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Fabrication and Characterization of Alumina Matrix Composites Reinforced with SiC whiskers

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Alumina matrix composites reinforced with up to 20vol% of aligned SiC whiskers were fabricated by tape casting and hot pressing. Alumina composites with randomly distributed SiC whiskers were also fabricated in order to investigate the effect of whisker alignment on properties of the composites. XRD and optical microscopy were used to examine the whisker orientation. The fracture toughness increased with increasing whisker content, and it was higher in the direction normal to the tape casting direction.

Key words : Alumina, SiC whisker, Tape casting, Hot pressing, Whisker orientation

I. Introduction

SiC whiskers have been used for reinforcing various monolithic ceramics including Al_2O_3, Si_3N_4, mullite and etc.[1-3] But, it is difficult to obtain fully dense composites containing whiskers because of whisker entanglement. It is important to avoid the contact among the whiskers and have homogeneous microstructure in order to achieve high density and excellent mechanical properties. Whisker entanglement can be reduced by aligning the whiskers. Furthermore, whisker alignment can provide excellent mechanical properties.

Lee *et al.*[4] showed the model predicting toughness anisotropy of whisker-reinforced ceramic composites and investigated Al_2O_3/SiC whisker composites with various textures by several techniques. However, like the other researchers, they hot pressed the composites that contained 2-D random whiskers. On the other hands, Wu and Messing[5] made mullite based composites with unidirectionally oriented SiC whiskers by using tape casting technique. They achieved 2-D aligned whiskers by tape casting and subsequently 3-D aligned whiskers by lamination, hot pressing

Tape casting is a method for producing flexible green sheet that can be subsequently stacked to form the laminated structure. It provides control over whisker orientation in ceramic tape and makes it feasible to align the whiskers with preferred orientation in 3-D composite structure after lamination. In the present study, we fabricated Al_2O_3 based composites with randomly oriented SiC whiskers by using conventional powder processing method and Al_2O_3/SiCw composites with preferred whisker orientation by a modified tape casting technique, lamination and hot pressing.

The influences of whisker content and orientation on the mechanical properties and toughness anisotropy of the composites were investigated.

II. Experimental Procedure

High-purity alumina powder (AKP-30, Sumitomo Chemical Co., Osaka, Japan), SiC whisker (SCW#1-0.8, Tateho Chemical Industries Co., Tokyo, Japan) and small amount of MgO powder (Junsei Chem. Co., Tokyo, Japan) were used in this study. Five kinds of specimens were fabricated: monolithic alumina, alumina composites with 10 vol%, 20 vol% of randomly oriented SiC whiskers which were labeled as R_{10}, R_{20} and alumina composites with 10 vol%, 20vol% of aligned SiC whiskers which were labeled as A_{10}, A_{20}.

Monolithic alumina sintered body with 0.5 wt% MgO was prepared by planetary ball milling, drying, sieving and hot pressing at 1450°C and 1800°C for 0.5 hr under 30 MPa in Ar for comparison with the whisker-reinforced alumina composites in terms of the mechanical properties and microstructures.

Alumina composites with randomly oriented SiC whiskers, R_{10} and R_{20} were fabricated by using the following procedures. Alumina powder and 0.5 wt% of MgO were mixed by planetary ball milling for 5 hrs. The solvent was ethyl alcohol and alumina balls of ϕ 5 mm in diameter were used as milling media. SiC whiskers were ultrasonicated in ethyl alcohol to reduce whisker agglomerates. After mixing the two slurries, second planetary ball milling was carried out for a short time, 10 min, in order to avoid breakage of SiC whiskers. The final mixed powder was obtained after drying and sieving. The sintered products were prepared by hot pressing at 1800°C for 1hr under 30 MPa in Ar gas.

The fabrication process of alumina composites contain-

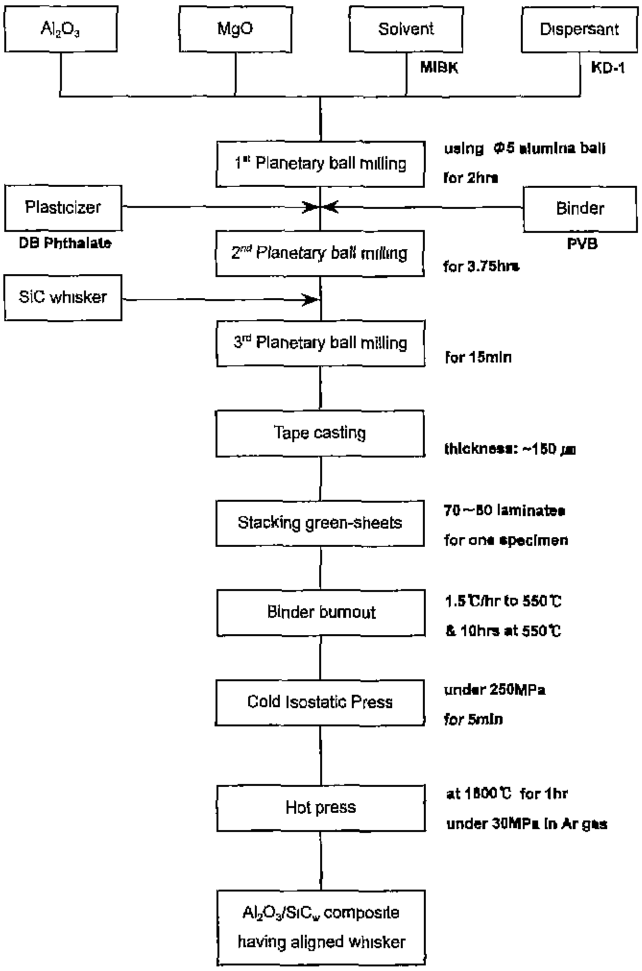

Fig. 1. Flow chart for fabrication of Al_2O_3/SiCw composite containing aligned whisker.

ing aligned SiC whiskers, A_{10} and A_{20} was shown in Fig. 1. It is divided into the three procedures; tape casting, lamination and hot pressing. Al_2O_3 and MgO powder were mixed with dispersant, solvent and they were planetary ball milled for 2 hrs in acetal jar by using high-purity alumina ball. MIBK (Methyl Isobutyl Ketone, Mallinckrodt Chemical Co., St. Louis, U.S.A.) was used as solvent and dispersant was KD-1 (ICI Chemical Co., Barcelona, Spain). Dibutyl phthalate (Aldrich Chemical Co., New York, U.S.A.) and Polyvinyl butyral (Aldrich Chemical Co., U.S.A.) were added as plasticizer and binder, respectively, and were planetary ball milled for another 3.75 hrs. The third planetary ball milling was carried out for 15 min after adding SiC whisker and mixed slurry was obtained. Rheological behavior of slurry is significantly dependent upon the optimal selection of the quantities and kinds of solvent/dispersant/plasticizer/binder system, which is important for making desirable green tape. The amount of solvent, dispersant, plasticizer and binder in this slurry was determined as the ratios with respect to the weight of added powders. The slurry was defoamed in the vacuum chamber and green tape was fabricated by a doctor blade. An array of pins was placed at the exit of the slurry reservoir in order to enhance the whisker alignment. Thickness of the tape was about 150 μm. Sheets cut from green tapes were stacked at 85°C for 30 min under 50 MPa. After binder removal, the laminated body was cold isostatic pressed for 5 min under 250 MPa and successively hot pressed at 1800°C for 1hr under 30 MPa in flowing Ar atmosphere in a BN- coated graphite mold.

The bulk densities of the sintered bodies were measured by water immersion method. The specimens were cut, ground and polished for characterization. Specimens were cut into $3\times4\times34$ mm^3 dimensions for three point flexural strength measurements. Span was 20 mm and crosshead speed was 0.5 mm/min. The microvickers hardness test was done under load of 2.9N. The fracture toughness value was obtained according to Charles & Evans' equations[6] after measuring indentation crack lengths that were generated under 196N.

X-ray diffractometer (Rigaku Co., Tokyo, Japan) was employed in order to investigate the degree of whisker orientation. The microstructure and crack propagation behavior were observed by scanning electron microscope (JSM-5800, Jeol, Tokyo, Japan) and optical microscope (EPIPHOT- 300, Nikon, Tokyo, Japan).

III. Results and Discussion

Fig. 2. shows the relative densities of monolithic alumina ceramics and whisker-reinforced composites R_{10}, R_{20}, A_{10} and A_{20}. Although the densities of composites were slightly lower than that of monolithic alumina, all of the composites exhibited high densification over 99.5% of theoretical density. Optical micrographs of the composites are shown in Fig. 3. Fig. 3(a) and (b) show the polished surface perpendicular to the hot pressing direction of the composites. The hot pressing direction was normal to

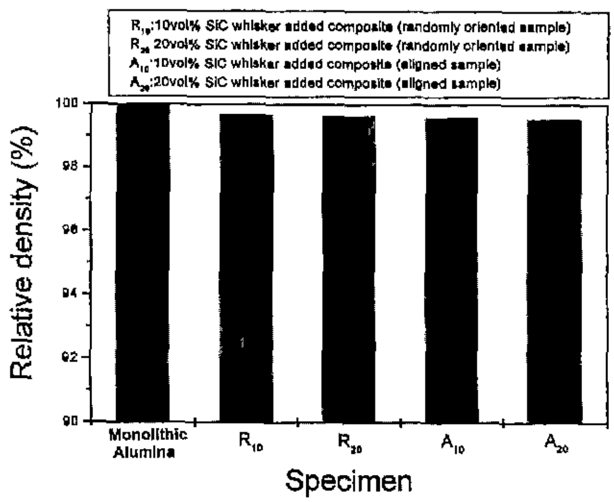

Fig. 2. The relative densities of the specimens.

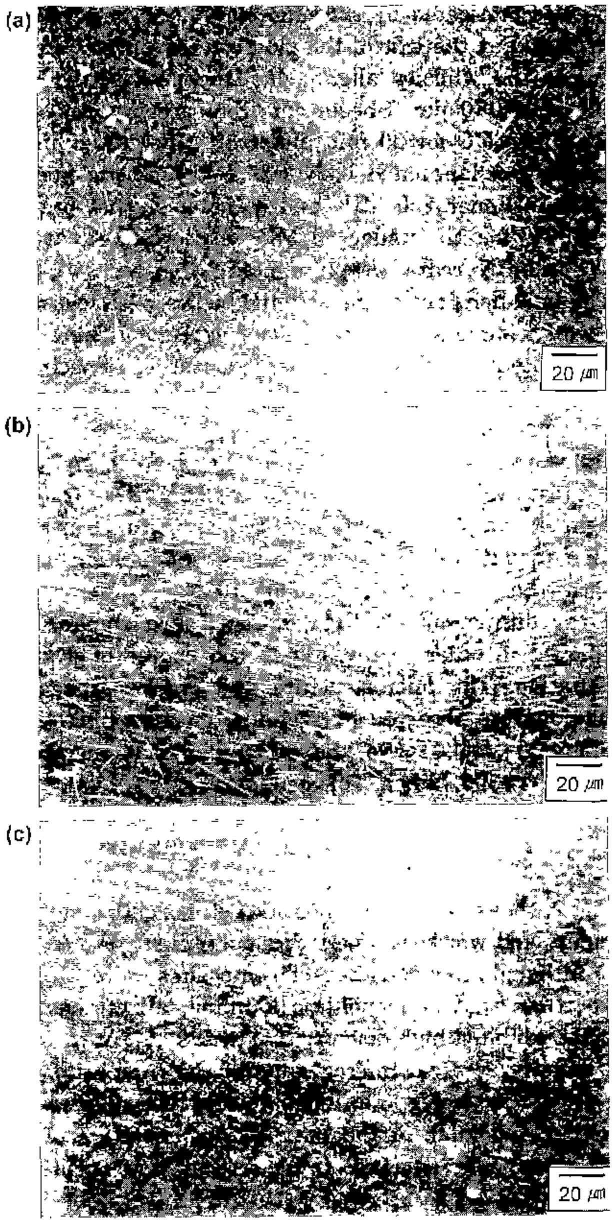

Fig. 3. Optical micrographs of the polished surfaces of the composites: normal to hot pressing direction of (a) R_{20}, (b) A_{20} and (c) normal to tape casting direction of A_{20}.

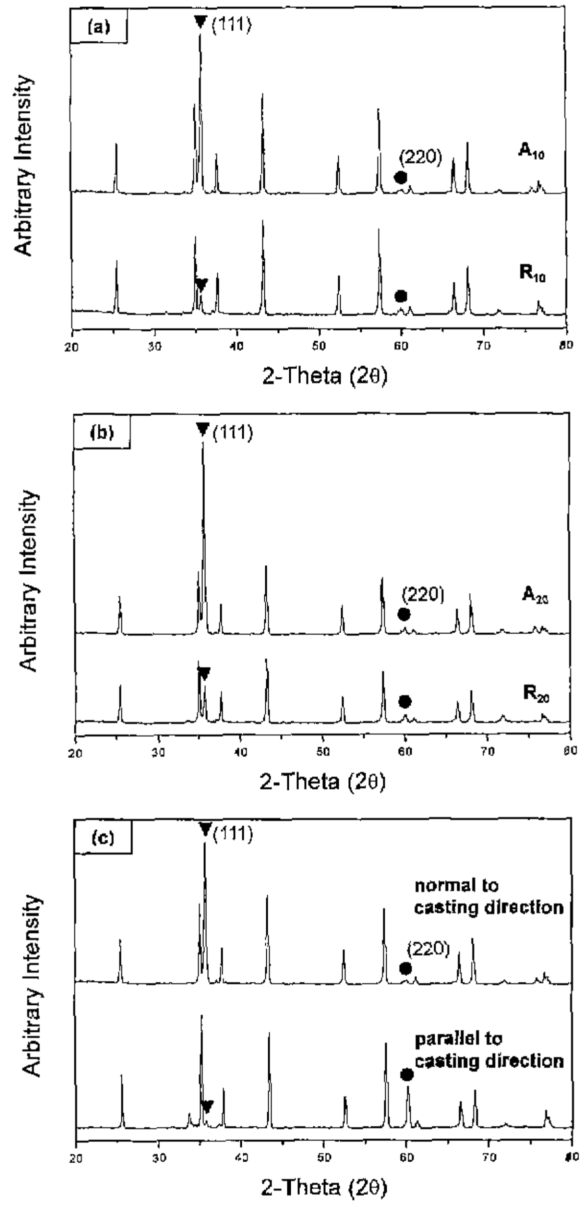

Fig. 4. The X-ray diffraction patterns of composites confirming the whisker alignment along the casting direction: ▼-(111), ●-(220) plane The other peaks indicate the monolithic alumina.

tape casting direction. The white, acicular shaped particles were SiC whiskers. It is evident that excellent whisker alignment as well as well-dispersed whisker was achieved by tape casting. The composites fabricated using powder-processing method have little whisker agglomerates due to ultrasonication treatment of whiskers before mixing. These facts result in the high densities shown in Fig. 2.

The as-received SiC whiskers used in the study were 0.6 to 1.0 μm in diameter and 5 to 35 μm long. Even if considering the facts that lengths of the randomly distributed whiskers in R composites appear shorter than their actual length on the ground surfaces, many whiskers in R_{20} composites are shorter than as-received whiskers in length. It is believed that many whiskers were damaged and reduced in length during mixing, sieving and hot pressing process. In case of whiskers in A_{20} composites, the degree of whisker breaking is less severe than that of R_{20} and still long whiskers are observed in abundance.

Fig. 3(c) is the micrograph of the surface normal to tape casting direction of specimen A_{20}. Unlike Fig. 3(a) and (b), the majority of the round-shaped white spots were observed. These seemed to be the cross-section of SiC whiskers and it means that whiskers are aligned well in the direction of tape casting. X-ray beams were irradiated on the surface normal to casting direction of specimens R_{10}, R_{20}, A_{10} and A_{20}. In addition, X-ray diffraction on the surface parallel to casting direction of specimen A_{10} was carried out. The results of X-ray diffraction analysis of the composites are plotted in Fig. 4. Fig. 4(a), (b) show the difference of the X-ray diffraction pattern between whisker aligned composites and randomly distributed whisker reinforced composites. While the peaks for (111) of specimen R_{10} and R_{20} are only a small fraction of the whole peaks, those of A_{10} and A_{20} are higher than any other peak. The SiC whisker used in this study is β-SiC which belongs to cubic crystallographic system, its long axis is toward <111> direction. As the (111) plane is the cross-sectional plane of cubic-structured β-SiC whisker and the X-ray was irradiated on the specimen surface normal to casting direction, it implies that many whiskers in specimen A_{10}, A_{20} are aligned toward the tape casting direction (Fig. 5). Since the peak represented (111) plane of the whiskers that were exactly aligned in the direction of casting, the actual degree of orientation should be improved by considering the whiskers slightly off the tape casting direction. XRD results of specimen A_{10} according to different X-ray irradiation directions are shown in Fig. 4(c). Although X-ray diffraction was carried out for the same sample, the results were different according to the irradiating direction. When X-ray was irradiated on the surface parallel to casting direction, the peak for (220) plane was stronger than any other peak of the SiC whiskers. When X-ray was irradiated on the surface perpendicular to casting direction, the highest peak was the peak for (111) plane. It reconfirmed that the SiC whiskers were aligned well along the casting direction in specimen A_{10}.

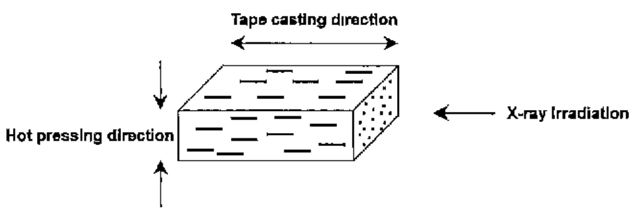

Fig. 5. Schematic of whisker aligned composites.

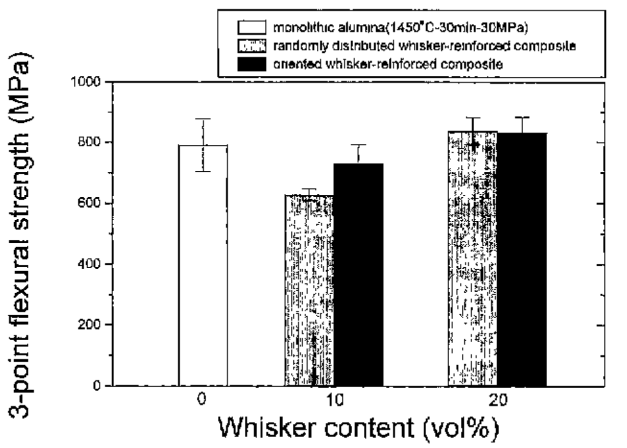

Fig. 6. The flexural strength of the specimen as a function of whisker content.

Fig. 6 shows the flexural strength of the specimens. The flexural strength of monolithic alumina was as high as 790 MPa. The highly fine alumina powder (median particle size of 0.36) was used and the added MgO inhibited the grain growth of monolithic alumina during hot pressing at 1450°C, resulting in fine-grained alumina. It seems likely that high strength of monolithic alumina was due to the fine-grained microstructure. In comparison between R and A samples, the strength of specimen A_{10} was higher than that of specimen R_{10}. The test bars for the strength measurements of specimen A_{10} were prepared to have tensile surface parallel to the casting surface and fracture was occurred along the direction perpendicular to SiC whisker orientation direction. It is thought that the aligned whiskers normal to fracture surface contributed to the higher strength of A_{10}. The composites with 20 vol% whiskers showed higher strength than the ones with 10 vol% whiskers, and they exhibited similar strength values regardless of whisker orientation.

Scanning electron micrographs of the polished surfaces of the specimens are shown in Fig. 7. Fig. 7(a) is the micrograph of monolithic alumina hot-pressed at 1450°C. In order to investigate effect of whisker content and orientation on the microstructures of the composites, five kinds of specimens were hot pressed at 1800°C. Fig. 7(b)~(f) show SEM micrographs of the specimens after thermal etching. The monolithic alumina sintered at 1450°C had a homogeneous microstructure and the grain size was smaller than that of any other specimen. In case of specimens sintered at 1800°C, the matrix grain size decreased as whisker content increased. It was reported that SiC whiskers inhibited growth of matrix grain in whisker-reinforced composites.[7]

The fracture toughness of the specimens obtained by using indentation method was shown in Fig. 8. The fracture toughness values were determined from measurements of crack lengths after Vickers indentation. The crack lengths were measured in longitudinal and transversal direction, namely, along the direction parallel and perpendicular to tape casting direction for the composites with aligned whiskers. The fracture toughness increased as the amount of whiskers increased. Fracture toughness increased from K_{IC}=2.5 MPa · $m^{1/2}$ for monolithic alumina to K_{IC}=4.8M Pa · $m^{1/2}$ for specimen R_{20}, and the toughness values are not different greatly according to the measurement direction. However, for specimens with oriented whiskers, the toughness anisotropy was remarkable. The toughness measured in the direction normal to the whisker orientation was larger than that measured along the whisker alignment direction; Maximum fracture toughness difference between the two directions was 1.75 MPa $m^{1/2}$. Fig. 9 shows the optical micrograph of the polished surface of specimen A_{20} after Vickers in-

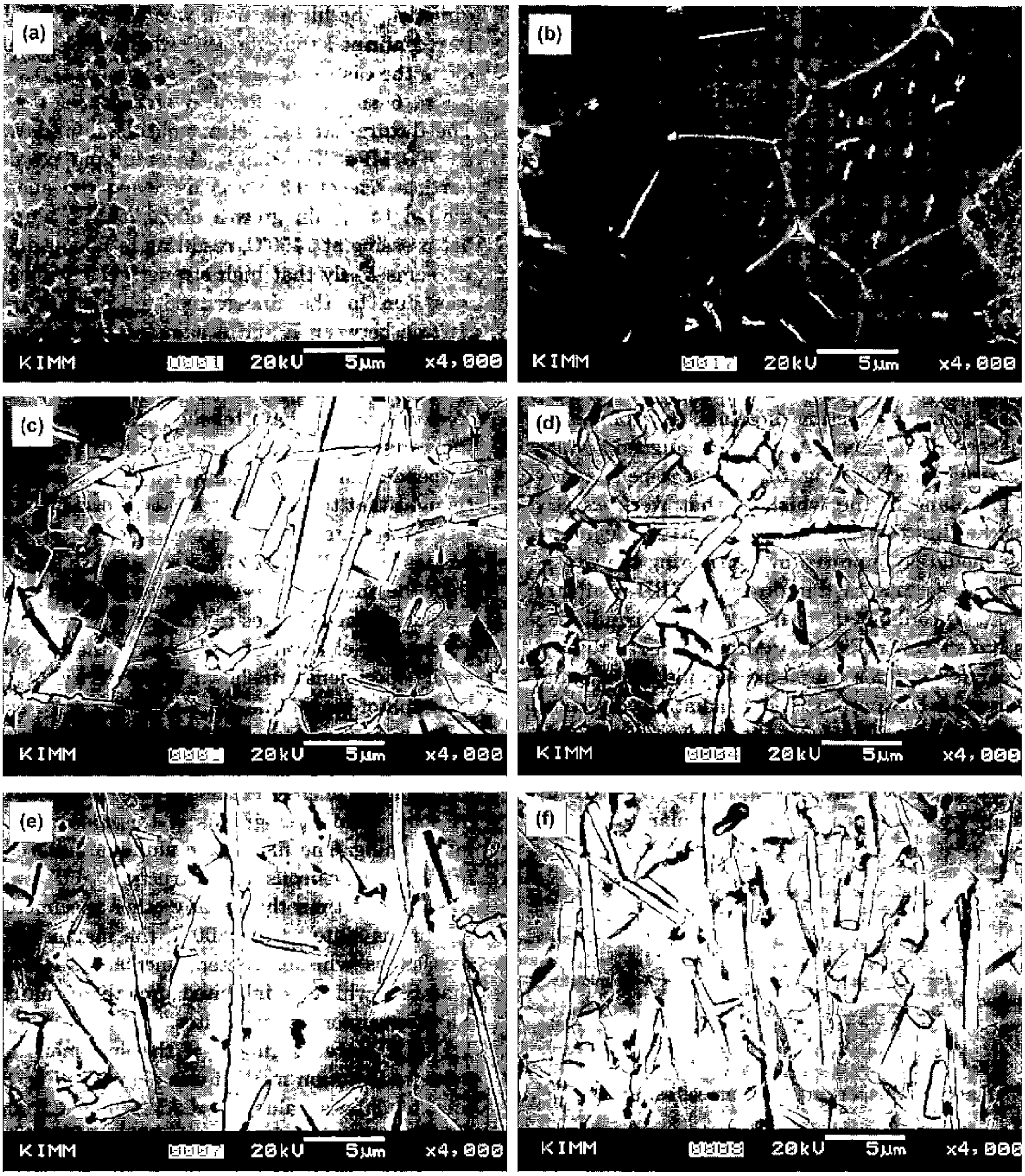

Fig. 7. SEM micrographs of the polished surfaces of the specimens: (a) monolithic alumina sintered at 1450°C, (b) monolithic alumina sintered at 1800°C, (c) R_{10}, (d) R_{20}, (e) A_{10} and (f) A_{20}.

dentation. An anisotropic crack propagation according to whisker orientation was clearly observed. The crack lengths were shorter and the fracture toughness values were higher in the direction normal to whisker alignment direction.

Toughening in whisker-reinforced composites occurs by a variety of mechanisms such as crack deflection, crack bridging and whisker pullout.[8] Each toughening mechanism can be combined with other mechanisms. In particular, the angle between the whisker and the crack plane was reported to be important for the toughening behavior of composites.[4] In this study, difference in toughening behavior is observed with crack propagation direction. In case of randomly oriented SiC whisker/Al_2O_3 composites, most of the whiskers were at small angles to the crack plane. Therefore, crack deflection and crack bridging accompanied by whisker/matrix interfacial debonding occurred predominantly (Fig. 10(a), (b)). In the direction normal to whisker alignment direction of specimens A_{10}, A_{20}, many whiskers were at high angle to the crack plane and toughening mainly occurred by crack propagation through whisker cut-off with pullout (Fig.

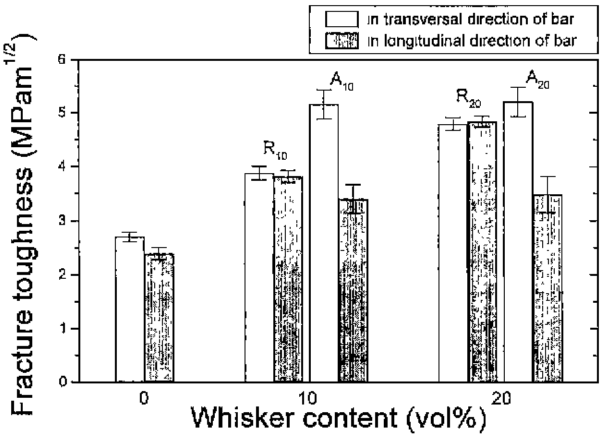

Fig. 8. The fracture toughness of the specimen with whisker content and crack propagation direction.

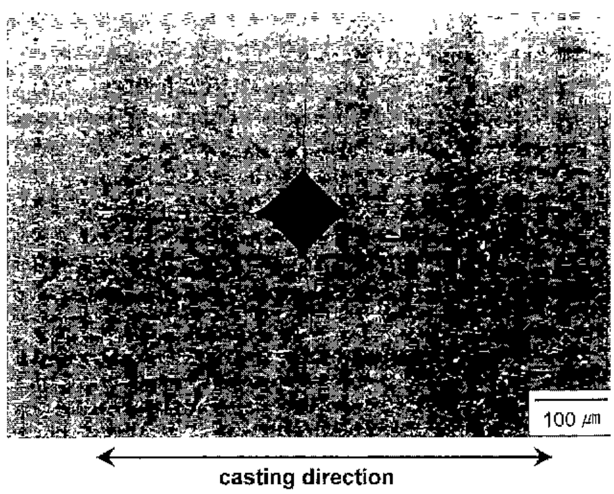

Fig. 9. Optical micrograph showing the crack propagation by vickers indenter for A_{20} composite.

10(c)). A small amount of crack deflection, bridging was also seen.

Fig. 11 shows the fracture surfaces of monolithic alumina and specimen R_{20}. The monolithic alumina was fractured along the grain boundary as well as within the grains, as shown in Fig. 11(a). Fig. 11(b) shows that a few whiskers were lying in the fracture surface, which indicates that the crack propagated along the whisker/matrix interface. Whisker protrusion and pullout were also observed.

IV. Conclusions

Al_2O_3 based composites with aligned SiC whiskers were fabricated by tape casting, lamination and hot pressing. The SiC whiskers were aligned along the tape casting direction, which was confirmed by X-ray diffractometer and optical microscopy. Al_2O_3 composites with 2-D randomly oriented SiC whisker were also fabricated by hot pressing. High densification was achieved for all

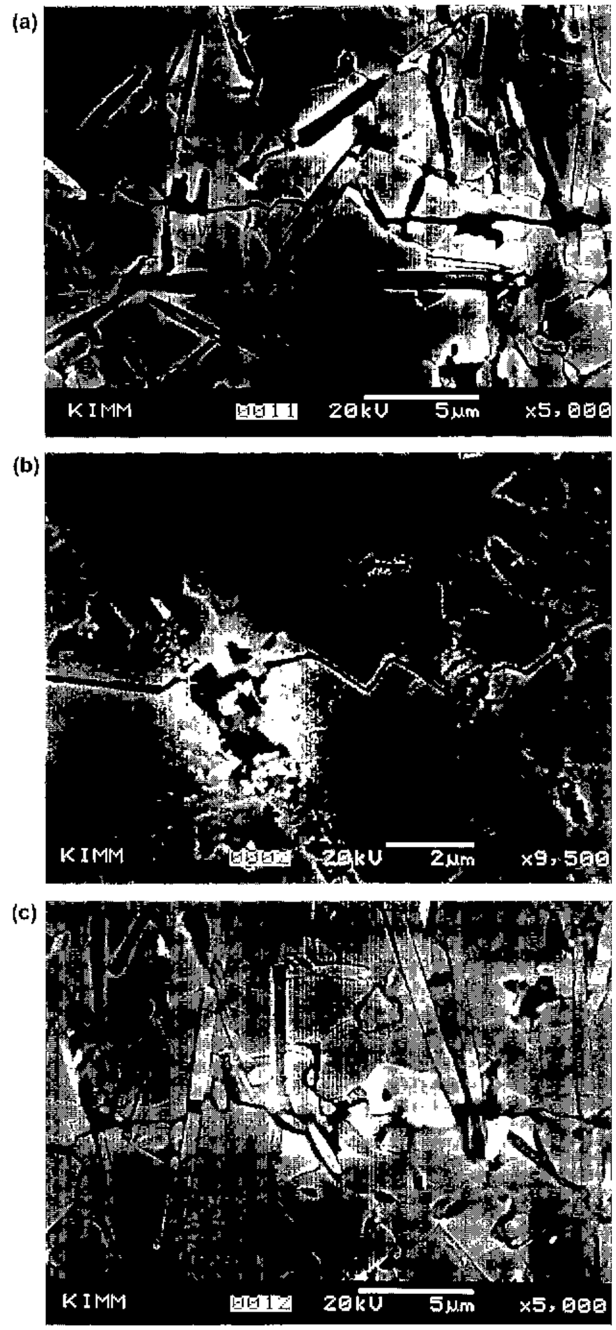

Fig. 10. SEM micrographs showing the toughening mechanisms by whiskers: (a), (b) R_{10} composite and (c) A_{10} composite.

the specimens. While specimen A_{10} showed higher strength than specimen R_{10}, the strengths of composites with 20 vol% SiC whiskers were similar regardless of whisker orientation. Addition of SiC whiskers promoted the fracture toughness, the toughness anisotropy was observed from the specimens with aligned whiskers. The fracture toughness value was the highest when the crack propagation was perpendicular to whisker alignment direction. Difference in the fracture toughness values according to the measurement direction was as large as 1.75 MPa · $m^{1/2}$. An evidence for crack deflection, bridging and whisker

(a)
(b)

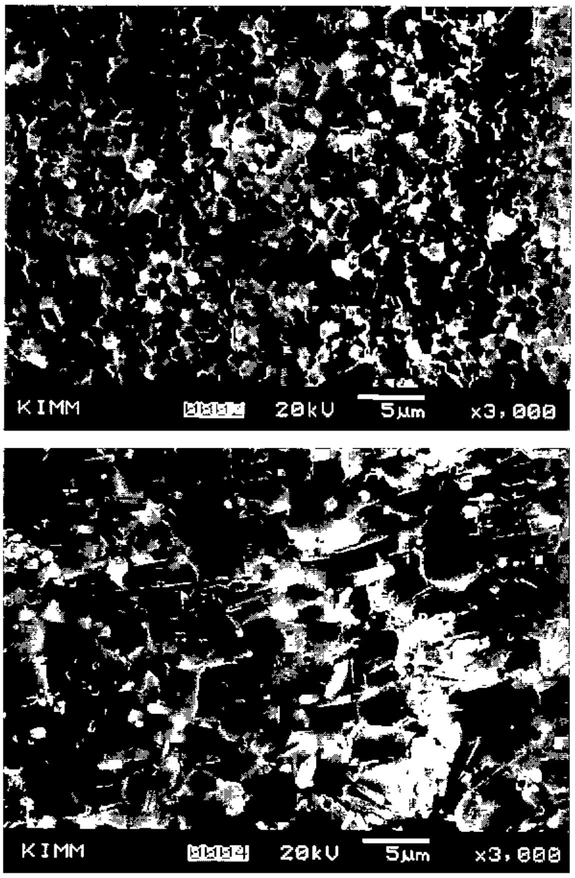

Fig. 11. SEM micrographs of the fracture surfaces of the specimens: (a) monolithic alumina sintered at 1450°C and (b) R_{20} composite.

pullout was observed from the polished and fracture surfaces of composites.

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