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Ammonium sulfate and PEG composite surfactant to promote dispersibility of precursors and Y_2O_3 powders for transparent ceramics

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ABSTRACT

Well-dispersed Y_2O_3 powders were synthesized by a precipitation method from yttrium nitrate solution using $(NH_4)_2SO_4$ and PEG4000 as the composite surfactant, and the dispersion mechanism of the composite surfactant was investigated in detail. Furthermore, the grain size, densification, mechanical and optical quality of as-prepared ceramics were also discussed. The dispersion state of both precursors and Y_2O_3 powders could be remarkably effected by the mass ratio of $(NH_4)_2SO_4$ and PEG4000 during the liquid phase reaction process. When the function of electrostatic repulsion and the steric hindrance were combined properly, the aggregation state of powders could be effectively inhibited. With the addition of the mass ratio of 3:2 ($(NH_4)_2SO_4$ to PEG4000), fine powders (130 nm) with high dispersion state were obtained. Meanwhile the Y_2O_3 ceramics with an average grain size of 3.65 μm were fabricated by sintering at 1750 °C for 8 h.

1. Introduction

Nowadays, much attention has been paid to the Y_2O_3 transparent ceramic due to its excellent potential for numerous optical applications, such as laser materials, host materials for scintillators, lighting devices, missile domes and high-temperature resistance windows [1–7]. To fabricate Y_2O_3 transparent ceramics with high optical and mechanical performance, the powders with fine homogeneity and dispersion state were essential. In the past few decades, a considerable number of preparation methods for Y_2O_3 powders have been investigated, such as precipitation [8–13], sol–gel [14], combustion [15,16], spray pyrolysis [17,18], emulsion synthesis [19], and hydrothermal [20]. Among them, the precipitation method was widely adopted by virtue of its fine morphology and high sintering activity of powders [21,22]. However, because of the tiny particle size (nanoscale) and large specific surface area of powders produced by the precipitation method, the aggregation was inevitable. In fact, the aggregation was extremely harmful for the densification and optical quality of ceramics [3,23–26]. In order to inhibit the formation of aggregation, $(NH_4)_2SO_4$ has been widely utilized in the precipitation process as the surfactant [10,11,27,28]. As profoundly investigated, the presence of the SO_4^{2-} ions is beneficial for controlling the dispersivity, shapes and particle size of Y_2O_3 powders

[29]. During the liquid phase reaction, precursor particles which adsorbed SO_4^{2-} ions could keep a certain distance from each other because of the repulsive force produced by negative charges of SO_4^{2-} ions. In addition, SO_4^{2-} ions could decompose and release gas to disperse the Y_2O_3 powders during the high temperature calcination process [30]. But an excessive addition of SO_4^{2-} ions could hardly benefit to the dispersivity of Y_2O_3 powders. In contrary, due to the high decomposition temperature (> 1180 °C) [27], it is extremely difficult to decompose the residual sulfate completely during the high temperature calcination process. The residual S could bring about the impurity of powders and this could notably affect the optical quality of transparent ceramics. Thus, the additive amount of $(NH_4)_2SO_4$ should be appropriately cut down. However, the dispersion state of the powders would decrease a lot even if the addition amount of SO_4^{2-} ions was only slightly reduced. In the previous report [3], the agglomeration state of powders with 4 mol.% sulfate dosage (molar ratio of Y_2O_3) was 1.4 times larger than that of samples with 7 mol.% sulfate dosage. Furthermore, after being calcinated at 1200 °C for 4 h, about 0.1 mol.% S still remained in Y_2O_3 powders. Therefore, only by optimizing the additive amount of single SO_4^{2-} ions, the dispersion state and purity of precursors and Y_2O_3 powders were hard to be ensured at the same time. Thus, a new composite surfactant for synthesizing Y_2O_3 powders should

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be investigated. We thought that the lost dispersion effect caused by the reduction of SO_4^{2-} ions addition amount could be compensated by the other new surfactant. In this way, the residual S in the final products could be reduced and the dispersion state of powders would not decline. Whereas, up to date, many papers have still focused on using $(\text{NH}_4)_2\text{SO}_4$ as the surfactant rather than researching the new composite surfactant in preparing Y_2O_3 powders by the precipitation method.

PEG4000 (polyethylene glycol4000) is a non-ionic surfactant, which has excellent water solubility and stability [31]. Meanwhile, it is not susceptible to electrolytes and acids or alkalis. It has been extensively used in many fields such as environmental remediation, nanofillers, cosmetics and surfactants [32–34]. Lots of ether bonds (–O–) in macromolecule chains and hydroxyl (–OH) of precursors will be combined to form hydrogen bonds. PEG4000 could be coated outside the surface of precursors, and the hydroxyl at the end of the macromolecule chains would enter the water and swim to keep the dispersibility [35]. During the formation and growing up of precursors, the long macromolecule chains provided the function of isolating effects, which could keep the distance of precursors from aggregating. Therefore, if the functions of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000 could be properly combined, the above problems about the dispersibility and impurity would be effectively resolved.

In this work, we reported the fabrication of Y_2O_3 powders and transparent ceramics by using $(\text{NH}_4)_2\text{SO}_4$ and PEG4000 as the composite surfactant during the liquid reaction process. The dispersion mechanisms of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000 were examined. Meanwhile, the effects of mass ratio of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000 on the morphology of precursors and powders were investigated in detail. The corresponding Y_2O_3 ceramics were sintered at 1750 °C for 8 h from the Y_2O_3 powders, and their densification, grain growth, mechanical and optical quality of ceramics were also discussed.

2. Experimental procedures

2.1. Synthesis of nano Y_2O_3 powders

$\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99.99%, analytical grades, Aladdin chemicals, Shanghai, China) was used as the starting material. Ammonia water ($\text{NH}_3 \cdot \text{H}_2\text{O}$, analytical grades, Aladdin chemicals, Shanghai, China) and ammonium hydrogen carbonate (NH_4HCO_3 , analytical grades, Aladdin chemicals, Shanghai, China) were used as the mixed precipitants. Firstly, $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ was diluted with distilled water until the Y^{3+} concentration reached 0.15 mol/L. Then, ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$, Sinopharm Chemical Reagent Co., Ltd, 99.0%) and polyethylene glycol4000 (PEG4000, analytical grades, Aladdin chemicals, Shanghai, China) was added into the $\text{Y}(\text{NO}_3)_3$ aqueous solution as the composite surfactant. The mass ratio of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000 ranged from 5:0–0:5, and all samples were respectively abbreviated as S5P0, S4P1, S3P2, S2P3, S1P4, S0P5, in which the number represented the mass ratio of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000. The total mass of the composite surfactant was kept at 5 wt% of that of $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$. After that, the mixed $\text{NH}_3 \cdot \text{H}_2\text{O}$ and NH_4HCO_3 precipitant (molar ratio 1:1.5) was sprayed into the Y^{3+} ions solution at a rate of 5 mL/min under the agitation rate of 440 rpm. The ultimate pH value of the reaction system was controlled at 8.0. After the precipitation reaction, the precipitate slurry was aged for 12 h, and then filtrated and cleaned with deionized water for 3 times, following with alcohol for 3 times. Then the as-prepared precursors were dried at 55 °C for 24 h in an oven. After being crushed and sieved, the precursors were calcined at 1250 °C for 4 h in air to synthesize Y_2O_3 powders.

2.2. Fabrication of Y_2O_3 ceramics

The as-prepared Y_2O_3 powders were uniaxially drying pressed into 16 mm diameter pellets at 25 MPa for 2 min. For the further densification, the green pellets were further cold isostatically pressed (CIPed)

at 200 MPa for 5 min. After removing the residue organics by being calcined at 800 °C for 4 h in a muffle furnace, the green bodies were sintered at 1750 °C for 8 h in a vacuum furnace under 10^{-3} Pa. Then the ceramics were air annealed at 1300 °C for 10 h. After annealing, they were all mirror polished on both sides to 2.0 ± 0.08 mm.

2.3. Characterization

Phases of Y_2O_3 powders were identified by X-ray diffraction method (XRD, D2, Bruker, Germany). Morphologies of precursors were performed by a field emission scanning electron microscope (FESEM, S4800, Hitachi, Japan). The polished discs were etched in H_3PO_4 at 85 °C for 5 min for the observation of the microstructures. The morphologies of Y_2O_3 powders and microstructures of sintered ceramics were observed by a scanning electron microscope (SEM, JSM-6510, JEOL, Japan). The particle size distributions of Y_2O_3 powders and grain size of ceramics were counted using the Nanomeasure. The hardness of ceramics was measured by the Vicker's indentation test machine (HVT-1000, Shanghai, China). The relative densities of Y_2O_3 ceramics were measured by the Archimedes method. The specific surface area of powders was measured by the BET method (Tristar II 3020, Micromeritics Instrument, America), and the equivalent particle size (D_{BET}) of Y_2O_3 powders can be calculated according to the following equation:

$$D_{\text{BET}} = 6/(\rho S_{\text{BET}})$$

where ρ is the theoretical density (5.031 g cm^{-3}) of Y_2O_3 and S_{BET} is the measured specific surface of the powder.

3. Results and discussion

3.1. Characteristics of precursors

Fig. 1 presents the morphologies of precursors adopting different mass ratios of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000. The precursors of the S5P0 and S4P1 sample (Fig. 1(a) and (b)) exhibited the severe aggregation state, in which non-uniform particle could be observed. In contrast, the S3P2 sample (Fig. 1(c)) showed the better dispersivity and homogeneity, i.e., more spherical particles with a uniform particle size distribution could be found, which was attributed to the combined effect of $(\text{NH}_4)_2\text{SO}_4$ and PEG4000. However, with further decreasing $(\text{NH}_4)_2\text{SO}_4$ and increasing PEG4000, the aggregation state of powders aggravated a lot. It was found that many flake-shaped films existed in the samples of S2P3 and S1P4 (Fig. 1(d) and (e)), and notably in the S0P5 sample (Fig. 1(f)). The reason is that, due to the reduction of $(\text{NH}_4)_2\text{SO}_4$, the repulsive force between precursor particles becomes weaker. Simultaneously, the twine of macromolecule chains produced by the redundant PEG4000 is also an important reason. When the single PEG4000 was added into the reaction system, due to the lack of the repulsive force and the excessive twine of macromolecule chains, the dispersion state became worse. To sum up, the optimal mass ratio of the two kinds of surfactants is in the middle of all samples (the S3P2 sample) rather than endpoints (the S5P0 or S0P5 sample), which shows the combination effect of PEG4000 and $(\text{NH}_4)_2\text{SO}_4$ in the process of precipitation.

3.2. Characteristics of Y_2O_3 powders

Fig. 2 depicts the XRD diagrams of the obtained Y_2O_3 powders. After being calcined, the sharp characteristic diffraction peaks corresponding to Y_2O_3 phase could be detected, indicating the good crystallinity of Y_2O_3 powders. Impurity phase could not be detected in the patterns. The grain sizes of calcined Y_2O_3 powders were calculated according to the Scherrer formula, and the corresponding results were listed in Table 1. As been seen from the Table 1, with decreasing $(\text{NH}_4)_2\text{SO}_4$ and increasing PEG4000, the grain size of powders mainly firstly rose and then declined. The reason was that, with the combination of $(\text{NH}_4)_2\text{SO}_4$

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