



Application of CaSO₄:Dy for high dose dosimetry

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Abstract

The present work was carried out on development of submicron/nano CaSO₄ phosphor using top down approach for high dose dosimetry. In top down technique high energy planetary ball mill was used at 400 rpm with Zirconium grinding bowl and 2 mm Zirconium balls. CaSO₄:Dy phosphor with 0.1 mole% Dy was synthesized using well known Yamashita method. X-Ray Diffraction (XRD) pattern of bulk and ball milled CaSO₄:Dy was well matched with ICDD file. The shape and size of ball milled powder sample was analyzed by a scanning electron microscope (SEM) technique. It shows nearly same glow curve structure and equal sensitivity as that of the standard CaSO₄:Dy phosphor. Ball milled, micron size particles show drastically reduced sensitivity. However there was no change in TL glow curve shape for the exposure ranging from 0.1 to 15 kGy. Bulk CaSO₄:Dy phosphor shows the dose linearity up to few grays but ball milled phosphor shows linearity up to 6 kGy. The top down approach may be useful as an alternative TL dosimeter in the field of food irradiation dosimetry.

Keywords: Planetary ball mill, High dose linearity, Food dosimetry

1. INTRODUCTION

The Dysprosium doped CaSO₄ thermoluminescent (TL) phosphor is widely used in different radiation areas like personnel monitoring, environmental monitoring due to its good dosimetric properties. In past, Burgkhardt et al. [1] and Piesch et al. [2] have investigated CaSO₄:Dy material for its application in the high dose region. They observed that CaSO₄:Dy phosphor shows complex glow curve nature at high doses. LiF:Mg,Cu,P [3], CaSO₄:Dy [4] and CaSO₄:Dy,Cu have been studied for high dose dosimetry. Co-doping of Cu atom in CaSO₄:Dy phosphor shows no growth of high temperature peak structure (350 °C) even for large doses [5]. For high dose dosimetry less number of phosphors were studied. The CaSO₄ phosphor has good dosimetric property hence we tried to develop phosphor for high dose dosimetry using top down approach [6]. If particle size of material decreases then number of surface states and proportion of recombination of charge carriers increases. Some researchers studied the TL properties of nano materials and they found that nano phosphor shows high TL sensitivity and their TL-dose response saturates at high dose levels [7,8,9]. Sawant et al. [10] reported that low

temperature peak at 110 °C get dominated over the 220 °C peak in nano form of CaSO₄:Dy phosphor prepared by bottom up approach. In this study, we used freshly prepared micro crystalline phosphor which has dosimetric peak at 220 °C and tried to convert it in submicron/nano size CaSO₄:Dy phosphor with the help of planetary ball mill.

The aim of the present investigation is to explore the possibility of using CaSO₄:Dy material in high dose gamma dosimetry, which is frequently required in industries related to chemical technology, food processing systems.

2. EXPERIMENTAL

CaSO₄:Dy phosphor was prepared using analytical reagent grade CaCO₃, H₂SO₄ and Dy₂O₃ reactants. They were calculated in stoichiometric proportion. H₂SO₄ was measured in excess quantity to that of stoichiometric value and Dy impurity was dissolved in hot concentrated H₂SO₄ acid at 300 °C. The required quantity of CaCO₃ dissolved in nitric acid and added drop by drop to the hot H₂SO₄. Acid was distilled out in a closed system similar to that described in the literature [11]. Phosphor thus prepared was

repeatedly washed with distilled water to remove the traces of acid and dried in an oven at 200 °C. The prepared sample was annealed at 700 °C temperatures for 2 hrs and used in the further experiments. The reduction in particle size of phosphor was done by top down approach. The high energy planetary ball mill was used at 400 rpm with 2 mm diameter balls and grinding bowl of zirconium. After some duration small quantity of ball milled phosphor was taken out for TL characterization.

The ball milled CaSO₄:Dy powder was characterized by X-ray diffraction, using Rigaku miniflex diffractometer with Cu K α radiation. Scanning electron microscopy (SEM) micrographs were obtained using JEOL, JSM-6390 scanning electron microscope. For taking TL, samples were exposed to gamma rays from a ⁶⁰Co source for various doses (100 Gy–7 kGy) at room temperature. The dose rate for gamma rays irradiation was 0.237 Gy/s. TL glow curves were recorded with 5 °C s⁻¹ heating rate on a Polltech TLD reader by taking 5 mg of sample each time.

3. RESULTS AND DISCUSSION

3.1 X-Ray Diffraction

Fig. 1 shows the XRD pattern for the ball milled CaSO₄:Dy powder. These patterns indicate that the ball milled material has maintained an orthorhombic phase of CaSO₄ and it is well matched with ICDD file no. 74-1782. The figure shows a clear broadening in the diffracted peaks, which is due to the reduction in the particle size of CaSO₄:Dy [12].

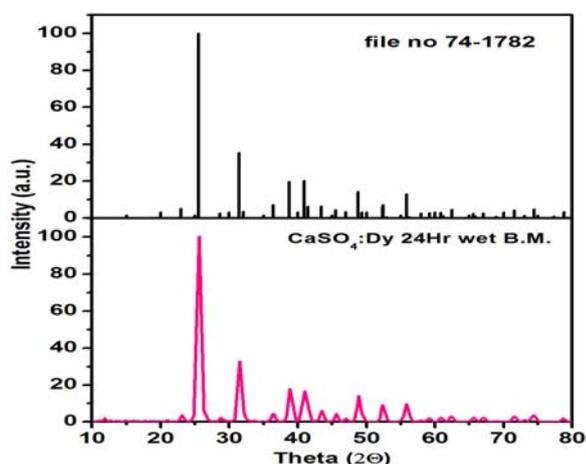


Fig. 1: XRD pattern of Ball milled CaSO₄ and ICDD file 74-1782

3.2 Grain Size And Morphology

The synthesized CaSO₄:Dy phosphor was treated for 24 Hr wet ball milling process. Fig. 2 shows the scanning electron micrograph pattern of the ball milled phosphor. It reveals that due to the hammering or collision of zirconium ball on phosphor, all grains acquired less than 1 micron size with random shape, whereas the conventional phosphor grains

prepared by re-crystallisation method are mostly above 75 micron in size and need to be ground for dosimetric (personnel, environmental) applications.

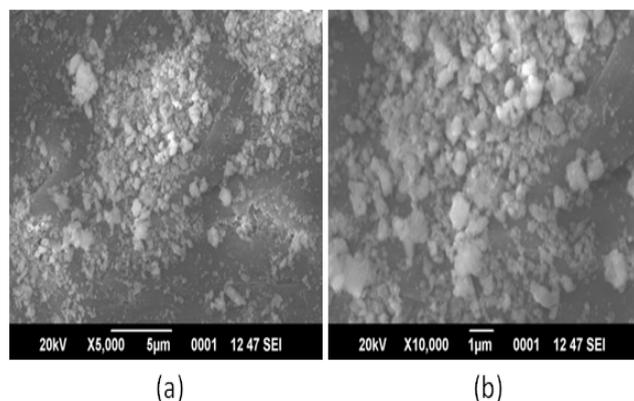


Fig. 2: SEM Images of ball milled CaSO₄:Dy (0.1 mole%) (a) 5 micron (b) 1 micron scale

3.3 TL Sensitivity Comparison

The reduction in particle size of phosphor was done by top down approach. The high energy planetary ball mill was used at 400 rpm with 2 mm diameter balls and grinding bowl of zirconium. After some duration small quantity of ball milled phosphor was taken out for TL characterization.

TL sensitivity of bulk and submicron ball milled CaSO₄:Dy phosphor were compared with standard CaSO₄:Dy. Fig. 3 shows the TL glow curve of bulk and ball milled phosphor. The sensitivity of standard and bulk CaSO₄ phosphor was found to be almost same and wet ball milled phosphor shows 144 times less sensitive than that of bulk CaSO₄:Dy phosphor.

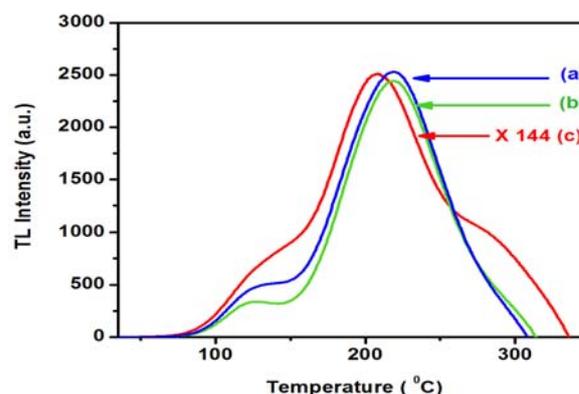


Fig. 3: TL glow curve of ball milled and without ball milled CaSO₄:Dy (test dose 50 Gy)

(a) As Prepared CaSO₄:Dy (0.1 mole%) (b) Standard CaSO₄:Dy (c) 24 Hr Wet ball milled CaSO₄:Dy.

Table 1 shows the sensitivity comparison between ball milled phosphor for different duration. From table it can be concluded that as the particle size of phosphor decreases, TL sensitivity of phosphor also decreases. The glow peak temperature of ball

milled phosphor slightly shifted towards the lower temperature.

Table 1: TL Sensitivity comparison of CaSO₄:Dy

S.N.	Specification (CaSO ₄ :Dy 0.1 mol%)	Intensity (a.u.)	Relative Intensity
1	Bulk Sample	574764.3	100.00
2	12 Hr Dry B.M.	37882.5	6.59
3	24 Hr Dry B.M.	7852.0	1.37
4	24 HrDry-24 Hr Wet B.M.	3992.9	0.69

3. 4 Dose Response

The TL linear response curve of submicron crystalline CaSO₄:Dy powder to different high doses in the range 100 Gy–7 kGy is presented in Fig. 4. The curve plotted by calculating the peak height of dosimetric peak 220 °C. From figure it can be illustrated that phosphor first exhibits linear behavior up to the 6 kGy and then get saturated above 7 kGy doses. This linearity range is useful for elimination of pathogenic organisms and micro-organisms from Fresh fruits, vegetables and for pasteurization of solid foods such as meat, poultry and sea foods [13].

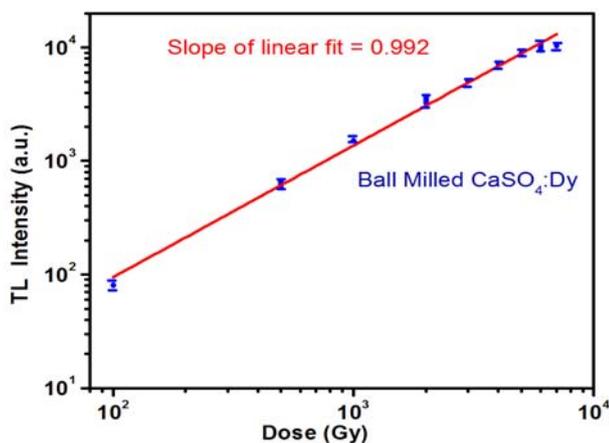


Fig. 4: TL-Dose response of ball milled CaSO₄:Dy

4. CONCLUSION

Sample prepared by Yamashita method is well matched with ICDD file. Particle size of the wet ball milled sample was decreased below 1 micron using planetary ball mill

machine. TL glow curve of ball milled phosphor shows same glow curve structure and temperature peak slightly shifted towards lower temperature scale. As the particle size of phosphor decreases then sensitivity of phosphor also decreases. The TL sensitivity of wet ball milled phosphor decreases drastically (144 times). Ball milled phosphor exhibits linearity upto the 6 kGy. Hence phosphor can be useful in application of high dose dosimetry.

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