

THERMOLUMINESCENT DOSEMETERS OF CaSO_4 : Dy + Teflon

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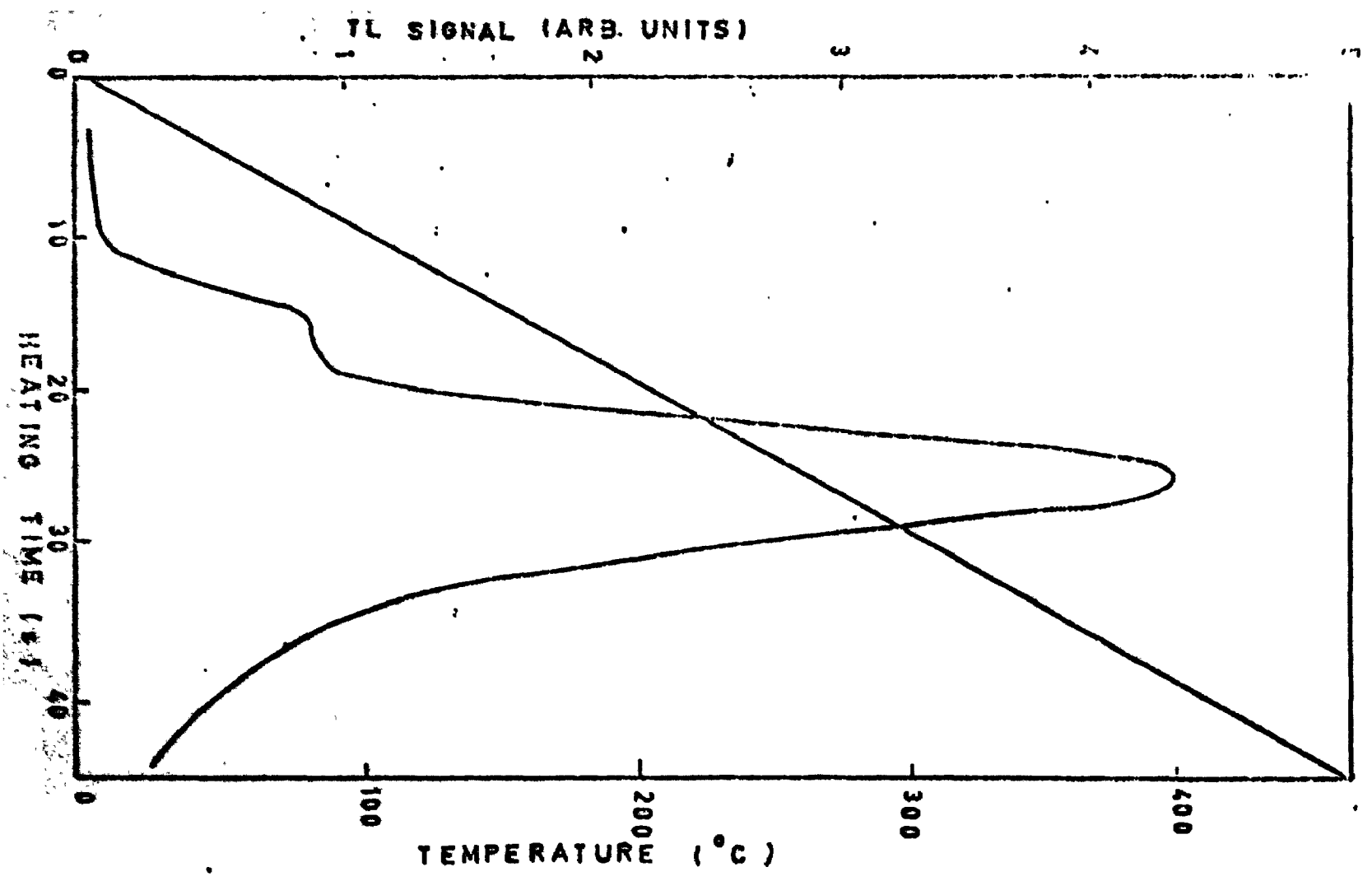
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Abstract

With the development of the nuclear programme in Brazil, the need of environmental and personnel monitoring becomes imperative. The present dosimetry system in use employs the photographic film and thermoluminescent (TL) dosimeters (LiF extruded ribbons) both of which are commercially available but imported. In view of saving foreign currency and becoming self sufficient in this field, a pellet dosimeter of CaSO_4 :Dy + Teflon was developed at IPEN. CaSO_4 :Dy thermoluminescent phosphor, grown in the Dosimetric Materials Production Laboratory was chosen, due to its high sensitivity, ease of preparation and comparatively low cost. Pellets were produced by cold pressing and sintering a mixture of CaSO_4 :Dy and Teflon powders. Extensive work was done to study in detail all CaSO_4 :Dy pellets characteristics from the point of view of dosimetry with the purpose of introducing it in the routine use. A filter combination providing an energy independent response from 20 KeV to 1,25 MeV was obtained. The dosimeter consists of three pellets sealed between two thin plastic sheets and placed under plastic and lead filters. The combination of these three filters allows the exposure as well as the energy determination of an unknown source. Field trials of this dosimeter have shown very good results.



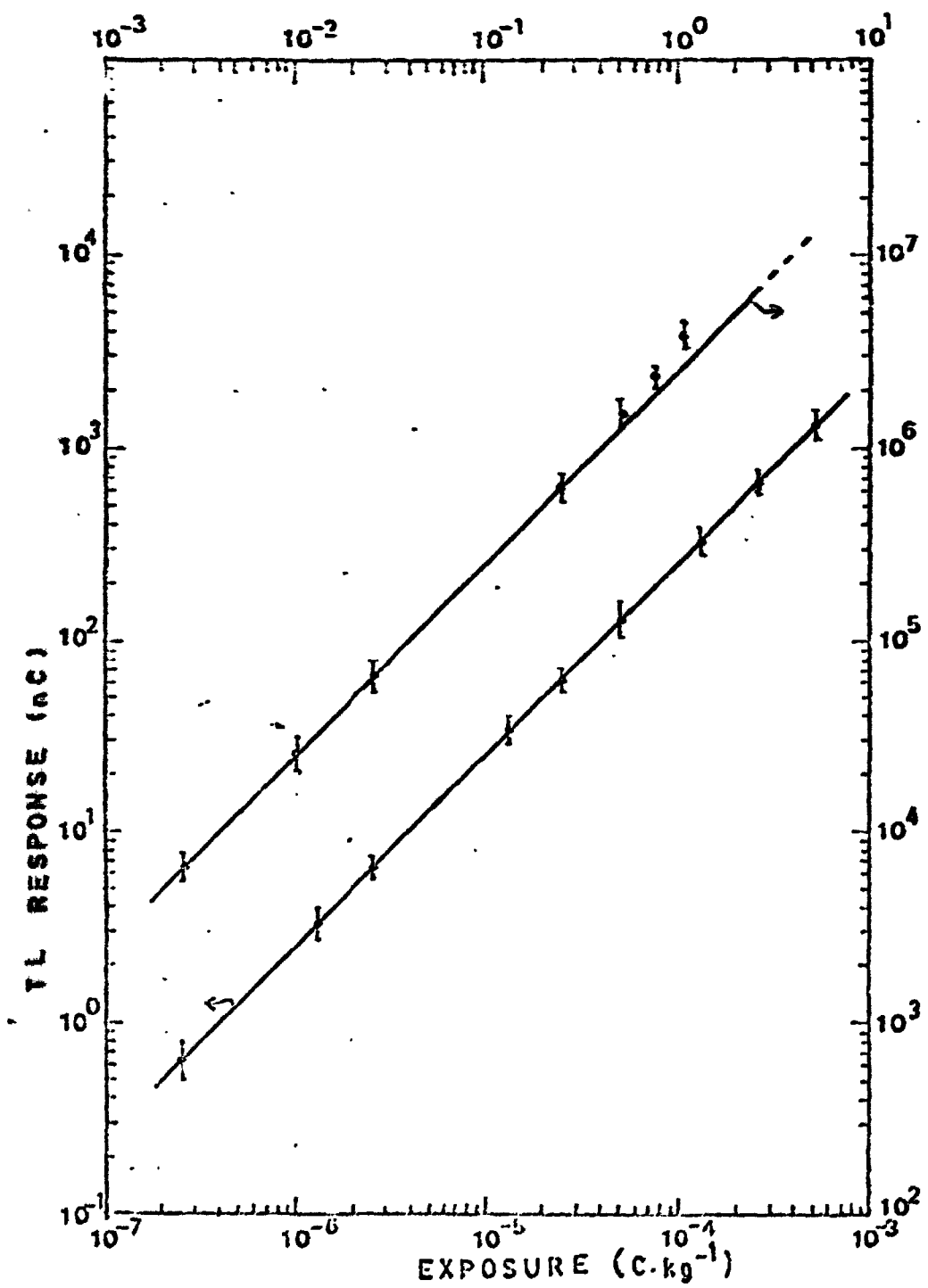


Fig. 2 - TL response of CaSO₄:Dy Teflon pellets for gamma radiation from ⁶⁰Co.

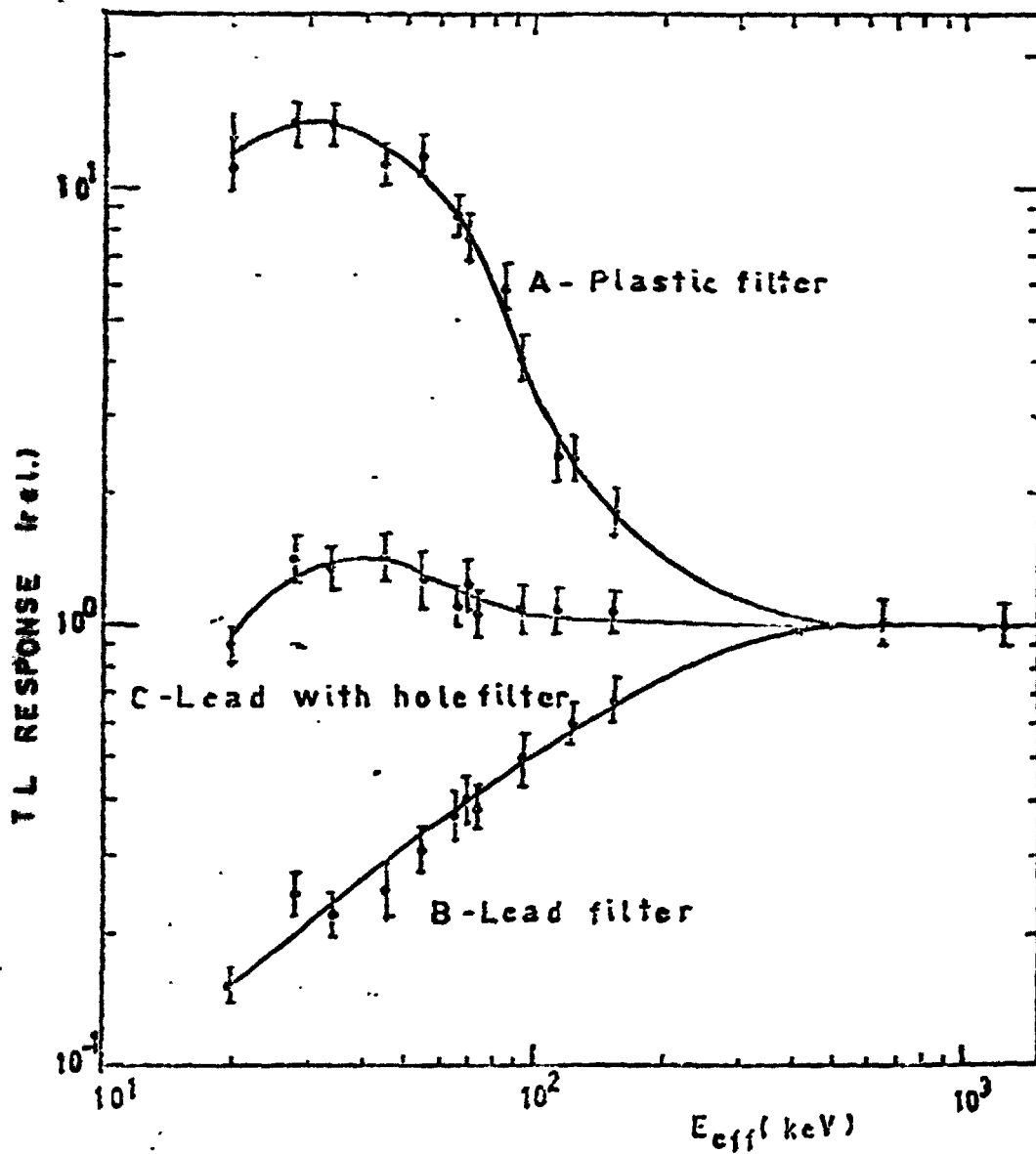


Fig. 3 - Energy response of $\text{CaSO}_4:\text{Dy}$ Teflon pellets under different filters.

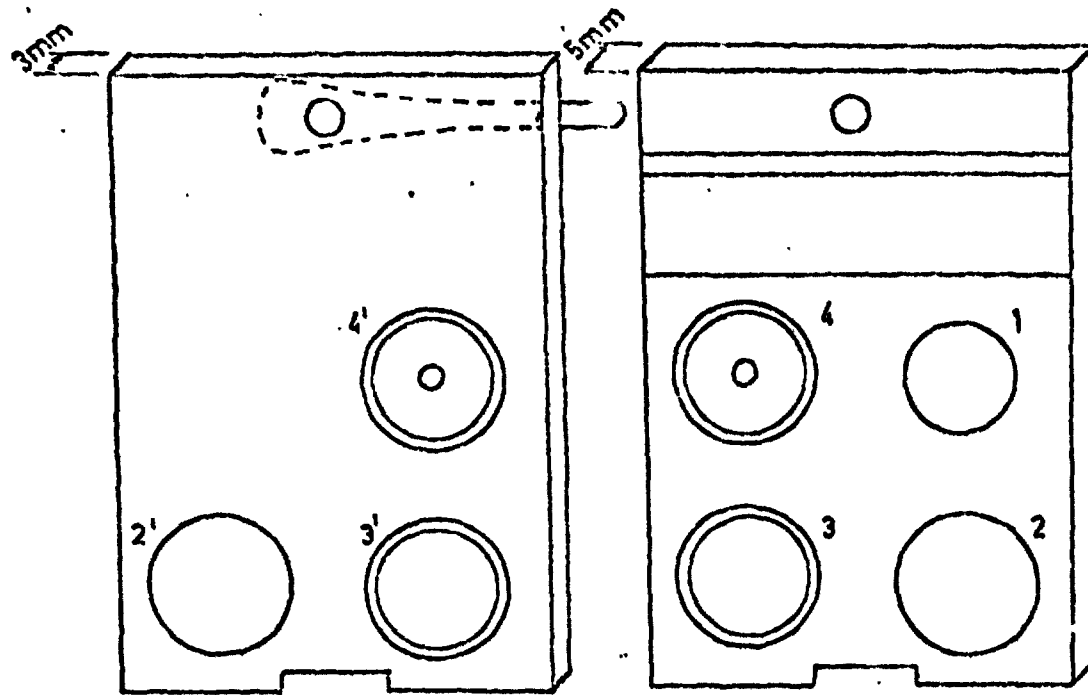


Fig. 4 - Plastic Badge with filters

1 - open window

2-2' - plastic filter

3-3' - lead filter

4-4' - lead with a central hole filter

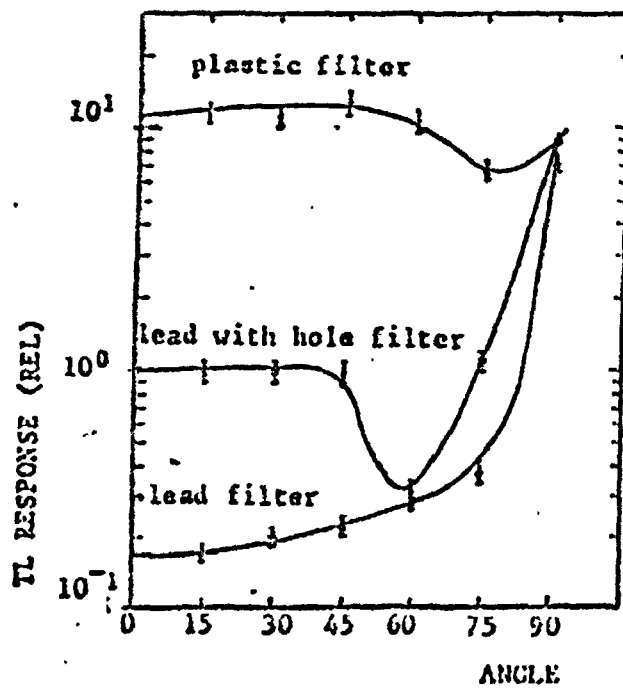


Fig. 5 Angular dependence of the TLD badge to 80kVp(Al filter)X-rays.

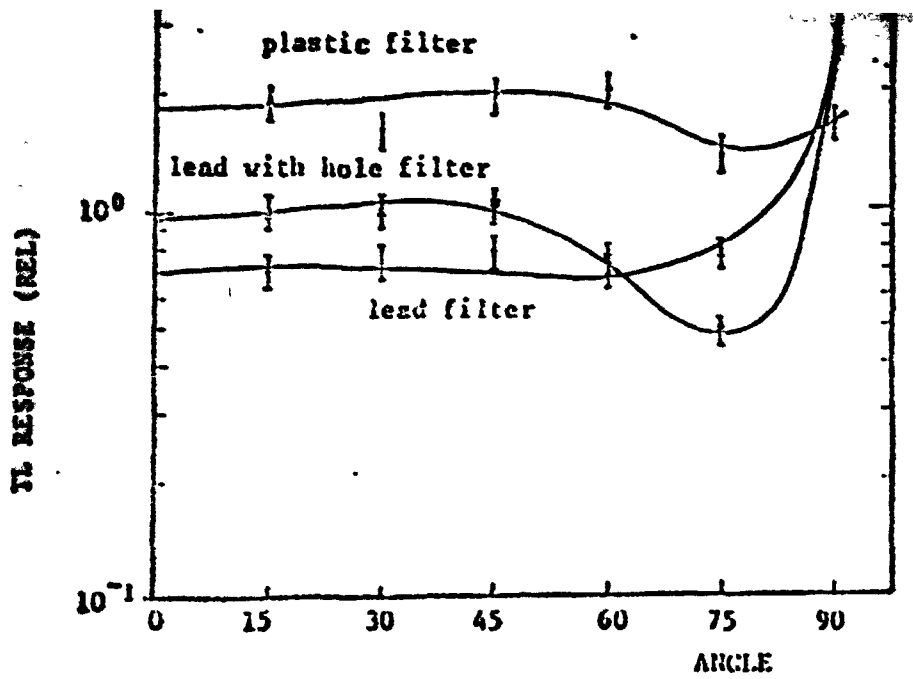


Fig. 6 - Angular dependence of the TLD badge to 300 kV_p (Thoraeus III) X-rays.

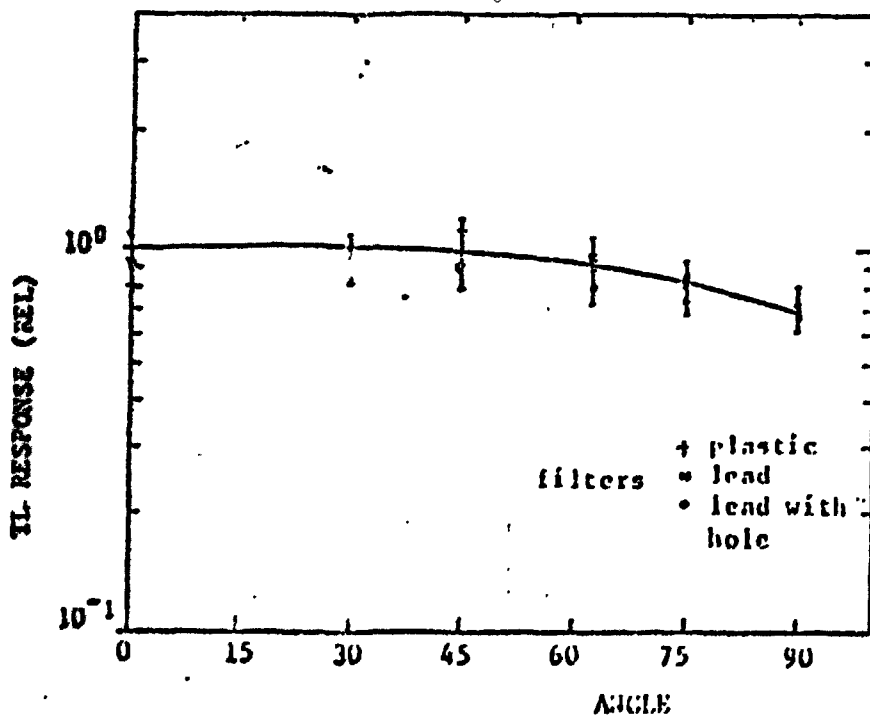


Fig. 7 - Angular dependence of the TLD badge to ⁶⁰Co radiation.

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INTRODUCTION

With the development of the nuclear programme in Brazil, the need of environmental and personnel monitoring becomes imperatives. The present dosimetry system in use employs the photographic film and thermoluminescent (TL) dosimeters (LIF extruded ribbons) both of which are commercially available but imported. In view of saving foreign currency and becoming self sufficient in this field, a new type of pellet dosimeter was developed at IPEN.

$\text{CaSO}_4:\text{Dy}$ thermoluminescent phosphor, grown in the laboratory was chosen, due to its high sensitivity, ease of preparation and comparatively low cost.

Pellets were produced by cold pressing and sintering a mixture of $\text{CaSO}_4:\text{Dy}$ and Teflon powders.

Extensive work was done to study in detail all $\text{CaSO}_4:\text{Dy}$ pellets characteristics from the point of view of dosimetry with the purpose of introducing it in the routine use.

PHOSPHOR PREPARATION

The laboratory phosphor preparation method used by Yamashita et al ⁽¹⁾ in a large scale production showed two great inconveniences: corrosion and environmental pollution due to the H_2SO_4 vapour. On the other hand the γ -ray TL sensitivity variation of the powder obtained with this method from batch to batch was $\pm 20\%$ due to contamination from external impurities. To avoid these problems a new preparation method for the obtention of TL dosimetric single crystals of $\text{CaSO}_4:\text{Dy}$ of optical quality for dosimetric and physical studies was developed.

Single crystals are now grown by the slow evaporation method from the solution of $\text{CaSO}_4 + \text{Dy}_2\text{O}_3$ in excess of concentrated H_2SO_4 , utilizing a sealed system with constant nitrogen gas flow as carrier vapour ⁽²⁾.

By this method crystallization can be controlled by varying the temperature and the gas flow rate.

The resulting sample of $\text{CaSO}_4:\text{Dy}$ (0,1% mol) is obtained in the form of single crystals with dimensions of 5.5 x 3.5 x 1mm, whose monocrystallinity was confirmed by the X-ray diffraction Laue method.

This material is washed repeatedly with distilled water to remove any traces of sulfuric acid, treated to dryness, crushed and sieved. Particles of sizes between 85 and 185 μm are selected. The powder is annealed at 600°C to

obtain optimum TL sensitivity⁽³⁾. The γ -ray TL sensitivity variation of the powder from batch to batch produced by this method is $\pm 3\%$.

The $\text{CaSO}_4:\text{Dy}$ powder is mixed with Teflon powder in the ratio of 1:1 by weight. Nine pellets of 50mg in the form of small discs with 6mm in diameter and 0.8mm in thickness are cold pressed simultaneously in a specially designed set-up and then sintered.

DOSIMETRIC CHARACTERISTICS

To study the dosimetric characteristics of the pellets, a Harshaw TL system, model 2000 A+B was used, with a constant flux of nitrogen (4 l/min). The heating rate was $5^\circ\text{C}/\text{sec}$ and the maximum temperature in the reader was 315°C . The emission light was integrated in the temperature interval 180 - 350°C . A typical glow curve is shown in Fig. 1. The main glow peak appears at 220°C .

ANNEALING

The annealing procedure to be used with the $\text{CaSO}_4:\text{Dy}$ pellets in order to eliminate the effects of previous exposures was firstly studied. It was found that, in our conditions, for the temperature of 300°C , a period of fifteen minutes is the best annealing time.

THERMAL FADING

Thermal fading characteristics of $\text{CaSO}_4:\text{Dy} + \text{Teflon}$ pellets were investigated and after storage periods of one and three months at room temperature, it was found a fading in the TL response of 3% and 7% respectively.

OPTICAL FADING

No evidence of optical fading was found on exposing pellets to laboratory light (not fluorescent).

REPRODUCIBILITY

The main advantage of the TL dosimetry system is its individual reusability. The effect of repeated heating cycle, irradiation (^{60}Co) and TL reading was investigated. None of the pellets showed a standard deviation value more than $\pm 3\%$ by repeated irradiation and heating cycles up to 20 times. Therefore $\text{CaSO}_4:\text{Dy} + \text{Teflon}$ pellets can be used repeatedly without recalibration at least 20 times.

EXPOSURE RESPONSE

The TL response linearity as a function of exposure to ^{60}Co radiation was verified between 2.58×10^{-7} C/kg (1 mR) and 12.90×10^{-7} C/kg (5000 R) and is

supra-linear.

ENERGY RESPONSE

The X-ray energy TL response of $\text{CaSO}_4:\text{Dy}$ pellets was measured from 20 keV to 155 keV and normalized for the ^{60}Co response. In this case, the experimental conditions are listed in Table 1 and the energy dependence is shown in Fig. 3 (curve A). The resulting strong energy dependency below 200 keV might be attributed to the high Z value (15.3) of the CaSO_4 .

LOWER DETECTION LIMIT

The lowest detectable value was chosen as $\bar{X}_0 + 3\sigma$ where \bar{X}_0 is the average background signal of unirradiated samples and σ the standard deviation from the mean value. The average TL response for 45 unirradiated pellets was $(0.060 \pm 0.005)/\text{nC}$. Therefore, the equivalent minimum measurable exposure from ^{60}Co was found to be about $3.87 \times 10^{-8} \text{ C.kg}^{-1}$ (150 μR).

DOSIMETER BADGE

In environmental and personnel monitoring a wide range of unknown radiation energies is present; therefore radiation dosimeters with energy independent response, relative to air, are preferred. In the case of CaSO_4 pellets the method of partial surface shielding filters was applied to reduce the energy dependence of its TL response.⁽⁴⁾ Detailed work showed that the use of 1.0mm thick lead filter in combination with 3mm thick plastic filter provides a cutoff in the 100 keV region and a uniform response above this energy. On the other hand, a 0.8mm thick lead filter with 2mm diameter central hole together with 3mm thick plastic filter provides a nearly energy independent TL response from 20 keV to 1.25 MeV. The responses of the dosimeter behind 3mm plastic filter, cutoff filter and energy independent filter are shown in Fig. 3.

Thus the final dosimeter badge proposed for radiation monitoring should have three dosimeter pellets, between pairs of the described filters (Fig. 4). This filter combination permits radiation quality estimation.

ANGULAR DEPENDENCE

Angular dependence studies were performed and showed that in the most unfavourable case (20 keV X-rays and TL response of the dosimeter behind the lead filter with hole) the TL response up to 45 degrees was independent (Fig. 5). Another cases are shown in Fig. 6 and 7.

Field trials for a period of one year in personnel and environmental radiation monitoring were performed in the Institute. The results were compared with those obtained using LIF (TLD-700). It was found that these results agree within $\pm 10\%$ in 100% of the cases in environmental monitoring and 80% of the

Table 1 - Energies used for CaSO₄:Dy pellets energy response

ENERGY keV	ADDED FILTRATION mm	HVL mm
20	2.0 Al	0.75 Al
28	2.0 Al	1.62 Al
34	2.0 Al	2.86 Al
45	4.0 Al	5.49 Al
55	0.2 Cu	0.35 Cu
66	0.5 Cu	0.57 Cu
70	0.5 Cu	0.7 Cu
74	0.5 Cu	0.76 Cu
96	1.0 Cu	1.42 Cu
116	Th II	2.07 Cu
126	Th I	2.4 Cu
155	Th III	4.0 Cu

National and International Intercomparisons results showed an agreement within $\pm 10\%$ in 95% of the cases.

CONCLUSIONS

In conclusion it can be stated that CaSO₄:Dy + Teflon pellets retain the original glow curve shape and sensitivity of CaSO₄:Dy powder. The characteristics of CaSO₄:Dy pellets studied in this work show its great usefulness in radiation dosimetry: low thermal fading, reusability until 20 times without recalibration, linear TL response to radiation exposure and possibility of obtaining energy independence. This material presents optimum qualities for personnel and environmental dosimetry purposes and started to be used by the principal Monitoring Services of Brazil.

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