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Full Length Article

Glow curve deconvolution of nano barium strontium sulfate and thermoluminescence trap centers behavior with gamma doses

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ABSTRACT

The kinetic parameters and the dose response of each trap in the nano $(Ba_{0.88}Sr_{0.12}SO_4)_{99.8\%}$:$Eu_{0.2\%}$ (B88) thermoluminescence dosimeter were investigated using the computerized glow-curve deconvolution (CGCD) algorithm. The glow-curve of nano B88 could be deconvoluted into five glow-peaks using general expressions derived from the one trap-one recombination (OTOR) level model. The analysis results showed that the activation energy E and the frequency factor s for each glow-peak remained approximately constant over a wide range of doses. The change of trapping-recombination ratio $\mathcal{R}$ parameter with gamma absorbed dose for all the deconvoluted peaks was studied. The glow curve shape of nano B88 changes with the absorbed dose, forasmuch the first shoulder of glow curve becomes most prominent beyond 1 kGy. The linearity of the dose-response of all the glow-peaks was proved except for peak 5. The CGCD analysis method reveals that the peak 1 can be considered as dosimetric peak by increasing the linearity range of dose response up to 4.5 kGy instead of 2 kGy.

1. Introduction

Many researchers have investigated a lot of nanophosphors having good luminescence properties that can be used in the field of dosimetry. One group of these nanomaterials, that considered as dosimeters of ionizing radiations, is mixed barium sulfates such as $Ba_{0.97}Ca_{0.03}SO_4$:Eu [1], $BaSO_4$:Eu [2], $Ba_{0.12}Sr_{0.88}SO_4$:Eu [3] and recently $Ba_{0.88}Sr_{0.12}SO_4$:Eu [4]. It has been reported that mixed sulfate nanophosphors have well-desired characteristics like a high-temperature glow peak, linear response with ionizing radiation exposure, low fading and the ease of preparation method.

The information about the physical parameters of traps present in the materials can be obtained by using the thermoluminescence (TL) technique. The characteristics of thermoluminescence materials depend on their kinetic parameters which quantitatively describe the trapping and luminescent centers. Computerized glow curve analysis is an essential tool in most applications of TL dosimetry otherwise “quick and dirty” dosimetry is obtained [5].

Recently, the effect of strontium additive on thermoluminescence properties of $(Ba_{1-x}Sr_xSO_4)_{99.8\%}$:$Eu_{0.2\%}$ nanophosphor series irradiated to gamma doses was investigated [6]. They found that the sample with $x=0.12$, nano- $(Ba_{0.88}Sr_{0.12}SO_4)_{99.8\%}$:$Eu_{0.2\%}$ (B88), with effective atomic number ($Z_{eff}=43.5$) has deeper trap centers with the best TL sensitivity. The complex glow curve of nano-B88 sample was analyzed to its components by using T_m-T_{stop} experimental method and the computerized glow curve deconvolution (CGCD) method using first order kinetics [6]. The comparison between these methods reveals that the glow curve (GC) consists of five overlapping peaks with a slightly difference in their T_m positions. The effect of different heating rates on the glow peaks of this sample showed that it follows the first-order kinetics. Another attempt of the CGCD for nano B88 using general order kinetics was done. According to general order kinetics, the glow curve consists of five peaks at 448, 475, 500, 545 and 597 K and the activation energies ranging from 1.05 to 1.74 eV [4].

2. Materials and methods

Nano-sample of B88 was prepared by the chemical co-precipitation method at NIS [4]. It was irradiated to different doses of Co-60 in the range from 0.4 Gy to 63 kGy which are measured using NIS secondary standard system (PTW UNIDOS electrometer, and 0.6 cc thimble PTW ionization chamber) with a combined uncertainty 0.88% for air kerma that is traceable to SI unit (calibrated at BIPM, France, 2012). Nano B88 samples were irradiated from cobalt source to intermediate level of gamma dose (0.4 Gy–1 kGy) and to high level in range 1–63 kGy. TL glow curves of powder samples were recorded directly after irradiation at a heating rate of 2 Ks^{-1}, using a Harshaw TLD reader (Model 3500) with suitable filter (density filter 2 from CVI laser optics) that

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allow us to read the irradiated samples with high doses. For reusing the prepared nano-B88, the sample is annealed in ceramic crucible using furnace at 673 K for 1 h, and then quickly quenched.

3. CGCD using general expressions derived from one-trap one recombination (OTOR) level model

General expressions based on the one trap-one recombination (OTOR) level model were employed in the deconvolution of the nano-B88 glow-curves. These equations were developed and their accuracy in calculating the activation energy was verified for the glow-peaks simulated by different TL models [7–10] and for the experimental glow-curves as well [11]. The main conclusion was that these equations are superior to the general-order kinetics (GOK) [12] and the mixed-order kinetics (MOK) [13] equations in the determination of the activation energy of the glow-peak. Moreover, unlike the GOK equation which is entirely empirical, these equations are based on the physical OTOR model. Hence, the physical parameter $\mathcal{R}$, which is the trapping-recombination ratio $\mathcal{R}=A_n/A_m$, where A_n and A_m are the trapping and recombination probability coefficients respectively, is obtained from the deconvolution process instead of the empirical kinetic-order b of the GOK equation. The relation behavior between the kinetic order b, and the trapping-recombination parameter $\mathcal{R}$, was investigated theoretically [7]. They showed that the glow-peak tends to the first-order-kinetics ($b\approx 1$) model as $\mathcal{R}\ll 1$. While, as $\mathcal{R}\to 1$, the glow-peak gets out from the first- to the second-order kinetics. The final form of these expressions for linear heating rate is formulated in Eqs. (1) and (2) for $\mathcal{R}<1$ and $\mathcal{R}>1$, respectively [11]:

$$I = I_m \frac{W[\exp(z_{1m})]+W[\exp(z_{1m})]^2}{W[\exp(z_1)]+W[\exp(z_1)]^2}\exp\left(-\frac{E}{k}\left(\frac{1}{T}-\frac{1}{T_m}\right)\right) \quad (1)$$

$$I = I_m \frac{W[-1,-\exp(-z_{2m})]+W[-1,-\exp(-z_{2m})]^2}{W[-1,-\exp(-z_2)]+W[-1,-\exp(-z_2)]^2}\exp\left(-\frac{E}{k}\left(\frac{1}{T}-\frac{1}{T_m}\right)\right) \quad (2)$$

Where E (eV) is the activation energy, I_m and T_m are peak maximum and peak maximum position, respectively, k is the Boltzmann constant in eV K^{-1}, $W[\exp(z_1)]$ and $W[-1,-\exp(-z_2)]$ are the principal and the second branches of the Lambert-W function, respectively,

$$z_1 = \frac{1}{c}-\ln(c)+\frac{E\exp\left(\frac{E}{kT_m}\right)}{kT_m^2(-1.05\mathcal{R}^{1.26}+1)}F(T,E), \quad (3)$$

$$z_2 = \frac{1}{|c|}+\ln(|c|)+\frac{E\exp\left(\frac{E}{kT_m}\right)}{kT_m^2(-3.24\mathcal{R}^{-0.74}+2.963)}F(T,E), \text{ and} \quad (4)$$

$$z_{im} = z_i(T=T_m),\quad i=1 \text{ or } 2 \quad (5)$$

$$F(T,E) = \int_{T_o}^{T}\exp\left(-\frac{E}{kT}\right)dT \quad (6)$$

where $\mathcal{R}$ is the trapping-recombination probabilities coefficient, and

$$c = \frac{n_o(1-\mathcal{R})}{N\mathcal{R}} \quad (7)$$

n_o is the initial number of the trapped electrons, and N is the total concentration of the trapping states.

The goodness of the fit was measured by the figure of merit (FOM) [14]:

$$FOM(\%) = \sum_{j_i}^{j_f}\frac{|y_i-y(x_i)|}{A}\times 100 \quad (8)$$

where j_i is the first channel in the region of interest, j_f is the last channel in the region of interest, y_i is the information content of channel j, $y(x_i)$ is the value of the fitting function in channel j, and A is the integral of the fitted glow-peak in the region of interest.

The best solution of the deconvolution process tends to achieve the minimum FOM value. However, a good deconvolution does not necessarily mean correct analysis. The correct analysis implies that the obtained results are not in conflict with the thermoluminescence theory in general and with the utilized model in particular. Generally, the correct analysis mainly tends to have activation energy and the frequency factor values in the usual limits $0.7\le E\le 2.0$eV, $10^7\le s\le 10^{13}s^{-1}$, respectively [15]. Moreover, in case of using the OTOR model in the deconvolution process, the position of the T_m depends on both E, s. Incorrect results may cause in a glow-peak with low T_m to have higher E and lower s than a glow-peak with high T_m. This should be avoided in the correct analysis. Thus, with a good FOM value, and reasonable analysis results, one can assume a good particular deconvolution results.

4. Results and discussions

4.1. Determination of optimum annealing conditions

Since the annealing process is one of the important parameters in TL dosimetry, the optimum annealing conditions for the nano-B88 dosimeter were investigated. After irradiation of the dosimeters by 0.5 Gy of Co-60 source and reading out using the usual conditions, two subsequent different annealing conditions were applied. The first is the isochronal annealing (different temperatures with constant period) and the second is the isothermal annealing (different periods at constant temperature) at the optimum temperature that resulted from the isochronal annealing. It is clear from Fig. 1(a) that the optimum temperature i.e. the lowest TL intensity for unirradiated sample after annealing and with the highest net TL intensity for irradiated sample, is 673 K. Fig. 1(b) illustrates that the optimum annealing period is one hour at 673 K.

4.2. The glow-curve deconvolution of the nano-B88 dosimeter

The glow-curves of nano-B88 were analyzed by the first-order kinetics model [6] into five peaks. However, it was found that this model was unable to describe the experimental glow-curve accurately. The resulted high values of FOM=1.22 leads us, in the current study, to use different two expressions in the CGCD algorithm. The first expression was the general-order kinetics which has the advantage to fit the intermediate cases between the first- and the second-order kinetics. However, many investigations indicated that this expression may fail to describe TL glow-peak in cases the trapping probability coefficient is greater than the recombination probability coefficient and the sample dose in the saturation level. These cases were successfully resolved by the new analytical expressions and their advantages are summarized as follows:

i. These expressions can, accurately, describe any glow-peak even in the cases in which the other expressions failed,
ii. These expressions use the frequency factor s with the unit of s^{-1} which has a physical meaning introduced by Randall and Wilkins (1945). While, the GOK expressions use the pre-exponential factor which has a unit of s^{-1} only in the cases of $b=1$ and $b=2$. For the intermediate cases, it has a unit of $cm^{3(b-1)}s^{-1}$ which is not related to any particular physical