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Journal of the European Ceramic Society 32 (2012) 2971–2979

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Co-precipitation synthesis route to yttrium aluminum garnet (YAG) transparent ceramics

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Abstract

Yttrium aluminum garnet (YAG) precursor was synthesized via a coprecipitation method with aluminum nitrate and yttrium nitrate as raw materials, using ammonium hydrogen carbonate (AHC) as the precipitant. Fine and low-agglomerated YAG powder was obtained by calcining the precursor at 1200 °C. The primary crystallites were measured to be ∼120 nm in size and weakly agglomerated to a particle size of ∼500 nm, indicating a high degree of sinterability. With 0.5 wt% tetraethyl orthosilicate (TEOS) and 0.1 wt% magnesia as sintering aids, transparent YAG ceramics were fabricated by vacuum sintering at 1730–1790 °C for various hours. The influences of sintering temperature and holding time on the microstructure and transmittance of YAG ceramics were discussed.

Keywords: Transparent ceramics; YAG powders; Vacuum sintering; Co-precipitation synthesis

1. Introduction

Coble first demonstrated that polycrystalline ceramics could be sintered to a visually transparent/translucent state and translucent alumina ceramics (Lucalox) were prepared.[1,2] Since then, a variety of transparent/translucent ceramics including MgO,[3] Y_2O_3,[4] $MgAl_2O_4$,[5] PLZT,[6] AlON,[7] SiAlON,[8] AlN,[9] YAG[10] have been successfully produced by sintering to exceptionally high densities and thus reducing/eliminating light scattering from residual porosity. Transparent YAG ceramics hold promise for certain optical applications such as solid-state laser host media, luminous pipes for high-intensity discharge lamps or heat-resistive windows because of their large doping concentrations,[11,12] increased compositional and structural versatility,[13–15] good chemical corrosion resisitivity,[16] and high mechanical property.[17]

Usually, two source powders are utilized to make YAG transparent ceramics: (i) mixtures of Y_2O_3 and Al_2O_3, which form YAG in situ during solid-state reactive sintering,[18–24] and (ii) wet-chemical synthesized YAG powder.[25–29] Solid-state reactive sintering is a relatively simple way to fabricate YAG transparent ceramics and a range of compositions is easy to implement by changing the reactant powder amounts during batching.[30,31] However, the incorporation of some impurities is unavoidable during ball milling. Wet chemical approaches such as precipitation,[32–36] spray pyrolysis,[37,38] hydrothermal (or solvothermal) synthesis,[39–41] sol–gel[42–44] and combustion synthesis[45–49] have many advantages such as atomic level mixing of high-purity precursors and low processing temperature. Among the wet chemical methods, precipitation is a relatively simple way to synthesize YAG powder with composition homogeneity, good crystallinity and pure phase at low temperature. And fully dense and transparent ceramics can be fabricated by vacuum sintering of YAG powders with high sinterability.

In this paper, carbonate precursors of YAG were synthesized via a coprecipitation method using ammonium hydrogen carbonate (AHC) as the precipitant. Nanosized YAG powders with high sinterability were obtained at 1200 °C and transparent YAG ceramics were fabricated by vacuum sintering at 1730–1790 °C with a considerable amount of additives of tetraethoxysilane (TEOS) and MgO.

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doi:10.1016/j.jeurceramsoc.2012.02.040

2. Experimental

2.1. Materials

Several raw materials were used to synthesize nanosized YAG powders: yttria (Y_2O_3, Shanghai Yuelong New Materials Co. Ltd., China, 99.99% purity), aluminum nitrate nonahydrate ($Al(NO_3)_3{\cdot}9H_2O$, Sigma–Aldrich Chemicals, USA, >98% purity), ammonium hydrogen carbonate (NH_4HCO_3, Sigma–Aldrich Chemicals, USA, >99% purity) and ammonium sulfate ($(NH_4)_2{\cdot}SO_4$, Sinopharm Chemical Reagent Co. Ltd., China, analytical purity).

2.2. Powder synthesis

In the present work, a wet-chemical synthesis route was utilized to produce nanosized YAG powders, employing $Al(NO_3)_3{\cdot}9H_2O$ and Y_2O_3 as the starting materials, respectively. Yttrium nitrate solution was prepared by dissolving the yttria powder in the heated nitric acid (Shanghai Lingfeng Chemical Reagent Co., Ltd., super-high purity). Aluminum nitrate solution was prepared by dissolving the $Al(NO_3)_3{\cdot}9H_2O$ in the deionized water. Al^{3+} and Y^{3+} concentrations of the nitrate solutions were assayed by the ICP (Inductively Coupled Plasma) spectrophotometric technique. The metal nitrates were mixed to meet the YAG stoichiometry and further adjusted the solution concentration to 0.19 M for Al^{3+}. As a precipitation solution, a 0.92 M solution of ammonium hydrogen carbonate (AHC) was used and ammonium sulfate (0.11 g in 100 ml of deionized water) was added to the solution. The precursor precipitate was made at the room temperature by dripping 2200 ml of ammonium hydrogen carbonate solution into 1100 ml of the mixed solution of mother nitrate salts at a speed of 2 ml/min under mild agitation and the final pH value of the slurry was about 8.0. The resultant suspension was agitated for 6 h and aged for 12 h. Then, the suspension was filtered using centrifugal filtration, washed four times with deionized water, rinsed twice with ethanol, and dried at 70 °C for 24 h. The dried cake was crushed with a corundum pestle and mortar, and sieved through a 200-mesh screen. The sieved precursor powder was calcined at different temperatures for 3 h to form garnet phase of YAG powders.

2.3. Powder characterization

Thermal gravimetric analysis and differential thermal analysis (TG–DTA) of the original precursor were recorded on a Netzsch STA 449C Instrument. Measurements were taken under a continuous flow of air (20 ml/min). Sample was heated at 10 °C/min to 1400 °C and then cooled to room temperature naturally.

Chemical analysis was made to confirm the chemical stoichiometry of YAG powders. Y and Al contents were determined by the chelate-titrimetric method.

Phase identification was performed by the X-ray diffraction (XRD) method on a HUBER Imaging Plate Guinier Camera G670 [S] ($CuK_{\alpha1}$ radiation, $\lambda = 1.54056$ Å, 40 kV/30 mA, Ge monochromator). The 2θ for all data ranged from 15° to 65° with 0.005° step size. The average crystallite size of the calcined powders was calculated from X-ray peak broadening using Scherrer formula. The XRD data are refined by Rietveld method using the Jade program (Version 5.0).

Specific surface area analyses were conducted at 77 K using a Norcross ASAP 2010 micromeritics, with N_2 as the absorbate gas. Powders were degassed at 150 °C until the air pressure was below 5 μm Hg. The specific surface areas were calculated using the BET multipoint method with 8 data points. The average particle size of the calcined powders was calculated from the specific surface area data.

Microstructures were observed on a JEOL JSM-6700 FESEM and the mean particle size was estimated by measuring diameters of 100 particles in the FESEM photographs. Powders were dispersed in ethanol using an ultrasonic horn. Drops of the dispersed materials were deposited on a copper stub and dried in nature. Samples were sputter coated with palladium using a JEOL JFC-1600 auto fine coater system.

The agglomeration particle size distribution was measured by a dynamic laser scattering (DLS) method on a Brookhaven ZetaPlus Zeta Potential Analyzer.

2.4. Ceramic fabrication and characterization

Using the powder calcined at 1200 °C as starting materials, 0.5 wt% TEOS (Alfa Aesar, 99.999% purity, USA) and 0.1at%MgO (Sinopharm Chemical Reagent Co., Ltd., spectral purity, China) as sintering aids, two powder batches were ball milled in anhydrous ethanol for 10 h using high-purity (99.7%) alumina balls and the balling media had no measurable weight loss after milling. The milled suspensions were dried at 90 °C in an oven and ground in an alumina mortar. After sieving through 200-mesh screen, two powder batches were uniaxially pressed in a 20 mm die at 10 MPa followed by cold isostatical pressing (CIP) at 250 MPa. The compacted disks were sintered at the temperature range of 1730–1790 °C up to 30 h in a tungsten mesh heated vacuum furnace (KZG-110F, Shanghai Chenrong Electrical Furnace Co., Ltd., Shanghai, China) under 3×10^{-3} Pa vacuum during holding. The heating and cooling rates were 5 °C/min and 10 °C/min, respectively. All the sintered specimens were annealed at 1450 °C for 20 h in air to remove the oxygen vacancies. For optical transmission comparison, YAG transparent ceramics were also fabricated by a solid-state reactive sintering method[20] using commercial Y_2O_3 and α-Al_2O_3 powders as starting materials.

Densities of the sintered specimens were measured by the Archimedes method, using deionized water as the immersion medium. Microstructures of the fractured surfaces and the polished and thermal-etched surfaces were observed by EPMA (Model JXA-8100, JEOL, Japan). Mirror-polished specimens on both surfaces with the thickness of 1.0 mm were used to measure the optical in-line transmittance (Model Cray-5000 UV-VIS-NIR Spectrophotometer, Varian, CA, USA).