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Short communication

Preparation of YAG powders and ceramics through mixed precipitation method

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Abstract

A novel precipitation method called mixed precipitation method for the fabrication of transparent YAG ceramics is reported. The YAG powder can be prepared at 1050 °C through the mixed precipitation method, and the particle size of YAG powder obtained is in the range of about 200–500 nm. The transmittance of ceramics prepared through the mixed precipitation method is about 60% in the visible region without using sintering aids. The grain size of sintered YAG ceramic is about 10 μm.

Keywords: Yttrium aluminum garnet; Transparent ceramics; Precipitation

1. Introduction

Yttrium aluminum garnet (YAG, $Y_3Al_5O_{12}$) exhibits cubic structure and the space group is *Ia*3*d*. Yttrium aluminum garnet ceramics with good optical properties can be prepared because of no birefringence [1]. And besides, transparent YAG ceramics have good chemical stability and creep resistance [2]. Especially, when doped with rare earth elements, such as neodymium, ytterbium, cerium, YAG ceramic can be used as ideal solid state laser materials and scintillating materials [3–5]. Many efforts have been put into the research of YAG based ceramic laser materials since the first laser oscillation in neodymium doped YAG ceramic was realized in 1995. The properties of YAG based transparent ceramics have been improved significantly.

Generally speaking, transparent YAG ceramics can be fabricated through two methods, one is called solid state reactive sintering technique [3] and the other is called nanopowder technology combined with vacuum sintering technique [6,7]. The latter process is more promising compared with the former [3,8]. As to the nanopowder technology, the YAG powder is derived from solutions using some methods, such as coprecipitation method [6,7] and citrate method [9]. The research of the co-precipitation technique has been conducted for quite a long time, and the mechanism of coprecipitation method has been studied initially and the improvement of that process has been tried [10–14].

During our initial research period, it was found that the alumina and yttria precursors prepared through precipitation method can be greatly affected by the concentration of precipitant and the mixing sequence. And the properties of alumina and yttria powders obtained will also be greatly affected [15,18]. The alumina precursor prepared under the condition of low precipitant concentration and through the normal striking method is colloid boehmite precipitate containing lots of crystal water and hydroxyl groups, and the hydroxyl groups could result in hard agglomerate of powders [16,17]. While the spherical yttria powders with good sinterability can easily be obtained under the condition of low precipitant concentration [18].

In this paper, YAG powders and ceramics are attempted to be fabricated by a novel precipitation method called mixed precipitation method. In mixed precipitation method, alumina precursor is first prepared and then two kinds of precursors are mixed through the precipitation of yttrium ions in the

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suspension consisting of yttrium ions solution and alumina precursor. Then YAG powders are obtained by calcining the mixed precursors in air. And YAG ceramics are sintered under vacuum circumstance.

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 ammonium hydrogen carbonate (NH_4HCO_3, analytical reagent) were used as raw materials. And the starting solutions were made by dissolving the corresponding raw materials into deionized water followed by filtering.

The flow chart of mixed precipitation method reported in this paper is presented in Fig. 1. As illustrated in Fig. 1, the alumina precursor was first synthesized by adding the aluminum nitrate solution (0.15 M) dropwise into the rapidly stirring ammonium bicarbonate solution (1.5 M). The prepared alumina precursor was dispersed in the yttrium nitrate solution after going through the steps of aging and washing to be made into uniform suspension. The ammonium bicarbonate solution (0.5 M) was added dropwise into the rapidly stirring suspension obtained above to prepare the uniform mixture of two kinds of precursors. The mixed precursor was aged (24 h), washed (three times using water and alcohol, respectively), dried and then calcined to convert into powders of yttrium aluminum garnet phase.

The YAG powders were moulded into disks using a steel die at 30 MPa followed by cold isostatic pressing at 200 MPa. The obtained green compacts were then 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 the precursors and calcined powders was performed via diffractometer (XRD, Model D/MAX-2550 V, Rigaku, Tokyo, Japan) using Cu Kα radiation at the scanning speed of 10°/min (2θ). The thermal analysis of the precursor was performed using a TG/DTA/MS (Model STA449C, Netzsch, Germany) analyzer in flowing air, and the heating rate was 10 °C/min. The morphologies of YAG precursors and YAG powders were characterized using field emission scanning electron microscope (FESEM, Model JSM-6700F, JEOL, Tokyo, Japan). The morphologies of fracture 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 Cary 5000, Varian, Salt Lake City, America).

3. Results and discussion

3.1. Characterization of YAG precursor

The XRD pattern of precursor prepared through mixed precipitation method is shown in Fig. 2. As shown in Fig. 2, diffraction peaks which correspond to the phase of yttrium carbonate can be observed in the diffraction pattern. No peaks corresponding to the alumina precursor are observed. That might be explained by either the fact that the peaks intensity for alumina precursor is quite weak relative to the yttria precursor or that the alumina precursor is amorphous.

Fig. 3 shows the thermal analysis curve of precursor. As seen in the figure, the total mass loss process is composed of four stages. The mass losses of four stages are 22.16% (room temperature to 270 °C), 11.64% (270–530 °C), 2.97% (530–940 °C) and 1.93% (940–970 °C), respectively. Only one obvious exothermal peak can be observed in the higher temperature region, and it can be attributed to the YAP/YAG transformation based on the XRD results. The discrepancy of phase transition temperatures is caused by the difference of soaking time during the heating process.

The mass spectrum of YAG precursor during heating from room temperature to 1000 °C is shown in Fig. 4. We can see two

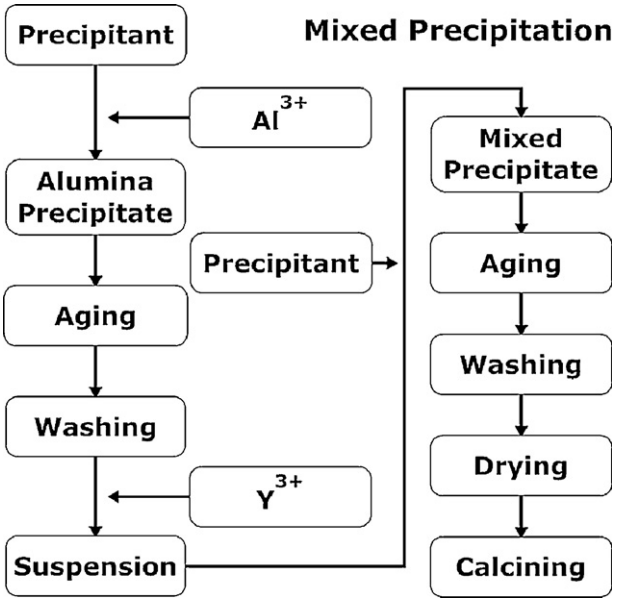

Fig. 1. The flow chart of mixed precipitation method.

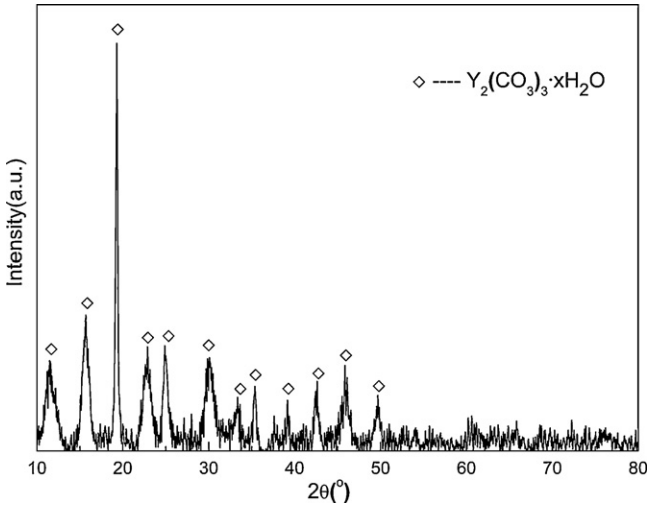

Fig. 2. The XRD pattern of as-synthesized precursor.

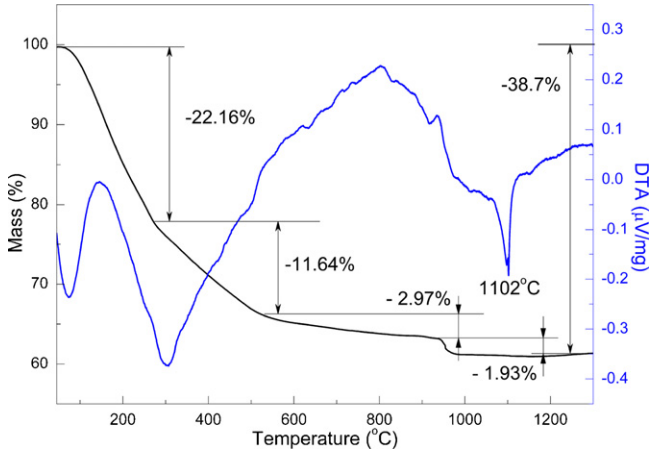

Fig. 3. TG-DTA curves of as-synthesized YAG precursor.

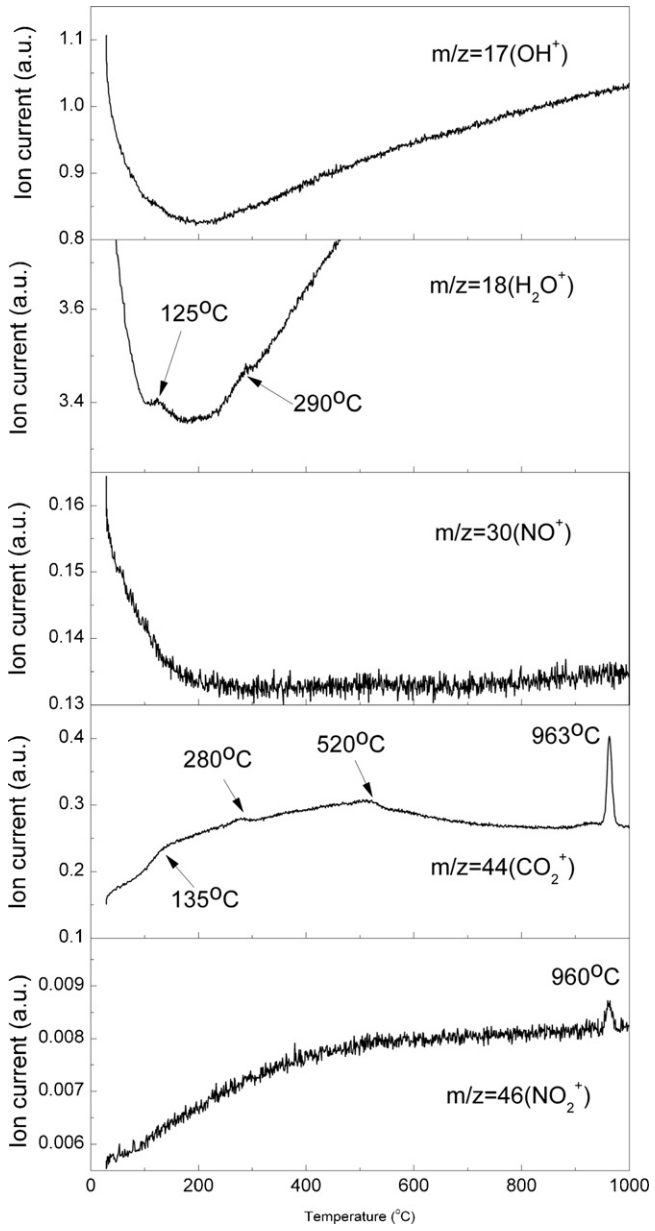

Fig. 4. The mass spectrum of as-synthesized YAG precursor.

weak ion current peaks corresponding to m/z value of 18 (H_2O^+) at 125 °C and 290 °C. As for m/z value of 44 (CO_2^+), four ion current peaks can be observed, three weak peaks at 135 °C, 280 °C and 520 °C and one sharp peak at 963 °C. A weak ion current peak corresponding to m/z value of 46 (NO_2^+) is also detected. No peaks corresponding to the m/z values of 17 (OH^+, NH_3^+) and 30 (NO^+) can be observed. According to the mass spectrum information, the four mass loss stages may be explained as follows. The mass loss in the first stage is caused by the evaporation of absorbed water and initial decomposition of carbonate. The second stage is caused by the evaporation of crystal water and further decomposition of carbonate. The third stage is caused by the further decomposition of carbonate. The fourth stage in a very narrow temperature range is also caused by the decomposition of carbonate, but the mass loss curve is so steep and the corresponding ion current peak at 963 °C is so sharp that it like that the preserved CO_2 is released in a very short time. The reason why that phenomenon occurs is not clear.

3.2. Characterization of calcined powders

The XRD patterns of calcined powders are presented in Fig. 5. The powders obtained at 900 °C are composed of two phases, yttrium aluminum monoclinic phase (YAM, $Y_4Al_2O_9$) and yttrium aluminum perovskite phase (YAP, $YAlO_3$). When the calcining temperature increases to 1050 °C, the diffraction peaks detected are all corresponding to the yttrium aluminum garnet (YAG, $Y_3Al_5O_{12}$) phase. It can be judged that the monoclinic phase and perovskite phase have transformed to garnet phase. With the further increase of temperature, no phase transformation is detected, and the only difference observed is the increase of peak intensity, which indicates better crystallinity of powders.

The YAG powder obtained at 1050 °C was chosen for the fabrication of YAG ceramics. The morphology of YAG powder obtained is shown in Fig. 6. The particle size of YAG powders is not very uniform. The size of large particles is bigger than 500 nm and that of the small particles is about 200 nm. The size of most particles is in the range of 200–500 nm. The powders

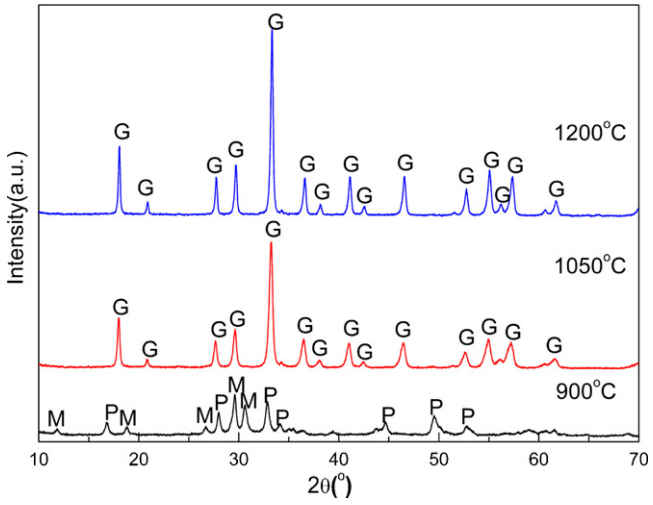

Fig. 5. The XRD patterns of powders obtained at different temperatures.

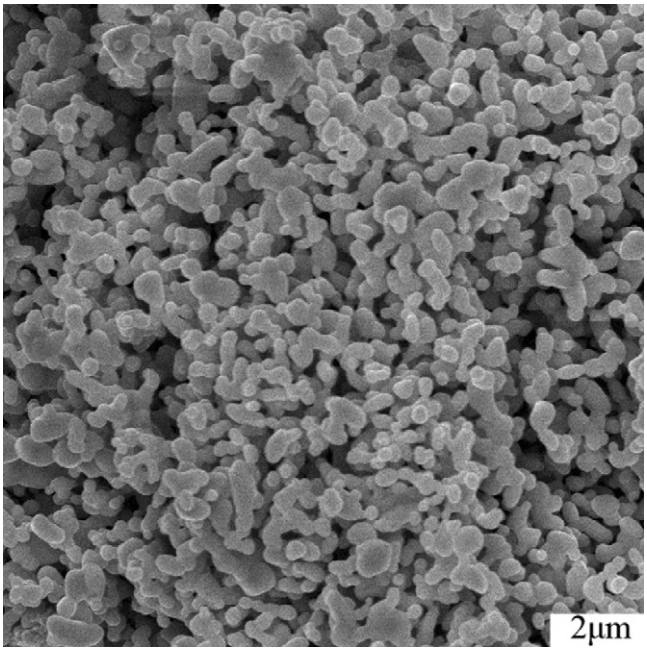

Fig. 6. The FESEM morphology of as-synthesized YAG powder.

are aggregated to some extent. The dispersion property of YAG powders might be improved by using some proper dispersant. And the use of proper dispersant will be studied in the further work.

3.3. Characterization of vacuum sintered YAG ceramics

The photo of YAG ceramic prepared fabricated using powder obtained through mixed precipitation method is shown in Fig. 7. The size of the sample prepared is Φ14 mm × 1 mm. As seen in the photo, the sample is transparent, and the words

Fig. 7. The photo of YAG ceramic prepared.

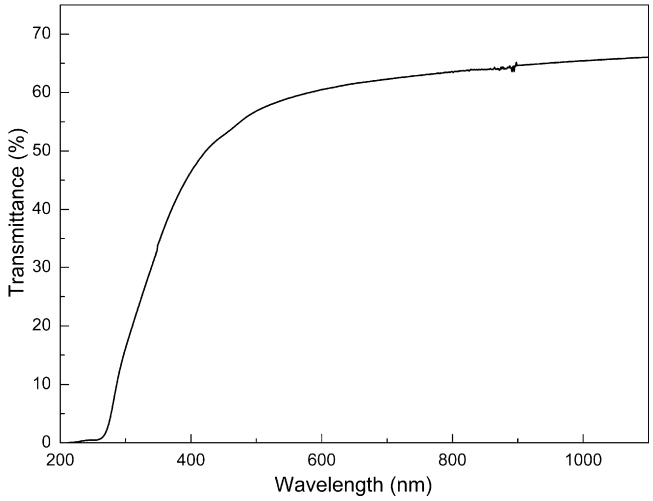

Fig. 8. The transmittance curve of YAG ceramic prepared.

on the paper can be identified clearly through the sample. The transmittance curve of YAG ceramic is shown in Fig. 8. As shown in Fig. 8, the transmittance of the YAG ceramic is 66% at 1100 nm and about 60% in the visible region.

The fracture morphology of ceramic prepared through mixed precipitation method is shown in Fig. 9. As shown in the picture, the fracture behavior of ceramic prepared takes place in the manner of both transgranular fracture and intergranular fracture. And the transgranular fracture could be resulted from the defects and pores existing in the grains. Fig. 10 shows the morphology of thermal etched surface of ceramic prepared. The pores existing in the grains and in the grain boundaries can be observed in the picture, which can result in the decrease of ceramic transmittance. And the average grain size is about 10 μm based on the image of thermal etched surface in Fig. 10.

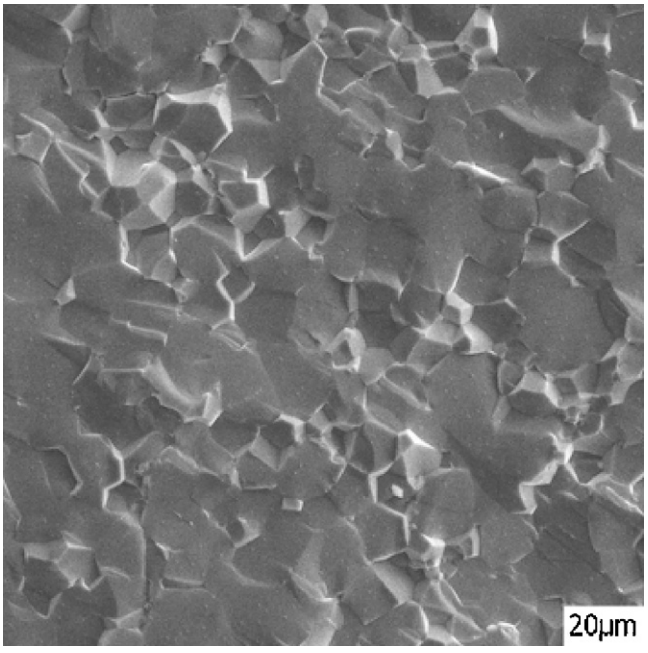

Fig. 9. The fracture morphology of YAG ceramic prepared.

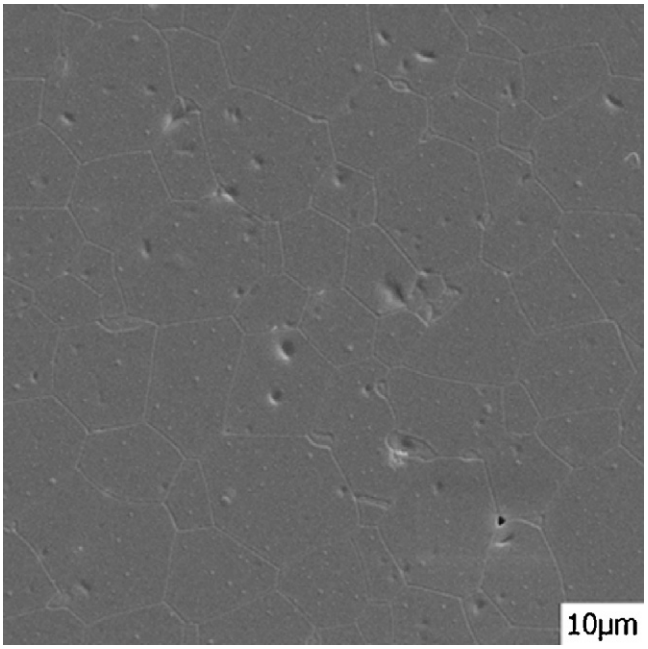

Fig. 10. The morphology of thermal etched surface of YAG ceramic prepared.

4. Conclusions

In this paper, a mixed precipitation method for the preparation of YAG ceramics is reported. The YAG powders can be obtained through calcining the precursors at 1050 °C for 2 h. The particle size of powders prepared is in the range of 200–500 nm. The YAG ceramic prepared is transparent and the transmittance is 66% at 1100 nm and about 60% in the visible region. The average grain size of the sample is about 10 μm.

Acknowledgements

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