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Effect of surfactant concentration and solvent used for washing in the preparation of Yb:Y_2O_3 transparent ceramics

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Nanoparticles of Yb:Y_2O_3 have been synthesized by co-precipitation synthesis routes. The $(NH_4)_2SO_4$ surfactant was added to $Y(OH)_3$ precipitate during synthesis to control the size, morphology, agglomeration and phase of nanoparticles. The dried precipitates were calcined at 900 °C to obtain the desired cubic phase Yb:Y_2O_3 nano particles. In the present study we reported the effects of surfactant concentration on size, morphology, agglomeration and phase of Yb:Y_2O_3 nanoparticles. However the addition of surfactant is not enough to get non-agglomerated nanoparticles. The extracted precipitate should be washed in proper solvent to avoid formation of hard agglomerate during drying. Hence the effects of washing solvent (i.e. water and methanol) on agglomeration and transparency were also reported. The transparency of the sintered pellets, prepared by varying the surfactant concentration and washing solvent, was evaluated. A transparency of $\sim$ 80 % at 1500 nm was achieved in 1 mm thick Yb:Y_2O_3 ceramic pellet by optimization of the surfactant and washing solvent.

Keywords: transparent ceramics, Yb:Y_2O_3, surfactant.

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1. Introduction

Nanotechnology and sintering techniques can be utilized in combination for the fabrication of rare earth-doped yttria transparent ceramics because the high surface area of the nanoparticles facilitates sintering, permitting one to reach full density more rapidly at temperatures far below the melting point of the material [1,2]. The fabrication of Yb doped Y_2O_3 transparent ceramic involves the synthesis of non-agglomerated nanoparticles of Yb doped Y_2O_3 and sintering of pressed pellets (prepared using a nanopowder) at high temperatures under low pressure [3]. Of the different methods adopted for the preparation of non-agglomerated nanoparticles, the co-precipitation method has been widely used for nanopowder synthesis because the particle size and morphology can be efficiently controlled [4].

However, in case for the synthesis of Yb:Y_2O_3 nanopowders using co-precipitation method, the $Y(OH)_3$ precipitate underwent severe agglomeration during drying. This agglomeration led to poor sinterability for the Yb:Y_2O_3 nanopowder obtained after calcination. Thus, special measures were adopted during precipitation, such as addition of surfactant to create non-agglomerated nanoparticles. It is reported that the surfactant $(NH_4)_2SO_4$, which adsorbs on the surface of the $Y(OH)_3$, hinders the bonding between particles [5,6]. It has also been shown that the morphology of precursors was affected by the $[(NH_4)_2SO_4]$/[Nd:Y_2O_3] ratio (measured by weight) [7]. However, the effect of surfactant concentration on the crystallographic phase of the prepared nanopowder has not been investigated yet. Similarly, the solvent used to wash the precipitate plays a crucial role in the agglomeration of nanoparticles. Some solvents remove the surfactant from the particle surface and cause hard agglomeration during drying of precipitate [8]. The chemical nature of the washing solvent may be different for dissimilar oxide nano particles.

Thus, the aim of this work is to study the effect of surfactant concentration on the size, morphology, agglomeration and phase of Yb:Y_2O_3 nanoparticles obtained using the co-precipitation method as well as its influence on the transparency of sintered pellets incorporating these nanoparticles. The effect of the solvent used for washing the precipitate on the transparency of the pellet is also evaluated.

2. Experimental

A 0.2 M mother solution of Yb doped (1 mol%) Y_2O_3 (using Yb_2O_3 and Y_2O_3 powders of purity 99.99 %; make: Alfa Aesar) was prepared by dissolution with dilute nitric acid. The mother solution was titrated with 1 M aq. ammonia by normal striking. A white jelly like basic Yb:$Y(OH)_3$ precipitate was formed during the titration which was controlled by measuring online pH. After aging, the hydroxide precipitate was divided into five equal parts. In order to investigate the effects of surfactant concentration on the transparency of sintered Yb:Y_2O_3 pellets, $(NH_4)_2SO_4$ surfactant having 5, 10, 15, 20 and 30 wt% concentration was added to each part of the precipitate separately i.e. 5 wt% of $(NH_4)_2SO_4$ solution were added into first part of hydroxide precipitate and so

on. Then, the samples were washed using methanol and dried in an oven. Calcination was performed at 900 °C for 4 h in air. Then, the calcined powders were pressed under a pressure of 150 MPa using a uniaxial press. To show only the effect of morphology and particle size distribution on pellet's transparency, a cold isostatic press (CIP) was not used for pellet pressing. The pellets were sintered at 1750 °C for 5 h using a high temperature high vacuum furnace with a tungsten mesh heating element (Hind High Vacuum High Temperature Furnace, Hind High Vac. Co (P). Ltd., Bangalore). Finally, the pellets were polished using 300 nm alumina powders. Phase identifications of the Yb (1 mol%):Y_2O_3 nanopowders prepared using different surfactant concentrations were performed using X-ray diffractometry (XRD) (Cu-Kα, Rigaku) and morphology was observed by field emission scanning electron microscope (FESEM, Zeiss).

3. Results and discussion

X-ray diffraction (XRD) patterns of Yb(1 mol%):Y_2O_3 powders prepared using different surfactant concentrations are shown Fig. 1. The XRD patterns of these powders are in good agreement with that of Y_2O_3 (Standard JCPDS, Card no. 41-1105) crystal [9, 10]. The results revealed that the precursor transformed to cubic phase of Y_2O_3 crystals after calcination at 900 °C for 5 wt% and 10 wt% of surfactant concentration but for 15 wt% and above surfactant concentrations, it transforms to a mixed phase i.e. combination of cubic and monoclinic phase Y_2O_3 crystal. The reflection from the monoclinic phase (space group C2/m) (Y_2O_3: PDF 44-0399) appeared in the diffraction patterns for 15, 20 and 30 wt% surfactant concentration as shown in Fig. 1 [10].

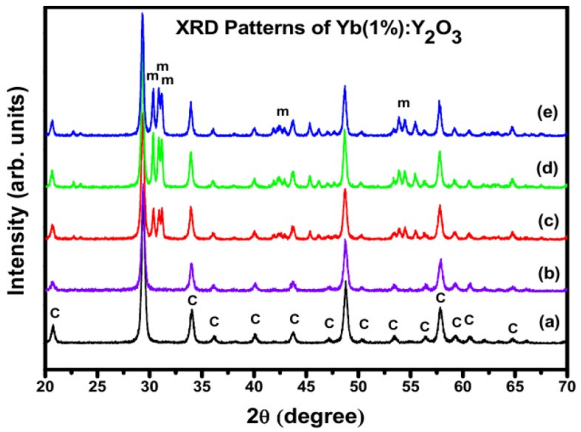

FIG. 1. X-ray diffraction patterns of Yb (1 mol%):Y_2O_3 nanopowders prepared using different surfactant concentrations: (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt % and (e) 30 wt%

Scanning electron micrographs of Yb (1 mol%):Y_2O_3 powder prepared using different surfactant concentrations are shown in Fig. 2. The morphologies of the Yb (1 mol%):Y_2O_3 nanoparticles are nearly spherical and are unchanged with variation in surfactant concentration. The existence of SO_4^{2-} on the surface of yttrium hydroxide precursors at comparatively high temperature inhibits volume diffusion and/or grain boundary diffusion, and then particle growth proceeds by surface diffusion or evaporation-condensation, which results in collapse of agglomerates into well-dispersed nanopowders [12].

Photographs of sintered pellets of Yb (1 mol%):Y_2O_3 prepared using different surfactant concentrations i.e. (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt% and (e) 30 wt% are shown in Fig. 3. As is clearly visible, the transparency of the sample (a) and (b) is so good, as to allow the easy reading of the underlying print, however, the transparency does seem to be decreased at higher surfactant concentrations. This may be due the transformation of the phase from cubic to a mixed one (as evident from Fig. 1).

Figure 4 shows the photograph of sintered pellet of Yb (1 mol%):Y_2O_3 prepared by using 5 wt% concentration of surfactant and water as the washing solvent. The simple change in washing solvent from methanol to water during synthesis would appear to lead to a drastic decrease in the transparency of the sintered Yb (1 mol%):Y_2O_3 pellet. This is due to the formation of high strength agglomerates after washing with the liquid having higher surface tension like water (greater than methanol) [13].

Using the above experimental conditions, we have successfully fabricated highly transparent Yb (1 mol%): Y_2O_3 ceramics pellets (transparency $\sim$ 80 % in visible-IR region without considering Fresnel's reflection).

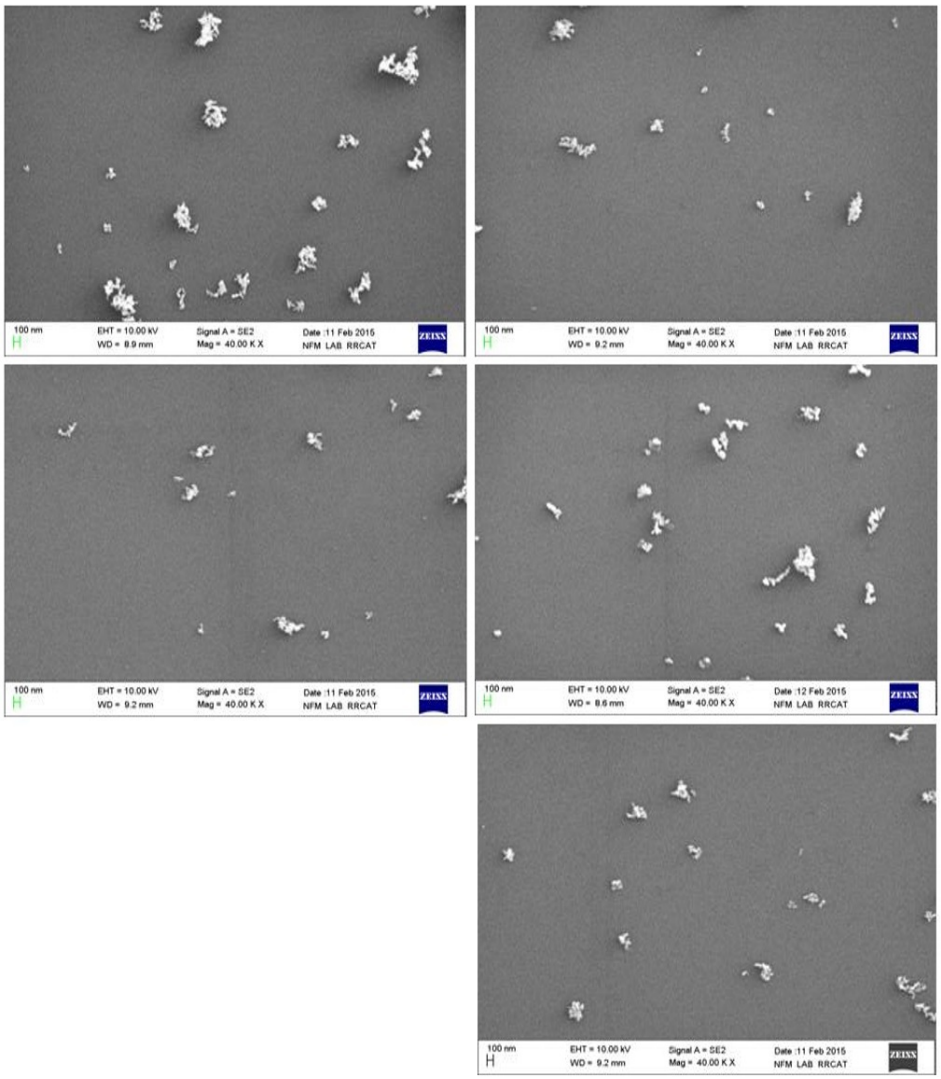

FIG. 2. FESEM images of Yb (1 mol%):Y_2O_3 nano powders prepared using different surfactant concentrations: (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt% and (e) 30 wt%

FIG. 3. Photograph of sintered pellets of Yb(1 mol%): Y_2O_3 prepared using different surfactant concentrations: (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt% and (e) 30 wt%

FIG. 4. Photograph of sintered pellet of Yb(1 mol%): Y_2O_3 prepared using 5 wt% of surfactant concentration and water as the washing solvent

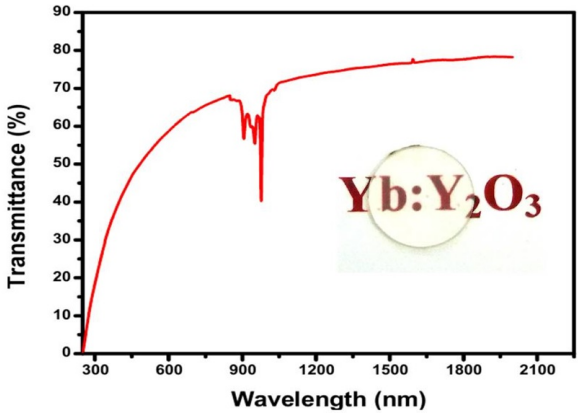

FIG. 5. Optical transmittance spectrum of a sintered pellet of Yb (1 mol%): Y_2O_3 and inset shows the photograph of transparent ceramic

4. Conclusion

The use of surfactant $(NH_4)_2SO_4$ during the synthesis of nanoparticles is helpful to control particle size and agglomeration. The excess concentration of it does not affect the particle morphology. Hence, the sinterability (which mainly depends on the size, agglomeration and morphology of the particles) of the all samples (prepared using different $(NH_4)_2SO_4$ concentrations) would remained same. Thus, the observed difference in transparency of Yb (1 mol%):Y_2O_3 sintered pellets is due to transformation of the phase from cubic to mixed (i.e. combination of cubic and monoclinic phase). Additionally, the surface tension of washing solvent affects the agglomeration during drying of the precipitate. The optimized synthesis parameters for Yb:Y_2O_3 nanoparticles i.e. 5 wt% surfactant concentration and methanol as washing solvent have been used to successfully fabricate highly transparent Yb (1 mol%):Y_2O_3 ceramics pellets.

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