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Feasibility study of CaSO₄:Tb,Yb as a thermoluminescent dosimeter

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HIGHLIGHTS

► A new dosimeter based on CaSO₄ and doped with Tb and Yb is proposed.
► The samples were grown using a production route based in the Yamashita method.
► CaSO₄:Tb,Yb glow curves exhibited a TL emission in temperatures from 100 to 270 °C.
► The TL responses of the composites produced are proportional to the dose absorbed.
► The CaSO₄:Tb,Yb has potential to be used as a thermoluminescent dosimeter.

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ABSTRACT

A new composite based on CaSO₄, using terbium as dopant and ytterbium as co-dopant (CaSO₄:Tb,Yb), was developed for employment as a thermoluminescent (TL) dosimeter. The crystals used in this work were grown using a production route based on the Yamashita method (Yamashita et al., 1968). Crystal powder was calcined at 600 °C for 1 h. Pellets were made by adding commercial and colorless glass to improve physical resistance and sintered at 700 °C for 6 h. All samples were irradiated by a beta source (90Sr/90Y) and received doses from 1 Gy to 5 Gy. TL analyses have been performed and characteristics such as sensitivity, reproducibility, linearity, and fading have been studied. The CaSO₄:Tb,Yb pellets glow curves presented two peaks, the first at around 115 °C, and the second at around 200 °C. The highest intensity was shown for CaSO₄:Tb,Yb with a concentration of 0.1 mol% of Tb and Yb together. In all the samples the TL response was proportional to the absorbed dose. Therefore, the CaSO₄:Tb,Yb has potential to be used as a thermoluminescent dosimeter.

1. Introduction

Calcium sulphate (CaSO₄) with different dopants is one of the most investigated materials to be used as a thermoluminescent dosimeter (TLD) (Campos, 1983). Thus, it is important to search for new doping elements for the base material in order to improve the sensitivity and the thermoluminescent (TL) response of the material to radiation (Mckeever et al., 1995).

Rare earth (RE) elements are often used as dopants for the production of CaSO₄ thermoluminescent detectors. Yamashita et al. (1968), (1971) developed a relatively simple and efficient method for doping CaSO₄ using rare earths. This method has been widely used and modified by various researchers (Kása et al., 2007; Yang et al., 2004; Junot et al., 2011).

In this work we consider a new composite based on CaSO₄, using terbium as dopant and ytterbium as co-dopant (CaSO₄:Tb,Yb), to be employed as a TLD for the measurement of ionizing radiation. The TL of CaSO₄:Tb,Yb has been studied with the aim of evaluating its applicability to dosimetry, considering characteristics such as sensitivity, dose response, reproducibility, and fading. In order to characterize the samples by this technique, composites were produced with the addition of glass.

2. Materials and methods

The crystals used in this work were grown using a production route based on the Yamashita method (Yamashita et al., 1968). Calcium carbonate (CaCO₃) (J.T. Baker 99%) was mixed with terbium oxide (Aldrich 99.9%) at a concentration of 0.1 mol% and with ytterbium oxide (Aldrich 99.9%) at concentrations of 0.1 mol%, 0.2 mol%, and 0.5 mol%, which will result in samples with terbium to ytterbium ratios of 1:1, 1:2 and 1:5, respectively. After this, the compounds were placed in a beaker with an excess of sulfuric acid (Vetec) and mixed by a magnetic stirrer at 350 °C.
under constant pressure of 1 atm until all the acid had evaporated (about 48 h), leaving only doped calcium sulfate.

After growing process, the doped crystals were rinsed three times with cold and hot distilled water, calcined at 600 °C for 1 h, crumbled, rinsed again, and grain-selected to obtain grain sizes between 75 and 180 μm.

Pellets were made using a 2:1 (wt) mixture of the phosphor and powdered commercial and colorless glass to improve physical resistance. The pellets were compacted by application of uniaxial pressing of 100 kGF and sintered at 700 °C for 6 h. After each irradiation-readout cycle the pellets were annealed for 1 h at 400 °C.

The samples were irradiated with a 90Sr/90Y source at a rate of 0.35 Gy per minute and received doses from 1 Gy to 5 Gy.

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

To study the fading of the composites, TL analyses were measured at the following times: immediately after irradiation, one week after irradiation, two weeks after irradiation, and one month after irradiation. Percentage decay of the TL intensity during storage period was calculated by comparison with the TL signal of the sample read immediately after irradiation.

3. Results

TL analysis of CaSO₄:Tb,Yb pellets, with terbium to ytterbium ratios of 1:1, 1:2, and 1:5, respectively, exhibited glow curves with TL emission in the temperature range from 100 to 270 °C. Fig. 1 presents a comparison among the glow curves of these three samples after exposure to 1 Gy of beta radiation. Two peaks can be observed: one at 130 °C, whose maximum intensity is obtained for the samples with 0.1 mol% of Tb and Yb, and the other at 240 °C whose maximum intensity is obtained for the samples with 0.1 mol% of Tb and 0.2 mol% of Yb. This demonstrates that the traps of CaSO₄:Tb,Yb with 0.2 mol% of Yb are deeper than those of the other compounds studied.

Fig. 2 shows the calibration curves of CaSO₄:Tb,Yb at doses from 1 Gy to 5 Gy deposited with beta radiation (90Sr/90Y). It can be observed that the thermoluminescent responses of the composites produced are proportional to the dose absorbed in this dose range.

With the goal of studying the reproducibility of the CaSO₄:Tb,Yb, five samples were irradiated with 1 Gy of beta radiation, read, annealed, and irradiated again, creating a cycle that was performed eight times. As can be seen in Fig. 3, all TL responses were similar with only around 5% deviation.

Table 1 shows the decay of the TL signal of the CaSO₄:Tb,Yb produced. Although the peak centered at 130 °C practically fades out after one month, the fading of CaSO₄:Tb,Yb appeared acceptable because in 30 days the peak centered at 240 °C which is the dosimetric peak, fades by 35% for a 1:5 ratio of Tb to Yb, 32% for a 1:2 ratio, and 19% for a 1:1 ratio.

It is important to consider that the fading is related not only to the composition of the dosimeter but also to the storage conditions during the time between irradiation and reading.

4. Conclusions

The CaSO₄:Tb,Yb phosphorous analyzed in this work showed TL glow curves with two peaks. The peak centered at 240 °C,
although not the most intense, could be used as the dosimetric peak due to its good sensitivity, reproducibility, and fading acceptable. The highest intensity was shown for CaSO$_4$:Tb,Yb with a concentration of 0.1 mol% of both Tb and Yb. In all samples the TL response was proportional to the absorbed dose. Commercial colorless glass was shown to be appropriate for incorporation with the phosphors for the production of dosimeters in the form of pellets. Due the previously mentioned properties associated with their low cost, CaSO$_4$:Tb,Yb has potential for use as a thermoluminescent dosimeter.

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References


