



Nanostructured yttrium aluminum garnet powders synthesized by co-precipitation method using tetraethylenepentamine

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Abstract: Tetraethylenepentamine ($C_8H_{23}N_5$, TEPA) has been used as a novel precipitant to synthesize yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) precursor from a mixed solution of aluminum and yttrium nitrates via a normal-strike co-precipitation method without controlling the pH value during precipitation process. The original precursor was analyzed by thermogravimetry/differential scanning calorimetry (TG/DSC). The evolution of phase composition and micro-structure of the as-synthesized YAG powders were characterized by X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy and transmission electron microscopy (TEM). Compositionally pure YAG nanostructured powders were directly obtained by calcination of the precursor at 900 °C without the formation of any intermediate phases. The average particle size determined from TEM micrograph for the powder obtained at 1000 °C was approximately 50 nm.

Keywords: yttrium aluminum garnet; powder technology; co-precipitation; ceramics; rare earths

Yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) is an ideal host material for phosphors and solid-state lasers when doped with transition or lanthanide element^[1,2]. Besides, because of its promising chemical stability and high-temperature mechanical properties^[3], YAG is also an important ceramic material. Neodymium-doped YAG (Nd:YAG) transparent laser ceramics have attracted much attention because of their several advantages, such as low cost, short preparation time, high doping concentration and large size, etc. Moreover, some properties of polycrystalline Nd:YAG ceramic laser materials have proved to be comparable or superior to those of single crystal^[4,5]. Given in such broad application potential of the YAG materials, the simple and practicable synthesis methods yielding phase-pure nanostructured YAG are highly acceptable.

Typically, YAG powders are prepared by a solid-state reaction from their respective oxide powders. This method usually requires high calcining temperature of over 1600 °C and long aging time^[6,7]. Compared with solid-state reaction, wet-chemical synthesis methods not only have the advantages of low synthesis temperature and short calcining time, but also can achieve homogeneous mixture of metal ions at the atomic level. Consequently, several wet-chemical methods have been extensively investigated in recent years for preparing pure phase YAG powders. These methods include

co-precipitation^[8–10], homogeneous precipitation^[11], sol-gel processing^[12], hydrothermal treatment^[13], spray pyrolysis^[14], combustion methods^[15,16], etc. Among them, co-precipitation is a relatively simple and cost-effective way for powder synthesis, and many precipitants, such as ammonia^[8,17,18] and ammonium hydrogen carbonate^[9,19], have been employed to produce YAG powder. However, pH values of the co-precipitation process with these precipitants have to be kept at some constant value ranging from 7.8–9^[8,17–19] due to amphoteric properties of Al, which makes the preparation procedures more complex and unmanageable.

In the present work, a novel precipitant, tetraethylenepentamine (TEPA), was adopted to synthesize nanostructured stoichiometric YAG powders via a co-precipitation method, and pH value of the reaction solution needs not be especially controlled. This method reported here simplified the manipulation procedures and was more feasible. The results on the synthesis and characterization of nano-sized YAG powder by the co-precipitation method carried out in this laboratory were elaborated in this paper.

1 Experimental

1.1 Synthesis

$Y(NO_3)_3 \cdot 6H_2O$ (>99.9% purity), $Al(NO_3)_3 \cdot 9H_2O$ (>99.9% purity) and TEPA (analytical grade) were used as raw materials for the synthesis of YAG powder. Concentrated solutions were obtained by dissolving $Y(NO_3)_3 \cdot 6H_2O$ and $Al(NO_3)_3 \cdot 9H_2O$ in distilled water, respectively. Then, $Y(NO_3)_3$ and $Al(NO_3)_3$ mixed solutions were prepared from the above concentrated salt solutions with appropriate volume to maintain the Y:Al molar ratio at 3:5. Subsequently, required amount of 0.5 mol/L TEPA solution (TEPA to metal ion ratio of 5) was added dropwise into the mixed solution (normal-strike method) while being stirred properly at 50 °C. After aging for 30 min, the resultant precipitate was filtered, washed with distilled water and ethanol, and dried at 60 °C for 1 d. The dried cake, which was the so-called original precursor, was put into an alumina crucible, and then calcined in a muffle furnace from 800 to 1000 °C for 2 h in air.

1.2 Characterization

The crystalline development of the powders heat-treated at different temperatures was identified by X-ray diffraction (XRD) on a MAC Science MXP21VAHF diffractometer. Thermogravimetry/differential scanning calorimetry (TG/DSC) of the original precursor was carried out on a SETARAM LabsysTM TG-DSC16 thermal analyser. Infrared (IR) spectra were recorded on a Bruker (SENSOR-27) FT-IR spectrometer by a KBr disk method. The particle size and morphology of the heat-treated powders were examined using transmission electron microscopy (TEM) (Model 200, JEOL, Tokyo, Japan).

2 Results and discussion

2.1 X-ray diffraction

The XRD spectra of the original precursor and powders calcined at various temperatures for 2 h is shown in Fig. 1. The original precursor remains amorphous. As the temperature increases to 800 °C, the powder exhibits diffraction peaks which are in agreement with that of YAG crystal structure with cubic structure (ICSD Card No. 20090). Nevertheless, the diffuse and weak peaks indicate the existence of a considerable amount of amorphous phases. With the heating temperature continuously elevating, the powder presents improved crystallinity due to further decomposition and crystallite growth. Calcining to 900 °C results in a complete conversion to YAG without any intermediate impurity phases detected, which indicates that phase-pure YAG is achieved and it is not necessary to dominate the pH value during the normal-strike co-precipitation process. However, most chemical precipitations are performed by reverse-strike

technique^[18,19] (adding salt solution dropwise to the precipitant solution) in co-precipitating multiple components. The possible interpretation may be proposed from the chemical property of TEPA. Belonging to polybasic amine, TEPA hydrolyzes and generates OH^- anion in aqueous solution until the hydrolysis achieves the equilibrium. When TEPA is dripped into the mixed solution containing metal ions, hydroxides of Y^{3+} and Al^{3+} occur and OH^- concentration in aqueous solution decreases in the meantime, which leads to the shift rightward of the hydrolysis balance. Consequently, OH^- anions are continuously released at the same time, and the pH in local areas is approximately considered at a relatively invariant value, which guarantees Y^{3+} and Al^{3+} undergoing precipitation with a ratio of 3:5, therefore no other intermediate phase, except YAG, occurs in the above XRD spectra. Further heating of the powder to 1000 °C shows no change in phase composition other than the intensity and sharpness of the diffraction peaks, which indicates the improved crystallinity at a higher calcination temperature.

2.2 Thermal analysis

Fig. 2 shows TG/DSC curves of the original precursor. Two major endothermic peaks located at approximately 91

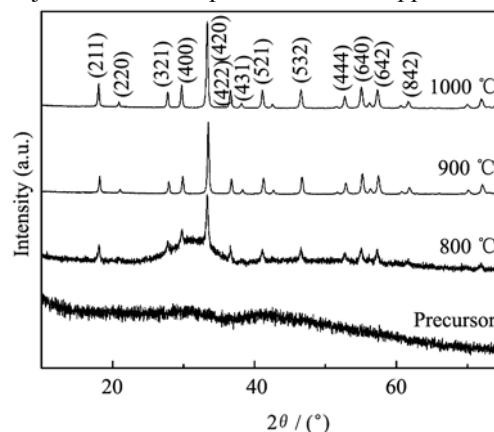


Fig. 1 XRD pattern of the YAG powders calcined at various temperatures

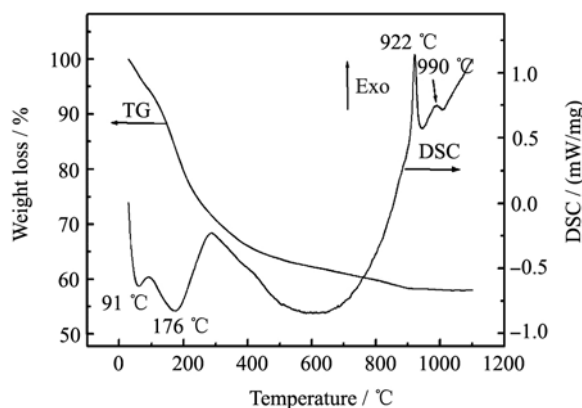


Fig. 2 TG/DSC curves of the original precursor

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