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TL properties of anhydrous $CaSO_4$:Tm improvement

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ABSTRACT

CaSO4:Tm is excellent phosphors for dosimetry of ionising radiations due to their high sensitivity, wide dose range (from 10^{-6} to 1 Gy), relatively simple trap structure, good chemical, thermal and physical stability and ease of preparation. Since 1968 have been developed new methods of preparing aim to increase the sensitivity and the linearity of the $CaSO_4$:Tm thermoluminescence. The objective of this work is to obtain single crystals of $CaSO_4$:Tm under controlled crystal growth conditions, such as temperature, atmosphere and velocity, aiming to improve the TL sensitivity of the crystals and investigate its dosimetric properties in order to evaluate the applicability of this material to dosimetry.

1. Introduction

Calcium sulphate doped with thulium is an excellent thermoluminescent phosphors for the dosimetry of ionising radiations due to its high sensitivity, wide dose range (from 10^{-6} to 1 Gy), relatively simple trap structure, good chemical, thermal and physical stability and ease of preparation.

At present, CaSO4:Tm is among the most sensitive thermoluminescence dosimeter (TLD) so that its application in the measurement of small doses is extensive (Poston, 2005; Nariyama et al., 1997). Four decades ago, Yamashita et al. (1968, 1971) developed an excellent and relatively simple method for preparing calcium sulphate doped with rare earths. The $CaSO_4 \cdot 2H_2O$ and Tm_2O_3 were dissolved in hot concentrated sulphuric acid and the acid was slowly evaporated. This method of production of $CaSO_4$:Tm was widely applied. Later, the method was modified by some researchers and new methods of preparing $CaSO_4$:Tm have been widely developed, including co-dopants, tending to increase the sensitivity on one hand and increase the upper limit of linearity on the other hand (Campos, 1983; Sohrabi et al., 1998; Lakshmanan, 2005; Kása et al., 2007).

The objective of this work is to obtain single crystals of $CaSO_4$:Tm under controlled conditions such as temperature, atmosphere and velocity, aiming to improve the TL sensitivity of the crystals and to investigate its dosimetric properties in order to evaluate the applicability of this material to dosimetry.

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2. Materials and methods

The dosimetric powder was prepared employing a variant of the method described by Yamashita and modified by Campos (1983) which consists of dissolving a mixture of calcium carbonate ($CaCO_3$), and thulium (III) oxide (Tm_2O_3), in an excess of sulphuric acid (H_2SO_4) that is dried at a temperature of 375 °C in a vacuum chamber, the vapor inside the chamber is taken in a controlled manner in order to prevent interference in the crystal growth process. Doped crystals were rinsed three times with cold and hot water which was previously distilled and deionized. After washing, the powder crystals presented pH 5. Powder samples, crumbled and grain-selected between 75 and 180 μm, were used in the dosimetric analyses. Before the gamma and beta exposures the powder was annealed at 600 °C for 1 h and cooled at room temperature in open air.

Samples were exposed to gamma source (^{60}Co) in a range from 0.01 to 10 Gy in air and electronic equilibrium conditions and to beta source (^{90}Sr+^{90}Y) with doses ranging from 1 to 1000 Gy. The powder was irradiated with UV and visible light. UV exposure was performed using two lamps of 20 W with a wavelength between 10 nm and 400 nm and energies between 3 eV and 124 eV. With the visible light, the samples were exposed to light from four 40 W lamps with wavelengths between 400 nm and 800 nm for 24 h. These exposures were performed after the irradiation with the beta source. Lamps were distant 1.0 m from the samples.

TL measurements were made with a Harshaw 3500 thermoluminescent reader using a heating rate of 10 °C/s. Uncertainty in TL measurements was lower than 6.5%.

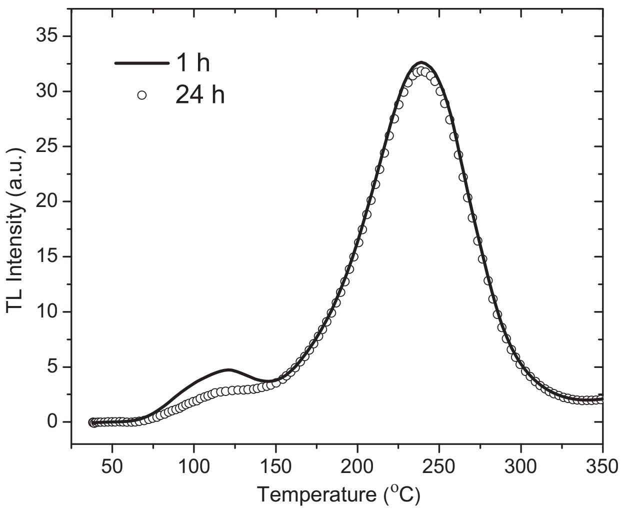

Fig. 1. Typical TL emission of $CaSO_4$:Tm exposed to gamma rays (^{60}Co).

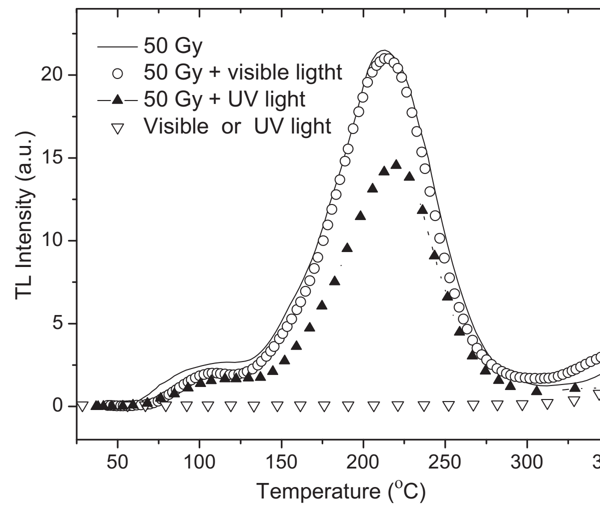

Fig. 3. Influence of visible light and UV light on TL emission of $CaSO_4$:Tm.

Measurements were made with a Harshaw 3500 thermoluminescent reader using a heating rate of 10 °C/s. The maximum shunting line found in the measures was of ±6.5%.

3. Results

Fig. 1 shows the TL emission of $CaSO_4$:Tm powder samples after exposure to 5 Gy of gamma rays and storage for 1 h and 24 h. It was observed that the first TL peak, around 120 °C, fades quickly, almost 37% after 24 h when stored at room temperature. Under the same conditions, TL intensity of the dosimetric glow peak at 220 °C did not exhibit significant change during storage; this peak decreases merely 1.5%. The samples exposed to 20 Gy of beta radiation showed TL glow curves similar to the samples exposed to gamma rays, as can be seen in Fig. 2; in this case the first TL peak decayed approximately 45% and the second 2.5%. The observed variation in peak position is probably due to small oscillations of electric current in the reading device. These variations induced changes in the temperature of the TL emission but no changes in TL intensity. Either way, the TL peak position was shown to be excellent for dosimetric applications.

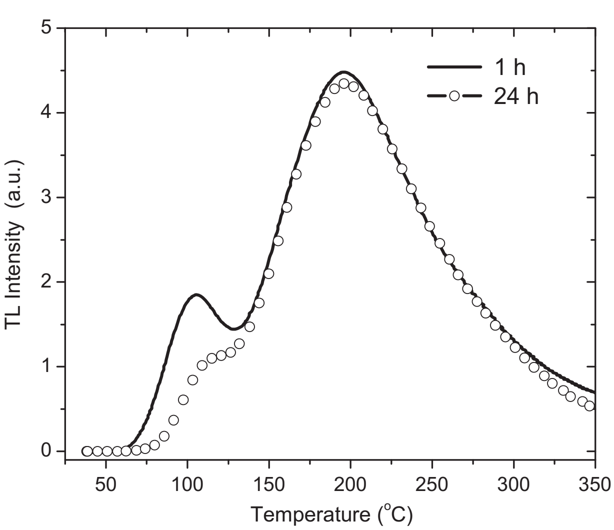

Fig. 2. Typical TL emission of samples exposed to beta rays ($^{90}Sr+^{90}Y$).

The analyses performed with samples that were thermally treated and then exposed to visible light and UV light for 24 h showed that these radiations do not induce TL emission (Fig. 3). This material has thermally stable traps resulting in a dosimetric peak at around 220 °C which is sensitive to low doses. However, the origin of TL peaks in the material can still be subject of further investigation.

Visible light produces no significant changes on TL signals of the samples pre-irradiated with beta source. TL emission of the pre-irradiated samples had a considerable decrease of the dosimetric peak when those were exposed to UV light; the decrease was approximately 67% in 24 h of irradiation. Such results indicate that the material can be exposed to artificial light wavelength higher than 400 nm, but not to sunlight or artificial UV light.

In order to analyze the TL response with the dose the samples were irradiated with gamma source in a range from 10^{-2} to 10^{1} Gy. TL response showed a linear behavior with the dose. The TL intensity has a monotonic increase as a function of the dose.

In order to study the influence of annealing temperature on the TL of $CaSO_4$:Tm we prepared three batches in which each one was thermally treated at a specific temperature, 350, 400 and 450 °C. After the thermal annealing the samples were irradiated with absorbed doses in the range from 1 to 10^3 Gy of beta rays. The TL response of the material is supralinear in this dose range to this radiation. Although a supralinearity on the TL response is not desired, that behavior does not preclude the use of this material in dosimetric procedures, provided that the TL is known at a broad range of doses.

4. Conclusions

The dosimetric powder of $CaSO_4$:Tm prepared employing a variant of the method described by Yamashita et al. (1971) and modified by Campos (1983) showed to be appropriate for dosimetric purposes. The TL peak around 200 °C was shown to be dependent on absorbed dose. Annealing before sample irradiation was found to influence the TL intensity. Sample thermally treated at 400 °C showed the highest TL intensity. In the dose ranges analysed the TL response of the material is linear between 10^{-1} and 10 Gy to gamma radiation (^{60}Co) and slowly supralinear between 1 and 10^3 Gy to beta radiation ($^{90}Sr+^{90}Y$). Visible and ultraviolet light are not efficient to produce TL emission; however UV exposure promotes the intensity reduction of the dosimetric TL peak. The material apparently is not affected by light at wavelengths lower than 400 nm, whereas a significant light induced fading is observed when exposing the material to UV light, i.e. daylight. It then means that $CaSO_4$:Tm dosimeters cannot be handled in daylight. The TL response as a function of absorbed dose suggests that $CaSO_4$:Tm can be a good alternative material for dose measurements.

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