Columnar inter-grain growth of SiC-AlN solid solutions

Chang-Sung Lim
Ceramic Materials Research Institute(CMI), Hanyang University, Seoul 133-791, Korea
Il Hyong Jung and Keun Ho Auh
Department of Inorganic Materials Engineering, Hanyang University, Seoul 133-791, Korea

1. Introduction

SiC and AlN become attractive materials with interesting mechanical and electronic properties. SiC is used as high strength and high temperature ceramics because of its excellent corrosion and erosion resistance in the field of promising candidates for structural materials at room as well as elevated temperatures. However, its low fracture toughness has to be overcome before promoting the application as engineering components. AlN has recently come into prominence as an electronic substrate material. SiC is a covalant compound which exists either in a cubic structure (3C) or in various hexagonal or rhombohedral polytypes (2H, 4H, 6H, 15R and 21R). The 2H polymorph of SiC is isostructural with AlN and Al_2OC with a strong covalent bond. The similarity between two structures and their properties suggests that alloying of one with other may provide the potential for property optimization[1-2].

Cutler et al.[3-7] reported the formation of a SiC-AlN solid solution in the range 2-100 % AlN has been formed at 1600℃ by a vapor phase process using carbothermal reduction of amorphous silica and aluminium hydroxide in nitrogen. Ruh and Zangvil[8] described the solid solution exists as a single phase above 2100℃ over the composition range 35-100 mol% AlN. Their flexural strengths were quite low due to inhomogeneities in grain size and composition and spinodal decomposition occurs on annealing below 1950℃. The covalent nature of the atomic bonding in SiC and AlN makes diffusion extremely slow, so that pressures of several hundred bars and temperatures of up to 2300℃ are required to obtain complete solid solutions with high-density bodies.

In this work, the fabrication of SiC-AlN solid solution is studied by the hot pressing under 2030℃ using seeds of α-SiC to achive the complete 2H solid solutions with a lowering of the transformation temperatures and an enhanced fracture toughness.

2. Experimental

The materials were used β-SiC (Beta Randum, Ibiden Cimpany), AlN (Grade F. Tokuyama Soda Company) and α-SiC (Du A-1 Showa Denko) powders. Nominal compositions investigated ranged between 2, 5 and 8 wt% α-SiC with 50% SiC/50%

AlN(Table 1). Specimens (3 cm in diameter by 0.5 cm thick) were uniaxially hot-pressed in graphite dies lined with graphite washer. Hot-pressing was conducted under nitrogen at 1870℃ for 4 h and 2030℃ for 1 h at 22.5 MPa(Table 1). Cooling was sufficiently rapid for this study, so that the high-temperature phase was quenched to room temperature.

The specimens were polished using diamond pastes of 30, 15, 3, 1 ㎛. The samples were ultrasonically cleaned in ethanol, rinsed with distilled water and dried. All of the samples were examined using X-ray diffraction(XRD) with CuKα. Polished samples were etched using Murakami's etch to reveal the microstructure. The etched sections were investigated using optical microscopy and scanning electron microscopy(SEM). The indentation fracture test method was used with a load of 10 Kgf, a loading speed of 0.1 mm/sec and a load time of 20 sec on the finished surfaces of the specimens for measurements of fracture toughness and Vickers hardness.

3. Results and discussion

The phase presents in experimental samples were determined by XRD analysis. On the compositions of 2W50A, 5W50A and 8W50A as hot-pressed at 1870℃ for 4 h were similar to those obtained on the compositions of A50 hot-pressed at 1870℃ for 4 h. When these compositions were hot-pressed at 2030℃ for 1 h, a single 2H solid solution was obtained for the compositions of 2W50A and 5W50A and with some 4H and 6H for the composition of 8W50A. These results indicate that the reaction of AlN and β-SiC powder transformed to the 2H structure appeared to depend on the temperature and amount of AlN and seeds present. The phase transformation is influenced by the seeding resulting in a lowering of the transformation temperature. The typical XRD patterns on the SiC-AlN solid solutions of the sample A50 and 5W50A pressed at 203 0℃ for 1 h are shown in Fig. 1. The hexagonal lines with various splittings on the sample A50 in Fig. 1(a) indicate the 2H solid solution with various polytypes, whereas the strong hexagonal lines with the absence of any splitting indicates that the sample 5W50A is a single SiC-AlN solid solution of 2 H in Fig. 1(b). The crystalline phases consist of two main phases : one is a SiC-rich solid solution phase and the other is an AlN-rich solid solution phase. The (110) reflection at 2θ=59.6° provides the maximum possibility of observing splitting (two phases of identical structure with slightly different lattice parameters) since it has largest 2θ difference between SiC and AlN of the major diffraction peaks. Fig. 2 shows the typical splitting at 2θ=59.530° and 60.205° in the sample A50, which indicates the high intensity value of Si-rich phase.

Results of SEM studies were good agreement with XRD results. Fig. 3 shows typical microstructures of the SiC-AlN solid solution of 2W50A, 5W50A and 8W50A as hot-pressed at 2030℃ for 1 h, which previously were etched using Murakami's etch. All of the sample had densities in excess of 99 % of theoretical density. According to the XRD results, for a composition of 50% AlN/50% SiC with a seeding of 2 wt% and 5 wt% α-SiC, the complete solid solution with a single phase of 2H could be obtained by hot pressing at 2030℃ for 1 h. The microstructures of the samples of 2W50A and

5W50A are equiaxed with a relatively homogeneous grain size of 2H phases (Fig. 3(a), 3(b)). Sample 2W50A presents a microstructure similar to that of 5W50A, with somewhat smaller grains. However the sample of 8W50A exhibited an inhomogeneous size distribution (Fig. 3(c)). Fig. 4 shows high magnification scanning electron micrographs of these samples of 2W50A, 5W50A and 8W50A. In Fig. 4(a), small amount of the columnar inter-grain growth in 2H structures was observed in equiaxed grains of the sample 2W50A. In Fig. 4(b), the sample 5W50A exhibits an uniform grains, which consists of a single phase of the solid solution. In contrast, a large proportion of heavy strained and faulted grains with exaggerated columnar inter-grain growth was observed in complicated grain mixtures of the sample 8W50A in Fig. 3(c). It is noted that a material with a high fracture toughness can be generated from those kinds of columnar inter-grain growth between grains. Table 2 and Table 3 show results of fracture toughness and Vickers hardness measurements. Sample 2W50A, 5W50A and 8W50A show renovational values in fracture toughness. Sample 5W50A which obtained relatively homogeneous columnar inter-grain growth was higher values of 6.9 MPa · $m^{1/2}$ than those of 2W50A and 8W50A. The fracture toughness of the specimens is enhanced by a reinforcement mechanism of the columnar inter-grain growth in 2H single solid solutions. The seeding weight dependences of the improvment of the fracture toughness are maximum in the case of 5W50A. In the 8W50A sample, somewhat exaggerated columnar growths decreased the fracture toughness. In the 2W50A sample, the fracture toughness decreased resulting from inhomogeneous columnar growths.

4. Conclusions

The reaction of AlN and β-SiC powder transformed to the 2H structure appeared to depend on the temperature and SiC/AlN ratio and seeds of α-SiC presents. For a composition of 50% AlN/50% SiC with a seeding of 2, 5 and 8 wt% for α-SiC at 2030℃ for 1 h, complete solid solution could be obtained. The phase transformation was influenced by the seeding resulting in a lowering of the transformation temperature and renovational values in fracture toughness. The crystalline phases consisted of SiC-rich solid solution phase and AlN-rich solid solution phase. The renovational values of 6.9 MPa · $m^{1/2}$ in fracture toughness are attributed to several thermodynamically induced columnar inter-grain growth in 2H solid solutions.

5. References

[1] D. Bloor, R. J. Brook, M. C. Flemings, S. Mahajan and R. W. Cahn, The Encyclopedia of Advanced Materials, Vol. 4 (1995) 2453.
[2] A. Zangvil and R. Ruh, J. Am. Ceram. Soc. 71[10] (1988) 884-890.
[3] Y. Sugahara, K. Kuroda and C. Kato, J. Am. Ceram. Soc. 69 (1984) C247.
[4] K. Tsukuma, M. Shimade and M. Koizumi, Ibid. 1 (1982) 9.
[5] Y. Sugahara, K. I. Sugimoto, H. Takagi, K. Kuroda and C. Kato, J. Mat. Sci. Lett. 7 (1988) 795.

[6] W. Rafaniello, K. Cho and A. V. Virkar, J. Mat. Sci. 16 (1981) 3479.
[7] I. B. Cutler, P. D. Miller, W. Rafaniello, H. K. Park, D. P. Thomson and K. H. Jack, Nature 275 (1978) 795.
[8] R. Ruh and A. Zangvil, Ibid. 65 (1982) 260.

Table 1 Compositions and hot-pressing conditions of powder mixtures

Sample name	SiC/AlN mole ratio	Composition (wt%)			Hot-press conditions		
		SiC	AlN	α-SiC	Temp (℃)	Time (h)	Press-ure (MPa)
2W50A	50/50	49.0	49.0	2	1870	4	22.5
					2030	1	22.5
5W50A	50/50	47.5	47.5	5	1870	4	22.5
					2030	1	22.5
8W50A	50/50	46.0	46.0	8	1870	4	22.5
					2030	1	22.5

Table 2 Vickers hardness of SiC-AlN solid solutions

Sample name	Hardness (GPa)
2W50A	17.085 ±0.15
5W50A	18.810 ±0.12
8W50A	16.683 ±1.34

Table 3 Fracture toughness and Vickers hardness of the 2W50A, 5W50A and 8W50A

Sample name	Fracture toughness (MPa · $m^{1/2}$)		
	Niihara	Ch & Ev	Lawn
2W50A	6.709±1.21	5.288±1.23	5.150±1.16
5W50A	6.885±1.17	5.427±1.11	5.310±0.98
8W50A	6.410±1.35	5.052±1.54	4.940±1.04

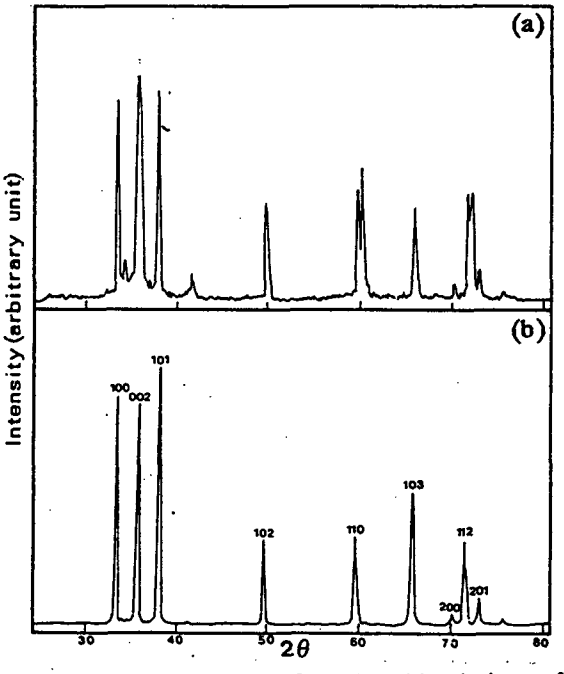

Fig. 1. XRD patterns of SiC-AlN solid solutions of the sample (a) A50 and (b) 5W50A hot pressed at 2030℃ for 1 h.

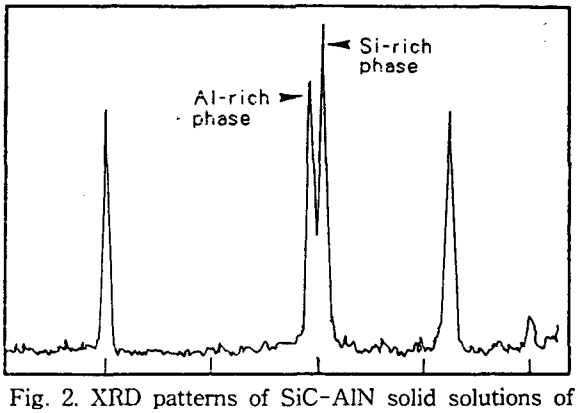

Fig. 2. XRD patterns of SiC-AlN solid solutions of the sample A50 indicates the splitting at 2θ =59.530° and 60.205°

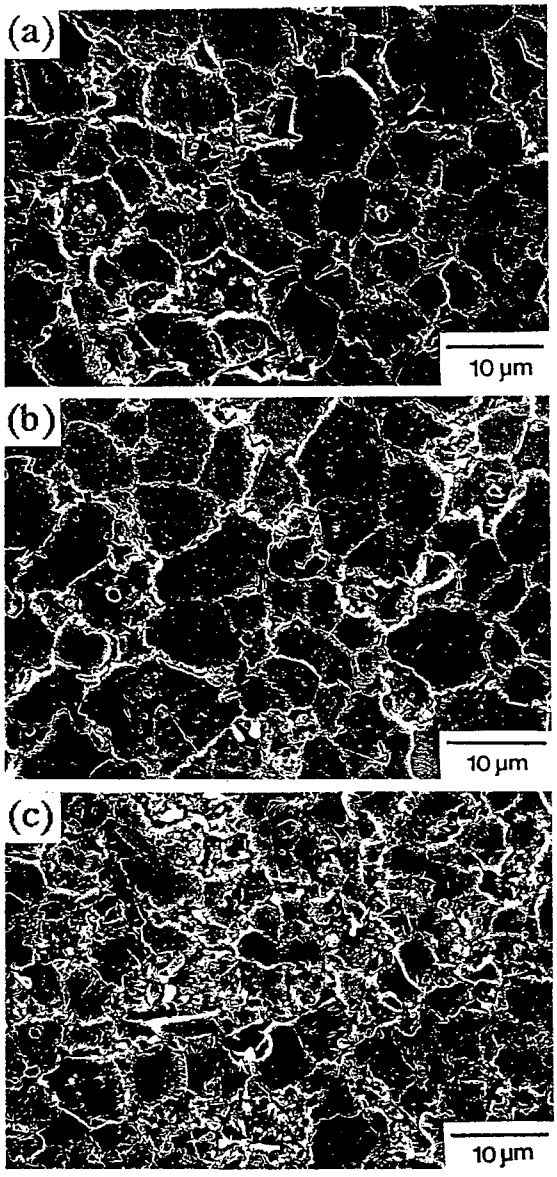

Fig. 3. SEM photographs of SiC-AlN solid solutions as hot-pressed at 2030℃ for 1 h : (a) 2W50A (b) 5W50A (c) 8W50A

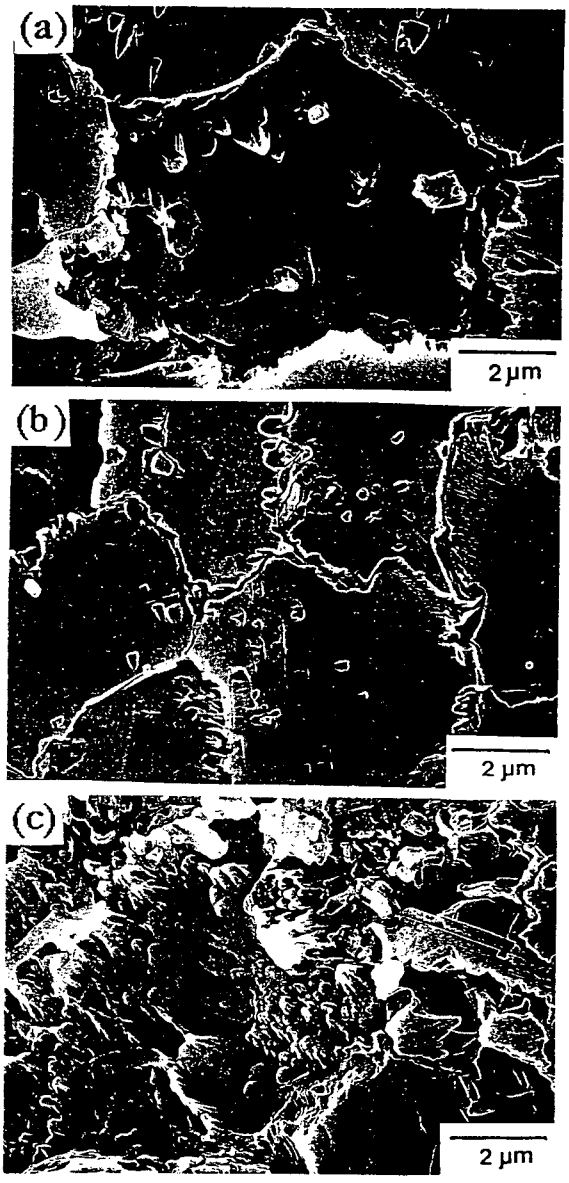

Fig. 4. High magnification SEM photograghs of SiC-AlN solid solutions as hot-pressed 2030℃ for 1 h : (a) 2W50A (b) 5W50A (c) 8W50A