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Bonding and Thermal Fracture of Silicon Nitride / Stainless Steal (SUS316)

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Abstract Diffusion bonding and thermal fracture between Si_3N_4 ceramic and stainless steel (SUS316) were investigated. Sintered Si_3N_4 ceramics were fabricated by hot-pressing of α-Si_3N_4 powder with 5wt% Y_2O_3 and 5 - 10wt% Al_2O_3. Bonding between SUS316 plate with 1 mm thick and Si_3N_4 specimens was carried out at 8 - 24 MPa and at 1000° - 1300°C for 1 h in vacuum. Thermal decomposition of the bonded specimen in air was followed by acoustic emission and high-temperature microscope. Bonding between SUS and Si_3N_4 was achieved by heating at 1150 and 1200°C in vacuum. When the bonded sample was heated in air, small cracking was formed at 700°C at the interface between SUS and Si_3N_4, followed by decomposition of the bonded sample at 900°C.
Key words: Diffusion Bonding, Silicon Nitride, SUS 316, Thermal Fracture

1. INTRODUCTION

Silicon nitride (Si_3N_4) is an engineering ceramic for high temperature applications, such as gas turbine, engines, because of its high hardness, thermal shock resistance and abrasive tolerance. [1] However due to poor machinability, a bonding between Si_3N_4 and machinable metals is needed to manufacture complex shaped components. It is known that SUS 316 is one of famous austenitic stainless steels suitable for high temperature corrosive environments because of its high oxidation and corrosion resistance comparing with other stainless steels. Bonding between Si_3N_4 and SUS has been usually carried out by brazing [2-6] or diffusion bonding [7-9]. Of these, diffusion bonding, which is usually carried out at ~1000°C, has a potential for use in high temperature environments. In diffusion bonding between Si_3N_4 and SUS, chemical reaction takes place to form compound containing Fe-Si silicides at the interface. Since the silicides are brittle and easily oxidized, it is important to control the reaction to restrict the formation of Fe-Si silicides with an adhesive and tough bonding. [8] It has been reported to control the reaction by changing the compositions of metals and ceramics. [10, 11] In this study, the bonding between Si_3N_4 and SUS 316 is carried out and the reaction at the interface is investigated with change of the amount of sintering Al_2O_3 additive. Thermal fracture behavior and acoustic emission (AE) during the heating are directly monitored by high temperature optical microscope (HTOM) and AE sensor system to evaluate the high temperature durability for this bonding.

2. EXPERIMENTAL PROCEDURE

α-Si_3N_4(SN-E10, average particle size of 0.5 μm; Ube Co.), Al_2O_3 (2 μm; Kanto Chemical) and Y_2O_3 (2 μm; Koujyundo Kagaku) powders were used. The α-Si_3N_4 powders with Y_2O_3 (4wt%) and Al_2O_3 (4wt% and 9wt%) were ball-milled in ethanol with plastic coated iron balls for 24 h. The Si_3N_4 ceramics containing 4wt% and 9wt%Al_2O_3 were abbreviated as 4wt% and 9wt% sample in the following section. The mixed powder of 15 g was hot-pressed (FVPHP-R-5, Fuji Denpa) at 24 MPa and 1900°C for 1 h in N_2 atmosphere. The sintered Si_3N_4 ceramic was cut to a specimen of 35×35×5 mm in size and characterized by density measurement, X-ray diffraction pattern (XRD, RIGAKU RINT-2000) and scanning electron microscope with energy dispersion analysis unit (SEM-EDS, JEOL JSM-6500F). A SUS 316 (Fe 67%, Cr 16%, Ni 14%, Mo 3%) plate (Niraco Co.) with 10×10×1 mm in size was sandwiched by two pieces of the sintered Si_3N_4 ceramics with 10×10×5 mm in size. The surface of the Si_3N_4 piece was mechanically polished and finished by 0.5 μm diamond paste. The SUS plate was also polished by the same paste just before the bonding experiments to remove surface oxide. The bonding was carried out at 1000°-1300°C in vacuum at 8 - 24 MPa for 1 h in the hot-pressing furnace. The bonded sample was cut into 4 pieces and each was characterized by XRD and SEM-EDS. Thermal fracture of the bonded sample was

Table 1. Results of diffusion bonding between Si_3N_4 and SUS 316 plate

Bonding temperature	Results	
	Al_2O_3 4wt%	Al_2O_3 9wt%
1000°C	No reaction	Bonded but peeled off
1100°C	Bonded but peeled off	Bonded but peeled off
1150°C	Bonded but peeled off	Bonded
1200°C	Bonded	Bonded but corrupt in the ceramic
1250°C	Bonded but corrupt in the ceramic	
1300°C	Bonded but corrupt in the ceramic	

monitored by high-temperature optical microscope (HTOM; ULVAC, VMC-700) and acoustic emission (AE) analyzer system (NF electronics Co.). [12] The bonding sample was heated at 20°C/min up to 1000°C in air. The image of the bonding interface was monitored by CCD camera and recorded by a video cassette recorder.

3. RESULTS AND DISCUSSION

3-1 BONDING BETWEEN Si_3N_4 AND SUS 316

The results of bonding experiments were summarized in Table 1. In the 4wt% sample, the reaction at the interface between ceramic and metal was not observed at 1000°C. At 1100°C and 1150°C, ceramics were bonded to metal, but one side of ceramic pealed off from SUS plate in the cutting process after bonding. At 1200°C, bonding was formed without crack as shown in Fig. 1 (a). At 1250° and 1300°C, cracks in the ceramic parallel to the interface were occurred like as shown in Fig. 1 (b) and sample peeled off at the crack. In the 9wt% sample, no reaction occurred between the ceramics and metals at 1100°C, but the bonding was formed at 1150°C. At 1200°C, the bonded sample was corrupted like as the 4wt% sample bonded at 1300°C.

Figure 2 shows the SEM photographs at the interface of the bonded ceramic-metal. In the 4wt% sample bonded at 1200°C, a reaction layer about 1 μm thick containing dark contrasted spots about 0.1 μm in size was observed. In the sample bonded at 1250°C, the interface became rough and the reaction layer grew to about 3 μm thick. The dark contrasted spots were also observed in the reaction layer and their size was slightly increased with increasing of bonding temperature. In the 9wt% sample bonded at 1150°C, the reaction layer had thickness of about 2 μm and contained dark contrasted spots smaller than in 4wt% sample. In the sample bonded at 1200°C, the reaction layer became thick about 6 μm in the fine dark contrasted spots. The sizes of dark spot were about 0.1 μm near the Si_3N_4 ceramic and became large up to about 0.5 μm toward to SUS substrate. Figure 3 shows the XRD patterns of the reaction layer formed on the SUS after bonding at 1200° and 1250°C for 1 h in the 4wt% sample. The sample peeled off was used for XRD measurements. From the XRD patterns, the large peaks of FeSi with Al_8Cr_5, $CrSi_2$ and AlN were observed in addition to SUS and Si_3N_4 ceramics. These compounds should have been formed during in diffusion bonding.

Figures 4 and 5 show SEM photographs at the interface with the results of EDS analysis in the 4wt% and 9wt% samples bonded at 1200°C, respectively. At position 1, Si with about 4atm% Al was detected, showing the composition of Si_3N_4 ceramic. Amount of Si was slightly decreased at the position 2. At position 3, which is reaction layer, about 50atm% Fe, 15atm% Si and 15atm% Cr were detected, suggesting that the area consists of Fe-Si silicide with Cr. At the dark contrasted spot of position 4, amount of Cr and Al was greater than the area around the spot of position 3, suggesting that the spots were Cr-Al compound. At the position 5 of SUS substrate, about 5atm% Si was observed in addition to the elements of SUS, the presence of the silicon showed that Si diffused into the SUS. EDS analysis result of the 9wt% sample was shown in Fig 5. The amount of Si was slightly decreased and Fe was increased by about

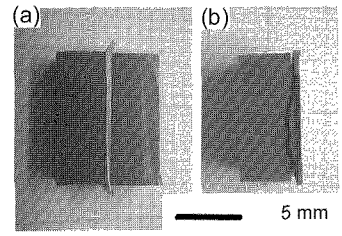


Figure 1. Photographs of samples bonded at (a) 1200°C and (b) 1300°C for 4wt% sample.

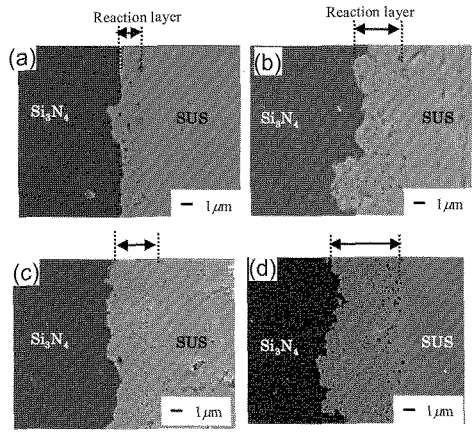


Figure 2. SEM photographs of the Interface between Si_3N_4 and SUS. The bonding was carried our at (a) 1200°C and (b) 1250°C for 4wt% sample and (c) 1150°C and (d) 1200°C for 9wt % sample.

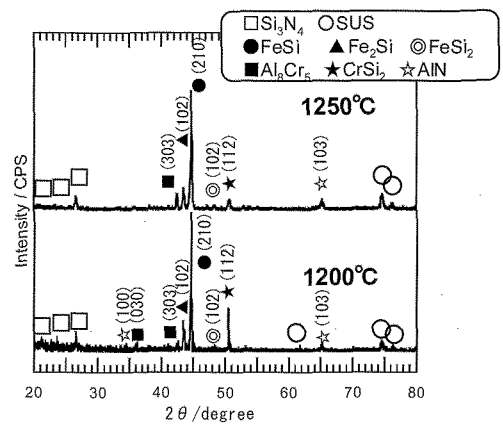


Fig 3. XRD patterns at the reaction layer of the 4wt% bonded sample. These pattern were obtained from the reaction layer formed on the SUS plate after bonding.

2atm% from position 1 to 2. At the reaction layer of position 3 and 4, the amount of Si, Fe, Cr and Al were constant at about 12atm%, 50atm%, 15atm% and 5atm%, respectively, but the amounts of Fe and Al changed to be 40atm% and 15atm%, respectively, at position 5, showing that a layer containing Al and Cr rich compounds was present near the interface between SUS and reaction layer.

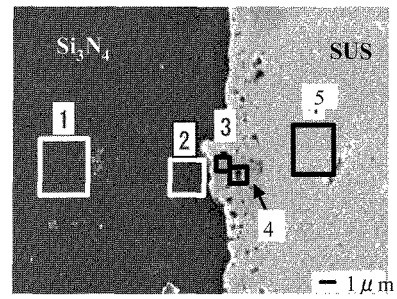
3-2 THERMAL DECOMPOSITION OF BONDING

Figure 6 (a) shows the results of AE events during the heating of the bonded sample in addition to the results for the sintered Si_3N_4 peace and SUS substrate. The Si_3N_4 ceramics showed no obvious AE events during the heating up to 900°C, while SUS substrate showed AE events at 730°-800°C and 900°-950°C. It is known that in the austenitic stainless of SUS 316, a small amount of carbon reacts with Cr to form chromium carbide at the grain boundary and the carbide is oxidized at higher temperatures. [13] Therefore, AE events observed in SUS plate were possibly monitored by the formation and oxidation of the chromium carbide. In 4wt% sample bonded at 1200°C, AE events were also observed at around 750°C and 850-950°C. Frequent occurrence of the events around 900°C was greater than that observed in SUS. After the AE observation, the metal in bonding sample was completely peeled off from the ceramics. From XRD analysis at the exposed interlayer by the thermal decomposition, NiO and Fe_2O_3 were observed, showing that the compound at the interlayer was oxidized by the heating. HTOM observation (Figs. 6 (b) and (c)) showed that fine cracks normal to the interface in the reaction layer were formed from 750°C. At 900°C, large cracks parallel to the interface occurred (Fig. 6 (c)). The results of HTOM, appeared that AE events at 750°C and 850°-950°C detects the fine cracking at the reaction layer and large cracking parallel to the interface, respectively, leading to the decomposition of the bonded sample. Occurrence of the small and large cracks was thought to be induced by the formation and oxidation of chromium carbide at the grain boundary in SUS. Once the cracking occurs in the reaction layer, the oxidation of the compound at the reaction layer accelerated and caused the breaking of the bonding.

4. SUMMARY

Diffusion bonding between stainless steel (SUS316) and Si_3N_4 ceramic containing 4wt% and 9wt% Al_2O_3 as a sintering aid was carried out in vacuum at applied pressure of 24 MPa at the temperature range from 1000° to 1300°C for 1 h. The relatively adhesive bonding without crack was formed at 1200° and 1150°C for the 4wt% and 9wt% bonding samples, respectively. At the interface, Fe-Si slicide with small particles 1 (~0.2 μm) consisting of Al-Cr alloys was formed and the layer thickness increased with increasing the bonding temperature and the amount of Al_2O_3.

HTOM observation of thermal fractural behavior of the 4wt% Al_2O_3 bonded sample in air showed that small cracks at the interface in the reaction layer formed at about 700°C and large cracks in parallel to the interface at about 900°C. AE monitoring showed the small and large AE events occur at about 700°and 900°C, respectively. Monolithic SUS 316 plate gave AE events at about 750°C, probably due to precipitation and



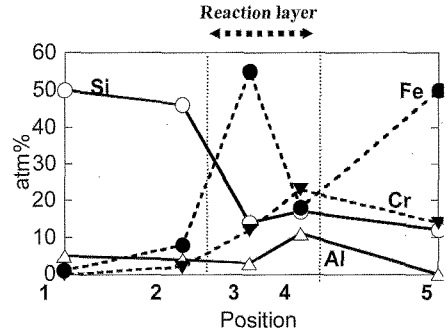
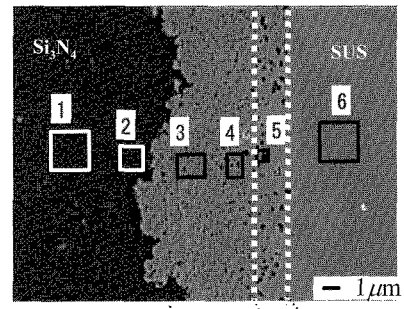


Fig 4. SEM photograph (upper) and EDS analysis (lower) at the interface in the sample bonded at 1200°C for 4wt% sample. Numbers in the SEM photographs denotes the position of EDS analysis.



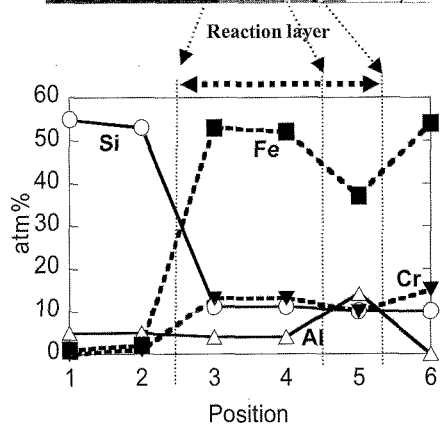


Fig 5. SEM photograph (upper) and EDS analysis (lower) at the interface in the sample bonded at 1200°C for 9wt% sample.

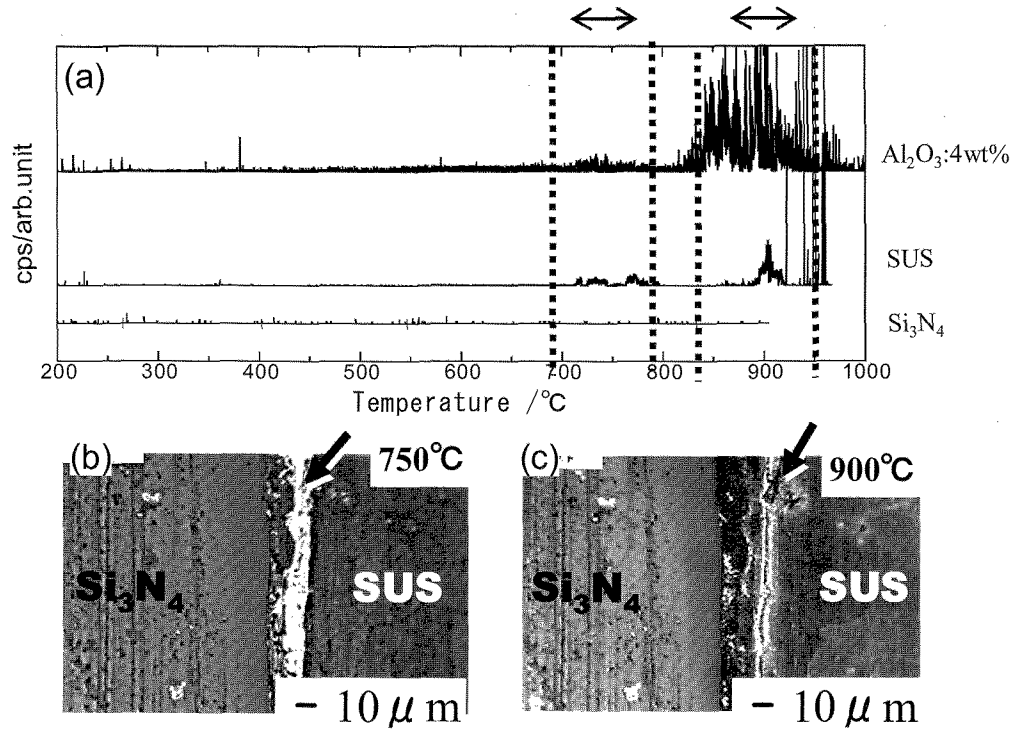


Fig 6. Plots of AE events with heating temperature (a) and the HTOM photographs of the 4wt% sample at (b) 750°C and (c) 900°C

oxidation of the chromium carbide in SUS. Therefore, it was suggested that small cracking at the reaction layer is induced by the precipitation and oxidation.

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