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Thermoluminescence (TL) in Eu doped Na_3SO_4F Fluoride Phosphor

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Abstract

Polycrystalline Na_3SO_4F: Eu phosphor prepared by a wet chemical method has been studied for its thermoluminescence (TL) characteristics. The TL glow curve of the compound has a prominent peak at 200 ^0C and may be useful for TL study. TL sensitivity of the Na_3SO_4F: Eu phosphor is found to be more sensitive than that of TLD-$CaSO_4$: Dy. TL glow curve of the Na_3SO_4F: Eu gives a better understanding of the TL mechanism (peaking at 200 and 290 ^0C) involved in the concerned phosphor. In this paper we report TL characteristics of Na_3SO_4F: Eu halosulphate phosphors first time.

Keywords: Fluoride phosphors; thermoluminescence; TLD material.

Introduction

The aim of study is to produce in simple way Na_3SO_4F: Eu sulphate phosphor and its TL characteristics with high intensity. The use of sulphates doped with rare earth (RE) ions as dosimeters have been investigated for many years. Some examples are $CaSO_4$: Eu, $CaSO_4$: Eu, P and $CaSO_4$: Tm. $CaSO_4$: Dy and $CaSO_4$: Tm phosphors have been widely used in environmental radiation monitoring due to their high dose response sensitivity and ease of preparation. At present, thermostimulated luminescence dosimeter (TLD) materials are widely used for personal and environmental radiation monitoring. Among the different types of TLD materials, RE-doped sulphates, especially $CaSO_4$: Dy or $CaSO_4$: Tm phosphors, developed are in use in many countries for dosimetry due to their high sensitivity, stability and low cost.

Fluorides were widely investigated in the early 1960s and 1970s to characterize the lattice defects associated with them. Among all of the fluorides, CaF_2 doped with rare earth impurity ions was extensively studied because of its high sensitivity and its ability to store the incident energy[1,2] giving it suitability for radiation dosimetry. The material has been marketed as a commercial TL dosimeter, CaF_2: Eu, under the commercial name TLD-200. On the other hand, luminescence studies of SrF_2 and BaF_2[3-6] doped with rare earths has received much less attention, despite their intense luminescence. For BaF_2 Lucas and Kapsar[7] studied the luminescence properties of BaF_2: Eu and the possibility of using this material in radiation measurements. Their results show that the BaF_2: Eu system exhibits intense thermoluminescence. Compared to the most commonly used TL dosimeter, it is 20 times more sensitive than TLD-100 and twice as sensitive as TLD-200. However, the problems of supralinearity and high fading preclude using this material in radiation dosimetry. Earlier TL models in fluorides at low temperature[8-10] are usually built on the inherent assumption of separated traps (intrinsic defects) and recombination centres (RE sites). This conclusion is mainly based on the observation that the glow peaks in the fluorite structure occur, more or less, at the same temperature irrespective of the RE dopant. Recently we have reported new halosulphate phosphors[11-13]. In the present paper we report Europium activated phosphor prepared by wet chemical method and characterized thermoluminescence (TL) properties.

Material and Methods

Na_3SO_4F (pure); and Na_3SO_4F: Eu phosphors were prepared by a wet chemical method[14]. Na_2SO_4 and NaF of analar grade were taken in a stoichimetric ratio and dissolved separately in double distilled de-ionized water, resulting in a solution of Na_3SO_4F (equation 1). Water-soluble sulphate salt of Europium was then added to the solution to obtain Na_3SO_4F: Eu. Confirming that no undissolved constituents were left behind and all the salts had completely dissolved in water and thus reacted.

$$Na_2SO_4 + NaF \rightarrow Na_3SO_4F \quad (1)$$

The compounds Na_3SO_4F (pure) and Na_3SO_4F: Eu in its powder form was obtained by evaporating on 80^0C for 8 hours. The dried samples were then slowly cooled at room temperature. The resultant polycrystalline mass was crushed to fine particle in a crucible. The powder was used in further study. Formation of the compound was confirmed by taking the x-ray diffraction (XRD) pattern that matched with the standard data available. For TL characteristics samples were exposed to gamma rays from a Co^{60} source at room temperature at the rate of 0.995 kGy/hr for 500 rad (5 Gy). After desired exposure, TL glow curves were recorded for 2 mg of sample each time at a heating rate of 2 ^0C/sec. TL glow curves were recorded on TLD reader. For comparison glow curves were also recorded under identical conditions for dosimetry grade commercially available TLD - $CaSO_4$:Dy.

Results and Discussion

TL glow curves of Na_3SO_4F: Eu: Figure 1 shows the X-ray diffraction (XRD) pattern of Na_3SO_4F material. The XRD pattern did not indicate presence of the constituents Na_2SO_4 or NaF and other likely phases. The XRD pattern of prepared material, matches with the standard data of $NaMgSO_4F$ from JCPDS file no. 75-1599. These results indicate that the final product was formed in homogeneous form. Figure 2 shows glow curves for Na_3SO_4F: Eu exposed to γ – rays for 500 rad at the rate of 0.995 KGy/hr for various concentrations of Eu i.e. i. Na_3SO_4F: Eu $_{0.5\%,}$ ii. Na_3SO_4F: Eu $_{0.2\%}$ iii. Na_3SO_4F: Eu $_{0.1\%}$ iv. Na_3SO_4F: Eu $_{0.05\%}$. It is observed that, TL intensity increases with the increase in concentrations of Eu. In order to investigate further significant effect of the RE doping on the intensity of the luminescence from the Na_3SO_4F: Eu materials, the measurement of thermoluminescence glow curves of this material was carried out. The maximum thermoluminescence intensity peak of Na_3SO_4F: Eu was observed at temperatures between 200 and 300^0C (figure 2). The maximum of the glow curve at 200 ^{0}C, beyond which the TL intensity decreased rapidly until the second weaker maximum at 290 ^{0}C. The curve consists of three maxima next to each other and thus the formation of new traps was confirmed. Curve (e) of figure 2 shows glow curves for the phosphor along with those of TLD-$CaSO_4$: Dy for an exposure of 500 rad of γ –rays taken for comparison and shows Na_3SO_4F: Eu is more sensitive than the standard TLD-$CaSO_4$: Dy phosphor. The peaks of Na_3SO_4F: Eu phosphor for all concentrations and peak of standard TLD-$CaSO_4$: Dy phosphors are located at the nearly same temperature site.

Figure 3 shows TL glow curves for γ –irradiated Na_3SO_4F: Eu phosphor with varying doses with a linear heating rate 2 ^{0}C.sec^{-1} which exhibits a glow peak at 200^0C and 290^0C. All the phosphors were exposed to a dose range of 1 Gy to 5 Gy. It is observed that TL intensity increases with the increase in dose. This change in the relative intensity of the peaks has been attributed to the change in the population of the luminescent/trapping centers. With a simple glow curve structure, easy method of preparation and no saturation in the exposed range Na_3SO_4F: Eu phosphor resembles the characteristics close to an ideal phosphor.

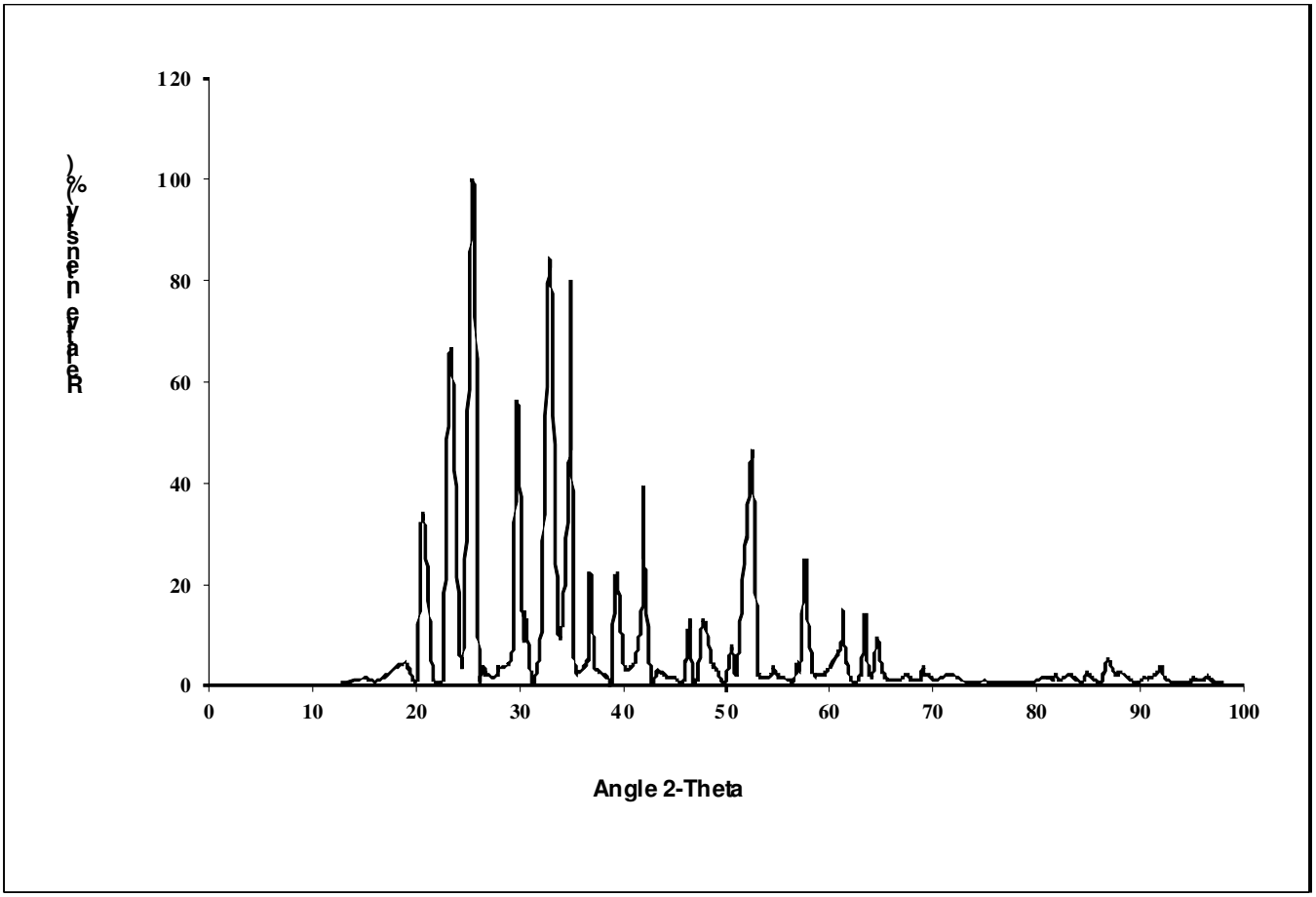

Figure-1
XRD Pattern of Na_3SO_4F[14]

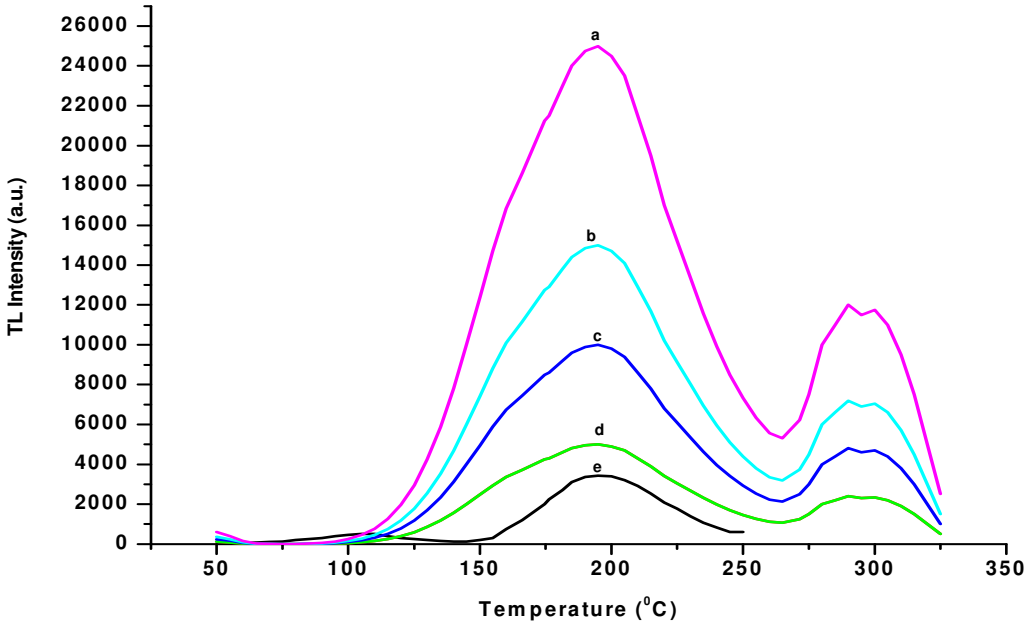

Figure-2
TL glow curves of various concentrations exposed to γ – rays for 500 rad at the rate of 0.995 KGy/hr i. Na_3SO_4F: Eu $_{0.5\%}$ ii. Na_3SO_4F: Eu $_{0.2\%}$ iii. Na_3SO_4F: Eu $_{0.1\%}$ iv. Na_3SO_4F: Eu $_{0.05\%}$ e) TLD-$CaSO_4$:Dy

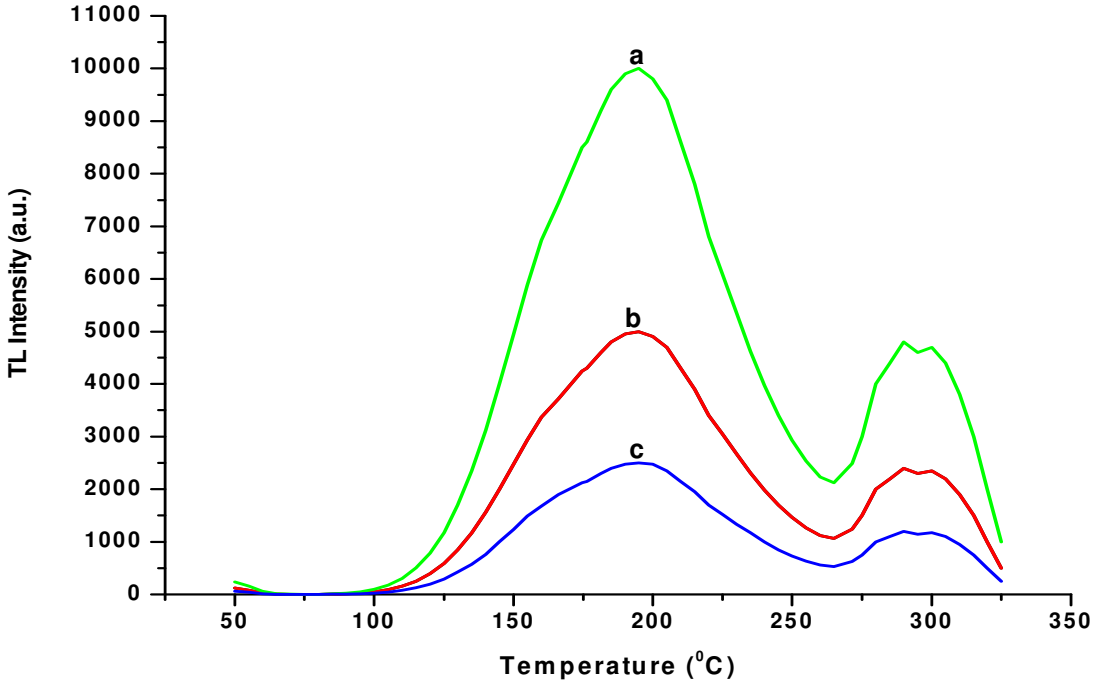

Figure-3
TL glow curves of Na_3SO_4F: Eu $_{0.1\%}$ for various doses exposed to γ – rays for i. 500 rad (5 Gy) ii. 300 rad (3 Gy) iii. 100 rad (1 Gy) at the rate of 0.995 kGy/hr

Conclusion

The TL glow-curves of very sensitive Na_3SO_4F: Eu phosphor for different concentrations have been recorded at a heating rate of 2 $^0Csec^{-1}$ and irradiated at a dose rate of 0.995 $kGyh^{-1}$ for 5 Gy and it was found that Na_3SO_4F: Eu is suitable for higher concentration of Eu. TL glow curves for γ –irradiated phosphor with varying doses at a linear heating rate 2 $^0C.sec^{-1}$ exhibit a single glow peak at 200 ^0C and 290 ^0C when exposed to a dose range of 1 Gy to 5 Gy. It is observed that TL intensity increases with the increase in dose. The phosphor is compared with standard $CaSO_4$: Dy phosphor and it is observed that presented phosphor is more sensitive than $CaSO_4$:Dy.However, more excellent results can be obtained for higher doses of γ – radiation.

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