

Development of Sintered Composites of CaSO_4 Doped With Eu^{3+}

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Abstract. The main purpose of this work is the developing of sintered composites in order to allow their use as thermoluminescent dosimeters using a simple route to preparation, employing high temperature. The composites were prepared with $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ doped with EuO_3 . Initially $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ was dehydrated in oven at 100°C for 1h. After addition of dopants, glass and homogenization, PVA was incorporated into the compounds to make their compaction easier. The pellets with 6 mm of diameter and 1 mm thick were sintered at 750°C for 6 h. The TL emission curves showed two peaks when the samples were irradiated with beta source (^{90}Sr - ^{90}Y) and X-ray. During calibration, TL responses were found to be proportional to the absorbed dose. The results showed that this material can potentially be used in dosimetry.

1. Introduction

Many authors have shown results from studies on the subject of doped detectors, revealing various of their properties. Those studies includes the thermoluminescent (TL) response to dose; TL emission kinetic; specific ionization density; enabled dopants and their concentrations, co-dopants; processes of energy transfer; thermally stimulated exoelectronic emission, results from of optical absorption and paramagnetic resonance measures[1-5]. The elements from the group of rare earth (RE) are frequently used as dopants of the CaSO_4 for the production of thermoluminescent detectors; amongst them, the most often used are: Dy, Tm and the Eu [5-10]. The model for the thermoluminescent emission of the doped CaSO_4 with RE is the one of energy transference. According to this model, there is no direct recombination of charge carriers due to the small quantity of the dopant (RE), but the energy of the electron-hole recombination is transferred to the RE^{+3} . However, no direct evidence has been published so far [3]. The europium is a soft, malleable and ductile silvery-looking metal that shows the electronic configuration $[\text{Xe}] 4f^4 6s^2$ [10]. The europium presents electronic configuration $[\text{Xe}] 4f^7 6s^2$, it is classified as a metal of internal transition and it is solid in its natural state [10].

In this work we are considering a new composite to be employed in the dosimetry of the ionizing radiations to the based on CaSO_4 , using the europium and glass for use in thermoluminescent of ionizing radiation dosimetry, mainly. We decided to use the glass in order to sintering the composites at a temperature lower than the melting point (1450°C). Normally, the thermoluminescent dosimeters based on CaSO_4 are produced with Teflon. However, there is a limitation for the use of this material as a binding because Teflon volatilizes at temperatures higher than 300°C .

2. Methodology and Materials

The samples were prepared in the Laboratory of Materials Preparation and Characterization Department of Physics of Federal University of Sergipe. It was used CaSO_4 with 98% of pureness (Labsynth Ltda) doped with 0.1, 1 and 2% of EuO_3 with 99.9% of pureness (Aldrich Chemical Company Inc.) and recycled window glass samples classified as transparent for industrial usage were utilized. The glass sample was crushed into pieces with diameter of 63 μm . After the milling step, the glass powder was dried and mixed with the topaz crystals (50% in mass). The samples were mixed with 10 ml of distilled water and taken to a magnetic agitator where they remained for 30 minutes. After that, drying took place in oven at a temperature of 100°C for 24 h. The dried powder was homogenized again in mortar with the polyvinilic alcohol (PVA) addition. The pellets were submitted to a uniaxial presage of 100 kgf/cm^2 and sintered at temperatures of 750°C for 6h, with a rate of heating of approximately 10°C per minute, and a free rate cooling down to room temperature in order to obtain pellets with 1.0mm thickness and diameter of 6.0mm.

A beta source ($^{90}\text{Sr}+^{90}\text{Y}$) of the dermatological applicator type and an x-ray device of clinical application (VMI Compact Plus - 500) was used to irradiate the pellets. The beta source had a $6.2 \cdot 10^{-3}$ Gy/s dose rate. The x-ray device has a 40 at 125 kV tube voltage and a 0.2 mAs to 150 mAs at 70 kV tube current time products. The x-ray beam was previously calibrated using an ionizing chamber. Before each exposition, the pellets were submitted at thermal annealing at the temperature of 400°C to 30min.

The measures of thermoluminescence were carried out at interval up to 6 h after irradiation. Measurements were made with a Harshaw 3500 thermoluminescent reader using a heating rate of 10°C/s. The maximum deviation found in the TL intensity was of $\pm 6.5\%$.

3. Results

Initially, was made a study of the possibilities of preparation of $\text{CaSO}_4:\text{Eu}^{3+}$ with different concentrations of dopants. The best results were showed to the composites doped with 2%. Figure 1 shows the TL glow curves of $\text{CaSO}_4:\text{Eu}^{3+}$ + glass performed using 2% of Eu^{3+} whiter that has been irradiated with 22.6 Gy from a beta source. TL emission exhibits two peaks with different intensities, one high TL peak with maximum of intensity at 195°C and a short one at 270°C. The other ones prepared with less dopant present similar TL peaks, but with reduced intensity. The glow curves of $\text{CaSO}_4:\text{Eu}^{3+}$ + glass are similar to the observed by Bernal at al [11]. The TL peaks there are at a high enough temperate so that it is not necessary a pre-annealing in the using of dosimetric procedures.

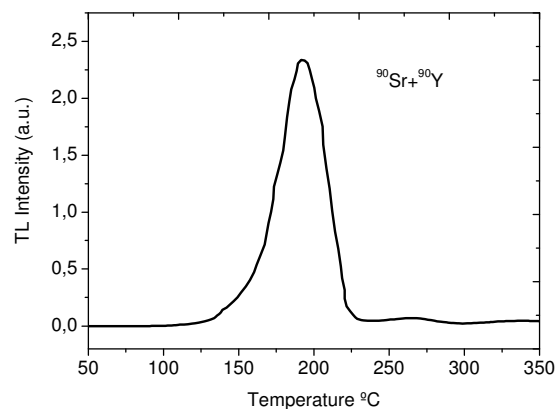


Figure 1. Typical TL emission curve of a $\text{CaSO}_4:\text{Eu}^{3+}$ + glass composites irradiated with $^{90}\text{Sr}+^{90}\text{Y}$.

The pellets exhibits a good rigidity and they showed no break or weight loss during the routine experimentation.

Pellets of $\text{CaSO}_4:\text{Eu}^{3+}$ + glass can be used in successive irradiation–reading–annealing cycles without change in their sensitivity.

Figure 2 shows the TL response of the $\text{CaSO}_4:\text{Eu}^{3+}$ + glass pellets irradiated with doses between 2.6 and 26.2 Gy of a $^{90}\text{Sr}+^{90}\text{Y}$ source. The TL intensity was proportional to the dose in all this range. The measurement uncertainties in those studies were always below 5%.

Figure 3 shows the glow curves of the pellets which were *x-irradiated* with $1.65 \cdot 10^{-4}$ Gy. The glow curve was similar at that one irradiated with beta source.

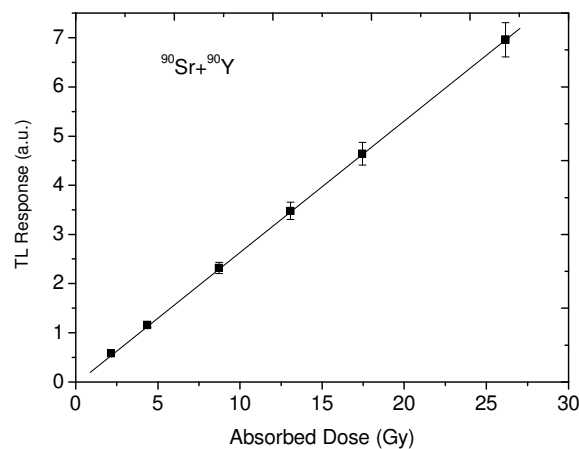


Figure 2. TL response of the $\text{CaSO}_4:\text{Eu}^{3+}$ +glass pellets as a function of absorbed dose in the range of 3.27 to 22.6 Gy ($^{90}\text{Sr}+^{90}\text{Y}$).

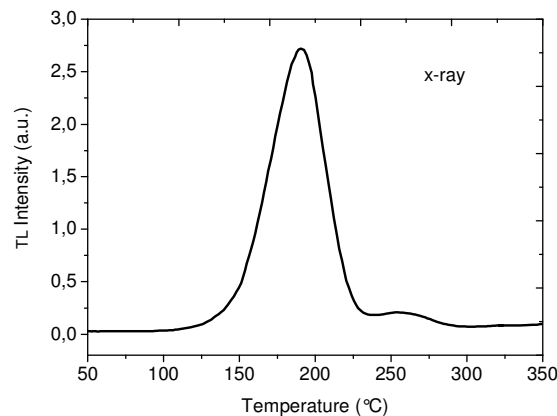


Figure 3. Typical TL emission curve of a $\text{CaSO}_4:\text{Eu}^{3+}$ + glass composites irradiated with x-ray.

In the figure 4 the TL response of the pellets was plotted as a function of the absorbed dose of x-rays. Calibration curves presented responses proportional to absorbed dose between 1.5 Gy and 6.0 Gy for the x-radiation.

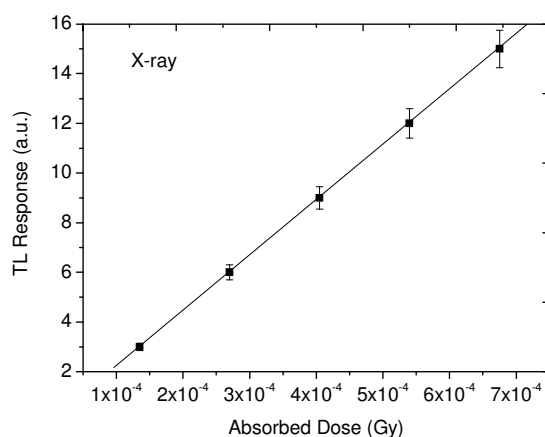


Figure 4. TL response of the $\text{CaSO}_4:\text{Eu}^{3+}$ glass pellets as a function of absorbed dose from x-ray.

4. Conclusions

The currently accepted procedures of radiological protection aim at reducing the exposition associated with practices that make use ionizing radiation, both for the occupationally exposed workers and for the environment. Therefore it is important to use detectors of high sensitivity in the personal and the environmental dosimetry. According to this study, it could be observed that the CaSO_4 detector doped with europium is appropriate for dosimetric purposes. Due the previously mentioned properties associated with their low cost, the described detectors could be used in teaching laboratories as a complement of didactic practices involving the use of other kinds of radiation detectors. Further techniques for preparation of the described composites, as well as other radiation sources for testing the pellets, could be within the scope of future works.

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References

- [1]. Cristovan F H, Eiras S P, W. Cruz O and Dias I. F. L 2005 *Quim. Nova*, **28**, (6) 964
- [2]. Hinatsu Y, Doi Y, Ito K 2003. *J. Solid. State Chem.* **172** (2) 438
- [3]. Lakshmanan A R 1999 *Progr. In Mater. Sc.* **44**: 1
- [4]. Q. Tang, C.X., Zhang, P.L., Leung, M., Li; D.L., Luo, 2005. *Acta Phys. Sinica* **54** (1): 64-69.
- [5]. K.N.R., Taylor; M.I., Darby, 1972. *Physics of Rare Earth Solids Chapman and Hall LTD.*
- [6]. Patil R R, Muthal P L, Dhopt S M, Kondawar V K and Moharil S V 2006 *J. Lumines.* **126** 517
- [7]. Cameron J R, N.; Suntharalingam, G. N., Kenney, 1968. *Thermoluminescent Dosimetry*. The University of Wisconsin Press.
- [8]. L. L., Campos; M. F, Lima, 1983. *J. Lumin.* **28** 3.
- [9]. M. A. P. Chagas, 2005. *Estudos espectroscópicos de cristais dopados com o íon Nd^{3+}* . *Dissertação de Mestrado*. Universidade Federal de Sergipe.
- [10]. D.R, Lide, 2006-2007. *Handbook of Chemistry and Physics* **87** Edition.
- [11]. Bernal R, García-Haro A R, Machi L, Brown F, Pérez-Salas R, Castaño V M and Cruz-Vázquez C, 2008. *Rad. Meas.* **43** 371.