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Thermoluminescence study of gamma irradiated BCNO nanophosphor

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ABSTRACT

Boron carbon oxynitride (BCNO) is a low Z_{eff} (≈ 6.8), near tissue equivalent material. Nanocrystals of the BCNO:Dy and BCNO:Cu were synthesized by solid state reaction method using different concentrations of dopants. TL characteristics of the synthesized BCNO and BCNO:Dy material doped with 2500 ppm concentration of Dy were studied and compared with each other. It is observed that the doping of BCNO with Cu or Dy leads to the quenching of its TL properties. BCNO exhibits a linear response from 1×10^2 to 2×10^3 Gy whereas BCNO:Dy exhibits a linear response from 1×10^2 to 1×10^3 Gy of gamma radiations. Fading and reproducibility of phosphors are also studied and it is found that the undoped BCNO shows better TL results than that of Dy doped BCNO phosphor. The main disadvantages of this material are its low thermal stability and high fading, so further studies are needed for this material before concluding it as the best dosimeter for gamma doses.

1. Introduction

Boron carbon nitride (BCN) compounds are the promising technological materials with many favorable physical and chemical (Badzian, 1981; Yu and Wang, 1998; Popov et al., 1998; Yao et al., 1999) semiconducting, intercalation and lubricating (Kawaguchi, 1997) properties. Different ways were used to synthesize hexagonal boron carbon nitride (h-BCN) materials (Kouvetakis et al., 1987; Bessmann, 1990; Moore et al., 1989; Sasaki et al., 1993; Hubacek and Sato, 1995; Riedel et al., 1991; He et al., 2000). It has been proposed that the compounds of BC_xN might be ideal candidates for solid state lightening materials but on the other hand such applications had disadvantages of low emission yield as well as difficulties in large scale production of BC_xN. So, recently boron carbon oxynitride (BCNO) phosphor with high yield was reported (Ogi et al., 2008). BCNO is an efficient and environmental friendly phosphor with emission covering the full visible and the near ultraviolet region (Liu et al., 2009). BCNO phosphor was synthesized using one step liquid process (Ogi et al., 2008), walker type multianvil apparatus (Leinenweber and Parise, 1995). Very recently chemical solid state reaction method was used by our group to synthesize BCNO without dopants (Singh and Chopra, 2011). Further TEM and EELS techniques were used for structural and chemical characterization of this material (Garvie et al., 1999). PL analysis under the excitation longer UV or blue-light wavelength was also reported (Kaihatsu et al., 2009). In the present work, dosimetric aspects of BCNO have been studied using thermoluminescence technique. The effective atomic number of BCNO is found to be 6.8 that is close to $Z_{eff}=7.4$ of biological tissue. Effective atomic number is a term that is similar to atomic number but is used for compounds and mixtures of different materials (such as tissue and bone) rather than for atoms. Z_{eff} can be calculated using the formula (Mayneord 1937)

$$Z_{eff} = \sqrt[2.94]{f_1\times(Z_1)^{2.94}+f_2\times(Z_2)^{2.94}+f_3\times(Z_3)^{2.94}+f_4\times(Z_4)^{2.94}}$$

where f_n=fraction of total number of electrons associated with each element, Z_n=atomic number of each element.

In the present study nanocrystalline BCNO phosphor was synthesized using solid state reaction method. The formation of material was confirmed by XRD and TEM. Thermogravimetric analysis (TGA) was performed to study the thermal stability of synthesized material. Its TL response to γ-rays has been studied. The TL properties of developed BCNO phosphor examined in this study includes annealing conditions, glow curve shapes, TL sensitivity, dose response, fading and reproducibility. The roles of dopants (Dy,Cu) on TL response have been examined. Moreover TL studies of Dy doped BCNO are also compared with that of pure BCNO. These results may be helpful in the development of tissue equivalent TL nanocrystalline detectors best suited for wide high range of radiation exposures.

2. Experimental

2.1. Synthesis

BCNO compound was prepared by using boric acid & melamine as the starting materials. The details of synthesis procedure are reported elsewhere (Singh and Chopra, 2011). BCNO:Cu was

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obtained by adding the appropriate amount of $Cu(NO_3)_2 \cdot 3H_2O$ to the starting mixture and to obtain BCNO:Dy, appropriate amount of $DyCl_3 \cdot 6H_2O$ was added. The synthesized phosphor thus obtained was then crushed to get the powder form. Finally the nanocrystalline powder was annealed at 300 °C for 30 min in the crucible in the presence of air and was quenched by taking the crucible out of the furnace and placing it on a metal block. The synthesized phosphor was then used for studying its TL properties.

2.2. Characterization

The formation of the compound was confirmed by X-ray diffraction pattern taken at room temperature by using Cu-target (Cu-$K_{\alpha 1}$ line, $\lambda = 1.54056$ Å) on Bruker AXS-D8 diffractometer at a scan step of 0.01°. The particle size and shape of the concerned phosphor was analyzed by using Transmission Electron Microscopy (TEM) Hitachi (H-7500) operated at 100 kV. The thermal stability of the concerned phosphor was obtained by thermogravimetric analysis (TGA) using Mettler-(4000) thermal analyzer coupled to a DSC-30S cell, in the nitrogen atmosphere at a heating rate of 10 °C/min.

To study thermoluminescence (TL) properties, the samples were exposed to γ-rays using ^{60}Co source for different doses (5×10^1–4×10^4 Gy) at room temperature. TL glow curves were recorded using a Harshaw TLD reader (Model 3500) fitted with a 931B photo multiplier tube (PMT), taking 5 mg of sample each time.

3. Results and discussion

3.1. Crystallite shape and size

The formation of synthesized compound was confirmed by studying the X-ray diffraction (XRD) pattern shown in Fig. 1 with (hkl) values. Scherrer's equation

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (1)$$

where D is average grain size of the crystallites, λ is incident wavelength, θ is the bragg angle and β is the diffracted full width at half maximum (in radians), was used to estimate the average crystallite size from the observed line broadening. The average crystallite size of the concerned compound was calculated to be approximately 30 nm.

The data was then fitted with the powder X-ray Data Analysis System. The XRD pattern was fitted well with hexagonal structure having lattice parameters parameters $a = 0.251$ nm, $b = 0.251$ nm and $c = 0.666$ nm. The shape and size of the particles of the concerned phosphor have also been determined by transmission electron microscopy (TEM). The TEM photograph shown in Fig. 2 reveals that the particles are of spherical shape with their average diameter 38 nm approximately. So the TEM results are found to be consistent with our XRD results.

3.2. Thermal stability

The thermal behavior of BCNO sample was studied using thermogravimetric analysis (TGA). Measurements were used to predict their thermal stability at temperature upto 900 °C. From TGA curve shown in Fig. 3, it was revealed that the weight of sample at 30 °C was 99.6%. When the sample was heated to 120 °C, it shows a weight loss of 14%. As the temperature was increased further to 390 °C, the total 28.4% of weight loss was observed and finally when the temperature reached 900 °C, the total weight loss was 37.5%, which may arise from the bond decomposition in the sample. At low temperatures, most of the bonds have energy less than the threshold energy that is required to decompose the bond. However as the temperature is increased, the bonds absorb energy and their energy becomes equal to or greater than threshold value, hence the bond dissociates that leads to weight loss of the sample. Since the weight loss is 37.5%

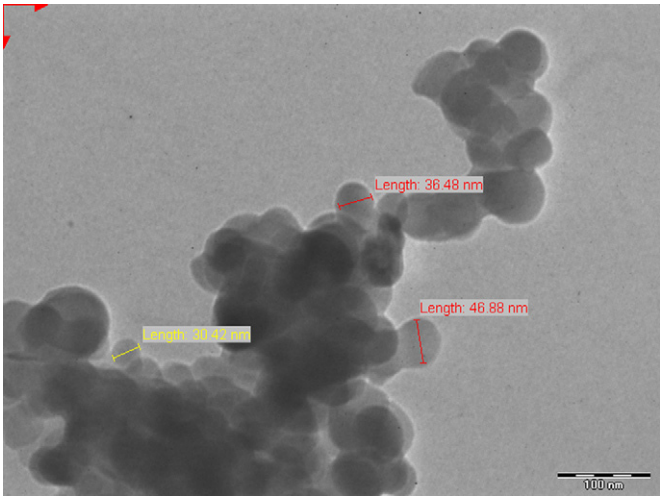

Fig. 2. TEM image of synthesized nanocrystalline BCNO compound.

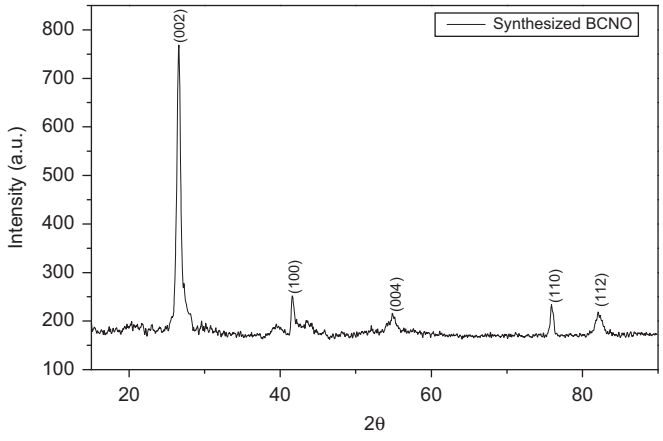

Fig. 1. XRD spectra of synthesized nanocrystalline BCNO compound.

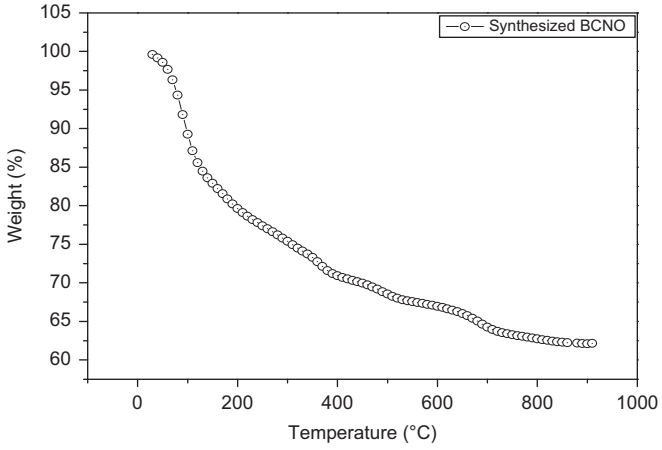

Fig. 3. TGA thermogram of synthesized nanocrystalline BCNO compound.