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Short Communication

Feasibility of preparing of silicon nitride ceramics components by aqueous tape casting in combination with laminated object manufacturing

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ABSTRACT

Homogeneous green silicon nitride sheets consisting of 48.7 vol.% ceramic particles were prepared by tape casting. The slurry, used to prepared green tapes, own obvious shear-thinning behavior. No noticeable precipitation during tape casting process was found revealed by the result of energy dispersion spectrum analysis. Components with complex shapes were fabricated by stacking layer by layer of the green sheets and a subsequent pressureless sintering at 1800 °C for 1 h. All α-Si_3N_4 has transformed into β-Si_3N_4 indicated by the result of X-ray diffraction. Due to the high homogeneous packing of the ceramic particles in the green bodies, the ceramic components could be sintered to full density with no noticeable distortions. Good flexural strength for the ceramic samples was observed.

1. Introduction

Silicon nitride ceramics are known for their excellent mechanical, thermal and physiochemical properties [1,2]. However, there is one problem that limits the wide commercialization of the silicon nitride ceramics. The low sinterability and the difficulty in machining of the silicon nitride ceramics are essential obstacles to the production of silicon nitride components having complex shapes at a commercially acceptable cost [3,4]. Although ceramic processing techniques, such as slip casting, freeze casting and gel casting, have been applied for the preparation of silicon nitride components [5–7], dense devises with no distortion remains a technical challenge.

Recently, Al_2O_3, SiC and Al_2O_3/ZrO_2 ceramics with complex geometries or improved properties were prepared by tape casting [8–10]. Tape casting in combination with laminated object manufacturing (LOM) is an effective approach in preparing ceramic components with controllable sintering shrinkage, which is critical in the production of ceramic substrates for high integration circuit boards [11]. Klosterman and Chartoff [12] investigated the interfacial characteristics of the composites fabricated by LOM. Cui et al. [13] used tape casting and LOM process to fabricate Al_2O_3 ceramics. However, reports on the Si_3N_4 components fabricated by aqueous tape casting are very few.

Tape casting can provide high density green tapes for LOM [14]. Aqueous slurries for tape casting are of obvious advantages because the use of water instead of the organic solvents usually used in conventional tape castings lowers processing cost [15,16] and avoids the environment threatening potentials.

In the present work, Si_3N_4 samples with complex geometries were prepared through aqueous tape casting and a subsequent pressureless sintering. The properties of the green tape and its effects on the sinterability, microstructures and flexural strength of the silicon nitride components were investigated.

2. Experimental details

The composition design of the slurry is listed in Table 1. Low cost commercial powders of α-Si_3N_4, yttrium oxide and alumina were used as the starting materials. The powder mixture of yttrium oxide and alumina in a weight ratio of 3:5 was used as the sintering aid to the silicon nitride ceramic components. The total concentration of the sintering aid was 10 wt% based on the Si_3N_4. Polyacrylic acid (PAA), polyvinyl alcohol (PVA), glycerol and n-butyl alcohol was added to the slurry as a dispersant, binder, plasticizer and defoamer, respectively. The characteristics of the various starting materials are listed in Table 2.

At the first stage of powder processing, the dispersant and the ceramic powders were dissolved in water in a plastic bottle and mixed for 12 h to secure homogenous dispersion. Secondly, the binder and the plasticizer were added to the slurry and mixed for another 2 h. In the third step, the n-butyl alcohol was added

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Table 1
Compositions of the slurry.

Materials	Function	Weight percent (%)
Si_3N_4	Powder	49.8
Al_2O_3	Sintering aids	3.44
Y_2O_3	Sintering aids	2.06
Polyvinyl alcohol (PVA)	Binder	2.75
Polyacrylic acid (PAA)	Dispersant	0.33
Glycerol	Plasticizer	5.5
Water	Solvent	36.12
n-Butyl alcohol	Defoamer	–

Table 2
Characteristics of the major starting materials.

Materials	Manufacturer	Characteristics
Si_3N_4	Yinuo Nitride Co. Ltd. China	99%, 3–5 μm
Al_2O_3	Fuguang Co. Ltd. China	99%, 3–5 μm
Y_2O_3	Yinuo Nitride Co. Ltd. China	99%, 3–5 μm
Polyvinyl alcohol (PVA)	Zhongjia Co. Ltd. China	1450, Analytically pure
Polyacrylic acid (PAA)	Zhongjia Co. Ltd. China	30000, Analytically pure
Glycerol	Zhongjia Co. Ltd. China	Analytically pure
n-Butyl alcohol	Zhongjia Co. Ltd. China	Analytically pure

and the slurry was degassed for 0.5 h in a 0.1 Pa vacuum, ready for tape casting. Finally, the slurry was casted on a glass slab by a blade (LYJ, Beijing Dongfang Co. Ltd. China) at a speed of 0.2 m/min. Drying of the tape was conducted in open air at room temperature. Dry tape of 150 mm in thickness was prepared for the subsequent stacking and production of the various silicon nitride components.

The dry tape was cut and stacked layer by layer to form green bodies of various geometries. The dry tapes were stacked under 50 MPa with isostatic pressing in room temperature for 2 min. After the binder was burnt out at 650 °C in air with a 0.5 °C/min

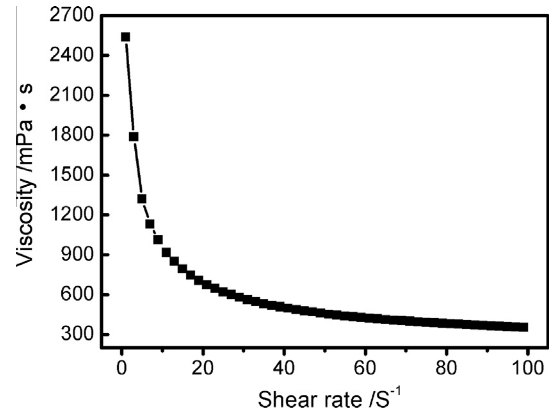

Fig. 1. Viscosity of the slurry changed as a function of the shear rate. The shear thinning behavior is desired for the aqueous tape casting.

heating rate, pressureless sintering of the ceramics was carried out in a Si_3N_4/BN powder bed at 1800 °C for 60 min under a nitrogen atmosphere of 50 Pa. The sintering was performed in a gas sintering furnace (SGM/VB/8-18, Shanxi Bohua Co. Ltd. China). Densification shrinkage of the samples was investigated by comparing the dimensions of the samples before and after sintering.

The viscosity of the slurry was measured by arotary viscometer (Model NDJ-7, Shanghai Balance Instrument Plant, Shanghai, China). Surface roughness of the dry tape was measured by laser confocal scanning microscopy (Zeiss LSM 5, Exciter, Germany). Bulk densities of the sintered ceramics were determined by the Archimedes principle. Relative densities were calculated.

Phase assembly of the sintered samples was checked by X-ray diffraction (XRD; Siemens, D5000, Japan) using Cu Kα radiation. Microstructure was observed on a scanning electron microscope (SEM; CamScan, Cambridge, UK) with an energy dispersive

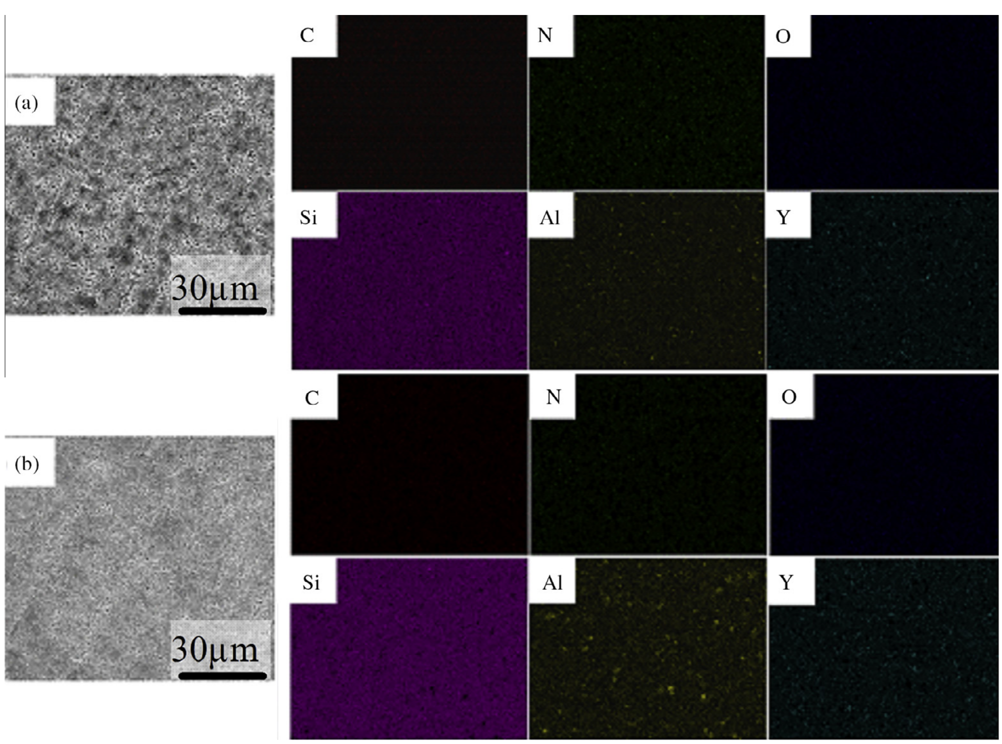

Fig. 2. Microstructure and EDS of the dry tape for (a) the top and (b) the bottom surface. The identical chemical composition suggests no noticeable differential precipitation of the ceramic particles during tape processing.