Preparation of Reaction-Bonded SiC

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Introduction

Reaction Bonded SiC[1-2] differs from conventional SiC in the point that its process uses liquid silicon to densify. Because the melting point of Si is relatively low(≈ 1450 °C), its sintering temperature is nearly the melting point of Si. Combined with its relatively low densifying temperature, it maintains its shape with a minimum dimensional change after completing process, which makes it possible that it needs minimum machining cost for various shapes. (near-net shape)

However, the relatively low strength and toughness[3-5] has been pointed out to be improved, which is mainly concerned with brittle silicon filling pores with 5~15 vol.%. The residual silicon also inhibits its uses at high-temperature above 1370 °C, nearly the melting point of silicon. Moreover, the mechanism of microstructural development (especially concerned with formation of β-SiC) is not clearly understood.

In recent researches[4-7], to enhance the mechanical properties, fiber composites composed of Si/SiC matrix and carbon/SiC fiber have been studied. Also, to replace the brittle matrix with silicides has been developed for application at high-temperature.

In this study, to improve the mechanical properties as well as microstructural development, the particle size of starting material is controlled. And the infilitration characteristics is also investigated as a function of particle size.

Experimental procedure

The starting materials used for fabricating preforms were α-SiC (Lonza-Werke GmbH, Switzerland) and graphite (SEC, Co., Japan) as given in Table 1,2. The particle sizes of graphite and SiC were varied to control pore diameter, which resulted in different microstructure. And the ratio of α -SiC : C was fixed by 6 : 4. After mixing SiC and graphite by ball milling, the preforms were dried and then granuled. The preforms were fabricated by uniaxial pressing in the range of 20~60 MPa. The relative density of preforms was nearly 60% of theoretical density of SiC as given in Table 3.

Fig. 1 shows overal experimental procedure. The reaction-bonding process was carried out under Ar atmosphere at 1550 - 1600 °C for 20 min.. After reaction bonding process, sintered bodies were cut into L40×W4×H3 mm bars

and then their surfaces were polished with an 1200-grit diamond wheel for flexural testing. The densities of sintered body were measured according to ASTM-C20.

Fracture toughness was measured using both indentation method by Lawn & Fuller[8] and Single Edge Notched Beam(SENB) method[9].

Microstructure and phase analysis

The density of reaction-bonded silicon carbide was nearly 3.0 g/cm^3 as given in Table 3. The density of sintered body[1-3] is known to be dependent on that of α-, β-SiC (3.21g/cm^3), residual silicon(2.33 g/cm^3) and pore. However, as the reaction-bonded SiC is fabricated with pore free, the low density is mainly due to resiual silicon filling pores. Calculating according to mixing rule, the amount of residual silicon is about 15-20 vol.%.

As given in Table 4, when a preform composing of fine SiC(0.5-1.5 μm) is infiltrated, cracking or formation of large defect is frequently obserbed. Also, the range of flexural strength is broad, which indicates possibility that the pore having small diameter can be chopped off by volume expansion during reaction. Thus, it is thought that the optimum composition of reaction bonded materials is in the range of coarse size (a few micrometer SiC).

The phases of reaction-bonded silicon carbide is analyzed by X-ray diffraction patterns as shown in Fig.2, whose phases are composed of α-, β-SiC and residual silicon. And the existence of graphite phase is negligible, which indicates that the reaction of graphite with molten silicon is fulfilled. The β-SiC[10-11] is known to be the newly formed phases around originl α-SiC, grown epitaxially.

Fig.3 shows typical SEM microstructure of reaction-bonded silicon carbide(× 1,500). A uniform and homogeneous microstructure is observed. The part of the white color represents original α-SiC used for starting material, and the part of the grey color is newly grown β-SiC around α-SiC. For the compostion having relatively large particle size of SiC, bimodal microstructure (composing of samll β-SiC between grains and epitaxially formed β-SiC around grains) are detected. As reported by many researchers[1-3,10-11], the mechanism of formation of fine grained β-SiC is mainly concerned with supersaturation of β-SiC. As the velocity of heat transfer is faster than that of material transfer almostly by 10^3, a temperature decrease is expected in the region of pore filled with silicon, which resulted in supersaturation of β-SiC due to low temperature. Concludly this supersaturation leads to formation of fine β-SiC by homogeneous nucleation and growth. Also, It is known that the formation of fine β-SiC does not always contribute to the improved mechanical properties because of the low joining strength between grains of β-SiC.

Mechanical properties of reaction-bonded silicon carbide

As shown Fig.4, the 3-p. flexural strength is in the range of 250-500 MPa. This strength is considered to be relatively high value as compared with conventional SiC(300-400 MPa for pressureless sintered SiC). However, for the case of small particle size of SiC(0.5-1.5μm), the mean strength is in the range of 250-300 MPa. And maximum strength is shown in the particle size of 7.9 μm SiC. Thus the optimum compostion is considered to extist in 7.9 μm (SiC).

Fig.5 shows fracture toughness. For a precise data, fracture toughness is measured by different method (Indentation method and SENB method). The difference between those of indentation method by Lawn & Fuller and SENB(Single Edge Notched Beam) is not appreciable. However, for composite of Si/SiC, SENB method is thought to be more acceptable, because crack length in the indentation method is irregular according to the part indentated.

Conclusion

The microstructure and mechanical properites are experimented as a function of particle size. It is found that bimodal microstructure is observed in the large particle size of SiC due to supersaturation of β-SiC. And the maximum strengh is about 500 MPa for composition of (SiC 7.9 μm and graphite 1 μm). And the mean fracture toughness is about 3.0 MPa · $m^{1/2}$.

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Table 1. Starting Materials

Materials		Supplier	Remarks
SiC	UF15	Lonza	Particle Size : 0.5 μm
	UF05	Lonza	Particle Size : 1.5 μm
	SiC(#4000)	Showa Denko	Particle Size : 3 μm
	SiC(#2000)	Showa Denko	Particle Size : 7.9 μm
	SiC(P800)	Showa Denko	Particle Size : 20 μm
Carbon	SGP 1	SEC, Japan	Particle Size : 1 μm
	SGP 5	SEC, Japan	Particle Size : 5 μm

Table 2. Composition of SiC/Graphite Preform

Graphite / SiC	SGP1	SCN1	SGP5
SiC #2000	SG1		
SiC P800	SG2		SG6
SiC #4000	SG3		
UF05	SG4		SG7
UF15	SG5		SG8

Table 3. Apparent Density of RBSC

Specimen	Preform Density (g/cm^3)	Reaction Bonded SiC Density (g/cm^3)
SG1	1.6	3.0
SG2	1.6	2.94
SG3	1.62	3.02
SG4	1.63	3.02
SG5	1.51	3.04
SG6	1.43	3.05
SG7	1.42	2.99

Table 4. Data for Characteristics of molten Si Infiltration

Graphite / SiC	SGP1	SCN1	SGP5
SiC #2000	○ SG1	○	
SiC P800	○ SG2		○ SG6
SiC #4000	○ SG3		
UF05	× SG4 cracking		× SG7 cracking
UF15	× SG5 cracking		× cracking

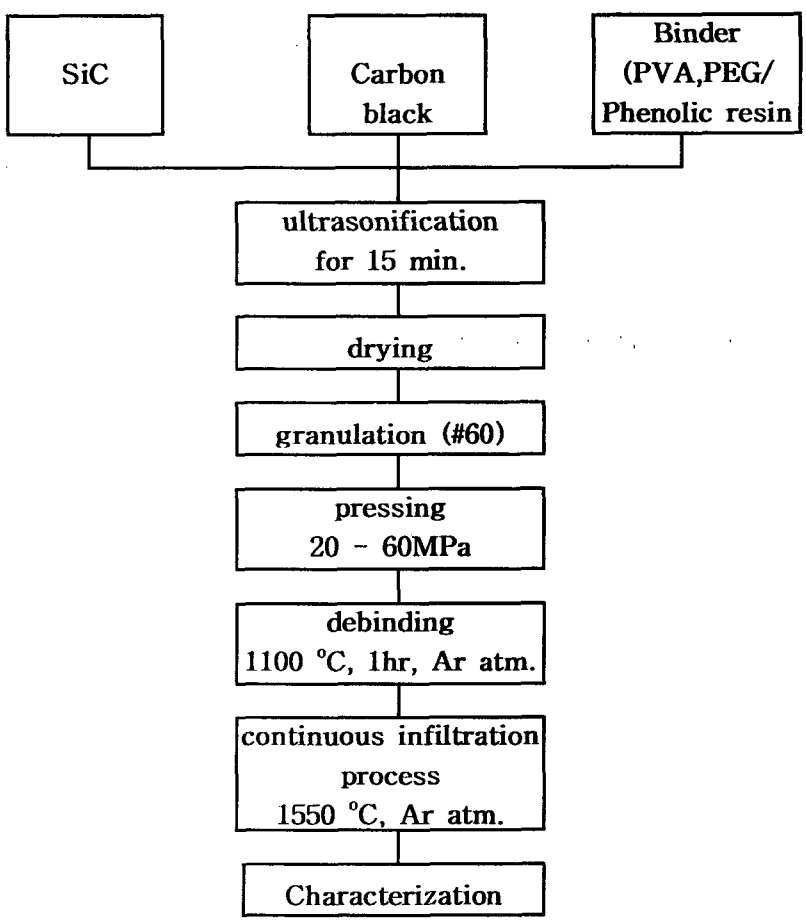

Fig.1. Overall experimental procedure

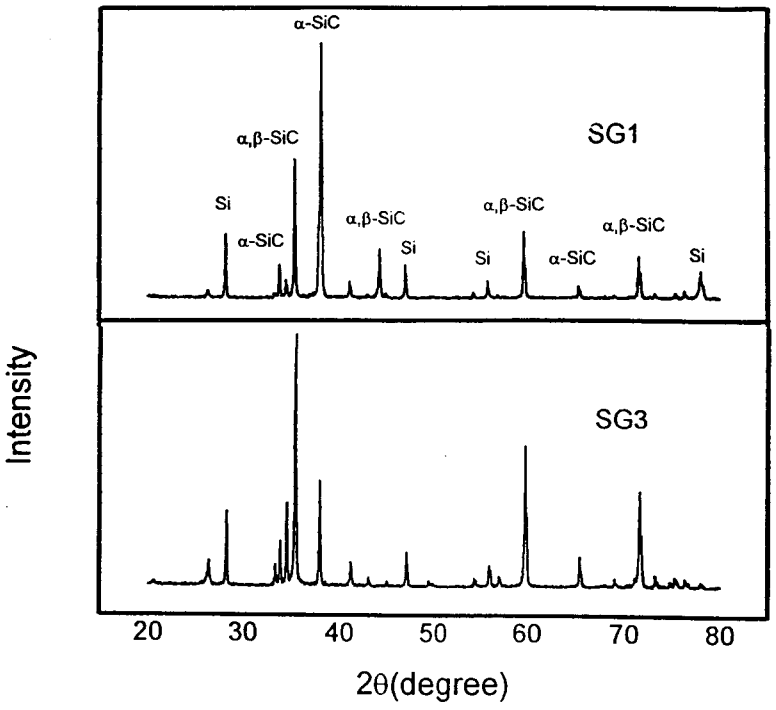

Fig.2. X-ray diffraction patterns for RBSC fabricated at 1550°C for 20 min.

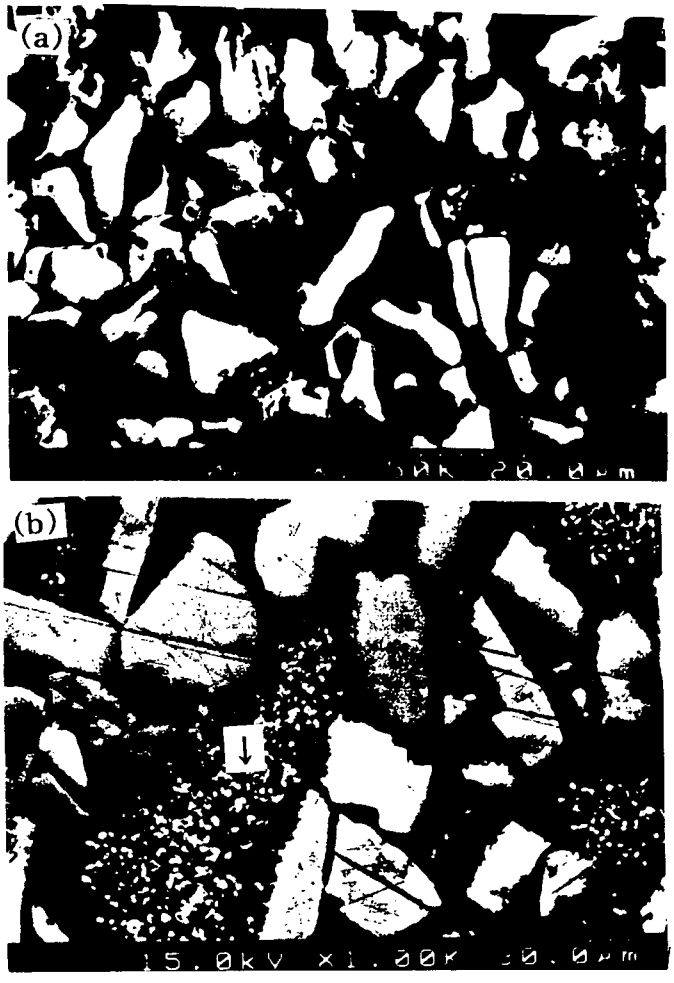

Fig.3. SEM microstructure of RBSC fabricated by a molten Si infiltration process ; (a)SG1 and (b)SG2

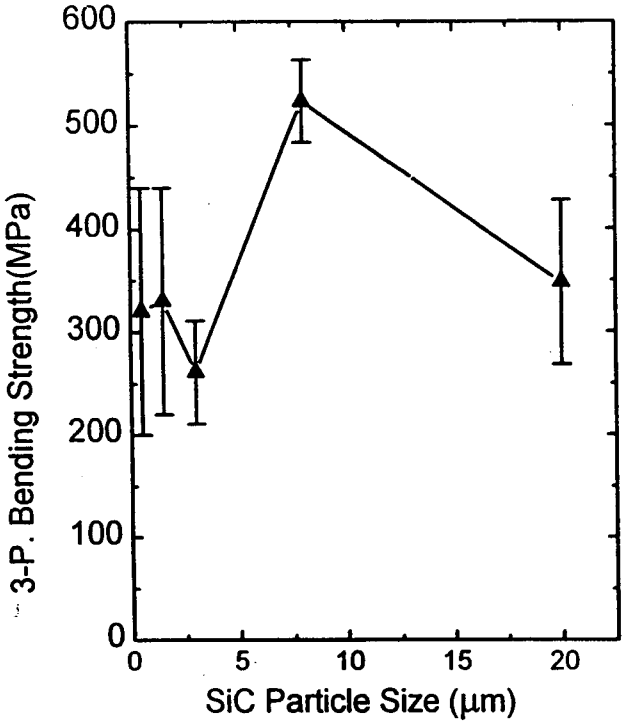

Fig.4. Variation of 3-P. bending strength as a function of SiC particle size used in the preform (graphite : 1μm)

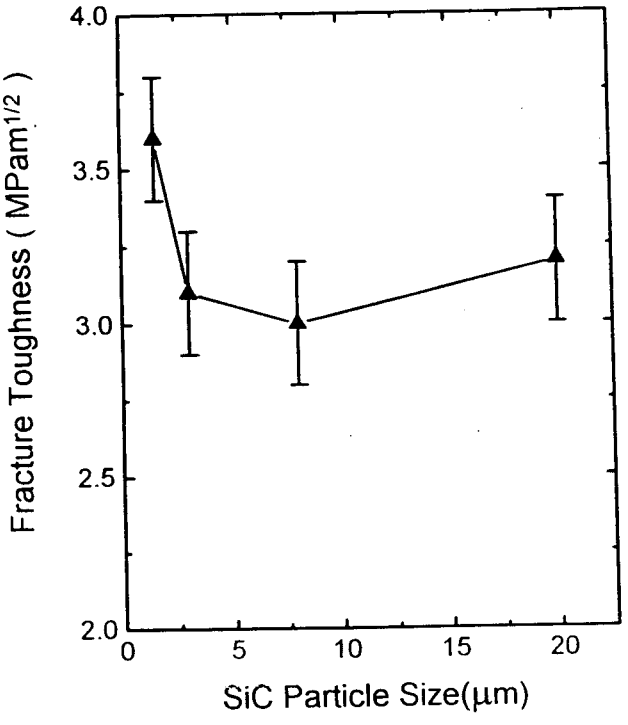

Fig.5. Variation of fracture toughness as a function of SiC particle size used in the preform (graphite : 1μm)