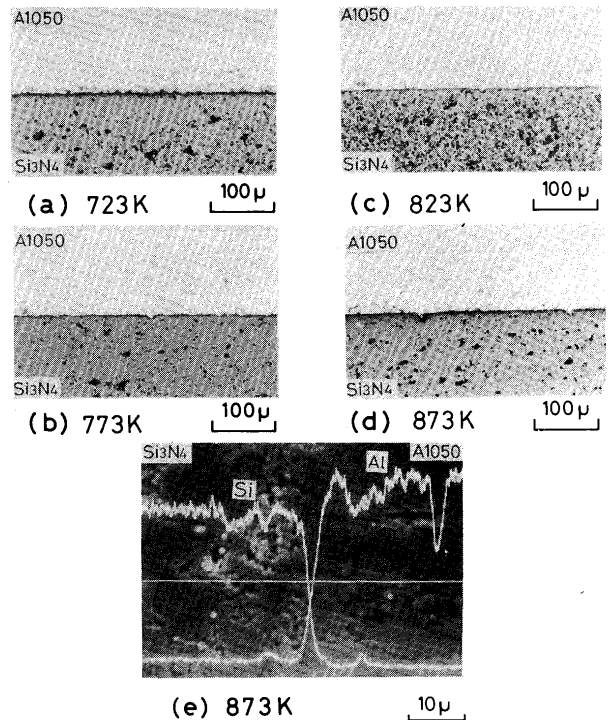


Fig. 7 Change in bonding interface part and EDX line analysis result of Si₃N₄ and A1050 with bonding temperature for bonding for 10 min under 100 MPa (20 kg/cm² oil pressure).

ature increased, the uneven parts increased at the interface of Si_3N_4 and A1050. From the line analysis of EDX at the interface part of the bonded joint bonded at 873K as shown in Fig. 7 (d), it may be shown that the reaction of Si_3N_4 and the aluminum occurs.

3.2 Bonding of Si_3N_4 to AC-8A

In order to know the influence of Si in Al alloy on bonding, bonding of Si_3N_4 to AC-8A was done. The results are shown in Fig. 8. Compared with the results of bonding of Si_3N_4 to A1050 bonded under same bonding conditions as shown in Fig. 8, the bonding to AC-8A at 773K was more difficult than the bonding to A1050. But, as shown in section 3.1, the bonding tendency for AC-8A was almost same as bonding for A1050. The bonding strength increases with the increase of bonding temperature, bonding time and bonding pressure, respectively. Bonding strength of Si_3N_4 to AC-8A bonded at 827K is almost same as the bonding strength of Si_3N_4 to A1050. For the bonding of Si_3N_4 to AC-8A, the bonding temperature influenced more effectively than the bonding time or the bonding pressure did. It may be due to easiness of the reaction of Si_3N_4 with aluminum at higher temperature of 823K inspite of existence of Si in AC-8A. From the results of sections 3.1 and 3.2, the influences of bonding conditions on the solid state reaction bonding of Si_3N_4 to Al alloys were considered as follows. As shown in Fig. 6, fracture does not occur in Si_3N_4 for the joint bonded under 350 MPa bonding pressure at 773K, but the joint bonded under 50 MPa bonding pressure at 823K fractured in Si_3N_4 . Similarly, the bonding strength of Si_3N_4 to AC-8A increased greatly with the increase of bonding temperature as shown in Fig. 6. At higher temperature, more easily metals deform, more intimately metals contact with ceramics. Therefore, the bonding of Si_3N_4 to Al alloys can be easily at higher temperature of 823K.

At the higher temperature of 823K, solid state reaction bonding of Si_3N_4 to AC-8A specimens of larger size was done. Figure 9 shows the appearance of a bonded joint of Si_3N_4 (25 mm X 25 mm X 5 mm) to AC-8A (20 mm in diameter). As shown in this figure, the bonding for such a large specimens is possible and strong joint is obtained.

3.3 Results of QMS analysis

To certify the reaction at the interface between Si_3N_4 and Al alloys during bonding, the mixed powders of Si_3N_4 and Al were heated in vacuum furnace. Mass spectrum of the gas evolved in reacting chamber was monitored during the heating by QMS. Figure 10 shows examples of mass spectrums measured at room temperature to check the residual gas. In Fig. 10, (a) and (b) show mass analysis results, (c) shows a mono element ion spectrum result obtained by giving the excess energy. In this study, to know the generation of nitrogen from Si_3N_4 during the

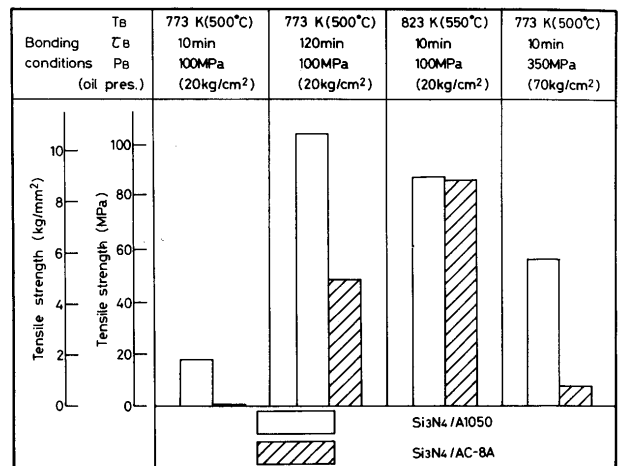


Fig. 8 Results of bonding of Si_3N_4 to AC-8A

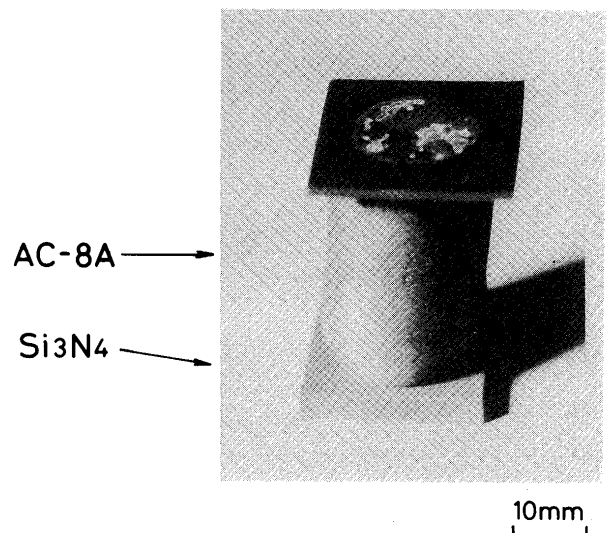


Fig. 9 Appearance of solid state reaction bonded joint of Si_3N_4 (25 mm X 25 mm X 5 mm) to AC-8A (20 mm diameter) bonded at 823K for 120 min under 27 MPa bonding pressure (60 kg/cm² oil pressure).

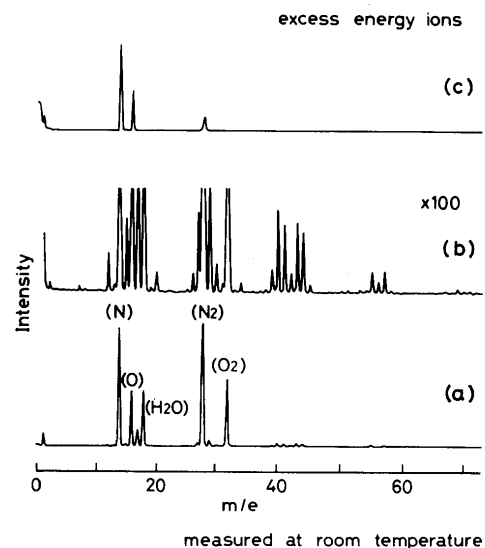


Fig. 10 Example of mass spectrum measured at room temperature by QMS. (a); mass spectrum, (b); 100 times magnified spectrum from (a) and (c); excess energy ions spectrum.

heating, the change in mass peak ratio of $m/e = 14$ (peak of nitrogen) and $m/e = 16$ (peak of oxygen) was obtained by calculating the measured spectrum results.

Figure 11 shows the changes in the temperature, the chamber pressure and the mass peak ratio given both by heating of Si_3N_4 powders to 1673K and for 60 min heating at the temperature. Similarly, results for the heating of Si_3N_4 -80wt%Al mixed powders at 1273K is shown in Fig. 12. When only Si_3N_4 powders heated at 1673K, the mass peak ratio became about 100% as shown in Fig. 11. At the temperature Si_3N_4 decomposed to give nitrogen. This fact is supported by the increase of chamber pressure as shown in this figure. From Fig. 12, however, the

mass peak ratio shows about 80%, and the generation of nitrogen was not seen. In this case, the increase of the chamber pressure was not acknowledged.

To identify the products, X-ray diffraction of powders after heating was done. Figure 13 shows XRD patterns of heated powders. From Fig. 13(b), Si_3N_4 powders heated at 1273K do not change. However the XRD pattern of Si_3N_4 powders heated at 1673K shows peaks of Si as shown in Fig. 13(a). From these analysis and XRD result, Si_3N_4 decomposes to generate nitrogen and Si by heating at 1673K, so Si was remained in the powder, but the decomposition of Si_3N_4 powders at 1273K is not seen. The XRD pattern of Si_3N_4 -80wt%Al mixed powders

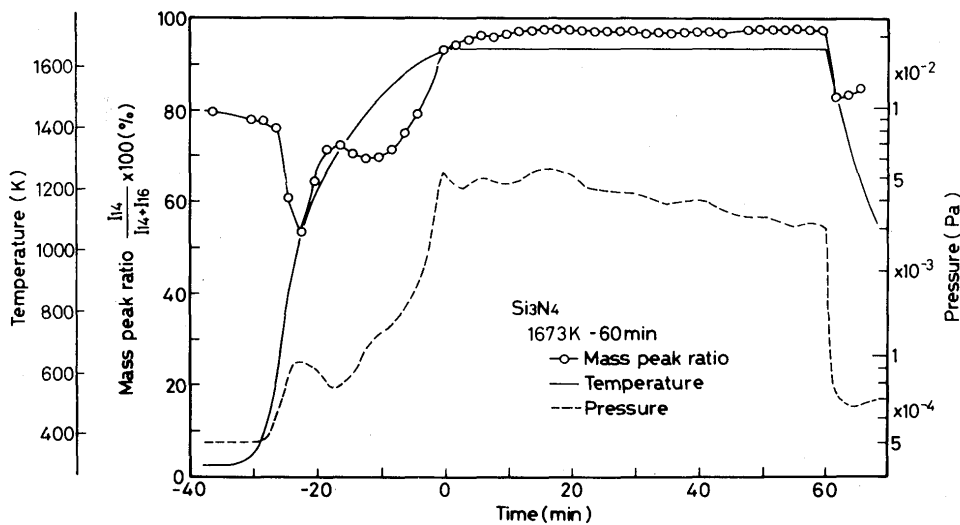


Fig. 11 Change in temperature, chamber pressure and mass peak ratio during heating of Si_3N_4 powders in vacuum. I_{14} and I_{16} respectively mean intensity of $m/e = 14$ (nitrogen) and $m/e = 16$ (oxygen) of mass spectrum.

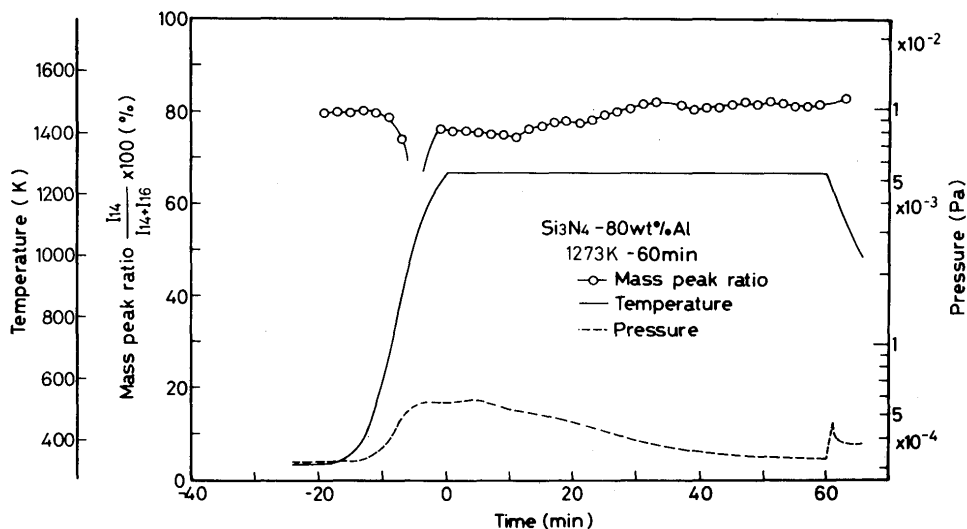


Fig. 12 Change in temperature, chamber pressure and mass peak ratio during heating of Si_3N_4 -80 wt%Al mixed powders in vacuum. I_{14} and I_{16} respectively mean intensity of $m/e = 14$ (nitrogen) and $m/e = 16$ (oxygen) of mass spectrum.

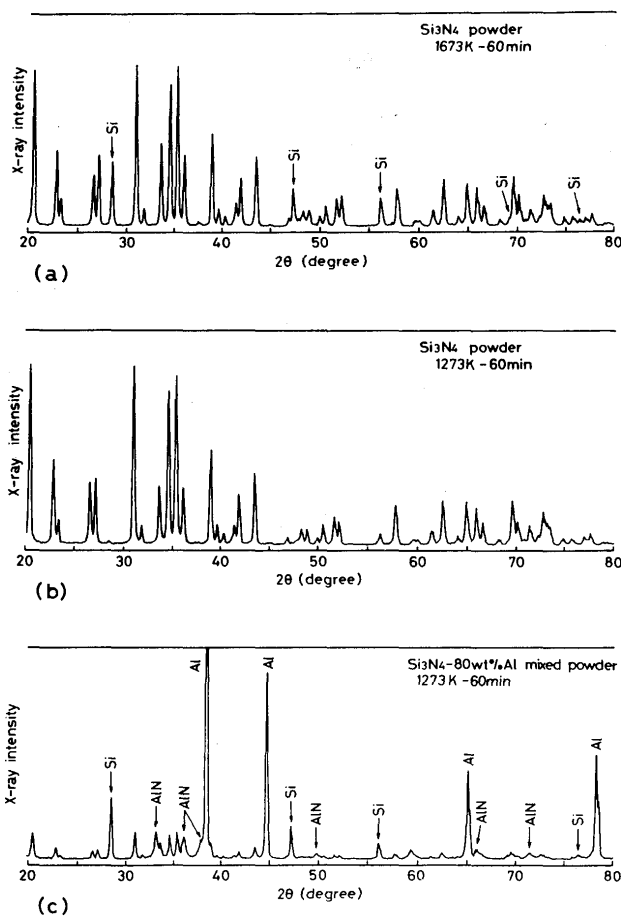


Fig. 13 Results of XRD after heating of Si_3N_4 -80wt%Al powders in vacuum.

heated at 1273K shows both Si and AlN peaks as shown in Fig. 13(c). From these results, Si_3N_4 reacted with Al to produce AlN even at 1273K.

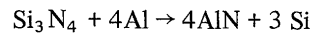
The results are summarized in Table 2 involving a weight loss of Si_3N_4 powders and the color change. The weight loss of Si_3N_4 powders heated at 1673K is about 10%, but the weight loss of Si_3N_4 -80wt%Al mixed powders is almost 0%. From these weight changes it is shown that Si_3N_4 powders themselves lose nitrogen, but Si_3N_4 -80wt%Al mixed powders do not lose nitrogen.

By a thermodynamic calculation, the following reaction may be considered to occur at 773K applied to the bond-

Table 2 Results of heating of Si_3N_4 and Si_3N_4 -80wt%Al mixed powders in vacuum.

Materials (color)	Conditions		Weight loss (%)	Color (after heating)	Products
	Temp. (K)	Time (min)			
Si_3N_4 -80wt%Al (light gray)	1273	60	0.1	black	Si, AlN
Si_3N_4 (light gray)	1273	60	0.7	light gray	—
Si_3N_4 (light gray)	1673	60	9.7	dark brown	Si

ing of Si_3N_4 to Al alloys,⁶⁾



$$\Delta G_{773\text{K}} = -125 \text{ kcal/mol} \quad (1)$$

Moreover, as shown in Table 2, the similar color change was seen at the interface of Si_3N_4 and Al alloys bonded at 773K. From these results, the reaction of eq.(1) may occur at the interface of Si_3N_4 and Al alloy when the bonding is done at 773K.

4. Conclusion

The bonding results in this experiment were summarized as follows;

- 1) Si_3N_4 was bonded directly to both A1050 and AC-8A by the solid state reaction bonding. The bonding strength of bonded joint increased with the increase of bonding temperature, bonding time and bonding pressure, respectively.
- 2) Bonding of Si_3N_4 to AC-8A was more difficult than bonding of Si_3N_4 to A1050. It may be due to the influence of Si in AC-8A.
- 3) From the reaction of Si_3N_4 -Al mixed powders heated in vacuum, the reaction to produce AlN and Si at the Si_3N_4 -Al alloy interface may occur during the bonding at a low temperature.

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