Environmentally benign processing of YAG transparent wafers

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Abstract:

Transparent yttrium aluminum garnet (YAG) wafers were successfully produced via aqueous tape casting and vacuum sintering techniques using a new environmentally friendly binder, a copolymer of isobutylene and maleic anhydride with the commercial name ISOBAM (noted as ISOBAM). Aqueous YAG slurries were mixed by ball-milling, which was followed by de-gassing and tape casting of wafers. The final YAG green tapes were homogenous and flexible, and could be bent freely without cracking. After the drying and sintering processes, transparent YAG wafers were achieved. The microstructures of both the green tape and vacuum-sintered YAG ceramic were observed by scanning electronic microscopy (SEM). Phase compositions were examined by X-ray diffraction (XRD). Optical transmittance was measured in UV-VIS regions with the result that the transmittance is 82.6% at a wavelength of 800nm.

Keywords: transparent, ceramic wafer, yttrium aluminum garnet, tape casting, rheological property

1 Introduction

With the increasing use of flat, thin and large scale ceramics for fabrication of substrates, packaging and multilayer technology[1-3] (capacitors[4], sensors[5], piezoactuator[6], laser composites[7], etc.), tape casting techniques have been very widely preferred over conventional ceramic processing techniques such as dry pressing, slip casting, and gel casting. Flat ceramic wafers with thicknesses ranging from a few microns to a few millimeters can be produced precisely by tape casting techniques.

Traditionally, tape casting is performed using organic solvents as liquid medium [4, 7-13], taking advantage of low boiling point, easy evaporation during the drying step, good wetting properties and

inertness to ceramic powders. However, due to environmental and health considerations, as well as high flammability and volatility of organic solvents, coupled with their high production cost, more research on water-based systems has developed in recent years. However, water-based systems are more sensitive than organic solvent systems to processing parameters, such as casting composition, drying conditions, and film thickness[14]. Issues such as the required use of slower drying rates, easy foaming and premature polymerization[15], require that more work on the aqueous tape casting techniques be done.

The typical tape casting system consists of well-dispersed suspension with water soluble dispersant, binder and plasticizer. During the tape casting process, particle size distribution, dispersion stability, and sedimentation play important roles on the quality of the final product in terms of the presence of cracks and deformation[16, 17]. Dispersant must be added to prevent micron/submicron size ceramic powders from agglomerating, a result of powders' electrostatic forces and high surface area/mass ratio[18]. Moreover, many binders have been evaluated for aqueous tape casting techniques, such as acrylic polymers[19, 20], cellulose ethers[21, 22], polyvinyl alcohols [23], latex emulsions[24], gelatin[25], etc. Interestingly, Reddy et al. reported the dispersant effect of binders in tape cast slurries[26, 27], which indicated the possibility of simplifying the tape casting process by removing the necessity of dispersant. ISOBAM is one binder which has been recently reported as a spontaneous gelling agent which also acts as an effective dispersant in aqueous systems [28-33].

ISOBAM is a water soluble copolymer (M.W. = 160 000~300 000) of isobutylene and maleic anhydride, with the appearance of white powder, density 1.3g/cm^3 ; which has application as a binder for ceramic processing techniques. In this work ISOBAM was adopted as both binder and dispersant for aqueous tape casting processes. After the ball milling and de-gassing processes, the slurry was casted on a plastic carrier. After debinding and vacuum sintering, transparent yttrium aluminum garnet (YAG) chips were finally achieved.

2 Experimental

2.1 YAG slurries preparation

High-purity powders of α - Al_2O_3 (Baikowski, Annecy, France), Y_2O_3 (99.99%, Sigma-Aldrich Corp, USA), and ISOBAM (Kuraray, Japan) were used as raw materials.

YAG slurries with solids loading of 63.5wt% were mixed by ball-milling with zirconia grinding media at a speed of 200-300 rpm for 3 hours. After the addition of plasticizer, the slurry was casted on a stationary Mylar carrier with a moving doctor blade at a speed of 10-35cm/min. After drying in air for 24 hours at room temperature, the green tapes were without deformation or cracking. After demolding and drying, the ceramic green tapes were debindered at 800-1000°C for 3 hours, at a ramp rate of 2-5°C/min. Final sintering was performed in a vacuum atmosphere (10^{-5} - 10^{-6} MPa) at a temperature of 1700-1800°C for 3 hours, at a ramp rate of 2~10°C/min.

2.2 Characterization for the slurries and ceramic wafers

The rheological properties of the produced slurries were measured with a rotational control stress rheometer (TA Instrument AR2000 Rheometer, USA) in continuous shear mode in the region of 0.01s^{-1} - 1000s^{-1} , at 25°C. The YAG green tapes were very flexible and could be bent and twisted freely. The surface and cross-sectional microstructure of the YAG green tapes and vacuum-sintered wafers were examined via scanning electron microscopy (SEM, Quanta 200F, FEI, USA). Phase compositions of YAG green body and vacuum-sintered transparent wafers were analyzed by X-ray diffraction (XRD, Bruker D2 Phaser, Germany) equipped with a Cu radiation ($\lambda = 0.154$ nm) in the range of $2\theta = 10\sim 75^\circ$. The in-line transmittance of the wafers was measured in the UV-VIS region using UV-VIS spectrometry (Perkin Elmer Lambda 950, Massachusetts, USA).

3 Results and discussion

3.1 Rheological property

The rheological behavior of the tape casting slurries was measured over shear rate range of $0.1\text{-}1000\text{s}^{-1}$, the results of which are shown in Fig. 1(a). The measured YAG slurries displayed shear thinning behavior over the entire measured region, which is desirable for tape casting process. High viscosities were achieved at low shear rates, which could prevent particle flocculation and sedimentation. At high shear rate, the measured viscosities were relatively low, which is desirable in the actual forming step of tape casting. For example, the viscosity is less than $10\text{ Pa}\cdot\text{s}$ at high shear rate ($>20\text{s}^{-1}$), which is useful to achieve homogeneous dispersion of the slurry system, and dimensional accuracy.

The shear thinning behavior of ceramic based suspensions used in tape casting could also be described by the equation below[34]:

$$\tau = k (r)^n$$

Where τ is the shear stress, r is the shear rate and n is a value unique to the material system which is less than one. Fig. 1 (b) shows the plot of the slip shear stress as a function of shear rate for YAG slip system. The value of n can be calculated from the slope of curves in Fig. 1(b), which is less than one, consistent with Eq.(1). Hence, the shear stress curve verified the shear thinning behavior as expected.

3.2 Tape casting of the YAG green wafer

In the tape casting step, the casting was performed at a speed of $10\text{-}35\text{ cm/min}$, similar to speeds reported in the literature for other water-based systems [14, 22, 24]. Without the addition of surfactant, all slurries wetted the Mylar carrier well and were “locked” onto the carrier to prevent the slip from receding and forming droplets. After drying for 2 hours on the Mylar carrier, the green tapes (Fig. 2, (a)) were easy to remove from the carrier film. No cracks, blisters or pinholes were observed in the green tape, and all had smooth surfaces. In addition, the tapes were smooth, flexible, and easy to handle (Fig.2 (b)). In conclusion, the binder ISOBAM can be used for aqueous tape casting systems to produce good quality tapes.

3.3 Microstructure research of green body and vacuum sintered YAG wafer

Shown in Figure 3 are the fracture surfaces of YAG tape after debinding (Fig. 3(a)) and vacuum sintering (Fig. 3(b)). The particles in the fracture surfaces of the debinded YAG green tapes are distributed homogeneously without any agglomerates. However, there is neck-formation after the debinding process (Fig. 3 (a)). As shown in Figure 3 (b), the sintered YAG wafers show good optical properties and fully densified microstructure. The grains densified uniformly with grain size distribution of 1.5-5 μ m, which was statistically calculated by Nano Measurer software. No differences in microstructure were observable between the casting and traverse direction. The fracture mode of the vacuum sintered YAG wafers appeared to be mainly transgranular for large grains, as well as minor intergranular for small grains within the grain size range of 1-3 μ m. There were no obvious bubbles in the fracture surface of the sintered YAG ceramic. According to Archimedes' measurements, the relative densities of the green body and vacuum sintered body were measured with the results ~54.5% for the green body and 99.5% for the vacuum-sintered sample.

3.4 Phase identification

Figure 4 shows the phase compositions of the tape-casted green body after the debinding in air at 800-1000 $^{\circ}$ C for 3 hours, and the YAG wafers sintered at 1700-1800 $^{\circ}$ C for 3 hours. Kinsman *et al.*[35] reported that three phases, including YAM ($Y_4Al_2O_9$), YAP ($YAlO_3$), YAG ($Y_3Al_5O_{12}$), could be formed during the different temperature region for the Al_2O_3 - Y_2O_3 system.

In Fig.3, after the debinding at 800-1000 $^{\circ}$ C for 3 hours, YAM was observed to exist with unreacted Y_2O_3 and Al_2O_3 . After a vacuum sintering at 1700-1800 $^{\circ}$ C, YAG is the only phase detected, indicating a complete reaction between Y_2O_3 and Al_2O_3 .

3.5 Optical transmittance of vacuum-sintered YAG wafer

A photo of a vacuum sintered sample with a thickness of 0.26mm is shown in the inset of Figure 5. The YAG chips also display excellent optical properties, and the in-line transmittance was measured to be about 82.6% at a wavelength of 800nm. These results verify that the ISOBAM tape casting system could be successfully used for the production of thin tapes with excellent preservation of optical properties.

4 Conclusion

Transparent YAG wafers were successfully processed by tape casting with ISOBAM as binder and the vacuum-sintering technique. Slurries produced with ISOBAM tape casting system show shear thinning behavior in the region of $0.1\text{-}1000\text{ s}^{-1}$. The final green tape is very flat, smooth, flexible, and easy to handle. The microstructure of the green tape is homogeneous without any observable agglomeration. The vacuum sintered YAG ceramic is uniform in both the casting and lateral direction with a grain size distribution of $1.5\text{-}5\mu\text{m}$ and transmittance of 82.6% at the wavelength of 800nm.

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Figure captions

Fig. 1 Rheology property as a function of shear rate (a) viscosity; (b) shear stress

Fig. 2 Photographs of the YAG green tape after demolding at room temperature

Fig. 3 Microstructure of fracture surfaces of YAG tape after (a) debinding at 800-1000°C for 3 hours and (b) vacuum sintering at 1700-1800°C for 3 hours

Fig. 4 X-ray diffraction pattern of YAG green body (800-1000°C, 3 hours) and vacuum-sintered wafer (1700-1800°C, 3 hours)

Fig. 5 Transmittance of vacuum-sintered (1700-1800°C, 3h) YAG chips formed by aqueous tape casting methods using ISOBAM as binder.

FIGURES

figure 1

figure 2

figure 3

figure 4

figure 5