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Invited Article

Highly transparent $Tb_3Al_5O_{12}$ magneto-optical ceramics sintered from co-precipitated powders with sintering aids

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ABSTRACT

Highly transparent terbium aluminum garnet ($Tb_3Al_5O_{12}$, TAG) magneto-optical ceramics were fabricated from co-precipitated nanopowders with tetraethoxysilane (TEOS) as sintering aid by vacuum sintering combined with hot isostatic pressing (HIP) post-treatment. The ball milled TAG powder shows better dispersity than the as-synthesized powder, and its average particle size is about 80 nm. For the ceramic sample pre-sintered at 1720 °C for 20 h with HIP post-treated at 1700 °C for 3 h, the in-line transmittance exceeds 76% in the region of 400–1580nm (except the absorption band), reaching a maximum value of 81.8% at the wavelength of 1390 nm. The microstructure of the TAG ceramic is homogeneous and its average grain size is approximately 19.7 μm. The Verdet constant of the sample is calculated to be $-182.7\ rad\cdot T^{-1}\cdot m^{-1}$ at room temperature.

1. Introduction

Faraday isolators are fundamental optical apparatuses of high-power laser systems as they can impede the unwanted feedback and guarantee the stability of the system [1–3]. As the core part of Faraday isolators, magneto-optical materials are usually garnets since they combine the advantages of excellent optical quality, good thermal-optical property and high Verdet constant [4–7]. Among them, terbium aluminum garnet ($Tb_3Al_5O_{12}$, TAG) is considered as a promising magneto-optical material applied in the visible and near-infrared spectral regions due to its higher Verdet constant than that of the commercial terbium gallium garnet ($Tb_3Ga_5O_{12}$, TGG) single crystal [8–10]. However, it's hard to grow TAG single crystal by the Czochralski method due to its incongruent melting nature [11]. Many approaches have been proposed to solve this problem, for example, some researchers utilized Tm^{3+} [12], Lu^{3+} [13], Yb^{3+} [14], Ga^{3+} [15], and Sc^{3+} [16] ions to partially substitute Tb^{3+} or Al^{3+} ions, which was aimed to obtain a stable TAG phase. In addition, many new techniques such as micro-pulling-down method [17] and laser floating zone method [18,19] have been applied to growing TAG single crystal. But still, the size of TAG single crystal is limited, which cannot meet the demands of optical devices.

In recent years, TAG transparent ceramics which can be manufactured with large size have become a preferable choice [20–24]. The advantages of TAG transparent ceramics are obvious. Firstly, it's not difficult to obtain a pure TAG phase as the fabrication process of ceramics is free from melting. In addition, with the development of transparent ceramic technology, TAG transparent ceramics can achieve an even better optical quality than single crystal [25], which is essential for their application in high-power optical isolators. In our previous work, we have successfully fabricated TAG transparent ceramics using the co-precipitated nanopowders for the first time [26]. The influence of ammonium hydrogen carbonate to metal ions molar ratio on the properties of TAG nanopowders and the resultant ceramics were also investigated [27]. However, the optical quality of the TAG transparent ceramics sintered from the co-precipitated nanopowders is not good enough for practical application. As is well known, the optical properties of transparent ceramics are sensitive to the agglomeration degree of raw powders, and the existence of agglomeration of powders will generate pores that are difficult to eliminate during the sintering. Many works have demonstrated that ball milling can effectively reduce the agglomeration degree of the raw powders and finally reduce the porosity of the obtained transparent ceramics [28–30]. In addition, selecting appropriate sintering aids is also important for the

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fabrication of transparent ceramics since they can promote the densification process and enhance the transparency of ceramics [31,32].

In this work, highly transparent $Tb_3Al_5O_{12}$ magneto-optical ceramics were fabricated using co-precipitated powders with TEOS as sintering aid. The morphologies of TAG powders before and after ball milling were compared. The influence of ball milling and sintering aids on the transparency and microstructure of TAG ceramics was also investigated.

2. Experimental

TAG nanopowders were synthesized by the co-precipitation method and the experimental details have been reported in our previous work [26]. Then the 1100 °C-calcined TAG powders were ball-milled in anhydrous ethanol for 12 h with 0.5 wt% tetraethoxysilane (TEOS, >99.999%, Alfa Aesar, Tianjin, China) as sintering aid. For comparison, TAG powders were also ball-milled without sintering aids for 12 h. After ball milling, the slurry was dried at 70 °C for 2 h, sieved through a 200-mesh screen and then calcined at 600 °C for 4 h to remove the organic ingredients. The as-prepared powders were then uniaxially pressed into pellets at low pressure followed by cold isostatically pressing at 250 MPa. The green bodies were vacuum sintered at 1720 °C for 20 h and further hot isostatic pressing (HIP) sintered at 1700 °C for 3 h under 200 MPa Ar atmosphere.

Dynamic light scattering (DLS) of the powders was performed using a ZetaPlus potentiometric analyzer (Brookhaven Instruments Corporation, USA). The measurement was conducted at 90° at a wavelength of 658 nm under the following conditions: particle refractive index 1.59, water refractive index 1.33, viscosity 0.89 Cp and at temperature of 25 °C. Morphologies of the powders and microstructures of the thermal-etched (1400 °C for 3 h) surfaces of mirror-polished ceramics were observed by a field emission scanning electron microscopy (FESEM, S-4800, Hitachi, Japan). Grain size of the ceramic sample was calculated by the linear intercept method. Specific surface areas (S_{BET}) of the powders were measured by Norcross ASAP 2010 micromeritics with N2 as the absorption gas. The ceramics were mirror-polished to 1.5 mm thickness for in-line transmittance measurement, and the measurement was conducted by a UV-VIS-NIR spectrophotometer (Model Cary-5000, Varian, USA). The Verdet constant was measured by the extinction method with an instrument consisting of a He–Ne laser, an electromagnet and two polarizers. The measurement was carried out at room temperature.

3. Results and discussion

Fig. 1 shows the FESEM micrographs of the as-synthesized and the ball milled TAG powders. It can be observed that the as-synthesized TAG powders are slightly agglomerated with dumbbell shaped particles and the average particle size is ~88 nm. After ball milling, the agglomerated and coarse particles are ground into smaller and approximately spherical particles with an average particle size of ~80 nm. The specific surface areas of TAG powders before and after ball milling are 10.8 and 12.1 m^2/g, respectively. The increase of S_{BET} of TAG powders indicates the improved dispersity of the powders, which is beneficial to the elimination of residual pores during sintering.

To better reveal the differences in agglomeration degree of the as-synthesized and the ball milled TAG powders, the dynamic light scattering (DLS) of both nanopowders was conducted, as shown in Fig. 2. It can be seen that the as-synthesized TAG powders exhibit a bimodal size distribution with the effective diameter of 499.9 nm. For the ball milled powder, it shows a relative narrow size

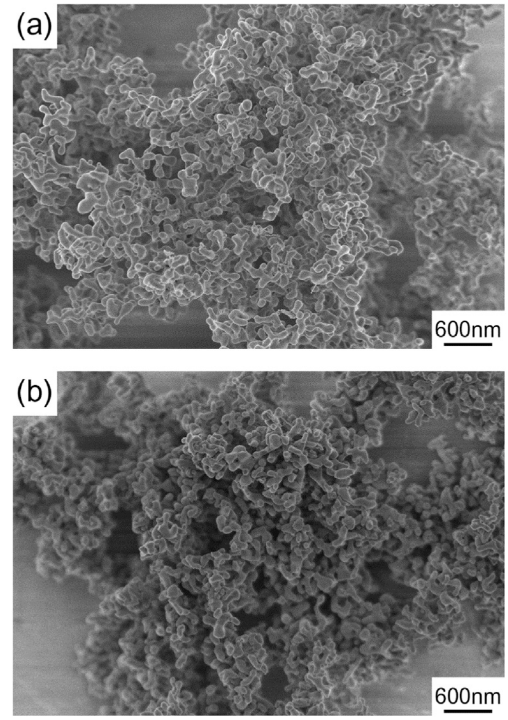

Fig. 1. FESEM micrographs of the (a) as-synthesized and (b) ball milled TAG powders.

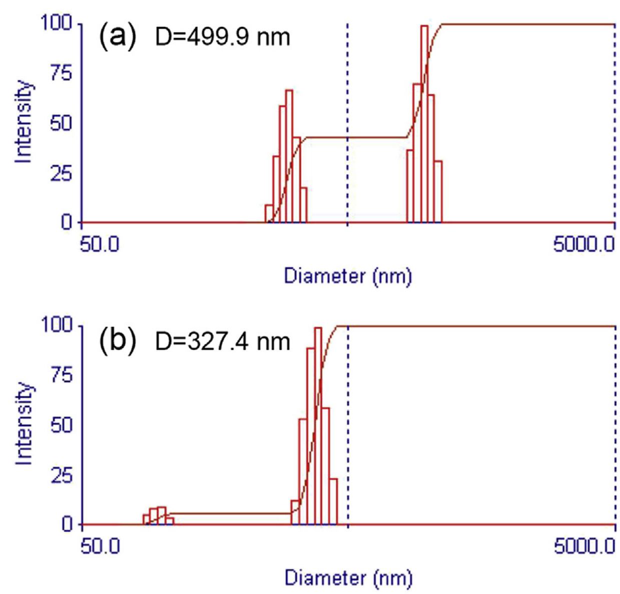

Fig. 2. Multimodal size distribution of (a) as-synthesized and (b) ball milled TAG powders.

distribution and the majority of the particles exist at 383.1 nm. The effective diameter of the ball milled TAG powders is 327.4 nm, which is smaller than that of the as-synthesized powders, indicating that the agglomeration degree is weaker after ball milling.

Fig. 3(a) shows the photograph of the double-polished TAG ceramics sintered from as-synthesized powders, ball milled powders, and ball milled powders with TEOS, respectively. As can be seen, all the samples exhibit relatively good transparency since the letters under them are clearly visible. The in-line transmittance curves of the corresponding samples are shown in Fig. 3(b). It is found that