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-Si₃N₄ 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 Si₃N₄ slurries were studied. The textured β-Si₃N₄ with 97% relative density could be obtained by slip casting in a magnetic field of 12 T and subsequent sintering at 1800 °C for 1 h. The textured microstructure is featured by the alignment of c-axis of β-Si₃N₄ 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 Si₃N₄ ceramics because of significant improvement in the mechanical properties and thermal conductivity [1,2]. One common method of producing textured Si₃N₄ is to align a small portion of rodlike β-Si₃N₄ 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. Al₂O₃, TiO₂, 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.). Si₃N₄ has a hexagonal structure and its single crystal is typically anisotropic, so this processing is suitable for producing textured Si₃N₄ ceramics. It has been reported that textured β-Si₃N₄ ceramics could be obtained by this processing [4]. Therefore, this work was intended to develop textured β-Si₃N₄ ceramics by this processing, using commercial α-Si₃N₄ raw powder and a mixture of Y₂O₃ and Al₂O₃ as sintering aids. Emphasis was placed on the preparation of well-dispersed Si₃N₄-sintering aids suspensions, including effects of dispersant, pH, stirring time, and solid loading.

2. Experimental and Results

Raw powders used are: α-Si₃N₄ (SN-E10, Ube Industries, Ltd., Japan), Y₂O₃ and Al₂O₃. The starting composition is Si₃N₄-Y₂O₃-Al₂O₃ = 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 normal sintering, despite the feeble magnetic property (e.g. Al₂O₃, TiO₂, 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.). Si₃N₄ has a hexagonal structure and its single crystal is typically anisotropic, so this processing is suitable for producing textured Si₃N₄ ceramics. It has been reported that textured β-Si₃N₄ ceramics could be obtained by this processing [4]. Therefore, this work was intended to develop textured β-Si₃N₄ ceramics by this processing, using commercial α-Si₃N₄ raw powder and a mixture of Y₂O₃ and Al₂O₃ as sintering aids. Emphasis was placed on the preparation of well-dispersed Si₃N₄-sintering aids suspensions, including effects of dispersant, pH, stirring time, and solid loading.

As shown in Fig. 1, the lower viscosity is observed at pH ≥ 10 in the absence of PEI. Since the IEP of this Al₂O₃ was reported to be at pH ~ 8.6, the IEP of Y₂O₃ 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 Y₂O₃ 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 Y₂O₃ particle from dissolving.

Fig. 1. Viscosity versus pH for 30 vol% Si₃N₄-Y₂O₃-Al₂O₃ slurries with PEI (dwb%: dry weight percentage of the Si₃N₄ powder basis).

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/cm³, corresponding to 97 % theoretical density and the α→β phase transformation is completed. This indicates the grain alignment occurs by the alignment of c-axis of β-Si₃N₄ 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 Si₃N₄-Y₂O₃-Al₂O₃ 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% Si₃N₄-Y₂O₃-Al₂O₃ slurries stabilized with 1.5 dwb% PEI at pH 10, the highly textured β-Si₃N₄, 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 °C for 1 h. The textured microstructure is featured by the alignment of c-axis of β-Si₃N₄ crystals perpendicular to the magnetic field.

4. References