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Short communication

Preparation of YAG ceramics through a novel process

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Abstract

Transparent YAG ceramics were fabricated by a novel process in which the yttria precursor and alumina precursor were synthesized separately, and then mixed through ball milling method. YAG powders were obtained by calcining the mixed precursors at 1350 °C for 2 h. YAG ceramics were fabricated via vacuum sintering at 1750 °C for 20 h. The transmittance of YAG ceramics fabricated was about 50.9–60.3% over the wavelength range of 200–1100 nm, and the average grain size was about 10 μm.

Keywords: Yttrium aluminum garnet; Transparent ceramics; Precipitation

1. Introduction

Transparent ceramics with yttrium aluminum garnet structure can be applied in many fields, especially in the field of host materials for solid state lasers. In 1995, Japanese scientist A. Ikesue fabricated highly transparent Nd:YAG ceramics successfully and realized the laser oscillation in ceramics with garnet structure for the first time in the world [1], and that report aroused the concern for all laser materials scientists in the world. In 1999, YAG ceramics with better properties were fabricated by Japanese scientists [2,3] using co-precipitation method and vacuum sintering technology, and the laser performance was improved significantly [4].

Generally speaking, transparent YAG ceramics can be fabricated through two processes, one was called solid state process in which the oxide materials were mixed through ball milling method and the ceramics were prepared by vacuum sintering [1,5], and the other was called wet process in which YAG powders were first synthesized using co-precipitation method or other chemical methods and the ceramics were also prepared through vacuum sintering [2,3]. Compared with the wet process, the solid state process performs much worse in the optical quality and laser performance of the samples prepared. For the solid state progress, out powers of 70 mW [1] and 1003 mW [6] were once reported, while for the wet process, out powers were increased greatly from 31 W [7] to 72 W [8], 1.46 kW [9]. But the wet process is complex and only a few researchers had mastered this technique. Many efforts had been put into the research of this process, and the mechanism and modification of the wet process were investigated [10–14], but the results were barely satisfactory.

During our initial research period, it was found that the optimal precipitation conditions for alumina and yttria precursors which were two major components of YAG precursors were different, especially for the mixing sequence. The alumina precursor synthesized by adding precipitant solution dropwise into the metal ion solution was colloid boehmite precipitate containing lots of crystal water and hydroxyl groups, and the hydroxyl groups could result in the hard agglomerate of powders [15,16]. While the yttria precursor synthesized by adding the precipitant dropwise into the metal ion solution was beneficial to preparation of spherical yttria particles with good sinterability [17]. So, it might be better that the alumina precursor and yttria precursor were synthesized separately and then mixed by some means of mixing. In this paper, YAG ceramics were fabricated by a new

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process in which the alumina precursor and yttria precursor were synthesized separately then mixed through the ball-milling method. After that, YAG powders were obtained by calcining the mixed precursors at a specified temperature. Then YAG ceramics were obtained via the processes of molding, sintering and post-treatments. The characterization of precursors, calcined powders and ceramic samples was conducted using X-ray diffractometer, thermal analyzer (TG-DTA), scanning electron microscopy (SEM) and spectrophotometer.

2. Experiment procedure

Yttrium nitrate hydrate ($Y(NO_3)_3\cdot H_2O$, purity $> 99.9\%$), aluminum nitrate hydrate ($Al(NO_3)_3\cdot H_2O$, purity $> 99.9\%$) and ammonia bicarbonate (NH_4HCO_3, analytical reagent) were used as raw materials. The starting solutions were made by dissolving the corresponding raw materials into deionized water following being filtered.

First, the alumina and yttria precursors were synthesized separately. The alumina precursor was synthesized by adding the aluminum ions solution (0.15 mol/L) dropwise into the ammonium bicarbonate solution (1.5 mol/L). The yttria precursor was synthesized by adding the ammonium solution (0.5 mol/L) dropwise into the yttrium ions solution (0.15 mol/L). Both precursors were three-time washed using water and alcohol respectively, then dried and sieved. Then both precursors were mixed uniformly by ball-milling. After that, the mixed precursors were dried, sieved and calcined at different temperatures to determine the temperature needed to obtain YAG powders. The obtained YAG powders were pressed into discs with the diameter

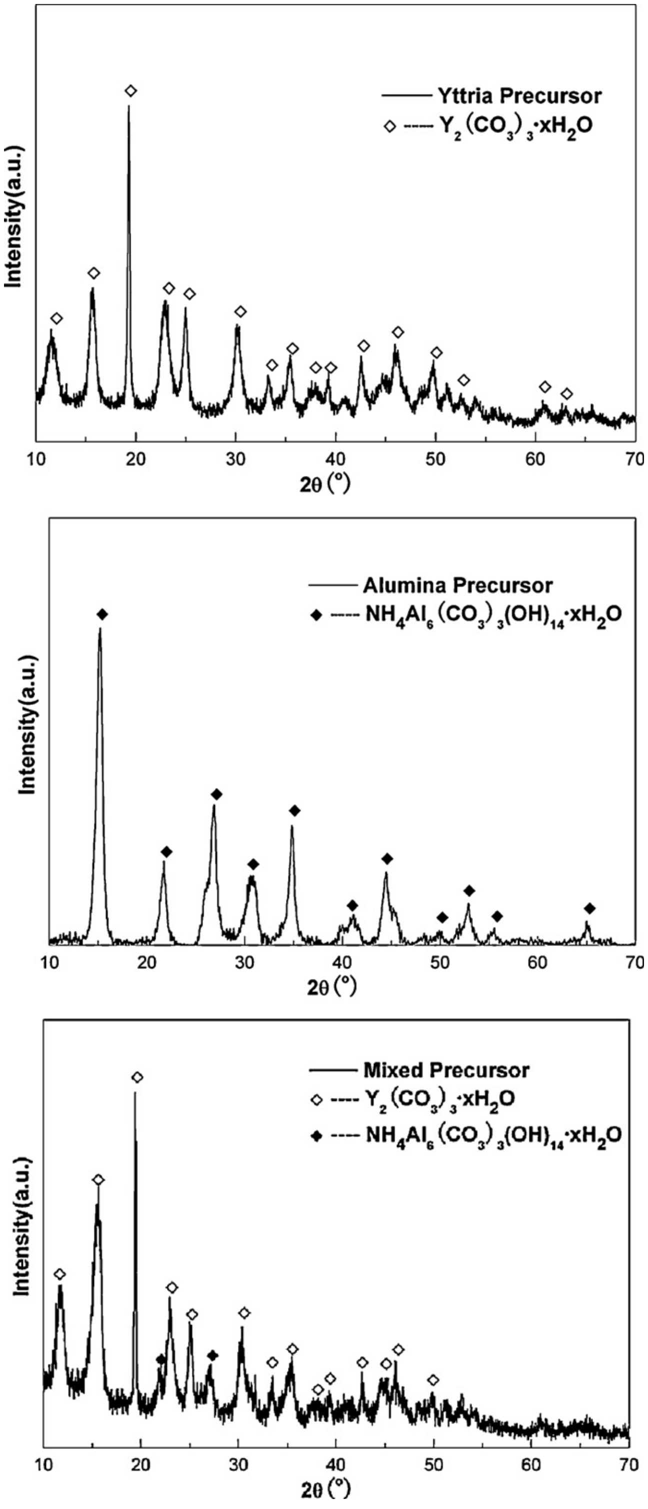

Fig. 1. The XRD patterns of separate precursors and mixed precursors.

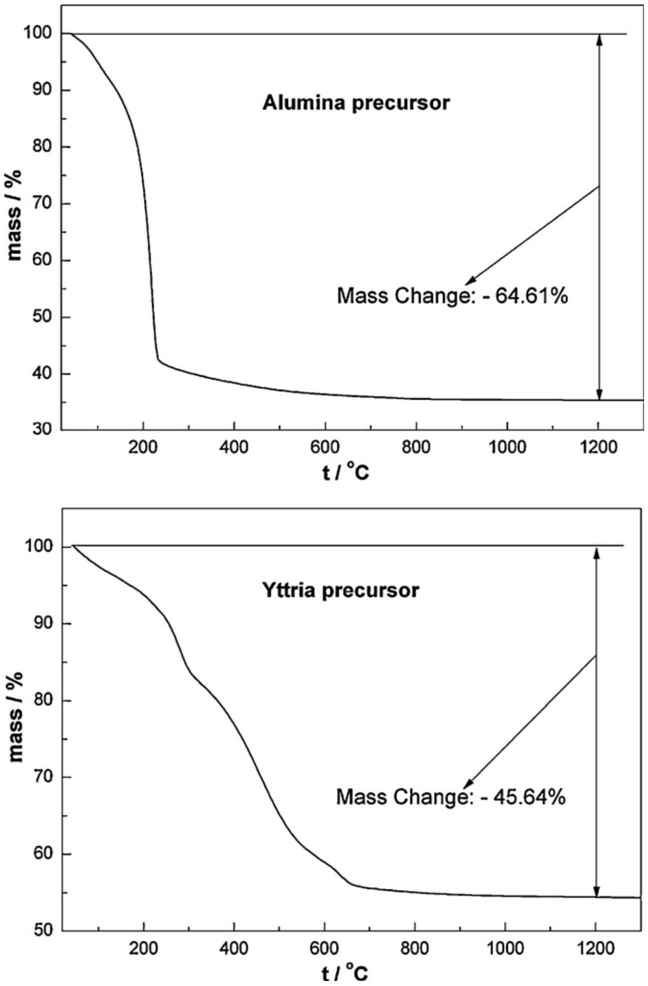

Fig. 2. The thermogravimetric curves of alumina precursor and yttria precursor.

of 18 mm and then treated with isostatic-pressing. The green compacts were sintered at 1750 °C for 20 h under vacuum condition of lower than 1.0×10^{-3} Pa. The vacuum sintered samples were annealed at 1450 °C for 10 h in air then mirror-polished and thermal etched for characterizations.

Phase identification of all precursors and calcined powders was performed via diffractometer (XRD, Model D/MAX-2550V, Rigaku, Tokyo, Japan) using Cu Kα radiation at the scanning speed of 10°/min (2θ). The weightlessness of two separate precursors was analyzed using thermal analyzer (TG-DTA, Model STA 449C, NETZSCH, Germany) with temperature programming from room temperature to 1300 °C by 5 °C/min. The morphologies of precursors, calcined powders and thermal etched surfaces of obtained ceramics were characterized using electron probe micro-analysis (EPMA, Model JXA-8100F, JEOL, Tokyo, Japan). The transmittance of polished ceramics was measured over the wavelength region from 200 nm to 1100 nm using a spectrophotometer (Model U-2800, Hitachi, Tokyo, Japan). The relative density of sintered sample was measured using the Archimedes method.

3. Results and discussion

3.1. Characterization of precursors

The XRD patterns of separate precursors and mixed precursors were shown in Fig. 1. It can be certified that the two separate precursors were yttrium carbonate and ammonium aluminum carbonate hydroxide. As seen in the XRD pattern of the mixed precursors, only two corresponding diffraction peaks of ammonium aluminum carbonate hydroxide could be found in the figure, which might be explained that the intensity of those peaks were so weak that most of them were concealed by the peaks corresponding to yttrium carbonate.

The weight loss of two separate precursors was measured using thermogravimetric analyzer, and the thermogravimetric curves were shown in Fig. 2. It can be learned from the curve that the weight losses of alumina and yttria precursors were 64.61% and 45.46%, separately. The stoichiometric amounts of two precursors based on their respective TGA were determined and mixed for the synthesis of YAG.

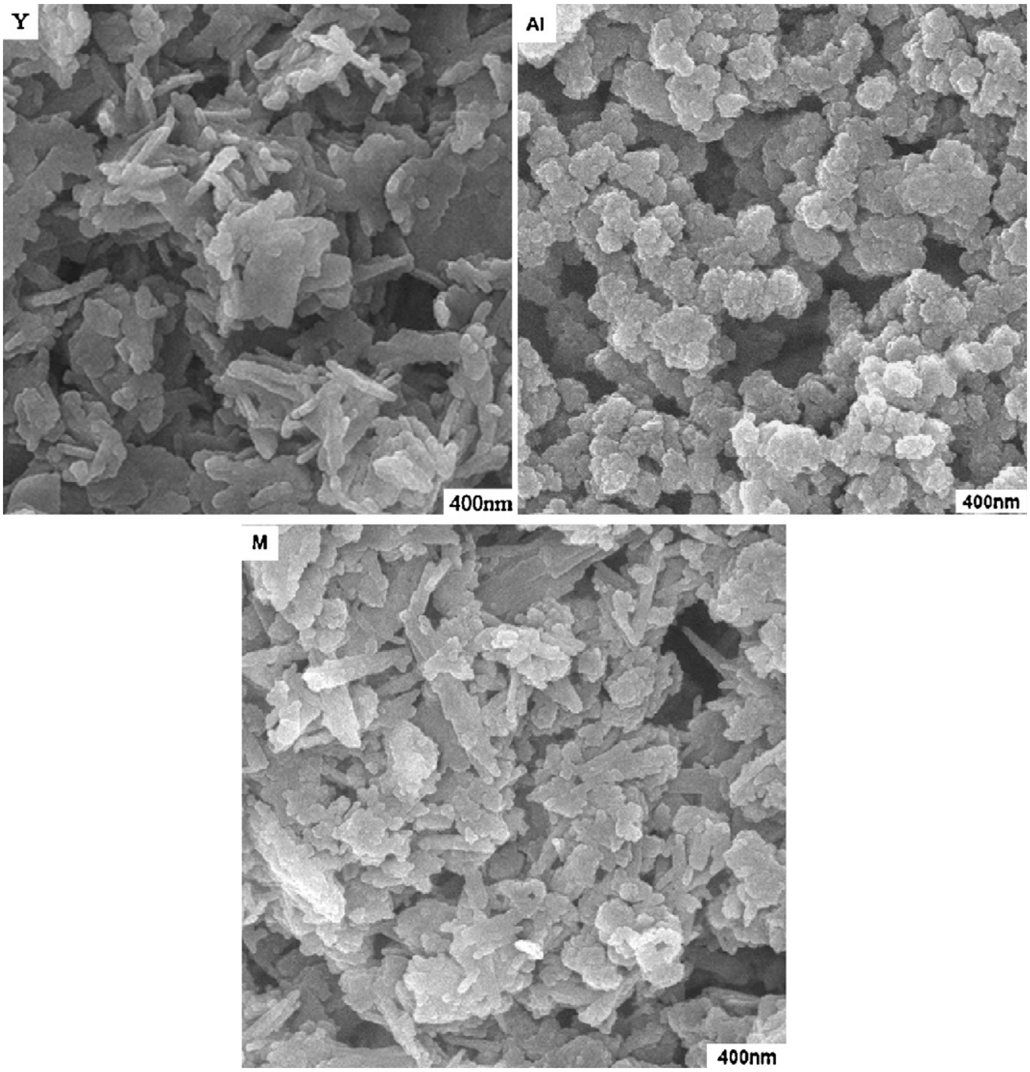

Fig. 3. The SEM morphologies of yttria precursor (Y), alumina precursor (Al) and mixed precursor (M).

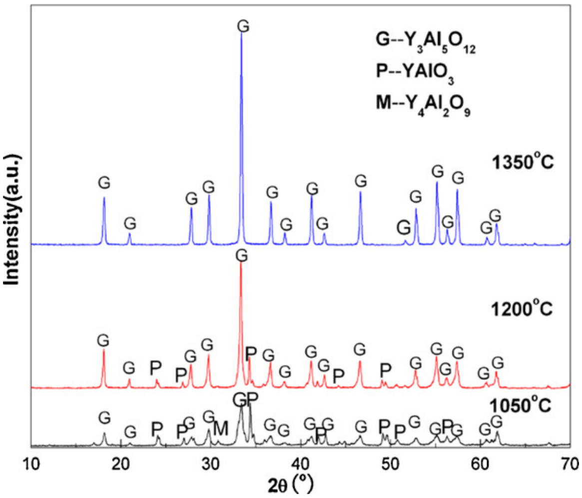

Fig. 4. The XRD patterns of powders obtained by calcining the mixed precursors at different temperatures.

The morphologies of separate and mixed precursors were presented in Fig. 3. As judged from the morphology of yttria precursor, small particles of several tens nanometers tended to aggregate into flakes of several hundred nanometers. While for alumina precursor, only clusters of small particles of 20–30 nm can be observed. And the uniform mixture of particles with two different morphologies, which indicates that yttria and alumina precursors were uniformly mixed, can be seen in the picture of mixed precursors.

3.2. Characterization of calcined powders

The XRD patterns of calcined powders were presented in Fig. 4. It can be seen that the phase composition of the powder obtained at 1050 °C was composed of three phases which were YAM($Y_4Al_2O_9$), YAP($YAlO_3$), YAG($Y_3Al_5O_{12}$), respectively and both YAM phase and YAP phase were intermediate phases. With the increase of calcining temperature, intermediate phases began to transform to the stable YAG phase. The powder obtained at 1200 °C was composed of phases of YAP and YAG, and YAM phase had disappeared. When the temperature was increased to 1350 °C, the powder obtained was composed of one single phase YAG. And the phases of YAM and YAP had finally transformed to YAG phase. So the condition of 1350 °C × 2 h was chosen to be the calcining condition for the synthesis of YAG powders.

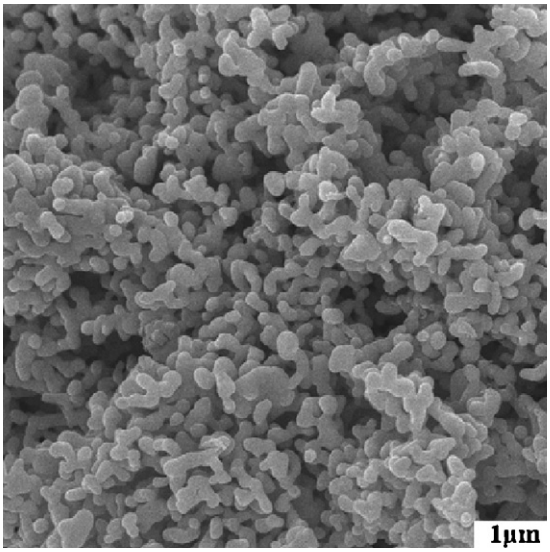

Fig. 5. The SEM morphology of YAG powders obtained under the condition of 1350 °C × 2 h.

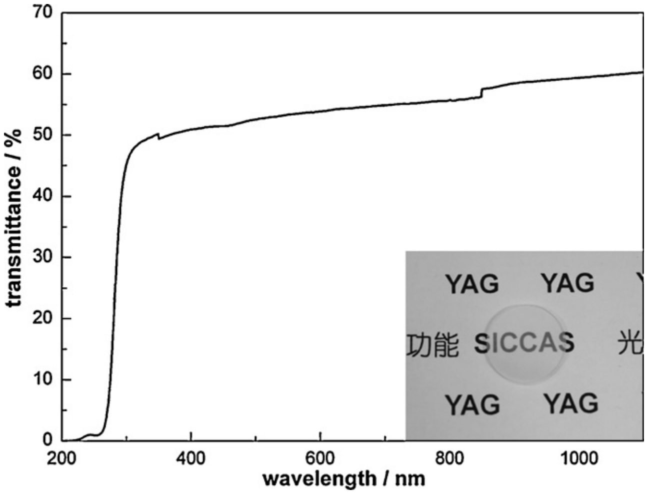

Fig. 6. The transmittance curve of vacuum sintered YAG ceramic.

The morphology of YAG powders obtained was shown in Fig. 5. As seen in the picture, the particles were agglomerated to some extent, and there were a certain amount of sintering necks existing between particles. And the average particle size was about 200 nm.

3.3. Characterization of vacuum sintered YAG ceramics

The relative density of polished sample which was measured using the Archimedes method was 99.68%. The transmittance curve of the vacuum sintered sample (14 mm in diameter and 1 mm in thickness) can be seen in Fig. 4, and the inset is the

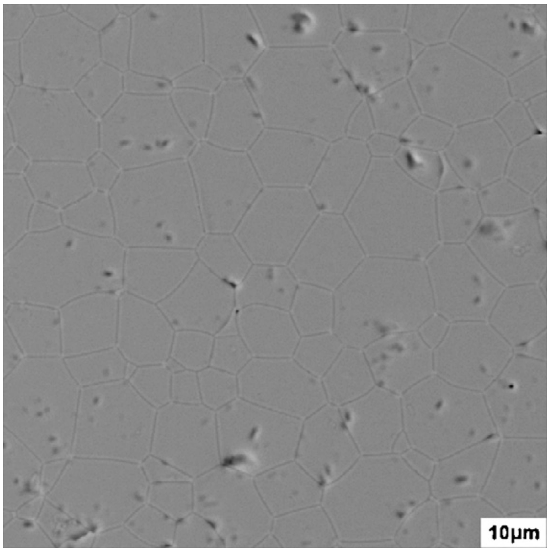

Fig. 7. The surface morphology of thermal etched YAG ceramics.

photograph of the ceramic tested. The inline transmittance of the sample varied from 50.9% to 60.3% over the wavelength region from 400 nm to 1100 nm. As seen in the inset, the words down the samples can be clearly identified.

The mirror polished ceramic samples were thermal etched at 1550 °C for 1 h. The surface morphology of thermal etched YAG ceramics was shown in Fig. 6. As shown in the picture, intragranular and intergranular pores which introduce the scattering of light and induce relatively low transmittance of YAG ceramics. It can be seen that the average granular size was about 10 μm (Fig. 7).

4. Conclusions

Transparent YAG ceramics were fabricated by a new process in which the yttria and alumina precursors were synthesized separately, and then mixed through ball-milling method. YAG powders were obtained by calcining the mixed precursors at 1350 °C for 2 h. YAG ceramics were obtained via vacuum sintering at 1750 °C for 20 h. The transmittance of YAG ceramics fabricated was about 50.9–60.3% over the wavelength range of 200–1100 nm. And the average granular size was about 10 μm.

Acknowledgements

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