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Preferred Orientation and Strength Anisotropy of Pressureless-Sintered Silicon Nitride

Yasuhiro Goto, Akihiko Tsuge

Research and Development Center, Toshiba Corporation, 1, Komukai Toshiba-cho, Saiwai-ku, Kawasaki-shi 210, Japan.

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Katsutoshi Komeya

Department of Materials Chemistry, Yokohama National University, Tokiwadai, Hodogaya-ku, Yokohama-shi 240, Japan

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Abstract

The relation between preferred orientation and mechanical properties of silicon nitride (Si_3N_4) is important for engineering applications in which the stress-direction is critical. Preferred orientation was confirmed even in pressureless-sintered Si_3N_4 by using a mixture of granular α- and needle-like β-Si_3N_4 particles. Strength anisotropy due to the crystal orientation was also observed. The anisotropy was affected by the size of the elongated β-Si_3N_4 grains.

Der Zusammenhang zwischen einer Textur und den mechanischen Eigenschaften von Siliziumnitrid (Si_3N_4) ist vor allem in den technischen Anwendungen wichtig, wo die Richtung der Belastung eine besondere Rolle spielt. Selbst in drucklos gesintertem Si_3N_4, das aus einer Mischung von körnigem α- und nadelartigem β-Si_3N_4 hergestellt wurde, konnte eine Textur nachgewiesen werden. Eine Festigkeitsanisotropie, hervorgerufen durch die Orientierung der Kristalle, wurde ebenso beobachtet. Die Anisotropie wurde durch die Größe der länglichen β-Si_3N_4 Körner beeinflußt.

Il est important de connaître la relation entre l'orientation préférentielle et les propriétés mécaniques du nitrure de silicium (Si_3N_4) pour applications mécaniques dans les cas où la direction de contraintes est critique. L'orientation préférentielle a été confirmée même pour le Si_3N_4 obtenu par frittage naturel en utilisant un mélange de grains sphéroïdes α et d'aiguilles β. On a également observé une anisotropie de la résistance causée par l'orientation cristalline, et dépendant de la taille des grains aciculaires β.

1 Introduction

Attention has recently been focused on structural applications of Si_3N_4 ceramics for engine parts. The relation between preferred orientation and the mechanical properties of Si_3N_4 ceramics is quite important for applications in which the stress-direction is critical. It is well known[1–5] that, in hot-pressed Si_3N_4 with additives, the *c*-axis of the hexagonal β-Si_3N_4 crystal has a preferred orientation perpendicular to the hot-pressing direction. Strength anisotropy due to the preferred orientation has also been reported.[1,2,5] However, results have not been reported for pressureless-sintered Si_3N_4 ceramics. Preferred orientation in pressureless-sintered Si_3N_4 has accordingly been investigated and related to strength anisotropy.

2 Experimental Procedures

Si_3N_4 raw powder, which contains over 99% α-type equiaxed grains, was mixed with sintering additives, 5 wt% yttria and 3 wt% alumina, by ball milling with alumina balls for 12 h. This powder mixture was heated at 1650–1700°C for 1 h under 1 atm nitrogen. Mixtures of granular α- and needle-like β-Si_3N_4 were obtained. These powder mixtures were then moulded by uniaxial cold-pressing. The size of the powder compacts was 40 × 40 × 6 mm. The powder compacts were sintered at 1800–1850°C under 1 atm nitrogen.

The powder mixtures and sintered materials were examined by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The α/β ratios of the powder mixtures were determined by X-ray diffraction (XRD).[6] The three-point bend strengths were also measured for the dense sintered bodies. The size of the bend test pieces was 3 × 3 × 30 mm, and the span was 20 mm.

A pressureless-sintered material prepared without prior heat treatment of the powder and a hot-pressed material were fabricated for comparison.

3 Results and Discussion

3.1 Preferred orientation

Figure 1 shows scanning electron micrographs of the heat-treated powders. Needle-like grains were formed by the heat treatment. Powder B, which was heated at 1700°C, had a larger size and larger amounts of the needle-like grains than powder A, which was heated at 1650°C. XRD charts of powders A and B (Fig. 2) show that the needle-like grains were β-Si_3N_4. The amount of β-Si_3N_4 was 7 wt% for powder A and 41 wt% for powder B. Figure 3 shows a TEM micrograph and an electron diffraction pattern of a needle-like grain. The growth direction of this needle-like β-Si_3N_4 grain was in the c-axis direction.

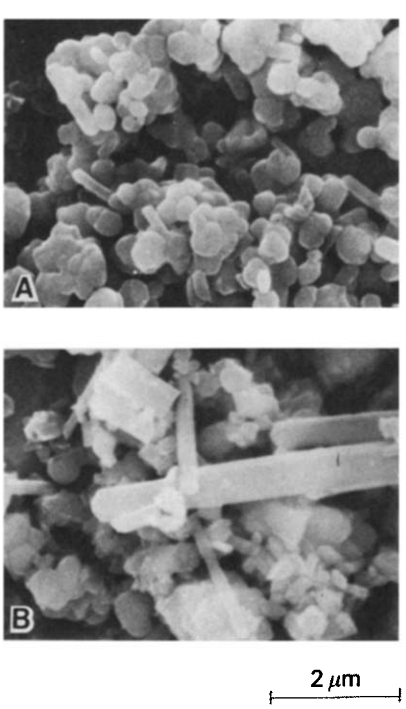

Fig. 1. SEM micrographs of heat-treated powders. A, 1650°C; B, 1700°C.

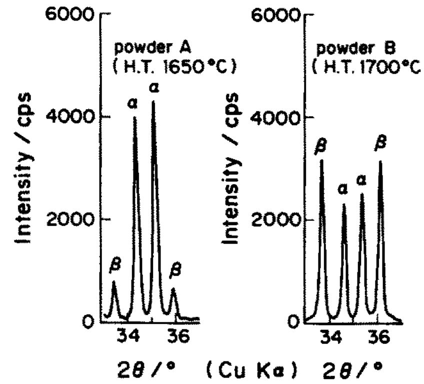

Fig. 2. X-Ray diffraction patterns of powder A and powder B. α, α-Si_3N_4; β, β-Si_3N_4.

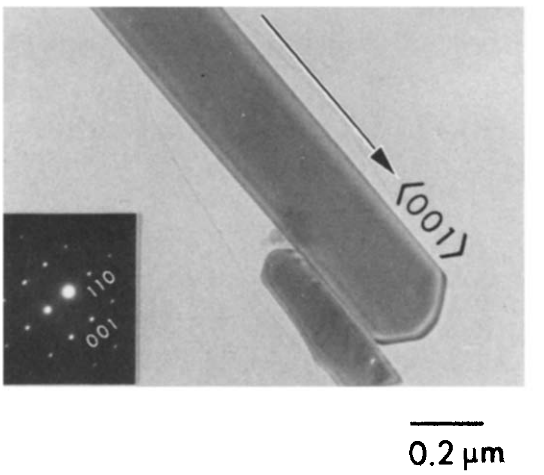

Fig. 3. TEM micrograph and electron diffraction pattern of a needle-like crystal.

Figure 4 shows the XRD results. The sintered blocks were examined by XRD in the two directions. One was perpendicular to the cold-pressing direction (shown by open symbols), and the other was parallel to it (shown by closed symbols). Because of the transformation of α- to β-Si_3N_4 during sintering, the crystal phase in the sintered bodies was the β-type. The X-ray intensity ratios of the β-Si_3N_4 (101)

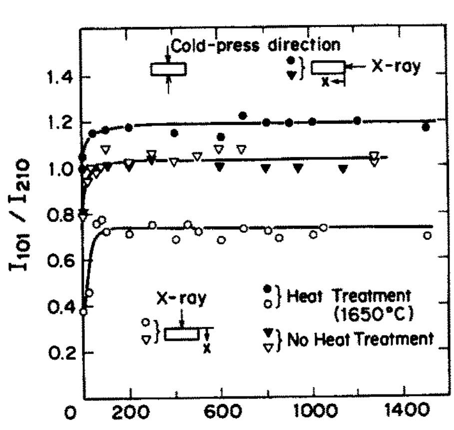

Fig. 4. X-Ray intensity ratio I_{101}/I_{210} as a function of distance x from the surface (sintering temperature 1830°C).

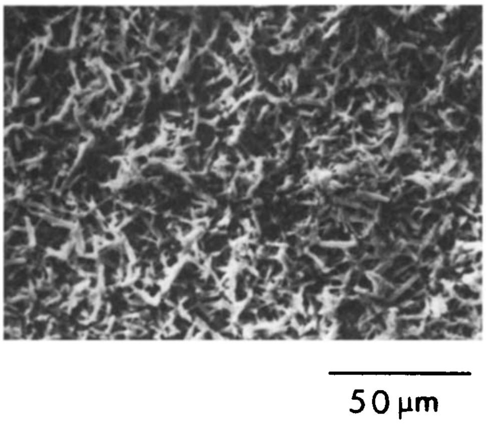

Fig. 5. SEM micrographs of sample surfaces.

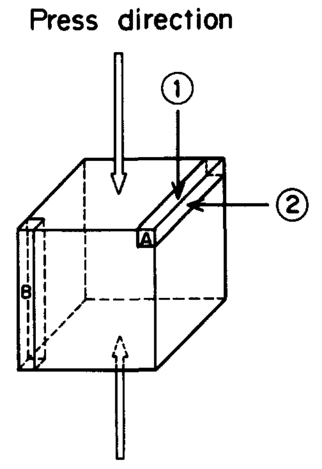

Fig. 6. Cutting and bending directions of test pieces.

planes and (210) planes (I_{101}/I_{210}) were measured at various depths from the sample surface. The X-ray intensity ratios of both directions were approximately 1·0 in the case without heat treatment. In random orientation, the X-ray intensity ratio is approximately 1·0. Thus, this sample had a random orientation of the β-Si_3N_4.

In contrast, in the samples prepared from the heat-treated powder which contained 7 wt% needle-like β-Si_3N_4, the X-ray intensity ratios were approximately 0·7 for the parallel direction and approximately 1·2 for the perpendicular direction. These results indicate that the c-axis of the β-Si_3N_4 is preferentially oriented perpendicular to the cold-pressing direction. It is thought that needle-like β-Si_3N_4 grains are already oriented in the powder compact and act as the nuclei for further β-Si_3N_4 grain growth during sintering.

The X-ray intensity ratio decreased sharply near the surface. The sample surfaces were accordingly observed by SEM (Fig. 5). Figure 5 indicates that a large number of elongated β-Si_3N_4 grains grew along the surface. This is the reason for the decrease of the X-ray intensity ratio near the surface. The β-Si_3N_4 elongated grain growth occurs through a liquid phase which is formed by the sintering additives. In the vicinity of the surfaces the liquid phase existed only along and behind the surfaces. Accordingly the proportion of the grains which grew along the surface increased in the vicinity of the surface.

3.2 Strength anisotropy

Figure 6 shows two kinds of test piece (A and B) and two kinds of bending direction (1 and 2) for the test piece A. Strength anisotropy due to preferred orientation in hot-pressed samples has been reported for test pieces A and B,[2,5] but the anisotropy for the bending directions 1 and 2 has not yet been reported. To make sure of this type of anisotropy, the three-point bending strength was measured for pressureless-sintered material with preferred orientation as already discussed and also for hot-pressed material. Figures 7 and 8 show the Weibull plots of bending strength at room temperature for pressureless-sintered samples and hot-pressed samples, respectively. The strength anisotropy by bending directions 1 and 2 was confirmed for

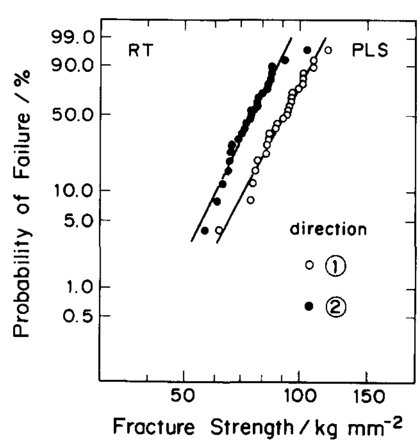

Fig. 7. Weibull plot of pressureless-sintered Si_3N_4.

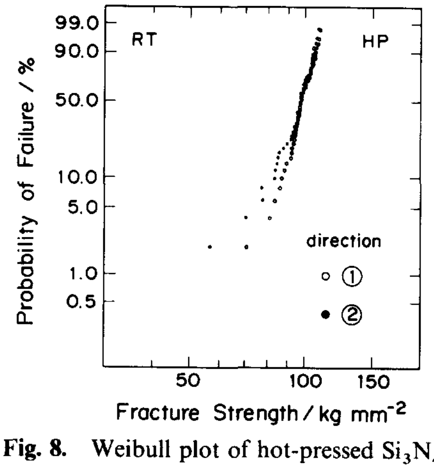

Fig. 8. Weibull plot of hot-pressed Si_3N_4.

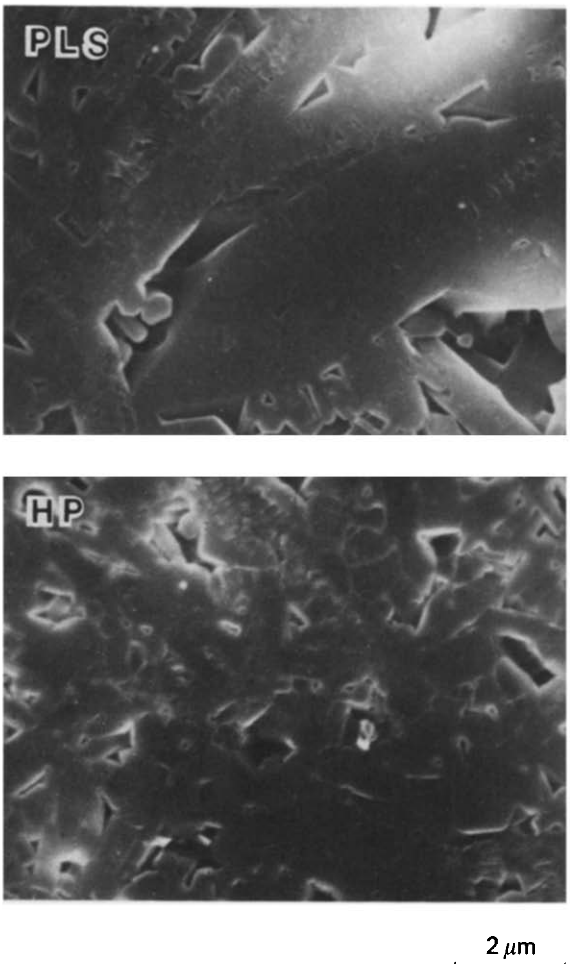

Fig. 9. Microstructure of Si_3N_4 ceramics. PLS, pressureless-sintering; HP, hot-pressing.

pressureless sintered material. However, for the hot-pressed material, the strength anisotropy was not confirmed, although a slight difference between the bending direction 1 and 2 was observed in the low strength area. According to XRD, the degree of preferred orientation for the hot-pressed sample was larger than that for the pressureless-sintered sample. The microstructures of both samples are shown in Fig. 9. The elongated β-Si_3N_4 grains of the pressureless-sintered sample are longer and larger in size than those of the hot-pressed sample. Therefore, it is thought that the strength anisotropy for bending direction 1 and 2 depends not only on the preferred orientation but also on the size of the β-Si_3N_4 elongated grains.

4 Conclusion

(1) The existence of preferred orientation for pressureless-sintered Si_3N_4 ceramics has been confirmed. This is caused by needle-like crystals and cold-pressing.

(2) Strength anisotropy as a function of bending direction has been confirmed. This type of anisotropy depends significantly on the grain size of the elongated β-Si_3N_4.

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