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Sol-gel synthesis of thermoluminescent Cd-doped ZnTe nanoparticles

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The Cd (3 wt%)-doped ZnTe nanoparticles with average size of 5 nm and band gap energy of 3.15 eV were synthesized by sol-gel method. The optical properties (such as band gap energy) and the size distribution of the nanoparticles have been investigated by using UV-Vis spectroscopy and transmission electron microscopy, respectively. Then, thermoluminescence property of the nanoparticles exposed to various doses of gamma radiation of Cs^{137} has been studied. The thermoluminescence glow peak of the nanoparticles showed a displacement from 290 to 230°C by increasing the dose of gamma radiation from 7.5 to 67.5 mSv. The thermoluminescence intensity of the synthesized Cd-doped ZnTe nanoparticles also showed a linear dose response.

Keywords: ZnTe, Sol-gel, Nanoparticles, Thermoluminescence

1 Introduction

During the last two decades, research on quantum size semiconductor particles has increased enormously due to their exciting novel properties. II-VI semiconductor nanoparticles are currently of great interest for their practical applications such as zero dimensional quantum confined materials and in optoelectronics and photonics. Numerous reports are available in the literature on synthesis techniques as well as potential applications of nano-sized semiconductor particles[1-5]. In the present paper, the characterization of Cd (3 wt%)-doped ZnTe nanoparticles have been studied and synthesized by sol-gel method. Most physical and chemical properties of these nanomaterials are sensitively dependent on their size and shape, and scientists are still focusing on developing of simple and effective methods for fabrication of nanomaterials with controlled size, morphology and doping[6,7]. Transmission electron microscopy is an irreplaceable method to characterize the structure of nanocrystals. Lattice image contrast and Z contrast provide complimentary information. Lattice imaging probes the crystalline core of particles with planes oriented perpendicular to the electron beam; Z contrast refers to the diffuse scattering of the electron beam being proportional to the atomic number (Z) of the element, and contrast provides contrast in the disordered/ misoriented regions. Thermally stimulated luminescence, commonly called thermoluminescence (TL), takes place when a material is exposed to ultraviolet (UV) or ionizing radiation, and later thermally stimulated. The radiation exposure of the material creates electron-hole pairs which eventually get trapped in localized trap-states existing inside the band gap of the material, and most of the charge carriers remain trapped afterwards. The localized trap states in a material are generated due to the existing intrinsic defects or the defects generated by radiation exposure. The thermal stimulation frees the trapped electrons (holes) which travel through the conduction (valence) band, opposite charges recombine, and light emission is produced through a radiative recombination process. The charge trapping and thermally stimulated recombination mechanisms are important for radiation dose assessment. In a good dosimetric material, the number of trapped charge carriers and in consequence the integrated intensity of the thermally stimulated light emission (recombination) is proportional to the exposure dose. In the present paper, the synthesis, TL properties, and dosimetric behaviour of Cd (3 wt%)-doped ZnTe nanoparticles, have been reported.

2 Experimental Details

Cd-doped ZnTe nanoparticles were prepared by sol-gel as a common method for synthesis of II-VI elements-based compounds. The method is based on the chemical replacement reaction between chemical compounds providing metal ions (here, Zn^{2+}) and those containing chalcogenide ions[3] (here, Te^{2-}). Zinc acetate ($[Zn(OCOCH_3)_2].2H_2O$) (E Merck Co., Germany) and sodium telluride (Na_2Te) (E Merck Co, Germany) were used as the source materials of Zn^{2+} and Te^{2-}ions. To synthesize Cd (3 wt%)-doped ZnTe nanoparticles, at first, 100 mL freshly prepared $[Zn(OCOCH_3)_2].2H_2O$ solution (0.2 M) was magnetically stirred for 30 min at room temperature. Then, 100 mL Na_2Te solution (0.1 M) in equal volume of methanol and water, 1.06 g poly vinyl pyrrolidone (PVP) and 3 mL cadmium chloride (0.1 M) were added into the zinc acetate solution while stirring. After 3 h, a cream coloured precipitate was obtained. To remove any type of unreacted chemicals or impurities, the precipitate was washed three times with methanol and dried in air at 60°C.

Optical absorption spectra of the Cd-doped ZnTe nanoparticles were obtained using a UV-Visible spectrophotometer (Lamba, model 45) in the wavelength range 300-800 nm. To analyze size and size distribution of the nanoparticles, transmission electron microscopy (TEM) was performed. Aliquots were taken from the growth solution. Then, methanol and butanol were added to induce flocculation of the nanoparticles and to help further mixing the methanol and the growth solution. Precipitated nanoparticles were separated from the supernatant by centrifugation and then redispersed in hexane. The obtained solution was drop-casted onto copper grids with carbon support and then allowed for a slow evaporation of the solvent in air at room temperature. TEM images were acquired using a Philips EM208 operating at an acceleration voltage of 100 kV. The thermoluminescence dosimetry of nanoparticles is taken with the help of Harshaw 4500 with a heating rate of 10 K/s. Equal amount of the nanoparticles (20 mg) was taken for each measurement. No irradiation was carried out before the experiments.

3 Results and Discussion

Fig. 1 shows the optical absorption spectra of Cd-doped ZnTe nanoparticles in the wavelength range 300-700 nm. It is seen that the spectra are featureless and no absorption occur in visible region (390-800 nm). An absorption edge was obtained at shorter wavelengths in UV region at 348 nm for the Cd-doped ZnTe nanoparticles.

Using the classical relation $\alpha h\nu = A(h\nu - E_g)^{n/2}$ (in which α is the absorption coefficient of sample, $h\nu$ the incident photon energy, A the constant and E_g is constant that is equal to 1 for direct band gap semiconductor[8]), the optical band gap energy of semiconducting nanoparticles can be evaluated. Fig. 2 shows the plot of $(\alpha h\nu)^2$ versus $h\nu$ for estimating E_g of the Cd-doped ZnTe nanoparticles through extrapolating the linear region of the curve to zero absorption coefficient. Using this method, the E_g of the Cd-doped ZnTe nanoparticles was estimated ~ 3.15 eV which is above the normal values of ZnTe (2.26 eV). It is known that the absorption edge systematically shifts to the lower wavelengths or higher energies with decreasing of size of the nanoparticles. An increase in the band gab is observed due to the quantum confinement effects in the nanoparticles[9]. In addition, using the obtained E_g, it is possible to estimate the size of nanoparticles through the following relation[10]:

$$E_g^* = E_g^{\text{bulk}} + \frac{h^2}{8er^2}\left[\frac{1}{m_e} + \frac{1}{m_h}\right] - \frac{18e^2}{4\pi\varepsilon\varepsilon_0 r} \quad \ldots(1)$$

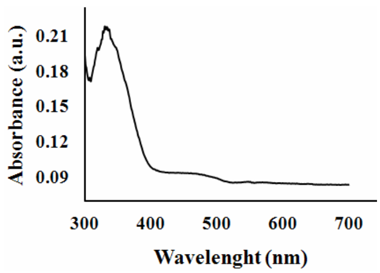

Fig. 1 — UV-Visible absorption spectrum of Cd-doped ZnTe nanoparticles in ethanol (~1 g/mL)

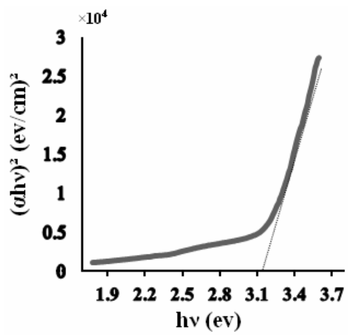

Fig. 2 — Plot of $(\alpha h\nu)^2$ versus incident photon energy ($h\nu$) for Cd-doped ZnTe nanoparticles

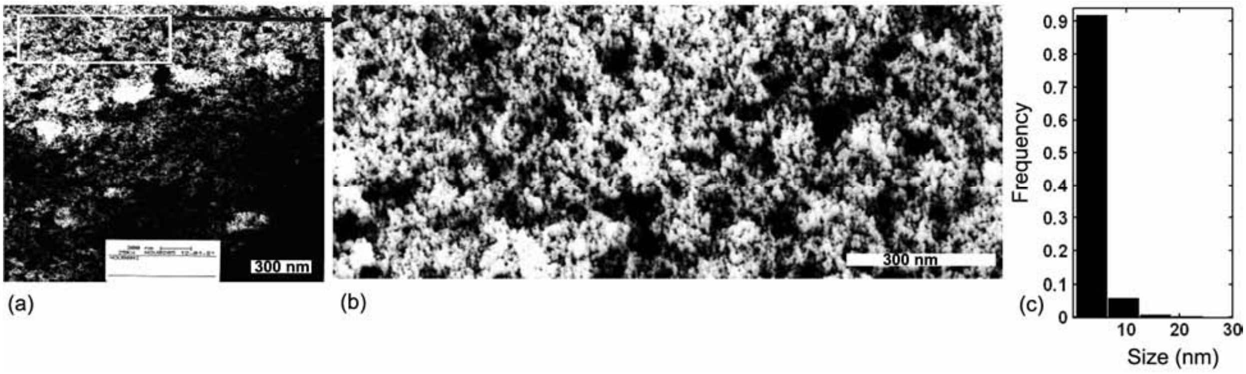

Fig. 3 — TEM images (a, b and c) particle size distribution of the Cd-doped ZnTe nanoparticles. (b) Shows a close window of the nanoparticles shown in (a). The number of particles considered in (c) was ~90

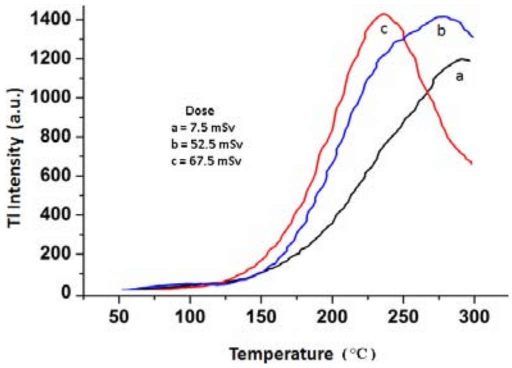

Fig. 4 — TL glow curve of Cd-doped ZnTe nanoparticles after exposing to (a) 7.5,(b) 52.5 and (c) 67.5 mSv doses of gamma irradiation of Cs^{137}

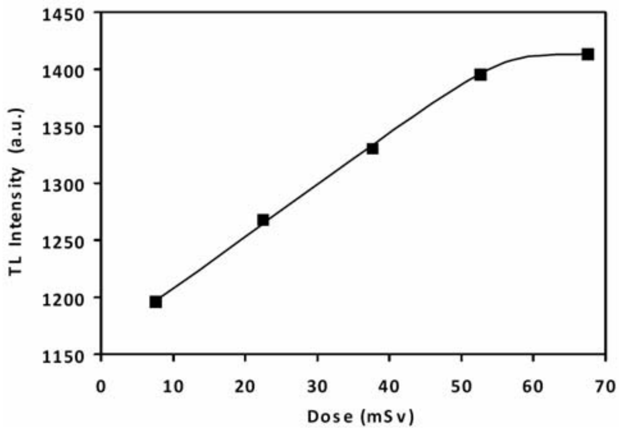

Fig. 5 — TL intensity of Cd-doped ZnTe as a function of doses of gamma radiation of Cs^{137}

where E_g^* is band gap energy of nanoparticles, h is the Planck's constant. Here, by substituting E_g^*=3.15 eV, E_g^{bulk} =2.26 eV, ε =8.65, m_e=0.24 m_0, m_h=0.45 m_0 the average size of the nanoparticles was estimated as ~ 1.7 nm.

The morphology of the nanoparticles and their size distribution have been investigated by using TEM, as shown in Fig. 3. It is seen that a homogeneous distribution of the nanoparticles prevails over a wide range of the sample. The mean diameter of the nanoparticles, obtained from the TEM images, was about 5 nm. In addition, most of the synthesized nanoparticles (~90%) showed sizes smaller than 7 nm and no nanoparticles with sizes larger than 20 nm was found.

Figure 4 shows the TL glow curve for the Cd-doped ZnTe nanoparticles exposed to various Cs^{137} gamma doses. The nanomaterial powder showed signal peak which moved from 290 to 230°C by increasing the dose from 7.5 to 67.5 mSv. This peak is very broad which may consist of several overlapping peaks. The glow curve structure and the peak shape did not change markedly by increasing the dose. This indicated the stability of the TL property of the Cd-doped ZnTe nanoparticles even at high doses (up to 67.5 mSv). Figure 4 also shows that the peak of TL spectra occurred at lower temperatures by increasing the dose of gamma irradiation. In addition, the TL intensity increased by increasing the dose of gamma irradiation. These indicate the ability of the nanoparticles in detection and discrimination of gamma sources having various doses.

In Figure 5, the TL intensity is plotted as a function of dose. A nearly linear response is observed for exposure doses ranging from 7.5 to 52.5 mSv.

4 Conclusions

The Cd (3 wt%)-doped ZnTe nanoparticles with average size of 5 nm and band gap energy of 3.15 eV were successfully synthesized by using sol-gel method. The synthesized nanoparticles exhibited thermoluminescent property after exposing to gamma irradiation of Cs^{137} source. It was found that by increasing the dose of gamma radiation from 7.5 to 67.5 mSv, the thermoluminescence glow peak of the nanoparticles significantly shifted from 290 to 230°C. The thermoluminescence intensity of the synthesized Cd-doped ZnTe nanoparticles also presented a linear response to variation of the dose of gamma radiation in the range 7.5-52.5 mSv. These properties indicate capability of the synthesized nanoparticles in reliable detection and discrimination of gamma sources with various doses.

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