Study on Brazing Properties of Metal/Ceramic Joints

금속/세라믹 결합부의 브레이징 특성에 관한 연구

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ABSTRACT 20 vol.% SiC를 포함한 두 층간의 Si_3N_4/SiC 나노 복합재료는α -Si_3N_4, 13 nm 크기의 나노탄소 분말 그리고 5 wt% Y_2O_3의 분말로 두 단계 소결을 통하여 제작된다. Si_3N_4 입계 사이의 결합은 소결 후 변하지 않고 남은 compact와 51~62%의 기공으로 얻어진 표면적 사이의 반응에 의해 생성된다. 이 연구에서는 Ti 합금을 SiC 층에 브레이징을 이용하여 제작하고 기계적 특성을 연구하였다. 다양한 변형율과 결합물의 강도, 변형율 증가에 따른 층간 변화를 연구하였다. 층간 파괴 형태는 금속과 브레이징 합금 사이의 파괴, 세라믹과 브레이징 합금 사이의 파괴, 그리고 세라믹 내부에서의 파괴를 보였다.

1. Introduction

The ceramic-metal joining is important to apply the ceramics to the structural components. The fracture strength of the ceramic-metal joint is, however, influenced by the residual stresses and the stress concentration caused by the difference between the thermal expansion coefficients and between the elastic moduli. Regarding the material combinations and the method of joining, many experimental studies have been conducted [1]. In the engineering ceramics, silicon nitride (Si_3N_4) ceramics and its composites, no matter they are dense or porous, are potential materials used at high temperatures. Various processing techniques have been developed to fabricate Si_3N_4 ceramics and its composite materials for structural or functional applications [2]. Porous Si_3N_4 ceramics with a microstructure of *rod-like β-Si_3N_4 grains shows* superior mechanical properties, such as high strength, good thermal shock resistance, and high strain and damage tolerance [3]. Thus, an α to β phase transformation with little or no densification is preferred when fabricating porous Si_3N_4 ceramics by sintering. Because densification by particle rearrangement usually occurs before the phase transformation, and the reconstructive α to β phase transition also provides a driving force for densification, ceramics with porosity of higher than 50% is difficult to obtain by sintering at high temperature with full phase transformation [4]. That how to increase the porosity while keep the fine pore size, good shaping behavior and sinterability is still an unresolved problem in the fabrication of porous ceramics.

In this study, Si_3N_4/SiC nanocomposites with 20 vol.% SiC were fabricated in free surface layers by the carbothermal reaction between Si_3N_4 and carbon. Effect of carbon addition on the sinterability, microstructure development and mechanical properties were investingated. And four-point bending tests were conducted for brazed Si_3N_4/SiC to 304 stainless steel joint with various strain rates and the variation in the deflection of Cu interlayer, the center of specimen, was studied in room temperature.

2. Experimental Details

The Si_3N_4/SiC nanocomposites were fabricated in the surface layers using the reaction process between Si_3N_4 and carbon powders. The used powders were high purity Si_3N_4 powder (SN-E10, UBE Industries Ltd, Tokyo, Japan; α ratio: >95%, mean particle size: 0.5 μm, main impurities by weight: O=1.6%; C<0.2%; Cl, Fe, Ca and Al_2O_3<50 ppm), carbon powder (No. 2600, Mitsubishi Chemical Corp., Tokyo, Japan; mean particle size: 13 nm) and sintering aid Y_2O_3 powder (99.9% purity, Kojundo Chemical Lab. Co, Ltd, Sakado, Japan). For fabrication of the Si_3N_4/SiC nanocomposites with 20 vol.% SiC, the following compositions of powder mixture were used: 89.63 wt% Si_3N_4, 5.37 wt.% carbon, respectively. 5 wt.% Y_2O_3 as the sintering additive was added in. The powder mixtures were then uniaxially pressed under 10 MPa to form rectangular bars measuring 15×20×20 mm. Some of the samples were then cold isostatic pressed (CIPed) under the pressure of 200 MPa. A pre-sintering of 1600℃/4 hr in argon atmosphere of gas pressure 0.6 MPa was used to obtain SiC particles through the reactions. Then the samples were sintered in a graphite resistance furnace at 1650, 1750 and 1900℃ for 2 hr in a nitrogen atmosphere of gas pressure 0.6 MPa with the temperature rising rate of 15℃/min.

The sintered Si_3N_4 and 304 stainless steel were joined by the active metal vacuum-brazing method in 800-850℃. A copper sheet was used as the interlayer and Ti-Ag-Cu alloy (Cusil-ABA, WESGO Metals, San Carlos, USA) was used as the brazing filler metal. Si_3N_4 specimens were cut into 3×4×20 mm bars. The bond surface of each bar was ground to average surface roughness of about 0.5 μm, and the faces of 304 stainless steel were finished with a 1200 grit SiC paper and macro-etched to remove any oxide films. The thickness of the copper interlayer and Ti-Ag-Cu brazing filler are 0.25-0.30 mm and 0.05-0.125 mm respectively. Four-point bending test procedures were conducted in accordance with the ASTM standard E855-90. The lower span distance was 30 mm and upper one was 10 mm.

3. Results and Discussions

3.1 Synthesis Properties

According to reactions, considering 1.6% oxygen on the surface of raw Si_3N_4 particles, weight loss after sintering are 9.28% theoretically for the composites with 20 vol.% SiC. The measured weight loss, relative density and porosity of sintered bodies were shown in Table 1. The weight loss of sample sintered at 1650 and 1750℃ was lower than the theoretical value, while that of the samples sintered at 1900℃ was almost the same. The low weight loss at low temperature of 1650 and 1750℃ was come from a fact that a mass of the carbon remained after the sintering and the reaction was not completed, indicating that it need a high temperature to transfer carbon to SiC for the bulk samples. The shrinkage of all samples after sintering was within 1%, indicated that the original dimensions of the powder compact remain virtually unchanged during sintering. The low shrinkage indicated a significant effect of carbon addition on the densification behavior, and the shrinkage was apparently restrained by the reaction between carbon and Si_3N_4. The densification of Si_3N_4 ceramics using Y_2O_3-Al_2O_3 oxide as sintering additives begin at temperature above 1400℃ [5], at which the glass phase formed and particle rearrangement was the main densification mechanism. The reaction between carbon and Si_3N_4 also begin at this temperature, and prior to it, reaction between carbon and surface SiO_2 begins. The occurrence of these reactions was at surface of Si_3N_4 particles, and the reactant located either at their surface or between the Si_3N_4 particles, which result in a reaction bonding between Si_3N_4 grains. It was well known that glass phase plays an important role in the Si_3N_4 particle rearrangement, and the densification of Si_3N_4 ceramics by liquid phase sintering dependent on the characteristics of glass phase, such as amount and viscosity [5]. At relatively low temperature of lower than 1600℃, the viscosity of glass phase is high, so the densification is limited [5]. As the reactions begin almost simultaneously with the glass formation, the bonding of the Si_3N_4 particles by the reaction formed SiC particles is very likely to obstacle the movement of Si_3N_4 particles and restrains their rearrangement. With increase in sintering temperature, the increased viscosity of glass phase indicated the densification tendency, however, as the bonding among Si_3N_4 grains

Table 1 Synthesis properties of sintered Si_3N_4/SiC Nano-composites

Sample	Relative density of green body	Sintering temperature[ ]	Weight loss [%]	Relative density after sintering	Total porosity	Open porosity
20% SiC	0.394	1650	7.2	0.387	0.613	0.611
		1750	7.9	0.384	0.616	0.615
20% SiC with CIP	0.548	1650	7.3	0.489	0.511	0.493
		1750	7.6	0.488	0.512	0.511

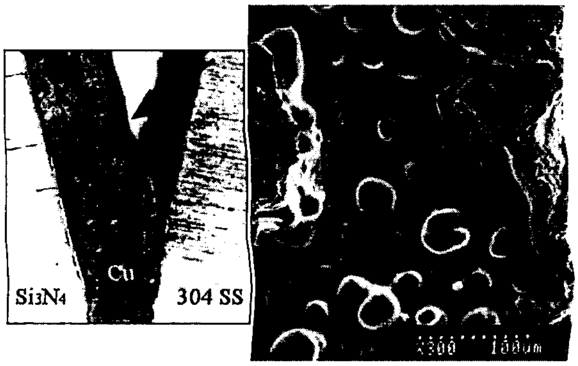

Fig. 3. SEM micrographs of brazed joint

was strong, hardly did the grain rearrangement take place. Finally low sintering shrinkage and high porosity after *sintering were resulted in. This method of* producing porous ceramics can be defined as the restrained sintering by reaction bonding.

3.2 Strength Properties

The relations between the bending strength and the various strain rates obtained by the four point bending tests at room temperature showed a linear increase of bending strength of 350-400 MPa from CHS = 0.005 up to 50 mm/min. This result is in reasonable agreement with other published works for Ti alloy and other metals [6]. The specimens were mainly fractured along the interfaces between the Cu interlayer and the metal side as shown in Fig. 3. The cracks initiated at reaction layer of the metal-brazing filler, so fracture behavior was similar to metals. The deflection level of Cu interlayer at maximum strength generally increased with increasing strain rates. This result might be influenced by increasing strength with high loading speed *in relatively high speed region. The downward* flex *of the specimens, namely, increased with* increasing applied load.

4. Conclusions

Si_3N_4/SiC nanocomposites with 20 vol.% SiC were fabricated in free surface layers by the carbothermal reaction between Si_3N_4 and carbon. And four-point bending tests were conducted for brazed Si_3N_4/SiC to 304 stainless steel joint. Results showed that controlled porosity is obtainable by varying the carbon content and green density. The samples exhibit the microstructure that is composed of fibrous Si_3N_4 grains and nanosized SiC particles, fine pore size, and a good permeability due to the high porosity. The bending strengths linearly increased with increasing strain rate at room temperature and distributed from 350 to 400 MPa.

Acknowledgments

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