Thermoluminescence properties of lab-prepared CaSrS: Dy nanophosphors

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Abstract: In this work the thermoluminescence properties of Calcium Strontium Sulphide: Dysprosium (CaSrS:Dy) prepared by the solid state diffusion reaction have been studied and their feasibility for radiation dosimetry applications is discussed. Three different pre-irradiation annealing programs were examined and found that the best program is 450°C for half an hour. The homogeneity of the $Ca_{0.65}Sr_{0.35}S$: Dy sample has been found as 12.1%, very good reproducibility of TL- measurements has been obtained. The TL characteristics have been examined through irradiation by gamma doses in the range 20mGy–10 kGy. The dose response was linear up to 10kGy without any observed saturation.

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Keyword: Thermoluminescence, pre-irradiation annealing, dose response and reproducibility**.**

1. Introduction

Thermoluminescence dosimetry has become a reliable and routine method for measuring ionizing radiation (1). As for this application there are many commercially available TL dosimeters (TLDs)by different trade names. In the past few years, nanostructured materials have drawn the interest of research workers because of their potential impact in many areas such as electronics, photonics, catalysis and sensing. It has been established that the optical properties of materials in the nanoscale can be very different from those of their bulk equivalent. These studies state that the aforesaid materials have a potential application in dosimetry of ionizing radiation where the conventional microcrystalline phosphors do not meet the requirements. Decrease in size of particles causes increasing of both the number of surface states and the proportion of recombination of the charge carriers. Studies on TL properties of nanomaterials show that they include some outstanding characteristics such as high sensitivity and saturation at very high doses (2). In those studies, it was found that the luminescence process in semiconductor nanoparticles is a very complex phenomenon and it was attributed to the deep traps of surface states whose energy levels lie within the band gap of semiconductor.

In this study we investigate some thermoluminescence properties of lab-prepared CaSrS: Dy nanophosphors such as pre-irradiation annealing, reproducibility of TL-signal and dose response.

2. Material and Method

In previous work (3) we established the preparation of CaSrS: Dy by using solid state diffusion method (4,5,6). The effects of changing in concentrations of Calcium sulphate and strontium sulphate have been studied. The optimum concentration has been found is 65%:35%. Different concentrations of Dy were added to $Ca_{0.65} Sr_{0.35} S$, the results showed that the optimum concentration of Dy doped to $Ca_{0.65} Sr_{0.35} S$ is (22wt%). Thermal treatment and High gamma dose sensitization have been used to increase sensitivity of CaSrS: Dy (0.22wt%). The optimum annealing temperature is 450°C for 1hr and pre- dose of gamma ray is 9KGy. This prepared$Ca_{0.65} Sr_{0.35} S$: Dy is subjected to the following TL features related to radiation dosimetry.

2.1 Pre-irradiation annealing

To define the optimum annealing courses many groups of CaSrS: Dy has been exposed to 10 Gy of gamma ray. Then, they are treated with three different annealing temperatures (350ºC, 400ºC and 450 ºC) for different periods of time (15 min, 30 min,40 min, 45 min and 60min). After that, the residual TL- intensity was measured by Harshaw 3500 series TLD reader. The result of annealing experiment is presented graphically.

2.2 Test of batch size homogeneity (Δ):

0.3gm of lab prepared CaSrS: Dy was treated with appropriate pre-irradiation annealing program, after that the sample are put in lightproof capsule and irradiated to test gamma dose of 10Gy. The capsule is divided into ten samples and TL-response is recorded for each sample.

The corresponding uniformity indices (Δ) were calculated according to the following equation**(7, 8):**

$$\Delta = \left[\frac{M_{max} - M_{min}}{M_{min}} \right] \times 100 \qquad (1)$$

Where (M_{max}) and (M_{min}) are representing the maximum and minimum values recorded, respectively.

2.3 Reproducibility of TL-signal:

Five similar TL samples are selected; heat annealed as before and impact inside five capsules which irradiated to one teat dose 10 Gy. These irradiated samples are subjected to individual and group TL-signal reproducibility. Average, standard deviation and standard deviation % are measured. The results have been recorded in table and determined the accuracy of the TL-measurement.

2.4 Dose Response

Many groups of CaSrS: Dy were packed in lightproof capsules and irradiated to different gamma doses from 20mGy to 10kGy. Then the TL response of CaSrS: Dy was measured, after that the dose response of CaSrS: Dy are presented graphically.

3. Results and Discussion

3.1 Pre-irradiation annealing

The reliable re-use of TLD materials often requires the use of strict thermal annealing procedures. High temperature annealing before use and re-use of phosphors has the purposes of minimizing the residual TL signal by emptying the high temperature traps and restoring the original sensitivity and glow curve structure (9). The results of the pre-irradiation annealing experiment are presented graphically at figure (3.1) as the residual TL-intensity against the heating time for different temperatures. From this figure we can show that when the material annealed at 350°C taken large period of time for reach to background. Also, when the material annealed at 400°C it reach to background at 60 min. However when annealed for 450°C its reach to background quickly at 40 min. So that annealing the material at 450 ºC for 30 min is the best annealing course.

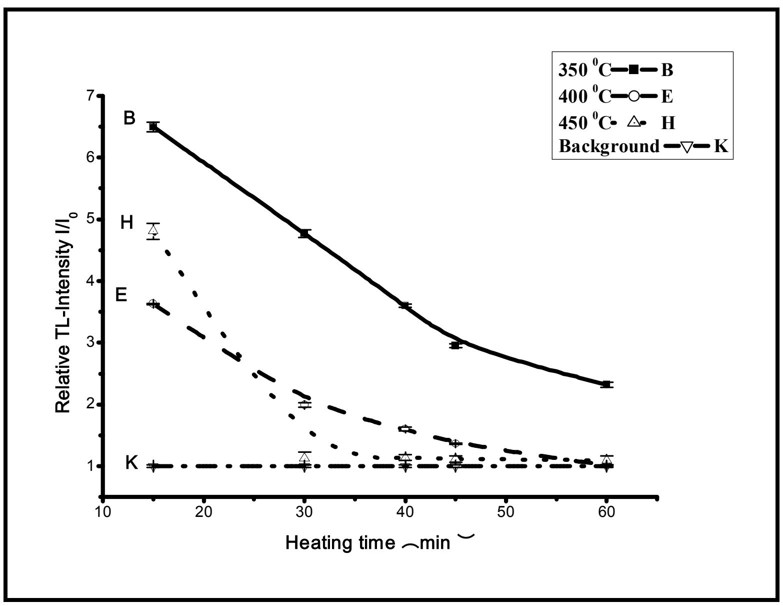

Figure (3.1). The effect of pre-irradiation annealing for CaSrS: Dy as a function of heating time at different heating temperatures.

3.2 Batch homogeneity (Δ):

The TL response of samples of same material that have undergone the same treatment may not necessarily be the same. The recommendation of the International Electrochemical Commission (IEC), however, is that the evaluated value for any one dosimeter in a batch shall not differ from the evaluated value of any other in the same batch by more than 30%(IEC) (10)

This has been verified for the investigated nanophosphor using 10 samples from the same batch which subjected to the same treatment and substituted in equation (1).The homogeneity factor, (Δ) has been calculated to be 12.1%, a value which is lower than the upper limit recommended by IEC.

3.3 Reproducibility

Reproducibility is another useful property that a sample should possess in order to be of any real use in dosimetry. In order to check the reproducibility of the dose measurements by such phosphor, five similar TL samples are selected and put in lightproof capsules then irradiated to one test dose 10 Gy. The average, standard deviation and standard deviation % are measured for both individual and group samples.

The results of this experiment are shown in table (3.1). It is clear from this table that the CaSrS: Dy nanophosphors is reproducible and accurate within **±$(\sigma_{n-1})_g$= 4.78%** and ± **$(\sigma_{n-1})_i$= 5.29%** for different capsules and individual detector respectively.

The accuracy of the TL- measurement is considered that the average of the two values of (σ_{n-1})g% and (σ_{n-1})i% which is given as

(σ_{n-1}) = 5.03 %

The reproducibility of the investigated TL-nanophosphor is expressed by another way as graph of scattered relative TL-signal, as shown in figures (3.2) and (3.3).

Table (3.1). Reproducibility of CaSrS: Dynano-phosphors for the same gamma dose 10Gy.

Sample no.	TL Intensity (nC) capsule1	capsule2	capsule3	capsule4	capsule5	$<x_i>$	$(\sigma_{n-1})_i$	$(\sigma_{n-1})_i$ %
1	18697	20895	20730	20120	22400	20568.4	1340.78	6.52
2	19243	20755	19145	21230	21350	20344.6	1074.22	5.28
3	20618	19950	22750	19890	20545	20750.6	1166.09	5.62
4	23800	20755	21000	20125	20020	21140.0	1543.38	7.30
5	22750	20727	19850	19845	21000	20834.4	1189.16	5.71
6	21562	21700	20055	20155	21350	20964.4	795.15	3.79
7	20750	19982	21280	21100	20120	20646.4	578.03	2.80
$<x_g>$	21060	20680	20687	20352	20969	20749.8		$<(\sigma_{n-1})_i>$%
$(\sigma_{n-1})_g$	1816	594	1167	569	830			5.29
$(\sigma_{n-1})_g$%	8.62	2.87	5.65	2.80	3.96	$<(\sigma_{n-1})_g>$%	4.78	
$<(\sigma_{n-1})>$%								5.03

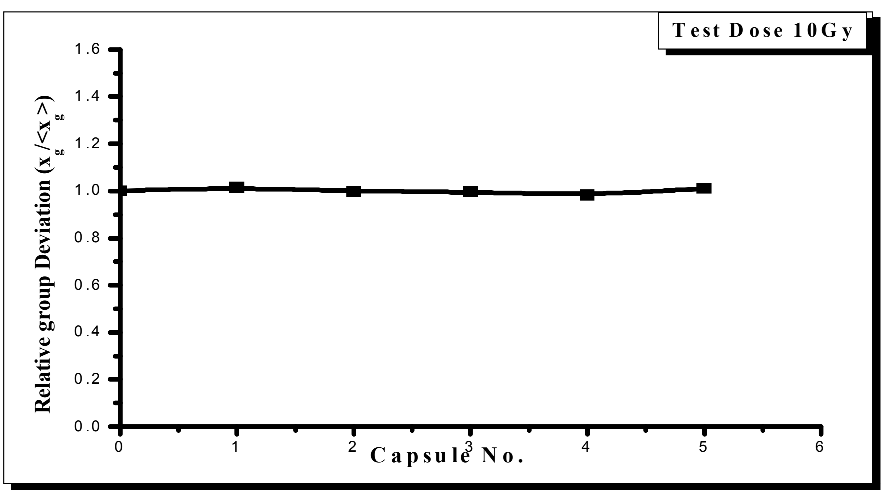

Figure (3.2). The relation between the relative group deviation ($x_g/<x_g>$) versus the capsule No.

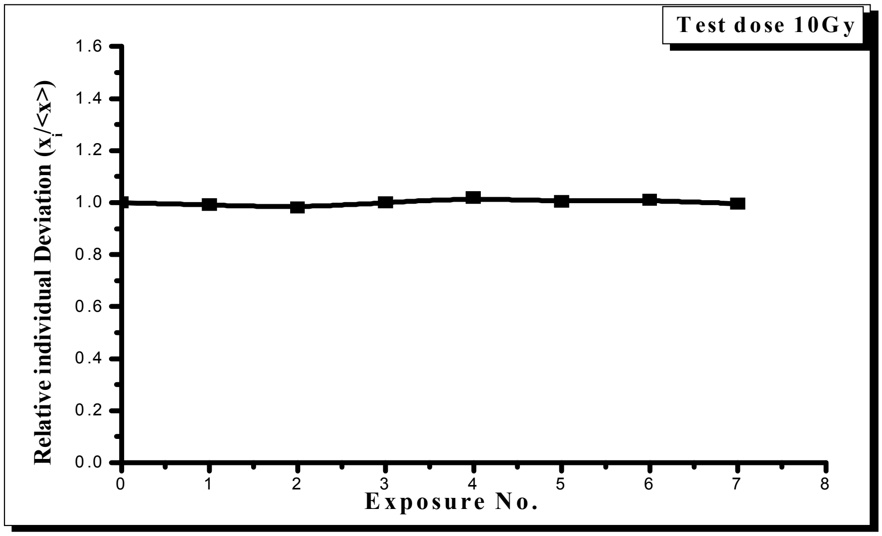

Figure (3.3). The relation between the relative individual deviated capsule batches (x_i/<x_i>) versus repeated exposure.

3.4 Dose Response

The TL response curves of CaSrS: Dy samples irradiated by gamma rays are shown in Fig. (3.4). The response curve of the irradiated material to a dose range 20mGy–10kGy on the log–log scale shows linearity to the full range without any tendency to saturation. This observation agrees with an earlier report on nanophosphors material (11-12)

The TL intensity of CaSrS: Dy increases linearly with dose. This might be due to high surface to volume ratio, which results in a higher surface barrier energy for the nanoparticles. On increasing the dose, the energy density crosses the barrier and a large number of defects are produced in the nanoparticles which ultimately keep on increasing with the dose (13).

The linear response of CaSrS: Dy nanophosphors differed than the response of microcrystalline phosphors material which often obtain dose response curve linear at low dose then sublinear after that superlinear and at last attain saturation. Therefore when these materials are used in high dose conversion factor must be applied but nanophosphors samples are often indicate linear response all over the response curve.

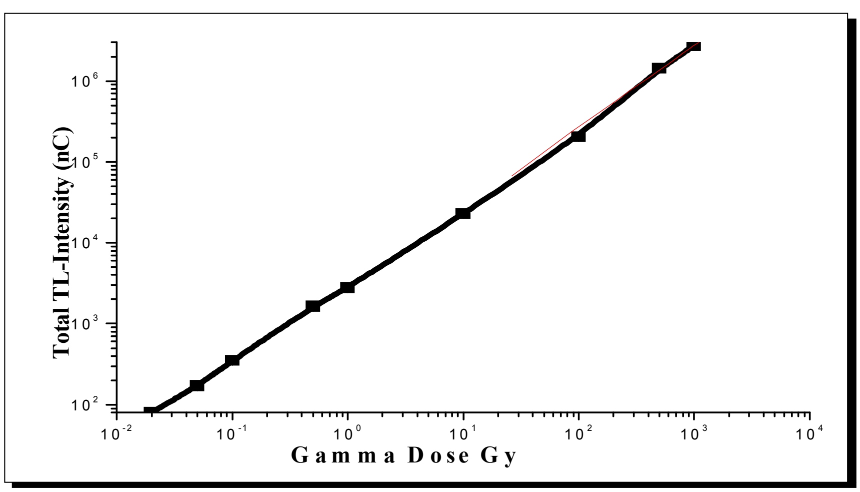

Figure (3.4). Dose response of CaSrS: Dy irradiated with ^{60}Co.

Conclusion

Thermoluminescence properties of lab prepared $Ca_{0.65}Sr_{0.35}$ S: Dy nanophosphors have been studied. The optimum pre-irradiation program was found to be 450°C for 40 min. The homogeneity of such samples as calculated was found 12.1%. Moreover, a very good reproducibility of TL- measurements have been obtained within an average of about **σ_{n-1} = ±5.03 %.** This investigated TL-nanophosphor show a linear gamma-induced response through the dose range 20mGy-10kGy without any observed saturation.

According to the obtained results, this nanophosphor may be used for estimating low and high gamma dose without need any conversion factor within a relatively high accuracy.

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