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Short communication

Transparent $Y_3Al_5O_{12}$ ceramics produced by an aqueous tape casting method

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Abstract

Composite structure $Y_3Al_5O_{12}$ transparent ceramics were prepared by an aqueous tape casting method. The slurry of Al_2O_3 and Y_2O_3 powders shows a shear thinning behavior. The powders were distributed homogeneously in the tapes. The average grain size of YAG ceramics sintered at 1750 °C for 10 h is 15 μm. The inline transmittance of the sample polished to 1.0 mm is 81.5% at the wavelength of 1064 nm.

Keywords: A. Tape casting; $Y_3Al_5O_{12}$; Transparent ceramics; Solid state reaction

1. Introduction

Transparent polycrystalline ceramics are widely used in applications such as solid-state lasers [1–3], scintillators [4], transparent armors [5] and high intensity discharge lamps [6]. $Y_3Al_5O_{12}$ (YAG) ceramics have excellent thermal and mechanical properties with a high melting point (1970 °C), cubic crystal structure and isotropic properties. The most important application of YAG ceramics is laser material.

In 1995, Ikesue prepared the first Nd:YAG transparent ceramics by the solid state reaction method [7]. Yanagitani's group developed a co-precipitation method with vacuum sintering and they obtained high quality YAG ceramics [8,9]. The two methods mentioned above are the main techniques for producing YAG ceramics. As we all know, there will be a great quantity of heat generation when the laser materials are working. The unwelcome consequences are heat stress, heat lens and the thermally induced birefringence effect. The heat effects can degrade the laser beam quality and the working life of laser materials [10–12]. Composite laser ceramics were designed and several configurations were developed in order to decrease the heat effects [13–15]. Among these configurations, gradient concentration structure is believed to be an outstanding solution route. Tape casting is an important technology in electronic industry. The single tape can be controlled at the micrometer level by adjusting the gap of the blade. Messing et al. and Tang et al. have produced Er:YAG and Yb:YAG transparent ceramics by the non-aqueous tape casting method and they both achieved laser output [16–18]. In consideration of the health problems of the workers and environmental protection, aqueous tape casting has been paid great attention and it has substituted for non-aqueous tape casting in many fields [19,20].

2. Experimental procedure

In the present research, we developed an aqueous tape casting and vacuum sintering method to prepare YAG transparent ceramics. This work is the basis for developing rare earth ions doped composite structure YAG ceramics. Yttrium oxide (Y_2O_3, 99.99%, Alfa Aesar), alumina

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(Al_2O_3, 99.99%, Alfa Aesar), tetraethyl orthosilicate (TEOS, 99.99%, Alfa Aesar), magnesium oxide (MgO, 99.99%, Alfa Aesar), polyacrylic acid (PAA, 25 wt%, Alfa Aesar), polyvinyl alcohol (PVA-124, Sinopharm Chemical Reagent Co., Ltd.), polyethylene glycol (PEG-400, AR, Sinopharm Chemical Reagent Co., Ltd.) were used as starting materials. The Y_2O_3 powders and Al_2O_3 powders were weighed according to the stoichiometry of $Y_3Al_5O_{12}$. TEOS and MgO were used as the sintering aids. The powder mixture was ball milled with the dispersant (PAA) and deionized water for at least 8 h. Then the binder (PVA) and the plasticizer (PEG-400) were added. The new slurry was ball milled for 24 h. The slurry was sieved and de-aired by using a vacuum system. The gap height of the blade was controlled at 300–500 μm. The casting speed is 10 mm/s. The tapes were left to dry at room temperature for 36 h. By using a mold with round shape, the dried tapes were cut into dozens of pieces. The tapes were packed for 30 layers. During this process, air between the tapes was eliminated from the bodies by high pressure. The binder was removed at 600 °C for 48 h in order to exclude the residual carbon (the byproduct of the decomposition of the organic compounds). The green bodies were cold isostatic pressed at 200 MPa for 2 min to obtain homogeneous bodies. And then the green bodies were sintered by a solid state reaction method in vacuum circumstance. The sintering temperature and the holding time were 1750 °C and 10 h, respectively. The pressure during sintering was below 1×10^{-3} Pa.

The rheological properties of the slurries were carried out using a shear controlled rheometer (Physica MCR301, Anton Paar, Germany). The microstructures of the tapes and the fracture morphologies of the green bodies were observed by using a field emission scanning electron microscopy (FESEM, Hitachi S-4800, Japan). The phase analysis was detected by a powder X-ray diffractionmeter (XRD, D8 Advanced, Cu Kα, Bruker, Germany). The microstructure characterizations of the YAG ceramics were performed by using an electron probe micro-analyzer (EPMA, JSM-6700 F, JEOL, Japan). The element distribution was detected by energy dispersive X-ray spectroscopy (EDS, Oxford, UK). The inline transmittances of the YAG samples were measured by an UV–vis–NIR spectrophotometer (Cary 5000, Varian, USA).

3. Results and discussion

Fig. 1(a) is the rheological curves of the slurry prepared from the mixture of Al_2O_3 and Y_2O_3 powders. From the relationship between shear stress and shear rate, it can be seen that the slurry exhibits shear thinning behavior. The shear stress begins from 0 Pa. The value of shear stress increases with the shear rate while the viscosity decreases. Both the shear stress curves and the viscosity curves show the existence of the thixotropic loops. The loop is an important characteristic for pseudoplastic fluid of tape casting slurry.

Fig. 1(b) represents the surface of the as synthesized tape of the oxides mixture by the aqueous tape casting method. It can be seen that there are two types of particles with different morphology characteristics in the photographs. The larger particles are Y_2O_3 powders with the particle size distribution from 1 μm to 3 μm. The finer particles are Al_2O_3 powders with the particle size distribution from 200 nm to 600 nm. The morphology of Y_2O_3 powders is irregular or flake shape. But Al_2O_3 powders are near spherical. These spheres are the aggregates of the fine Al_2O_3 powders whose particle sizes are about 100 nm. The photographs indicate that the starting powders distribute homogeneously in the tapes.

Fig. 2(a) shows the tape, single slices of the tape, stacked slices with 30 layers and pressed slices with 30 layers, respectively. The thickness of the tapes were controlled at 100 μm. The diameter of the slice is designed at 20 mm. Fig. 2(b) shows the green body and the sintered YAG transparent ceramics. The green body underwent the binder removal and cold isostatic pressing procedures. As a result, the diameter shrunk from 20 mm to about 18 mm. During the sintering process, shrinkage occurred again and the diameter decreased from 18 mm to about 15 mm. Composite structure YAG ceramics with the thickness of 1.0 mm have high optical quality. The words behind the ceramics can be seen clearly.

Fig. 2(c) is the fracture morphology of the green body. It can be seen that the body has a dense structure. All of the slices cut from the tape were pressed closely and no gap was found across the fracture surface. Fig. 2(d) is the fracture morphology of the polished surface of YAG ceramics. The ceramics are almost full dense body and

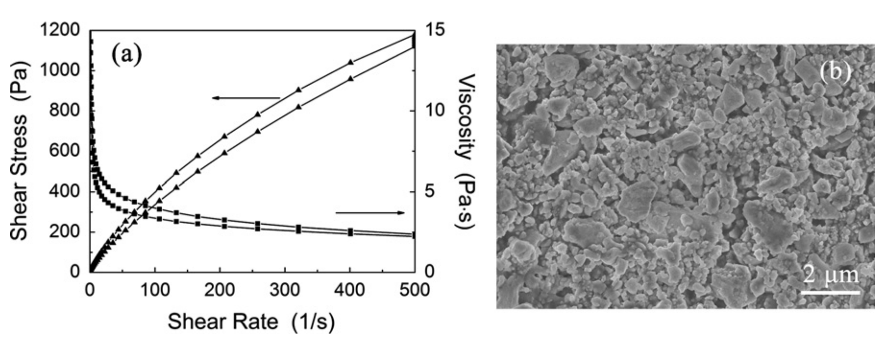

Fig. 1. (a) Rheological properties of the slurry and (b) SEM image of the tape.

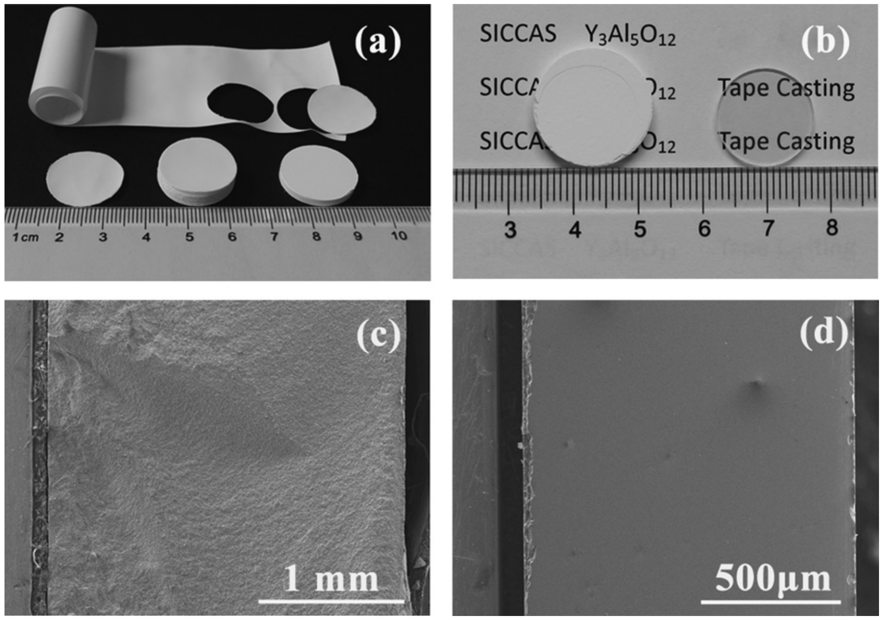

Fig. 2. (a) Photographs of the tape, as-cut slice, stacked slices and as-pressed slices; (b) Photographs of green body and sintered YAG composite structure ceramics; (c) Fracture morphology of the green body and (d) EPMA picture of the fracture morphology of YAG ceramics.

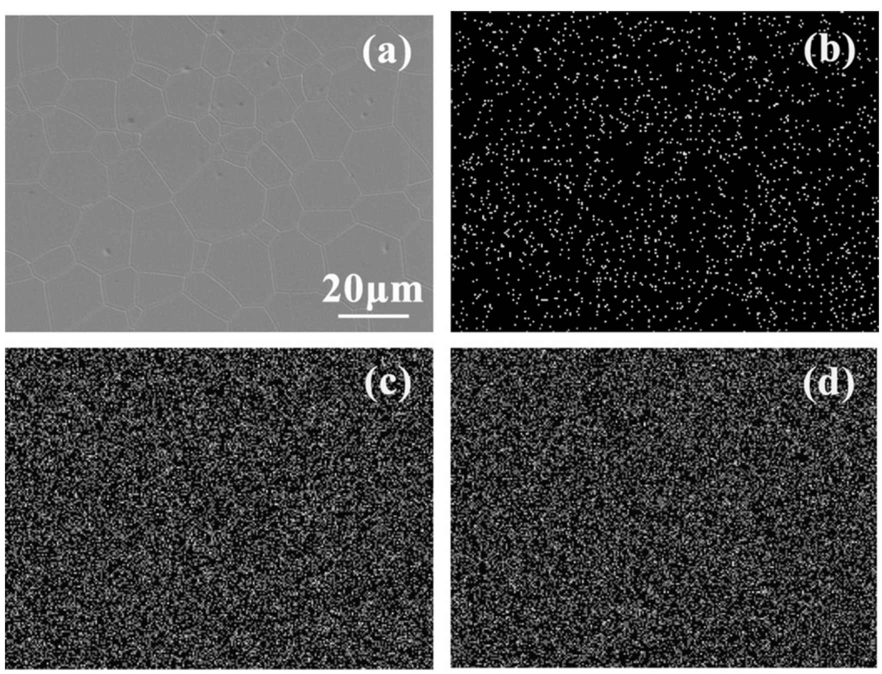

Fig. 3. Microstructure and element area scanning figures of the as-sintered YAG ceramics.

there is no crack on the surface. The results indicate that there is no delamination occurred during the binder removal process and the sintering process.

Fig. 3(a) is the EPMA image of the as-synthesized YAG composite structure ceramics. The YAG fine grains grow with each other closely and no secondary phase was found. There are some pores in the grains. The sizes of the pores are less than 1 μm. The grain size was determined with the EPMA by freely drawing a line on a high resolution image of the sample. The average grain size was calculated by the formula below [21,22]:

$$D = \frac{1.56 \times L}{N \times M} \tag{1}$$

In the formula, D is the average grain size, N is the number of grains on the line, L is the length of the line, M is the magnification. The calculated average grain size of the YAG composite structure ceramics are about 15 μm.

Fig. 3(b), (c) and (d) are the area scanning figures for O, Al and Y, respectively. The elements have homogeneous

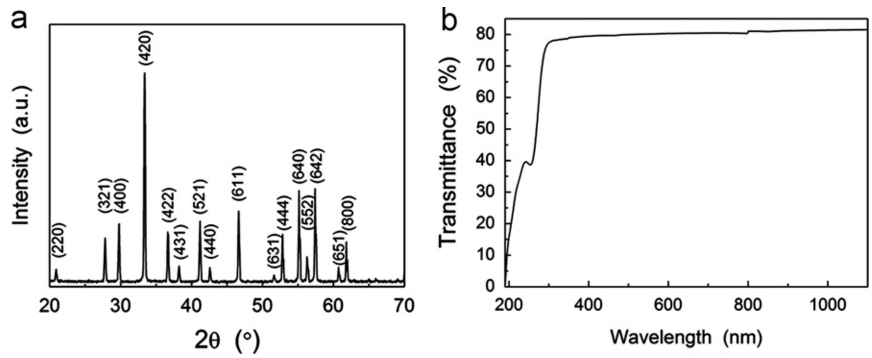

Fig. 4. (a) XRD pattern of YAG ceramics sample and (b) Transmittance curve of YAG composite structure ceramics.

distribution in all of the pictures. The results indicate that the as-sintered YAG ceramics achieved a homogeneous microstructure. The homogeneous microstructure is an evidence that the ceramic is of high quality.

Fig. 4(a) is the X-ray diffraction pattern of the $Y_3Al_5O_{12}$ composite structure ceramics sintered at 1750 °C for 10 h. The peaks in the pattern match well with yttrium alumina garnet ($Y_3Al_5O_{12}$, JCPDS 88-2048). This result indicates that the composition of the ceramics is $Y_3Al_5O_{12}$ with garnet structure and no secondary phase was found.

Fig. 4(b) is the inline transmittance curve of the YAG composite structure ceramics polished to 1.0 mm thick. High inline transmittance was obtained from 300 nm to 1100 nm. The transmittance at 400 nm is 79.5%. And the transmittance at 1064 nm (the laser emitting wavelength) is 81.5%. The peak at 253 nm in the curve can be attributed to the absorption of the Fe^{3+} ions [23]. The absorption band was believed to be associated with the oxygen ion charge transfer band due to the accidental introduction of a very low concentration of iron [24,25]. The impurity including Fe^{3+} ions should be introduced from the starting materials or the pressing process because the mould material is steel.

The results show that the aqueous tape casting method is a promising route to prepare transparent YAG composite structure ceramics. The quality of the ceramics can be improved by adjusting the technology and optimizing the parameters. The most important role of this method is that complex structure laser ceramics and ceramics with gradient composition can be obtained if different doping compounds are designed and added in the ceramics. The heat distribution among the laser ceramics would be arranged more homogeneously. As a result, the laser quality would be improved significantly.

4. Conclusions

In summary, the aqueous tape casting method was used to prepare YAG composite structure ceramics with high optical quality. The average grain size of the YAG ceramics sintered at 1750 °C for 10 h is about 15 μm. The inline transmittance of the sample is 81.5% at 1064 nm. The results indicate that the aqueous tape casting method is a promising technology for preparing transparent ceramics. By using this technology, laser ceramics with composite structures can be obtained. Recently, Nd:YAG and Yb:YAG composite ceramics are being developed in our group. The information of the new works will be reported soon.

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