Study of optically stimulated luminescence dosimetric properties of submicron BaSO₄: Eu Phosphor

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Abstract. In this context dosimetric properties of submicron BaSO₄: Eu phosphor have been studied. To reduce the grain size of the material to submicron level, top-down approach (ball milling) has been adopted. The phosphor showed good optically stimulated luminescence (OSL) properties. The OSL properties of the phosphor were altered due to the particle size reduction on ball milling. Ball milling affected the kinetics of OSL Processes. OSL dose response of the phosphor has been increased up to 13 kGy on ball milling which makes it useful in high dose dosimetric applications.

Keywords: High dose dosimetry, Optically Stimulated Luminescence Dosimetry, BaSO₄: Eu

1. Introduction

Optically stimulated luminescence (OSL) is the recent and advanced technique for radiation dosimetric applications like personnel and environmental monitoring, retrospective dosimetry which includes geological dating and accident Dosimetry, space Dosimetry and many other applications. In the applications where high doses of radiations are to be delivered, the phosphor needs to have high dose response. Many materials have been studied so far to be the potential candidates for OSL dosimetry however sulphates fail to attract many of the researchers because of its high effective z value which makes them unsuitable for personal dosimetry. For high dose applications like food irradiation or even for low environmental doses phosphor needn’t have to be tissue equivalent. Yamashita et al. [1] reported highly intense TL in several Tb and Eu doped sulphate samples. Dhoble et al. and other researchers have studied sulphate for thermoluminescence dosimetry (TLD) [2,3,4,5,6]. Numan Salah et al. studied BaSO₄: Eu nanoparticles for their TL dose response to gamma radiation over a very wide range of exposures [7]. Bhatt et al. reported the TL and OSL of BaSO₄: Eu [8]. Recently Anant Pandey et al. successfully prepared nanocrystalline Eu doped BaSO₄ using chemical co-precipitation technique claiming that phosphor to have TL dose linearity up to 2 kGy [9].

In this context the efforts were made to study dosimetric properties of submicron BaSO₄: Eu phosphor prepared by simply ball milling the bulk sample.
2. Experimental
The BaSO₄: Eu phosphor was synthesized using recrystallization method by transferring aqueous solution of GR grade BaCl₂ in double distilled water and stoichiometric amount of Eu₂O₃ dissolved in nitric acid to the flask containing excess amount of distilled H₂SO₄. The solution was then allowed to evaporate slowly in acid distillation plant. The powder thus obtained was washed thoroughly with double distilled water and dried under lamp. The dried powder was then annealed at 850°C in air and quenched to room temperature. This sample was referred as bulk sample. The Eu concentration was optimized to 0.5 mol%. Eu incorporated in the lattice in Eu²⁺ form which was confirmed from the PL study of the phosphor. The top-down approach was employed for particle size reduction to the submicron level using high energy planetary ball mill (RETSCH GmbH Germany make). The bulk material was dry ball milled with 10 mm zirconium balls for 8 hours. The mass of balls was taken in the ratio of 25:1 g of the phosphor. Particle size analysis was carried out on Master sizer 2000 E particle size analyzer with water as a dispersant.

The OSL measurements were carried out on PC Controlled TL/OSL Reader (TL/OSL 1008) manufactured by Nucleonix systems fitted with Blue LEDs cluster, each of 3-watt output placed 180 degrees opposite which gives stimulation output with peak wavelength emission of 465 nm, having Luminous flux radiometric power of 30 mW & emission wave length band ranging from 460 to 470 nm. 60Co source was used for irradiating the samples.

3. Result and Discussion
After 8 hours of dry ball milling, the particle size of the BaSO₄ phosphor was found to be reduced to 3-10 μm with an average grain size of 5 μm which was confirmed by particle size analysis. The phase purity of the samples was confirmed from the XRD patterns obtained using Cu Kα x-rays with wavelength 1.5046 Å (figure 1). The XRD patterns of the bulk and ball milled samples matched with the ICDD data 83-2053. From XRD data it was observed that ball milling does not modify the host lattice structure or even the phase of the material.

![Figure 1. XRD pattern of bulk and ball milled BaSO₄: Eu phosphor](image-url)
Continuous wave optically stimulated luminescence (CW-OSL) was studied in these samples. Figure 2 shows the CW-OSL response of the samples exposed for 100 Gy of $^{60}$Co gamma radiation. The readings were normalized to 1 mg of the sample. The CW-OSL curves were recorded after one day of irradiation in order to remove phosphorescence and to get a stabilized OSL response. Slow CW-OSL decay was observed in bulk BaSO$_4$ phosphor while in ball milled sample the CW-OSL decay was much faster than the bulk sample which can be seen from normalized CW-OSL decay curve given in the inset of figure 2. The CW-OSL sensitivity of the bulk sample was found to be 25% of the OSL sensitivity of standard phosphor lithium magnesium phosphate (LMP) [10] by considering the integral area under curve while the sensitivity of the ball milled phosphor was found to be only 3% of the OSL sensitivity of LMP. However on averaging out the initial counts for first 3 seconds, the CW-OSL sensitivities of the bulk and ball milled samples were found to be 15% and 0.1% of the OSL sensitivity of standard phosphor LMP respectively. Hence it can be concluded that on ball milling the sensitivity of the sample reduced to 3% of the bulk sample.

Figure 2. CW-OSL curves for Bulk and Ball milled BaSO$_4$: Eu phosphor compared with standard LMP Phosphor.

Figure 3 shows the CW-OSL decay curve of the samples along with the fitted components. The CW-OSL signal of both the samples can be exactly fitted with sum of three exponentials given by the equation 1,

$$I_{OSL} = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + A_3 \exp(-t/\tau_3)$$

Where $I_{OSL}$ is the initial OSL intensity and $\tau_1$, $\tau_2$, $\tau_3$ are the decay constants of the respective OSL traps.

The values of coefficients, photo-ionization cross section ($\sigma$) and decay constant ($\tau$) for each component are summarized in Table 1.
Figure 3. CW-OSL decay curve of the BaSO₄:Eu phosphor along with the fitted components

Table 1. CW-OSL parameters of BaSO₄:Eu

| Sample          | CW-OSL Components | Coefficient A | Decay Constant τ (s) | Photo ionization cross section σ (cm²) | COD (R²) | FOM (%) |
|-----------------|-------------------|---------------|----------------------|----------------------------------------|----------|---------|
| Bulk Sample     | Fast              | 190125        | 1.02                 | 1.39 x 10⁻¹⁵                           |          |         |
|                 | Medium            | 121700        | 8.27                 | 1.72 x 10⁻¹⁶                           | 0.99989  | 0.38    |
|                 | Slow              | 71000         | 26.05                | 5.45 x 10⁻¹⁷                           |          |         |
| Ball milled     | Fast              | 6828          | 0.27                 | 5.26 x 10⁻¹⁵                           |          |         |
| Sample          | Medium            | 1751          | 1.58                 | 9.00 x 10⁻¹⁶                           | 0.99993  | 1.13    |
|                 | Slow              | 450           | 6.45                 | 2.20 x 10⁻¹⁶                           |          |         |

From table 1 it is clear that, for bulk sample the ratio of fast to slow components was 2.67 while in ball milled sample it was found to be 15.17. Similarly, the ratio of medium to slow component in bulk sample was 1.71 and that for ball milled sample was found to be 3.89.

Figure of merit (FOM) for the OSL deconvolution was calculated using formula

\[
FOM(\%) = \frac{\sum_i |y_{sum}^i - y_{exp}^i|}{\sum_i y_{sum}^i} \times 100. \tag{2}
\]
Where \( y_{\text{sum}} = \) fitted OSL intensity coordinates of sum of exponentials and \( y_{\text{exp}} = \) experimental OSL intensity coordinates. The FOMs for OSL deconvolution of bulk and ball milled samples are found to be 0.38% and 1.13% respectively.

OSL dose response of the samples was studied for the wide dose range from 25 Gy to 13 kGy using \(^{60}\)Co gamma source as shown in Figure 4. Sub-linear response was observed in the bulk sample. However in ball milled sample linear dose response was observed up to 13 kGy. Hence it can be concluded that ball milling enhanced the dose linearity of the phosphor.

![Graph showing OSL dose response of Bulk and Ball milled BaSO\(_4\):Eu phosphor](image)

**Figure 4.** OSL dose response of Bulk and Ball milled BaSO\(_4\):Eu phosphor

4. Conclusions
OSL properties of the submicron BaSO\(_4\):Eu phosphor were studied. The phosphor shows good OSL properties for gamma irradiation. OSL properties of the phosphor were altered due to the particle size reduction on ball milling. OSL intensity in ball milled sample was only 3% of the OSL intensity in bulk sample. This may be due to the surface accommodation of the charge carriers.

Ball milling affected the kinetics of OSL Processes. The CW-OSL curves for both the samples were exactly fitted with three components. In bulk sample the contribution of fast and medium components was 2.67 and 1.71 times that of slow component respectively while in ball milled sample the contribution of fast and medium components was 15.17 and 3.89 times that of slow component respectively.

The OSL dose response for gamma irradiation was studied in both the samples over the dose range of 25 Gy to 13 kGy. OSL dose response was found to be increased from 3 kGy to 13 kGy on ball milling.

Hence from the results so obtained, it is concluded that BaSO\(_4\):Eu is the potential candidate which can be used as a tool for dosimetric applications. On ball milling the dose response of the phosphor enhanced to higher doses in kGy range which makes it useful in high dose dosimetric applications.

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6. Acknowledgement
We are thankful to department of Chemistry, R.T.M.N.U campus, Nagpur for providing irradiation facility.