Aqueous Processing of Textured Silicon Nitride Ceramics by Slip Casting in a Strong Magnetic Field

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Abstract

This work will report a highly textured β-Si_3N_4 ceramic by aqueous slip casting in a magnetic field and subsequent pressureless sintering, Effects of the sintering aids, polymer dispersant, pH and stirring time on the stability of the Si_3N_4 slurries were studied. The textured β-Si_3N_4 with 97 % relative density could be obtained by slip casting in a magnetic field of 12 T and subsequent sintering at 1800 ^oC for 1 h. The textured microstructure is featured by the alignment of c-axis of β-Si_3N_4 crystals perpendicular to the magnetic field, and the Lotgering orientation factor, f, is determined to be 0.8.

Keywords : Silicon nitride, Textured ceramics, Magnetic field, Slip casting, Colloidal stability

1. Introduction

Recently, texture development has received particular interest in the processing of advanced Si_3N_4 ceramics because of significant improvement in the mechanical properties and thermal conductivity [1,2]. One common method of producing textured Si_3N_4 is to align a small portion of rodlike β-Si_3N_4 particles in fine powder matrix by tape casting or extrusion. Another method is the superplastic sinter-forging (SPSF) by superplastic deformation and alignment of rodlike β-grains along the extruding direction during sinter-forging. Recently, a novel processing has been successfully developed to produce textured ceramics by aligning and consolidating particles in "colloidal state" under a strong magnetic field, followed by normal sintering, despite the feeble magnetic property (e.g. Al_2O_3, TiO_2, HAP, etc.) [3]. Compared with TGG and SPSF, this novel processing offers the potential to produce versatile textured ceramic materials ranging from ceramic films to bulk parts, depending on the consolidation methods (e.g. slip casting, electrophoretic deposition (EPD), etc.). Si_3N_4 has a hexagonal structure and its single crystal is typically anisotropic, so this processing is suitable for producing textured Si_3N_4 ceramics. It has been reproted that textured β-Si_3N_4 ceramics could be obtained by this processing [4]. Therefore, this work was intended to develop textured β-Si_3N_4 ceramics by this processing, using commercial α-Si_3N_4 raw powder and a mixture of Y_2O_3 and Al_2O_3 as sintering aids. Emphasis was placed on the preparation of well-dispersed Si_3N_4-sintering aids suspensions, including effects of dispersant, pH, stirring time, and solid loading.

2. Experimental and Results

Raw powders used are: α-Si_3N_4 (SN-E10, Ube Industries, Ltd., Japan), Y_2O_3 and Al_2O_3. The starting composition is Si_3N_4:Y_2O_3:Al_2O_3 = 90:5:5 at molar ratio. The distilled water was used as dispersing medium and Polyethylenimine (PEI) (Mw = 10 000) was chosen as a dispersant. The slurries were prepared by mixing the dry powder and the solution with predetermined amounts of PEI, followed by ultrasonic treatment 5 min, then the slurries were kept stirring prior to stability evaluation and / or slip casting. The suspension pH was adjusted using HCl and KOH solutions. The electrokinetic measurements were conducted using an acoustic & electroacoustic spectrometer on the 5 vol% suspensions. The rheological measurements were conducted with a cone-type viscometer at 25 ^oC. The well-dispersed Si_3N_4-Y_2O_3-Al_2O_3 slurries were used for slip casting experiments without and with a magnetic field of 12 T, using glass ring set on the porous alumina blocks. The direction of magnetic field is parallel to that of slip casting (gravity). After drying and burning out polymer, the samples were sintered in a graphite-resistance furnace at 1800 ^oC-1 h-0.2 MPa N_2. The density was determined by the Archimedes method. The texture development was characterized by X-ray diffraction analysis.

As shown in Fig. 1, the lower viscosity is observed at pH ≥ 10 in the absence of PEI. Since the IEP of this Al_2O_3 was reported to be at pH ~ 8.6, the IEP of Y_2O_3 is most likely below pH 10; otherwise, the occurrence of hetero-coagulation possibly results in the higher viscosity at pH ≥ 10. In the presence of PEI, as the PEI amount increases from 0.75 to

1.5 dwb%, the pH value for achieving the minimum viscosity shifts from pH 9 to 10 within the pH range examined. This seems to suggest that the stabilization of the slurries with sintering aids is dominated by the electrosteric effect. Nevertheless, the stabilization of the slurries can not be obtained near the IEP (pH ~ 10.8). The reason is possibly due to the unsaturated adsorption.

Figure 2 shows that the slurries without PEI shows lower value in the minimum viscosity than those with PEI, but the former is more unstable than the latter with stirring time. Evidently, the slurry without PEI shows a large increase in the viscosity with time, particularly after ~ 10 h. The reason is mainly due to the accelerated dissolution of Y_2O_3 with decreasing pH to below pH 10. However, the slurry with PEI shows no change in the viscosity with time, indicating a better stability. This suggests that the adsorption of PEI play a positive role in preventing the Y_2O_3 particle from dissolving.

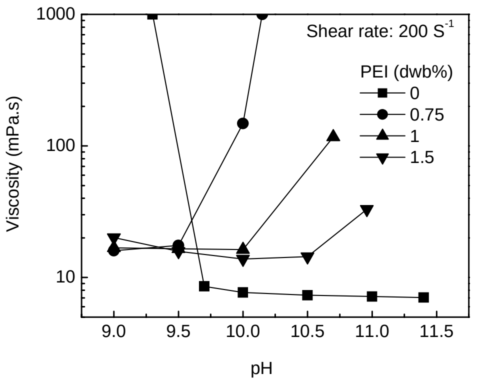

Fig. 1. Viscosity versus pH for 30 vol% Si_3N_4-Y_2O_3-Al_2O_3 slurries with PEI (dwb%: dry weight percentage of the Si_3N_4 powder basis).

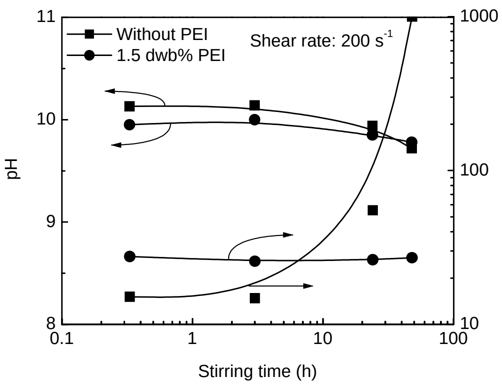

Fig. 2. Viscosity versus pH for 35 vol% Si_3N_4-Y_2O_3-Al_2O_3 slurries without and with 1.5 dwb% of PEI at pH 10.

As shown in Fig. 3, XRD reveals that the use of magnetic field results in the texture development in the sample, which shows strong intensities of (hk0) peaks, particularly (200) and (210) peaks, but weak intensities of (101) and (002) peaks at the top surface, and vice versa at the side surface. Note that all the samples were densified to the densities of ~ 3.22 g/cm^3, corresponding to 97 % theoretical density and the $\alpha \rightarrow \beta$ phase transformation is completed. This indicates the grain alignment occurs by the alignment of c-axis of β-Si_3N_4 crystals perpendicular to the magnetic field. The Lotgering orientation factor, f, is determined to be ≈ 0 and 0.8 for the samples prepared without and with a magnetic field, respectively [5].

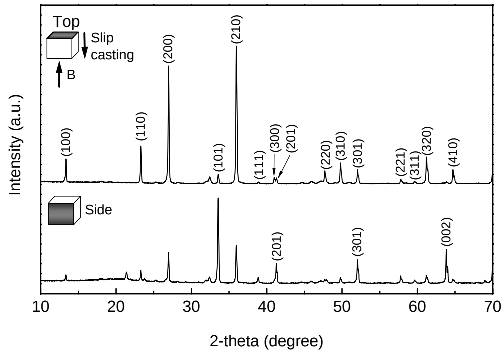

Fig. 3. XRD patterns of the sintered samples prepared by a magnetic field, using 30 vol% slurries with 1.5 dwb% PEI at pH 10.

3. Summary

It is shown that polyethylenimine (PEI) is an effective dispersant for stabilizing the Si_3N_4-Y_2O_3-Al_2O_3 slurries that does not show a time-dependent behavior at an optimum pH ≈ 10, compared to the case in the absence of PEI. Using the 30 vol% Si_3N_4-Y_2O_3-Al_2O_3 slurries stabilized with 1.5 dwb% PEI at pH 10, the highly textured β-Si_3N_4, showing 97 % relative density and Lotgering orientation factor (f) of 0.8, could be produced by slip casting a magnetic field of 12 T and subsequent sintering at 1800 ^oC for 1 h. The textured microstructure is featured by the alignment of *c*-axis of β-Si_3N_4 crystals perpendicular to the magnetic field.

4. References

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