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Synthesis and properties of ultrafine YAG powder via low-temperature microwave hydrothermal method

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Yttrium aluminum garnet (YAG) has good optical properties and has been widely used in engineering. The preparation of conventional YAG powder is costly due to its high synthesis temperature, so lowering the synthesis temperature is the most effective way to save cost. The ultrafine oxide composite powder was rapidly synthesized using the microwave hydrothermal method at a low temperature. The effect of temperature and its performance. Differential thermal analysis and XRD phase analysis showed the mixed solution of $Al(NO_3)_3$ and $Y(NO_3)_3$ at pH=9 and $C(Al^{3+})=0.42$ mol·L^{-1}. SEM, zeta potential, and particle size analysis revealed that the microwave hydrothermal reaction temperature was 170 °C, and the high-purity ultrafine YAG powder was calcined at 928 °C, which was lower than conventional calcination temperature. The synthesized ultrafine spherical YAG powder had a small particle size and a uniform distribution.

Key words: Ultrafine YAG powder, Synthesis temperature, Composite powder, Microwave hydrothermal method.

Introduction

Ceramics, as an important part of industrial ceramics, has been extensively investigated because of its high-temperature performance. The most common ones are Al_2O_3 ceramics, SiC ceramics, and Si_3N_4 ceramics [1-3], but the sintering temperature of several ceramics is usually high, accompanied by high cost, long preparation period, and limited existing research. YAG is a complex oxide formed by the reaction of Y_2O_3 and Al_2O_3, which belongs to the cubic system and has a garnet structure [4]. YAG ceramics have excellent high-temperature mechanical properties and chemical stability. YAG powder, which is the main material of YAG ceramics, has been widely explored. In optics, YAG is the best choice for the selection of laser matrix materials [5-6].

YAG is prepared mainly through hydrothermal, coprecipitation, mixed calcination, and sol-gel methods, et al. [7-10]. These synthetic methods have their own advantages and disadvantages. Although the cycle of ultrafine YAG powder preparation via coprecipitation is short, it results in large locally precipitated particles because of the influence of diffusion time when a precipitant is added during precipitation. The hydrothermal method yields ultrafine YAG powder with a small particle size and a uniform distribution, but the preparation cycle is longer and the error is larger than those of other methods.

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Coprecipitation and hydrothermal methods result in YAG powder with good dispersibility. However, in comparison with coprecipitation, the hydrothermal method has the advantages of yielding YAG powder with a small particle size and a uniform distribution, but the preparation cycle is long and costly [11-12]. Therefore, finding a method with a short period, low cost, and excellent powder performance is the current research direction concerning ultrafine YAG powder preparation. In terms of the shortcomings of the hydrothermal method, this study adopts the microwave hydrothermal method to help prepare ultrafine YAG powder. This method results in ultrafine YAG powder with a small particle size, a uniform distribution, and a considerably shortened preparation cycle [13-15].

In this paper, ultrafine YAG powder was prepared through microwave hydrothermal synthesis technology with a considerably shorter preparation cycle than the traditional hydrothermal method, and the process conditions in the YAG ultrafine powder synthesis process were started. The lowest synthesis temperature in the range was used to analyze the variables and determine the optimum conditions for the lowest synthesis temperature of ultrafine YAG powder to reduce the sintering temperature and cost of YAG ceramics. The findings are also the innovation of this paper.

Experimental Materials and Methods

The main raw materials were aluminum nitrate $Al(NO_3)\cdot 9H_2O$ (analytical grade), yttrium nitrate $Y(NO_3)\cdot 6H_2O$ (analytical grade), and ammonia water. Analytically

pure $Al(NO_3)_3 \cdot 9H_2O$ (purity: 99.99%) was weighed with a beaker, deionized, dissolved, and stirred thoroughly with a magnetic stirrer to obtain Al $(NO_3)_3$ at a concentration of 2.0826 mol·L^{-1}. The pure $Y(NO_3)_3 \cdot 6H_2O$ (purity: 99.99%) solid was weighed into a beaker, dissolved in deionized water, and fully dissolved by stirring with a magnetic stirrer to obtain an $Y(NO_3)_3$ solution with a concentration of 2.4866 mol·L^{-1}. The precursors of YAG, $Al(OH)_3$, and $Y(OH)_3$ were obtained by adjusting the pH of the mixed solution of $Al(NO_3)_3$-$Y(NO_3)_3$ by using the aqueous ammonia solution to carry out precipitation reaction and obtain $Al(OH)_3$ and $Y(OH)_3$. Different concentrations of $Al(NO_3)_3$ and $Y(NO_3)_3$ mixed solutions were prepared according to a Y:Al ratio of 3:5 and adjusted to different pH values by using a pH-adjustable aqueous ammonia solution. The mixed precipitates of the YAG precursors $Al(OH)_3$ and $Y(OH)_3$ obtained after precipitation were subjected to microwave hydrothermal decomposition by using a microwave hydrothermal synthesizer to obtain a mixed precipitate of Al_2O_3-Y_2O_3. Thereafter, the obtained mixed precipitate was filtered to obtain white composite powder that was then subjected to differential thermal analysis. The temperature of the YAG crystal phase was determined using a DSC-TG curve via a STA 449 F3-type comprehensive thermal analyzer (NETZSCH, Germany) and calcined in a programmed furnace to analyze the temperature. After calcination, the phase was conducted by using a D8 Advance X-ray powder diffractometer (Bruker, Germany). Surface morphology and particle size (D_{50}) were examined using a Zetasizer Nano Zeta potential and particle size analyzer (Malvern, UK), respectively.

Results and Discussion

Effect of pH on the properties of YAG ultrafine powder

The prepared mixed solution of $C(Al^{3+})$=0.42 mol·L^{-1} and $C(Y^{3+})$=0.25 mol·L^{-1} was adjusted to different pH values. The microwave hydrothermal temperature was 180 ºC and the holding time was 5 min (Table 1). White precipitated powder was filtered and dried to obtain a composite powder.

Fig. 1 shows the XRD pattern of the YAG precursor in a Cu target with a source of Ka-ray (λ = 1.5405 nm) and a scan range of 2θ from 10° to 80°. The XRD pattern of the YAG precursor after thermal decomposition by microwave. The YAG precursor did not decompose into an oxide at a hydrothermal temperature of 180 ºC (Fig. 1).

Fig. 2a is a DSC curve of YAG precursor after microwave hydrothermal decomposition in an air atmosphere at a heating rate of 15 K/min from 25 ºC to 1200 ºC. An endothermic peak appeared at approximately 150 ºC, and this observation was caused by the dehydration decomposition reaction of Y and Al hydroxides

Table 1. Microwave hydrothermal synthesis conditions at different pH conditions.

pH	7, 8, 9, 10
Concentration of Al^{3+}/(mol·L^{-1})	0.42
Hydrothermal temperature/(?)	180
Holding time/(min)	5

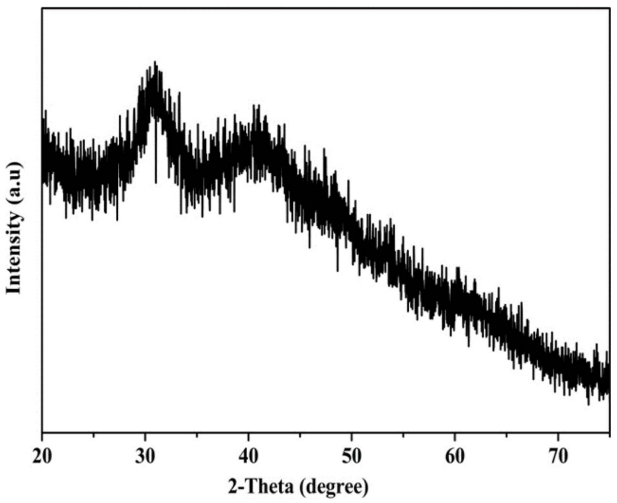

Fig. 1. XRD pattern of YAG precursor after microwave hydrothermal decomposition.

and the evaporation of adsorbed water. An exotherm occurred at approximately 940 ºC. The peak did not lose weight because of the crystallization of the amorphous powder into the YAG phase. Fig. 2b illustrates the synthesis temperature and pH of YAG obtained by preparing the ultrafine YAG powder under different pH conditions. The relationship curve shows that the temperature of the YAG crystal phase initially decreases and then increases as the pH of the mixed mother salt solution increases. At pH=9, the temperature of the YAG crystal phase is relatively low, and the temperature is 933.9 ºC.

Fig. 3 is the XRD pattern of the YAG precursor after calcination. At pH=8 and 9, pure cubic phase ultrafine YAG powder can be synthesized, but the low calcination temperature is observed at pH=9. ultrafine YAG powder and high crystallinity. At this time, Y:Al=3:5 may result in the lower potential energy of YAG synthesis and lower synthesis temperature. When pH=10, the intermediate phase $Y_4Al_2O_9$ (YAM) appears, and Y:Al= 2:1 mainly because a certain content of AlO^{2-} is formed at a high concentration of OH^- so that the ratio of Y:Al is greater than 1, leading to the appearance of the intermediate YAM phase of YAG during calcination. When pH=7, the YAM phase appears because of the low concentration of OH^-, and Al^{3+} cannot be completely precipitated, so the $Al(NO_3)_3$-$Y(NO_3)_3$ mixed salt solution is not completely converted into hydroxide precipitate [16-18]. The ratio of Y:Al does not reach 3:5, resulting in the occurrence of the intermediate YAM phase of

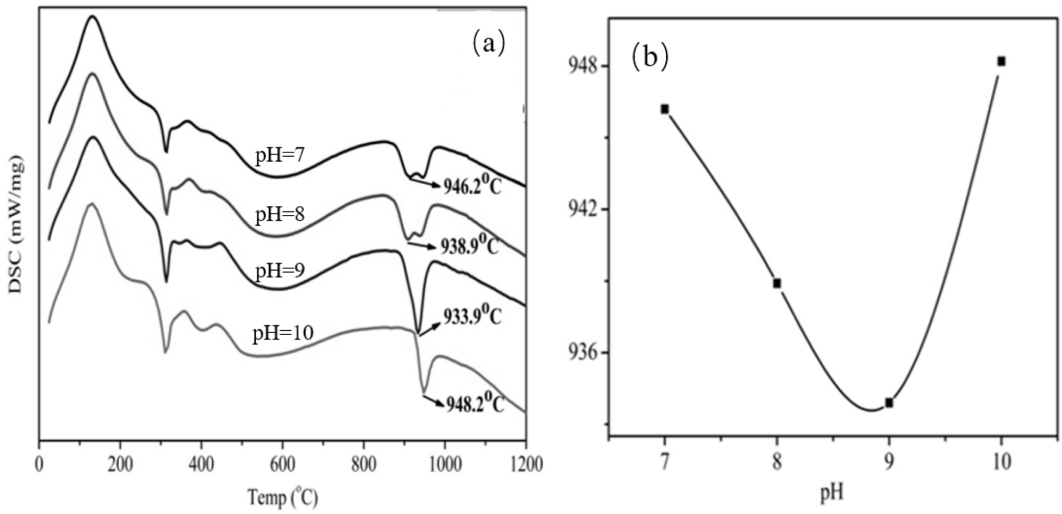

Fig. 2. DSC curves of ultrafine powders (a- prepared at different pH; b-the relation of synthesis temperature and pH curve).

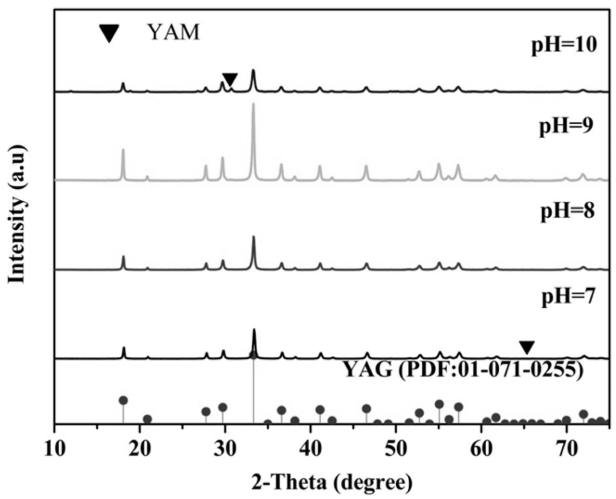

Fig. 3. XRD patterns of calcined ultrafine powders prepared at different pH.

Table 2. Microwave hydrothermal synthesis conditions of YAG mixed solutions at different Al^{3+} concentrations.

Concentration of Al^{3+} / (mol·L^{-1})	0.27, 0.42, 0.52, 0.62, 0.83
pH	9
Hydrothermal temperature / (?)	180
Holding time / (min)	5

YAG during calcination. At pH=9 of the mixed mother salt solution, the pure cubic phase ultrafine YAG powder was synthesized at a low temperature (933.9 ºC) under the condition of Y:Al=3:5.

Effect of Concentration on the Properties of YAG Ultrafine Powder

Fig. 4a shows the DSC curve of the synthetic powder. An endothermic peak appears at approximately 150 ºC, and this peak is caused by the decomposition of the hydroxide of Y and Al and the adsorbed water. An exothermic peak appears at approximately 940 ºC, and no loss of weight is observed because of the crystallization of the amorphous powder into the YAG phase. Fig. 4b shows the synthesis temperature of YAG and the Al^{3+} concentration in the mixed parent salt solution. The relationship curve (Fig. 4b) illustrates that the temperature of the YAG crystal phase initially decreases and then increases as the concentration of Al^{3+} in the mixed mother salt solution increases. At 0.52 mol·L^{-1} concentration, the YAG crystal phase is produced at a lower temperature, and the temperature is 929.8 ºC.

Fig. 5 shows the XRD pattern of the YAG precursor after calcination. When the Al^{3+} concentration in the mixed salt solution is less than 0.52 mol·L^{-1}, a pure cubic-phase ultrafine YAG powder can be produced, and the Al^{3+} concentration in the mixed salt solution is 0.42 mol·L^{-1}, which is synthesized. The ultrafine YAG powder has a strong peak and a high degree of crystallinity. When the concentration of Al^{3+} in the mixed salt solution is greater than or equal to 0.52 mol·L^{-1}, the YAM phase is produced, and the synthesis temperature of the ultrafine YAG powder increases as the concentrations increase. In the reaction of a dilute solution system, Al^{3+} precipitates earlier than Y^{3+} does because Ksp of the solubility products Y^{3+} and Al^{3+} is different. However, the formed $Al(OH)_3$ cannot be nucleated, making Y^{3+} precipitate in the outer layer of Al^{3+} to form a unique inclusion structure in high concentration systems. This phenomenon may be due to the incomplete conversion of Al^{3+} to $Al(OH)_3$ precipitation or AlO^{2-}, resulting in the precipitation of $Al(OH)_3$ in the solution [19-21]. Less, eventually Y:Al=2:1, and the YAM phase is produced after calcination. Therefore, under the same conditions, when the concentration of Al^{3+} in the mixed salt solution is 0.42 mol·L^{-1}, a pure cubic-phase ultrafine YAG powder can be synthesized at a calcination temperature of 931.9 ºC.

Fig. 6 is a comparison of the particle size of the

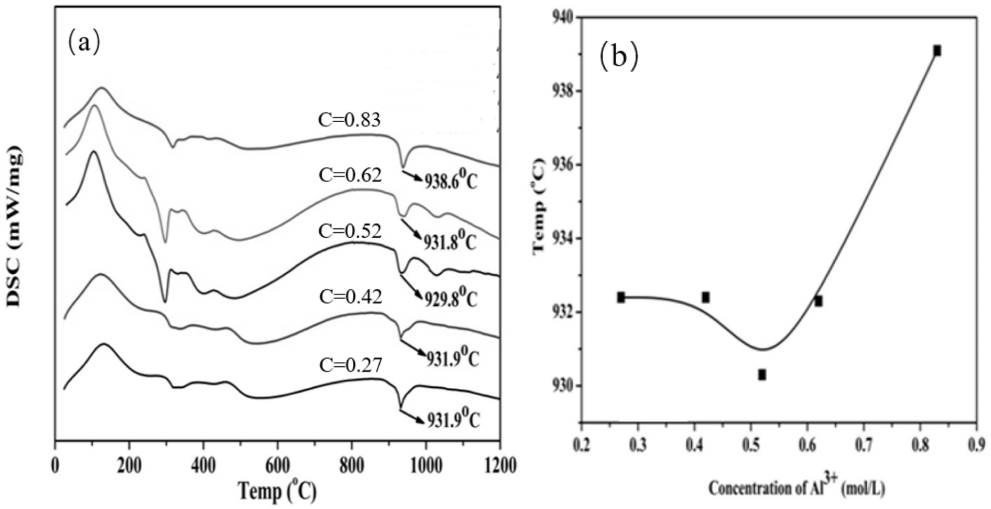

Fig. 4. (a) DSC curves of ultrafine powders prepared at different concentrations;(b) YAG synthesis temperature-concentration curve.

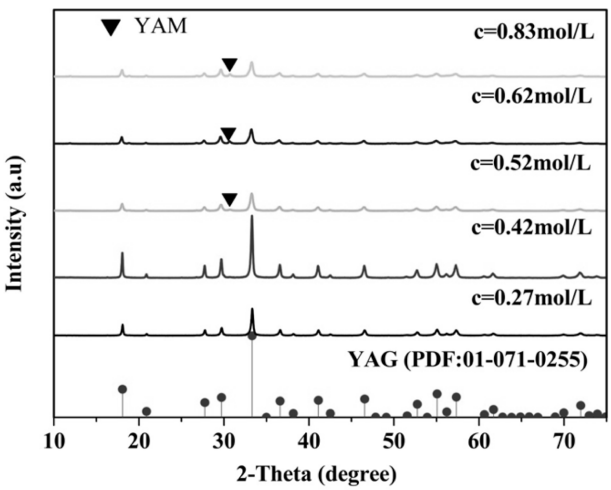

Fig. 5. XRD patterns of calcined ultrafine powders prepared at different reaction concentrations.

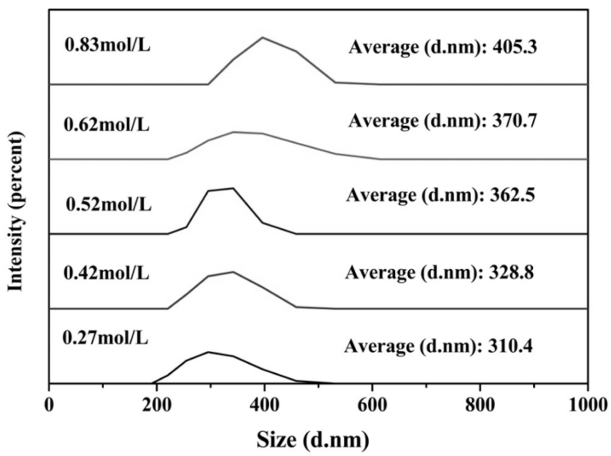

Fig. 6. Effect of different concentration of Al^{3+} on particle size of YAG ultrafine powder.

Effect of hydrothermal temperature on the properties of YAG ultrafine powder.

Table 3. Microwave hydrothermal synthesis conditions of YAG at different hydrothermal temperatures.

Hydrothermal temperature / (?)	160, 170, 180, 190, 200
pH	9
Concentration of reactant / ($mol \cdot L^{-1}$)	0.42
Holding time / (min)	5

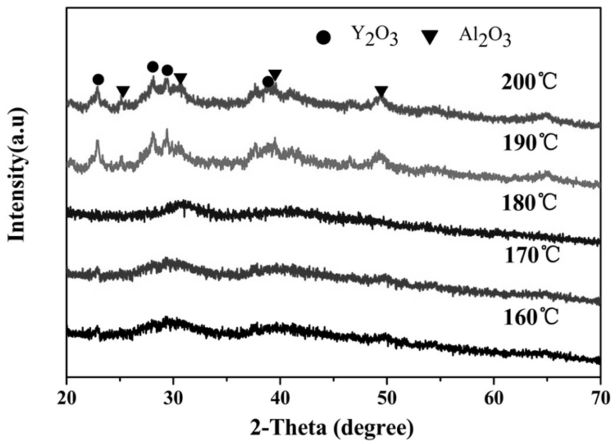

Fig. 7. XRD spectra of YAG precursor after microwave hydrothermal decomposition.

ultrafine YAG powder prepared at different Al^{3+} concentrations. Fig. 6 shows that the particle size of the prepared ultrafine YAG powder also increases as the concentration of the reaction mother salt solution increases. Thus, the higher the concentration is, the higher the formation temperature of the YAG crystal phase will be.

Fig. 7 shows the XRD pattern of the YAG precursor after microwave hydrothermal decomposition. YAG precursors have crystal phases of γ-Al_2O_3 and Y_2O_3 under the experimental parameters of the microwave hydrothermal temperatures of 190 ºC and 200 ºC and the holding time of 5 min, respectively.

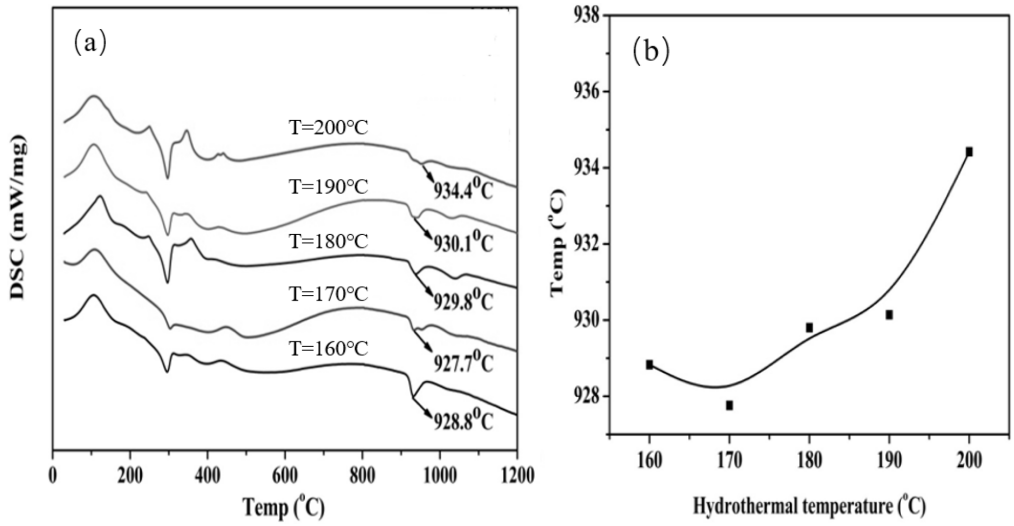

Fig. 8. (a) DSC curves of powders prepared at different hydrothermal temperatures; (b) YAG synthesis temperature-hydrothermal temperature curve.

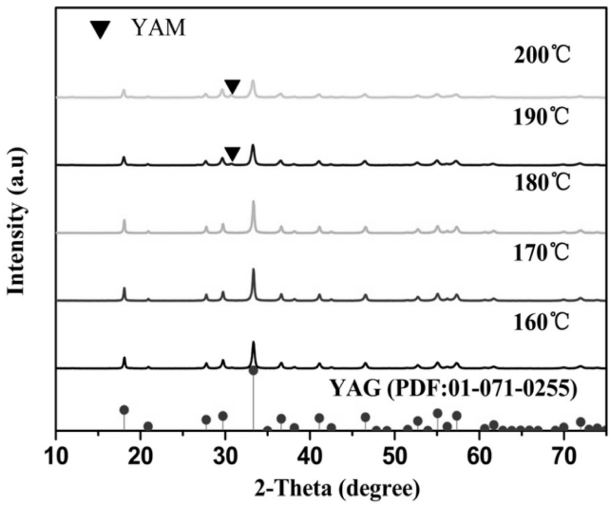

Fig. 9. XRD patterns of calcined ultrafine powders prepared at different hydrothermal temperatures.

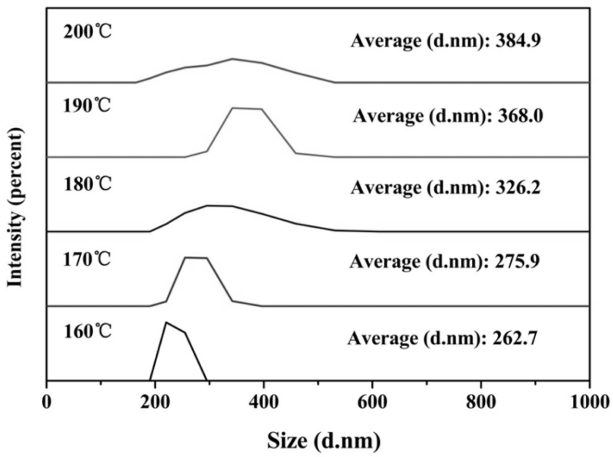

Fig. 10. Effect of different hydrothermal temperature on particle size of YAG ultrafine powder.

Fig. 8a is the DSC curve of the powder. An endothermic peak appears at about 150 ºC because of the decomposition of the hydroxide of Y and Al and the evaporation of the adsorbed water. An exothermic peak appears at approximately 940 ºC, and no weight loss is observed because of the crystallization of amorphous powder into the YAG phase. Fig. 8b shows the relationship between the synthesis temperature of YAG and the temperature of microwave hydrothermal reaction. When the temperature of microwave hydrothermal reaction is 170 ºC, the YAG crystal phase produces a low temperature of 927.7 ºC.

Fig. 9 illustrates the XRD pattern of the YAG precursor after calcination via microwave hydrothermal decomposition. The intermediate-phase YAM of YAG occurs when the hydrothermal temperatures are 190 ºC and 200 ºC, and only the pure cubic-phase ultrafine YAG powder can be produced at the hydrothermal temperatures of 170 ºC and 180 ºC, respectively, and the water is hot. The ultrafine YAG powder prepared at 170 ºC has a lower synthesis temperature and a calcination temperature of only 927.7 ºC. Therefore, at 170 ºC, it can provide the energy required for the decomposition of the YAG precursor hydroxide precipitate into the corresponding oxide to form a crystal phase [22-24]. As the temperature continues to increase, the energy also increases, so the formed grains grow and the particle size becomes coarse. Consequently, the calcination temperature of the ultrafine YAG powder further increases. When the hydrothermal temperatures are 190 ºC and 200 ºC, the intermediate-phase YAM of YAG appears because the crystal phases of Al_2O_3 and Y_2O_3 appear in microwave hydrothermal reaction in the YAG precursor. Large amounts of Al_2O_3 and Y_2O_3 decompose, and solid phase reaction is formed during calcination. The intermediate-phase YAM of YAG

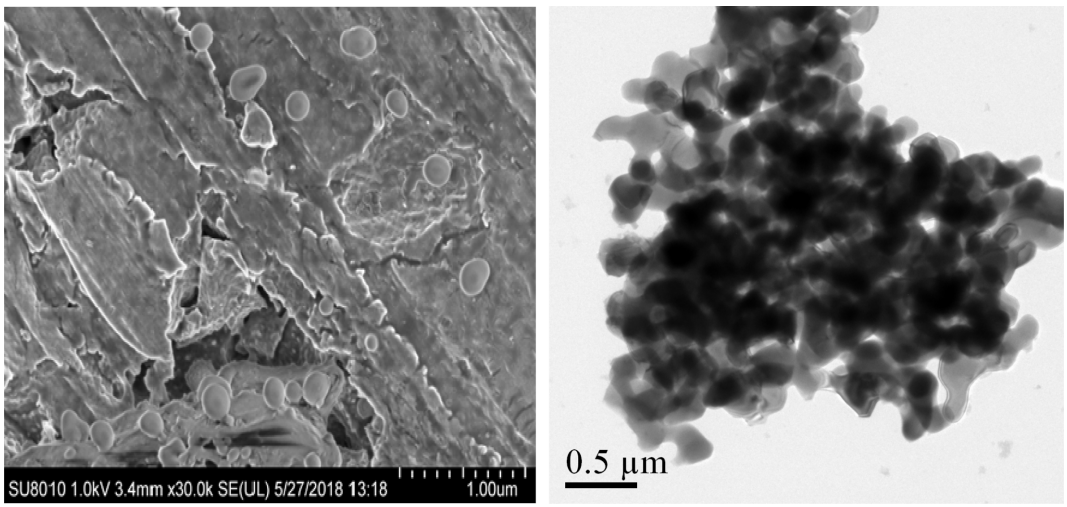

Fig. 11. SEM and TEM patterns of YAG ultrafine powder.

inevitably occurs because the solid phase reaction in YAG is gradual. Under other conditions, at the microwave water temperature of 170 ºC, a pure cubic-phase ultrafine YAG powder can be synthesized at a low calcination temperature (927.7 ºC).

Fig. 10 shows a chart of the analysis of the particle size of the ultrafine YAG powder, indicating that the particle size of the prepared ultrafine YAG powder also continuously increases as the microwave water temperature increases because in the microwave. During the hydrothermal process, an increase in temperature causes the crystal grains to grow continuously, resulting in a large particle size of the synthesized ultrafine YAG powder and a high temperature of the crystal phase.

Fig. 11 illustrates a mixture of $Al(NO_3)_3$ and $Y(NO_3)_3$ calcined at 927.7 ºC under the experimental conditions of pH=9, $C(Al^{3+})$=0.42 mol·L^{-1}, and a microwave hydrothermal reaction temperature of 170 ºC. Fig. 11 also shows the scanning electron micrograph of the pure cubic-phase ultrafine YAG powder. The ultrafine YAG powder synthesized under the experimental conditions is approximately spherical, and the particle diameter is approximately 200 nm. It can be seen from the figure that the particle size of the powder is very small and has a spherical shape. Due to the small particle size, the powder has a large specific surface area, a high specific surface energy, and is prone to agglomeration.

Conclusion

YAG precursor oxide was synthesized using the microwave hydrothermal method. After calcination, the powder was analyzed, and the following conclusions were drawn: as the Al^{3+} concentration in the mother salt solution increased, the particle size of the prepared ultrafine YAG powder also increased. As the microwave hydrothermal temperature increased, the particle size of the prepared ultrafine YAG powder increased. With microwave hydrothermal synthesis, the process conditions of high-purity ultrafine YAG powder were pH=9, $C(Al^{3+})$=0.42 mol·L^{-1}, microwave hydrothermal reaction temperature=170 ºC, and calcination=928 ºC. The body had a particle size of about 200 nm, and its shape was almost spherical. This study provided a reference for the low-temperature synthesis of high-purity and high-performance ultrafine YAG powder.

Acknowledgments

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