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STUDY OF DOSIMETRIC PROPERTIES OF $CaSO_4$:Mn PELLETS PRODUCED AT IPEN

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ABSTRACT

The manganese activated crystals of calcium sulfate ($CaSO_4$:Mn) are thermoluminescent and the dosimetric properties of this material are known. Depending on the heating rate, one TL emission peak is showed at about 100 ºC and the wavelength is 500 nm. These crystals present high sensitivity to ionizing radiation, but the emission in that temperature region implies in a pronounced fading of the TL signal at room temperature. The parameters to obtain $CaSO_4$:Mn crystals were established in a previous work, where the optimum Mn concentration is 1.0 mol %. In practice, the use of these crystals is made easier in pellet shape, with defined mass of the component materials. The $CaSO_4$:Mn pellets were produced by pressing and sintering crystals with grain size between 75–180 μm mixed with polytetrafluoroethylene (PTFE). The aim of this work is the study of dosimetric properties of $CaSO_4$:Mn + PTFE pellets produced at IPEN. The studied parameters were: glow curve, TL emission peak temperature, fading, thermal treatment for reutilization, dose response, lower detection dose and reproducibility. Because of inherent features the $CaSO_4$:Mn pellets must be utilized under well controlled conditions, observing especially the storage of irradiated pellets within constant temperature, as well as the elapsed time after irradiation. Following a established procedure, the reproducibility of the TL measurements of $CaSO_4$:Mn pellets is better than ± 3%.

1. INTRODUCTION

The dosimetric properties of manganese activated crystals of calcium sulfate ($CaSO_4$:Mn) are known and described [1, 2]. This material is very sensitive to ionizing radiation, UV and visible light; it presents one emission peak at about 100 ºC and the wavelength is 500 nm.

The TL emission peak positioned in the low temperature range results in the fast fading of the TL signal when irradiated crystals are maintained at room temperature. The use of these crystals is not recommended for personal dosimetry, when the dosimeter is used by workers under several climatic conditions, becoming the dose assessment very complex or impossible.

Considering that the TL emission peak is simple and located in the region that practically is not influenced by heating incandescence, $CaSO_4$:Mn is an excellent material for the study of dosimetric properties[2, 3]. It can be used for research and low doses measurements, if some parameters are controlled.

The aim of this work is the study of dosimetric properties of $CaSO_4$:Mn + PTFE pellets produced at IPEN. It includes glow curve, TL emission peak temperature, fading, thermal treatment for reutilization, dose response, lower detection dose and reproducibility.

2. MATERIALS AND METHODS

The $CaSO_4$:Mn crystals were prepared by the dissolution of high purity $CaCO_3$ and $MnSO_4.H_2O$ in hot concentrated sulfuric acid, followed by slow evaporation of the solvent in a well closed system to avoid that acid vapors escape to the atmosphere. The acid vapors were dragged by an air flux by means of a suction pump, and collected in gas washer bottles with NaOH solution.

The optimum concentration of Mn^{2+} is about 1.0 mol %. The parameters to obtain $CaSO_4$:Mn crystals were established in a previous work [4].

After the sulfuric acid evaporation the $CaSO_4$:Mn crystals were washed, ground and sieved. The 75-180μm crystal powder was mixed with polytetrafluoroethylene (PTFE) and pressed in pellets of 6mm diameter and 0.050g weight. The pellets were sintered in air at 400ºC for 1 and a half hour in a microwave furnace.

The pellets were submitted to annealing and to gamma irradiation of a ^{60}Co or ^{137}Cs source. The dosimetric properties were analyzed by TL emission.

The main utilized equipments were J. L. Shepherd pneumatic TLD dosimeter irradiator with ^{137}Cs sealed source whose activity was 0.231 10^{10}Bq on the 16th of Jun, 2007, irradiation system assembled at IPEN with ^{60}Co sealed source whose activity was 0.214 10^{9}Bq on the same date, Harshaw TL reader 2000 A and B model with data acquisition system and CEM Corporation microwave muffle furnace MAS-7000 model.

3. RESULTS

3.1. Glow Curve and Emission Peak Temperature.

The glow curve of $CaSO_4$:Mn has only one peak. The temperature of TL peak (T_m) depends on the heating rate, and can appear between 100 and 160 ºC in the crystal form [2]. T_m is higher in the pellets than in the crystal form, mainly due to the PTFE present in the pellet. Fig.1 shows the glow curves of irradiated pellets with three different doses. The TL peaks are situated around 138 ºC, using a heating rate of 9.9 ºC.s^{-1}.

Fig.2 shows the peak shift to higher temperature when the heating rate was increased. The peak temperature (T_m) variation with the heating rate (β) was used to determine the activation energy or trap depth (E) and frequency factor (s) using the equation (1) for a straight line.

$$\ln(\beta / T_m^2) = \ln(sk / E) - E / kT_m \qquad (1)$$

where k is Boltzmann's constant. The determined values in the method indicated by Campos [5] were E = (0.49 ± 0.70) eV and s = (0.35 ± 0.59) x 10^6 s^{-1}. These preliminary measurements could be compared with the results of other methods such as initial rise and glow curve shape.

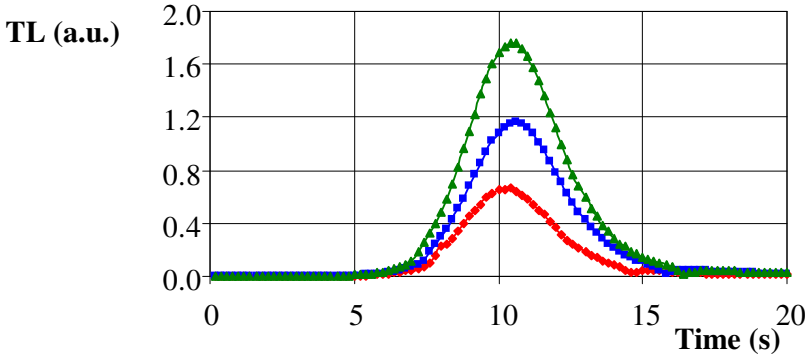

Fig.1. $CaSO_4$:Mn glow curves of irradiated pellets with three different gamma doses. The temperature of the peaks is around 138ºC.

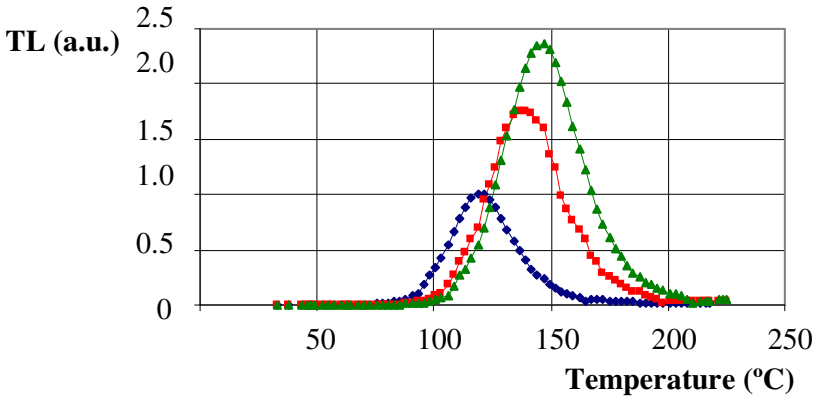

Fig.2. Glow curves obtained with heating rates of 4.3, 9.9 and 12.8 ºC.s^{-1}. The highest is the heating rate, the highest is the peak temperature.

3.2. Fading

The $CaSO_4$:Mn TL peak presents a pronounced fading at room temperature. The fading depends on the storage temperature of irradiated pellets. It is more pronounced if the storage temperature is higher. The fading values obtained under room conditions are shown in Table 1.

Table 1. Fading of $CaSO_4$:Mn TL signal

Room temperature (ºC)	Elapsed time (hour)	Fading (%)
24 to 27	24	47
20 to 25	24	38
21	1	2

3.3. Thermal Treatment for Reutilization

The purpose of this treatment also designated annealing is to remove the TL signal which could interfere in the radiation dose determination. Furthermore the pellets submitted to the thermal treatment should give reproducible TL readings. Usually the treatment consists in heating the pellets in air at a determined temperature during a period of time.

The $CaSO_4$:Mn pellets were treated at 200 and 300ºC, changing the duration time from 15 until 60 minutes. They were irradiated with gamma dose and the TL readings were proceeded. The results are presented in Table 2. The treatment at 200ºC during 1 hour presented more precise results.

Table 2. TL response of $CaSO_4$:Mn pellets for different thermal treatments

Treatment	Temperature (ºC)	Duration (min)	Rel. Dev. (%)
1	200	15	5.1
2	200	30	5.9
3	200	60	4.2
4	300	15	6.4
5	300	60	5.1

3.4. Dose Response

After the data cited by Cameron [2], it is acceptable that $CaSO_4$:Mn could be utilized in the dose range from about 10^{-3} to 10^5 mGy. The phosphor saturation occurs at 5. 10^5 mGy. The dose response at radiation protection level obtained by irradiating the pellets with ^{60}Co sealed gamma source are measured and the results were presented in Fig.3. The obtained calibration factor (f_c) was (24.19 ± 0.15) nC/mGy.

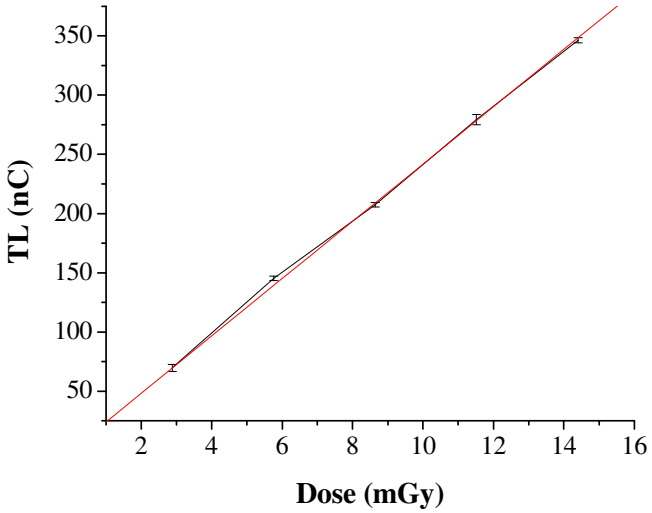

Fig.3. $CaSO_4$:Mn pellets dose response at radiation protection level.

3.5. Lower Detection Dose

$CaSO_4$:Mn is one of most sensitive TL materials and it can be used to detect small radiation doses. In practice, very small doses result in TL readings influenced by the dark current of the equipment. The lower detection dose (D_{min}) for $CaSO_4$:Mn pellets was calculated using the expression:

$$D_{min} = (TL_{0Gy} + 3\sigma) / f_c \qquad (2)$$

where TL_{0Gy} is the TL response of non irradiated pellets, σ is the standard deviation of the mean and f_c is the calibration factor calculated from Fig.3. The obtained value was $D_{min} = (1.5 \pm 0.2)$ μGy, according to that reported by Cameron [2].

3.6. Reproducibility

The reproducibility of TL readings was evaluated by submitting the $CaSO_4$:Mn pellets to the following procedure:

a) Selection of pellets.
b) Thermal treatment at 200ºC during 60 min, followed by fast cooling to room temperature.
c) Irradiation with 3,88 mGy of gamma dose.
d) Control of elapsed time after irradiation of the pellets.
e) TL reading in a temperature controlled room.
f) Repetition of the same procedure several times.
g) Analysis of the data.

The reproducibility of the TL response was better than ± 3%.

4. CONCLUSIONS

The obtained results from $CaSO_4$:Mn pellets produced at Ipen-Cnen/SP agree with those reported in the literature and indicate that they can be useful for TL measurements of low doses. When selected pellets are utilized under well controlled conditions, the measurements can be accomplished within ± 3% reproducibility. The $CaSO_4$:Mn is available in powder or pellet form.

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