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Synthesis and Characterization of Nanocomposite ZnO on SiO_2 Glass by Sol-Gel Method

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Nano-ZnO embedded SiO_2 glass was prepared by sol-gel method. During this study, zinc oxalate was prepared in aqueous medium using zinc acetate and oxalic acid as precursors. The thermo-gravimetric analysis was showed formation of ZnO at 400 ºC. Nano-size zinc oxide was obtained by thermal decomposition of aqueous mediated zinc oxalate at 450 ºC. The phase purity was confirmed by X-ray diffraction. Crystal size was determined from transmission electron microscopy and was found to be 15-25 nm for the aqueous mediated ZnO. Scanning electron microscope also revealed different nature of surfaces and microstructures for zinc oxide obtained in aqueous medium.

Key Words: Nano-ZnO, Nanocomposite, SiO_2 glass sol-gel.

INTRODUCTION

Zinc oxide is an important semiconductor, which has a direct band gap (3.37 eV at room temperature), large bond strength, large excitation binding energy about 60 meV and high melting temperature (2248 K)[1]. Zinc oxide is an exceptionally important material having applications in pigments, rubber additive, gas sensor, varistors and optical devices[2]. Zinc oxide nanoparticle embedded SiO_2 composites have attracted extensive research interests. It has been found that these materials have improved luminescence efficiency bulk ZnO material. Excellent nonlinear optical properties, saturable absorption and optical bistability have also been reported for these composites[3-9]. Various techniques have sol-gel, impregnation and magnetron sputtering, *etc.*[4,10,11]. The aim of this paper is to synthesize nano-ZnO embedded SiO_2 glass by a sol-gel process and to investigate its ultraviolet emitting characteristics.

EXPERIMENTAL

Sample preparation: The preparation of nano-ZnO embedded SiO_2 glass involves three steps: (1) formation of colloidal suspension of zinc hydroxide $Zn(OH)_2$, (2) mixing $Zn(OH)_2$ colloidal suspension with silica sol and (3) annealing of the precursor glass to obtain the nano-ZnO embedded glass.

The colloidal suspension of zinc hydroxide was formed by solution process at 90 ºC using zinc acetate dihydrate and sodium hydroxide, as source materials. All the chemicals were purchased from Aldrich Chemical Corporation and used without further purification. For synthesis, 6.57 g zinc acetate dihydrate was dissolved in 100 mL of de-ionized water under stirring at room temperature. Simultaneously, 3 M sodium hydroxide solution was added drop wise while stirring it continuously, resulting in a white milky precipitates.

$$Zn(CH_3COO)_2{\cdot}2H_2O + 2NaOH \rightarrow Zn(OH)_2 + 2CH_3COONa + 2H_2O$$

$$Zn(OH)_2 \rightarrow ZnO + H_2O$$

The solution was then transferred in a three-necked refluxing pot and refluxed at 90 ºC for 0.5 h. Before refluxing the solution, pH was measured as 13.2 by the expandable ion analyzer (EA 940, Orian). Then the colloidal suspension of $Zn(OH)_2$ and the silica sol were mixed and stirred for 20 min at different temperatures between 30 and 70 ºC. The final molar ratio of $Zn(OH)_2$ to SiO_2 was 1:20. Then the transformation of the sol to gel took place due to the interaction between negative and positive charge. Finally, silica gel containing $Zn(OH)_2$ was aged at room temperature for several days in air; after aging, the white powder was washed with methanol several times, followed by heat treatment process at 250 ºC for 12 h to form the nano-ZnO embedded SiO_2 glass.

Characterization: The thermal study of nano-ZnO embedded SiO_2 glass synthesized was carried out using Thermo Gravimetric Analyzer TGA-DTA, Perkin-Elmer Tyris diamond (SII) up to 600 ºC in air at the heating rate of 10 ºC/min. Nano-ZnO embedded SiO_2 glass prepared by decomposition of zinc hydroxide at 450 ºC was employed for the powder

XRD studies. A scanning electron microscope with EDAX (Philips-XL-30) was used to study the surface morphology of the zinc oxide powder. A transmission electron microscope (TEM) (JEOL-1200EX) was used to measure the particle size.

RESULTS AND DISCUSSION

XRD analysis: The $Zn(OH)_2$ colloidal suspensions were processed at different temperatures to form nano-ZnO particles[12]. Fig. 1 shows the XRD spectra of the $Zn(OH)_2$ colloidal suspensions processed at 250 ºC in the air. X-ray diffraction peaks of ZnO are observed for the sample processed at 200 ºC or above, which indicates that $Zn(OH)_2$ undergoes a decomposition process to form ZnO at temperatures above 200 ºC. The sharp peaks indicate that the products were well crystallized. The average size of the particles can be calculated by the Scherrer formula, *i.e.*,

$$d = \frac{0.9\lambda}{\beta \cos\theta}$$

where d is the average size of the particle, λ is the wavelength of the X-ray, β is the FWHM width of the diffraction peak and θ is the corresponding diffraction angle of the diffraction peak. According to the data in Fig. 1 and formula (1), the average particle size of the nano-ZnO is 15 nm for the sample processed at 200 ºC.

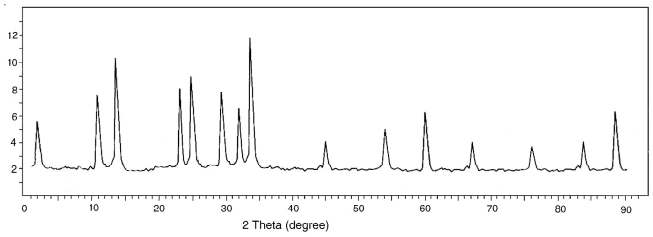

Fig. 1. XRD spectra of nanocomposite ZnO on SiO_2 glass

Thermal analysis: Fig. 2 depicts the TGA/DTA curves for decomposition of the nano-ZnO embedded SiO_2 glass synthesized in aqueous solvent. TGA showed a weight loss in two steps at 110 and 450 ºC and correspondingly DTG and DTA showed two endothermic peaks at these temperatures. The endothermic peak at 110 ºC was due to removal of water and the peak at 450 ºC was due to decomposition of zinc acetate as per reaction. More accurate decomposition temperature of $Zn(OH)_2$ is determined to be 110 ºC as from the result of the TGA data shown in Fig. 2.

Particle morphology by SEM: A typical SEM image of the nano-ZnO embedded SiO_2 glass is shown in Fig. 3. The average size of the nanocomposite is 20 nm. It is clearly, indicating that such nano-ZnO embedded SiO_2 glass was quite dense.

TEM microscopy: Fig. 4 represents the transmission electron microscopy (TEM) of aqueous mediated ZnO images showed agglomerated spherical like morphology mediated ZnO, where as nanocomposite were regular and spherical in shape. TEM study also shows that the solvent plays a key role in controlling the morphology of nanocrystalline zinc oxide[13]. In water, the reaction disperses more homogeneously and the growth of crystal nucleus is subjected to less

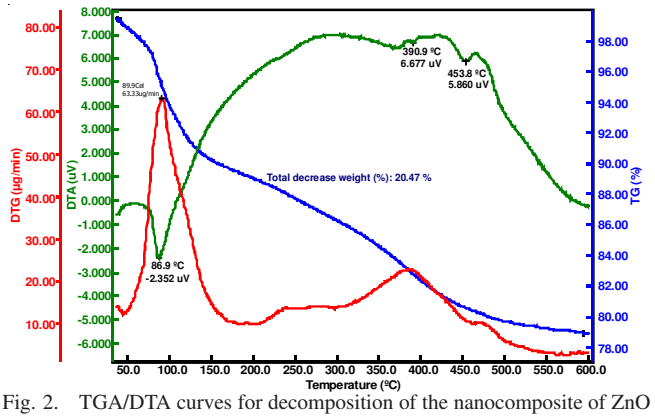

Fig. 2. TGA/DTA curves for decomposition of the nanocomposite of ZnO embedded SiO_2 glass

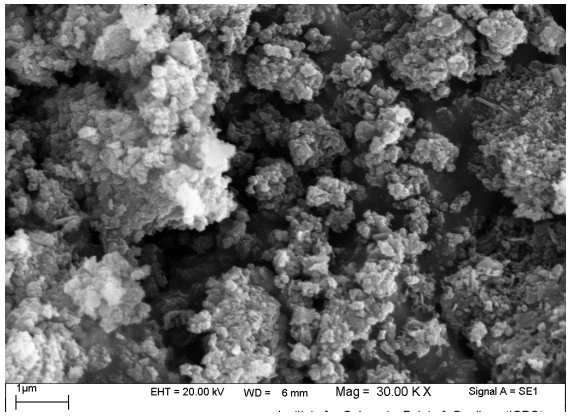

Fig. 3. SEM image of the nano-ZnO embedded SiO_2 glass

confinement in boiling droplet of solvent. Therefore, it is liable to form mixed rectangular and trigonal morphology in water. Further oriented growth of ZnO crystal is apparent with slightly larger size.

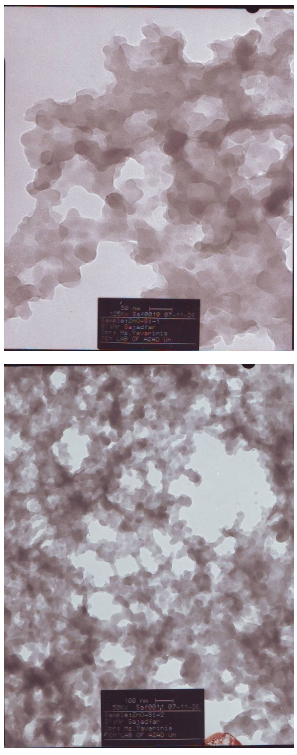

Fig. 4. Micrographs (TEM) of nano-ZnO embedded SiO_2 glass

Conclusion

Nano-ZnO embedded SiO_2 glass is fabricated by sol-gel chemical method. It is found that pure and intense ultraviolet emission from nano-ZnO embedded glass can be obtained by controlling the preparation conditions and parameters, such as the pH value of the gel, the gelation temperature and the processing temperature.

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