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INTERMEDIATE NEUTRON DETECTION BY THERMOLUMINESCENCE*

Eliana Navarro dos Santos and R. Muccillo**

ABSTRACT

Thermoluminescent (TL) studies were carried out in cold-pressed $CaSO_4$:Dy + Dy_2O_3 + KCl ([redacted]) and CaF_2 + Dy_2O_3 + KCl ([redacted]) polycrystalline samples exposed to mixed neutron-gamma fields, ~~The purpose~~ [redacted] for the detection of intermediate neutrons which is based on the evaluation of the TL signal of the specimens stored for 24 hours after being exposed to a mixed neutron-gamma field and thermally annealed to erase the total radiation-induced TL.

The addition of Dy_2O_3 to $CaSO_4$:Dy in the proportion 1:2 increased the neutron response by a factor of 180 relative to that of $CaSO_4$:Dy.

180 mg of $CaSO_4$:Dy + Dy_2O_3 + KCl in the proportion 2:1:3 showed to be an appropriate detector of intermediate neutrons; the minimum detectable fluence was estimated to be 3.5×10^5 neutrons/cm^2. ([illegible])

I – INTRODUCTION

The thermoluminescence (TL) technique has been widely used in radiation dosimetry. In this work we are mainly concerned with TL neutron detection, which could either be direct – requiring the use of two materials with different neutron/gamma sensitivities – or indirect – the TL material is self-irradiated during the decay of activated nuclides. The latter has the advantage of the easy discrimination of the gamma components always present in neutron fields.

Several thermoluminescent materials have already been proposed to the detection of fast and thermal neutrons by the self-activation technique(1,2,5-9). We decided to undertake a research work which could lead to the detection of intermediate neutrons, namely, neutrons with energies ranging from the Cd cut-off energy to about 100 KeV(3). This is usually done by activation of metal foils but it is time-consuming in the determination of intermediate neutron flux densities from isotopic sources which range from 10^4 to 10^7 n/cm^2.s.

We have used the self-activation technique because it allows easy discrimination of gamma radiation and also can have its sensitivity highly improved by mixing a suitable TL phosphor with a convenient material having isotopes with large thermal neutron activation cross sections(2).

Readily available natural Calcium Fluoride and easily prepared $CaSO_4$:Dy(0.1%)(12) were choosen as TL phosphors due to their known radiation sensitivity, little fading and high light yield(11); Dysprosium Oxide was used as activator because it has 28.18 % ^{164}Dy, an isotope with a thermal (and consequently also intermediate in the tail of the Maxwellian distribution of energy) neutron activation cross section of 2600 b. Mixing these two materials did not make good pellets so KCl was choosen as an aggregant.

We report here a systematic experimental work to choose the best proportion of the components of the pellets (phosphor:activator:aggregant), to detect intermediate as well as thermal neutrons from an

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isotopic source and the possibilities of using the self-irradiation technique to determine intermediate neutron fluences.

II – MATERIALS AND METHODS

Two kinds of TL phosphors were used: Brazilian natural Calcium Fluoride (greenish fluorite) and $CaSO_4$: 0.1% Dy prepared by the method described by Yamashita and cols.(12).

Pellets of 10 mm of diameter and 1 mm thick were prepared by adding Dy_2O_3 (as activator) and KCl (as additive) to the TL phosphors, mixing and cold-pressing at 1200 lb/in^2 for 3 min. The TL phosphors, the additive and the activator were grinded and sieved to 85-185 microns. Natural CaF_2 as well as $CaSO_4$: Dy were annealed at 600 C/2 h prior to usage.

Thermoluminescence measurements were done in a commercial Harshaw 2000 A/B TL reader; TL peak heights were taken as a measure of the response of the pellets.

Irradiations were made using a ^{252}Cf source immersed in a water tank. Its thermal neutron flux density was measured by conventional activation foil analysis and determined to be 1.65 x 10^6 n/cm^2.s. In all irradiations two similar pellets were used: one bare and another wrapped in Cd foil. The Cadmium ratio was 6.43. The Cd cover used was 1 mm thick, so it was sufficient to absorb the thermal neutrons of energies lower than the Cd cut-off energy(4). One of the irradiation facilities of IEAR-1 (swimming pool-type reactor) was also used. The thermal neutron flux was determined as above: 6.7 x 10^{11} n/cm^2.s, with a Cd ratio of 17.36.

III – RESULTS AND DISCUSSION

Figure 1 shows the results that gave the best proportion of the components in specimens I and II to neutron detection. Ten bare samples of each composition were exposed to a mixed neutron-gamma field in the Reactor, annealed at 600 C/10 min, allowed to be self-irradiated for a period of 24 hours (which corresponds to approximately ten half-lives of ^{165}Dy) and had their TL output determined. This best proportion is in both cases phosphor:activator:additive::2:1:3, showing that the self-shielding effect due to the thickness of the specimens is the same for both mixtures. The shape of the curves is to be expected because of the competition between the TL sensitivity (for decreasing the mass of the phosphor) and the internal irradiation source (for increasing the mass of the activator). The TL intensity reaches a maximum and then decreases due to several factors: decrease of phosphor concentration, self-absorption of the TL phosphor due to a reduction in the optical transmission of the pellets, and self-shielding.

The ratio of the TL output of the best proportion-specimen to the one without addition of Dy_2O_3 is 160 (specimen I) and 60 (specimen II). A memory effect has been observed: the samples which gave rise to the results of Figure1 were re-stored to undergo self-irradiation, and then had their TL outputs re-measured yielding similar results. Then the optimized proportions for neutron detection were found to be 60 mg of the phosphor ($CaSO_4$:Dy or CaF_2) + 30 mg Dy_2O_3 + 90 mg KCl. Hereafter all experiments were performed using pellets with these compositions.

The next step was to find out the internal radiation source responsible for the self-irradiation of the specimens. The simplest procedure is to follow the decay of the self-induced TL at equal time intervals(5-7). The results are shown in Figure 2 where the measurements were performed every 0.5 hour using three samples: specimen I (line A), specimen I covered with Cd (line B), and bare specimen II (line C). A, B, and C have the same slope and the half-life was determined to be 2.3 h, showing that ^{165}Dy is actually the contributor to the self-induced TL. It has already been shown that activation due to other nuclides(^{34}S, for example) makes contribution to the TL only for self-irradiation times longer than 24 h(6).

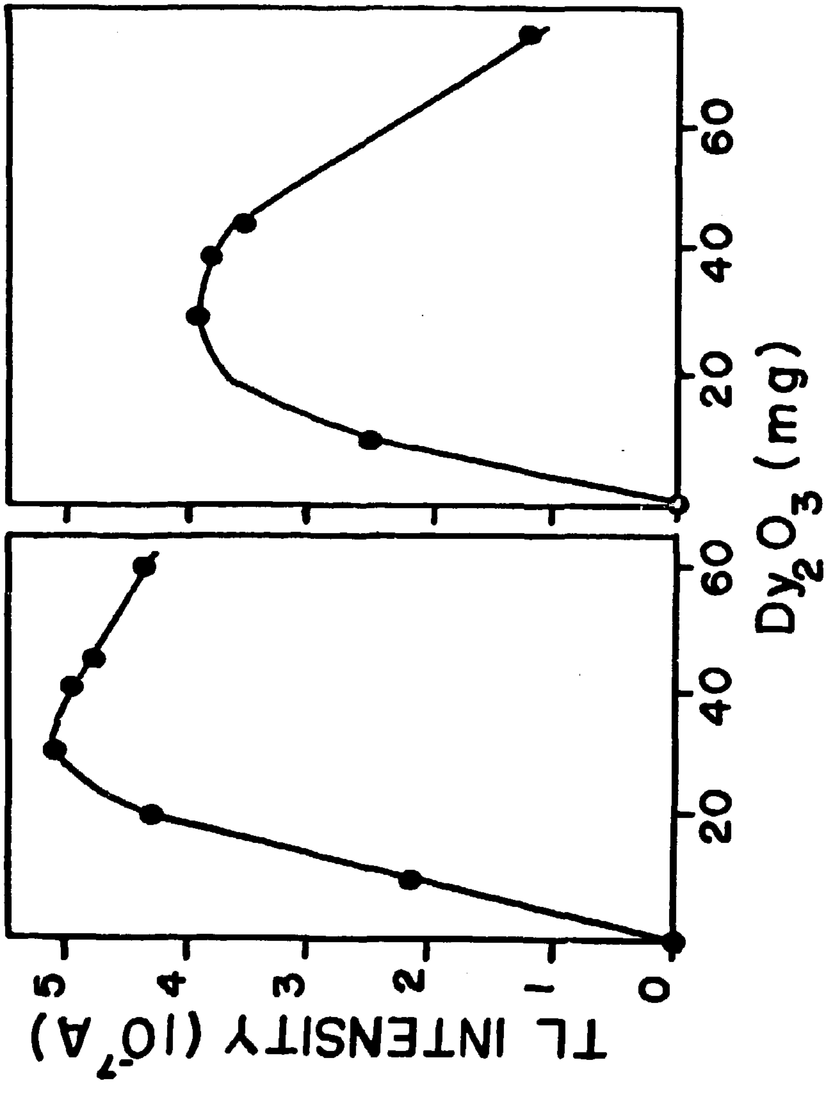

Figure 1 – Self-Induced Thermoluminescence as a Function of Dy_2O_3 Concentration; Left: $CaSO_4$:0.1% Dy + Dy_2O_3 + 90 mg KCl; Right: Natural CaF_2 + Dy_2O_3 + 90 mg KCl. Mass of the Pellets: 180 mg

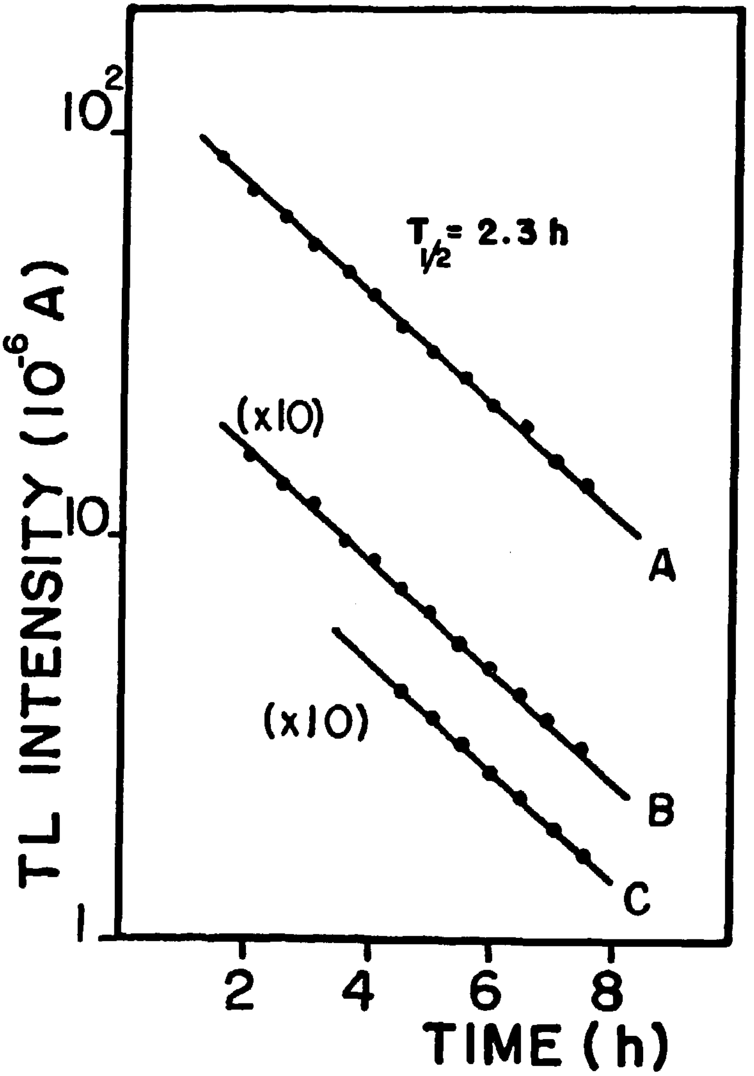

Figure 2 – Decay of the Self-Irradiation Induced TL in $CaSO_4$: 0.1% Dy + Dy_2O_3 + KCl (Specimen I) and CaF_2 + Dy_2O_3 + KCl (Specimen II); A: Specimen I, B: Cd-covered Specimen I, and C: Specimen II

Several specimens were then prepared to study the possibility of detection of intermediate neutrons. For this purpose two pellets at a time were exposed to the moderated neutron field of the ^{252}Cf source. The Cd cover in one of the pellets assured the absence of thermal neutrons. The results are shown in Figure 3 where the TL intensity (self-induced TL; storage time: 24 h) is plotted as a function of exposure time. The upper curves are supralinear because the TL here represents the sum of the activities due to thermal and to intermediate neutrons. On the other hand, the lower curves show a sublinear response in agreement with the foil-activation technique results already well known. These results found to be reproducible within 5% suggesting the use of the technique to the determination of intermediate neutron flux densities from isotopic sources with known spectrum of neutron energies.

The results shown in Figure 3 were also used to give values of TL intensity as a function of intermediate neutron fluence (see Figure 4). The intermediate neutron flux density ϕ was estimated by using the following equation$^{(10)}$:

$$\phi = \frac{(A/N - A_c/N_c) \int_{E_1}^{E_2} dE/E}{(1 - e^{-\lambda t})(CR - 1) \int_{E_c}^{\infty} \sigma(E)\, dE/E}$$

where

A and A_c are the activities of the bare and Cd-covered specimens, N and N_c the numbers of target nuclei in the bare and in the Cd-covered specimens, E_1 and E_2 the minimum and the maximum energies E of the neutron in the ^{252}Cf source spectrum, E_c the Cadmium cut-off energy, λ the disintegration constante of the target specimen, CR the Cd ratio at the irradiation position, and σ (E) the neutron activation cross section of the target specimen. Gold foils were used allowing the determination of the intermediate neutron flux density in the irradiation position. The minimum detectable fluence was estimated to be 3.5×10^5 n/cm^2 taking into account the sensitivity of our TL detection apparatus. This figure could probably be lowered by improving the thermoluminescence detection system and/or increasing the mass of the pellets.

IV – CONCLUSIONS

1) Cadmium-covered cold-pressed pellets of 60 mg $CaSO_4$: 0.1% Dy + 30 mg Dy_2O_3 + 90 mg KCl showed to be easy to handle detectors of intermediate neutrons.

2) The minimum detectable fluence of intermediate neutrons was determined to be 3.5×10^5 n/cm^2.

3) The main advantages of this new detector are the easiness of fabrication, its sensitivity to intermediate as well as to thermal neutrons, and the discrimination against the gamma radiation field.

RESUMO

Foram feitos estudos relativos às emissões termoluminescentes de fósforos dosimétricos, visando a detecção de nêutrons na faixa de energia intermediária. Este intervalo foi definido como sendo desde a energia de corte do cádmio até 0,1 MeV, por analogia ao que é adotado no campo de metrologia nuclear. A medida da termoluminescência foi feita em amostras policristalinas de $CaSO_4$: Dy + Dy_2O_3 + KCl e de CaF_2nat + Dy_2O_3 + KCl compactadas a frio. A detecção de nêutrons é feita indiretamente pela auto-irradiação provocada pela ativação neutrônica.

Essas pastilhas apresentam diferentes dependências de amplitude de emissão TL com o tratamento mecânico, mas o efeito de auto-blindagem ~~foi constatado~~ tem a mesma importância nos dois tipos de amostras.
(Constatou se que)

Estudos de sensibilidade foram feitos comparativamente à de pastilhas de ^{6}LiF: Mg; Ti (TLD-600), obtendo-se que, para uma mesma dose de radiação, as amostras que contém $CaSO_4$: Dy (CaF_2nat) apresentam uma amplitude de emissão TL 24 (3) vêzes superior à das pastilhas de TLD-600.

Os resultados que se seguem estão relacionados às pastilhas que contém como fósforo o $CaSO_4$: Dy, visto que sua elevada sensibilidade relativamente às amostras de fluorita.

O comportamento da emissão TL nesses materiais foi também verificado através de irradiações nos campos mistos n-gama do reator IEAR-1 e de uma fonte de ^{252}Cf, tendo sido estimada a fluência mínima detectável: ~3,5 x 10^5 $n.cm^{-2}$.

Foi identificado, através da determinação da meia-vida efetiva da emissão TL, o $^{165}_{66}Dy$ como sendo o principal isótopo responsável pela termoluminescência auto-induzida nestas amostras.

Os resultados obtidos sugerem a utilização de pastilhas de $CaSO_4$: Dy + Dy_2O_3 + KCl, na determinação de fluências de nêutrons intermediários em fontes isotópicas.

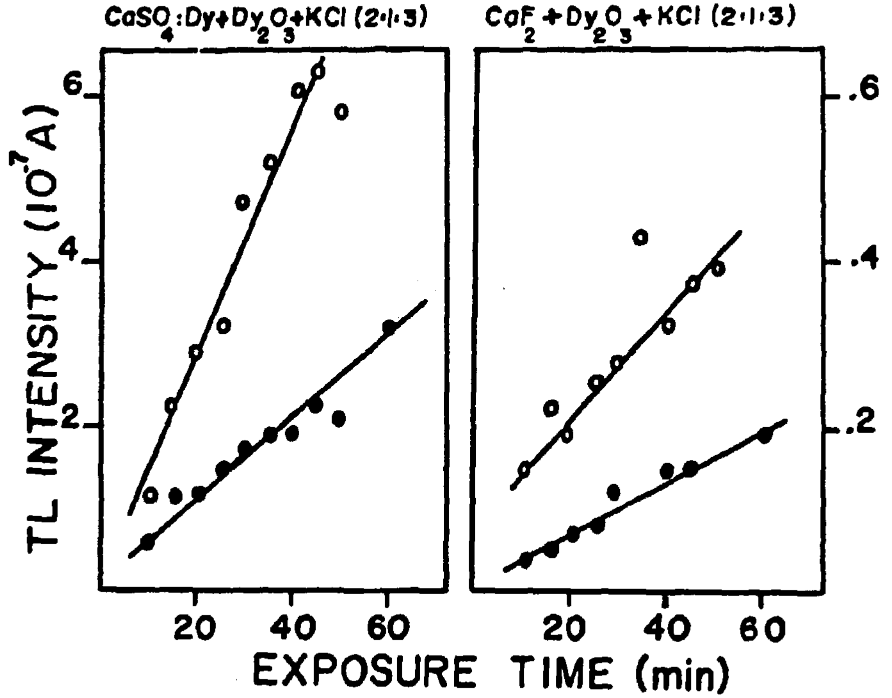

Figure 3 – Dependence of the Self-Induced TL Intensity on the Exposure Time of Specimen I and II to Neutrons of a ^{252}Cf Source; Upper Curves: Bare Samples; Lower Curves: Cd-covered Samples

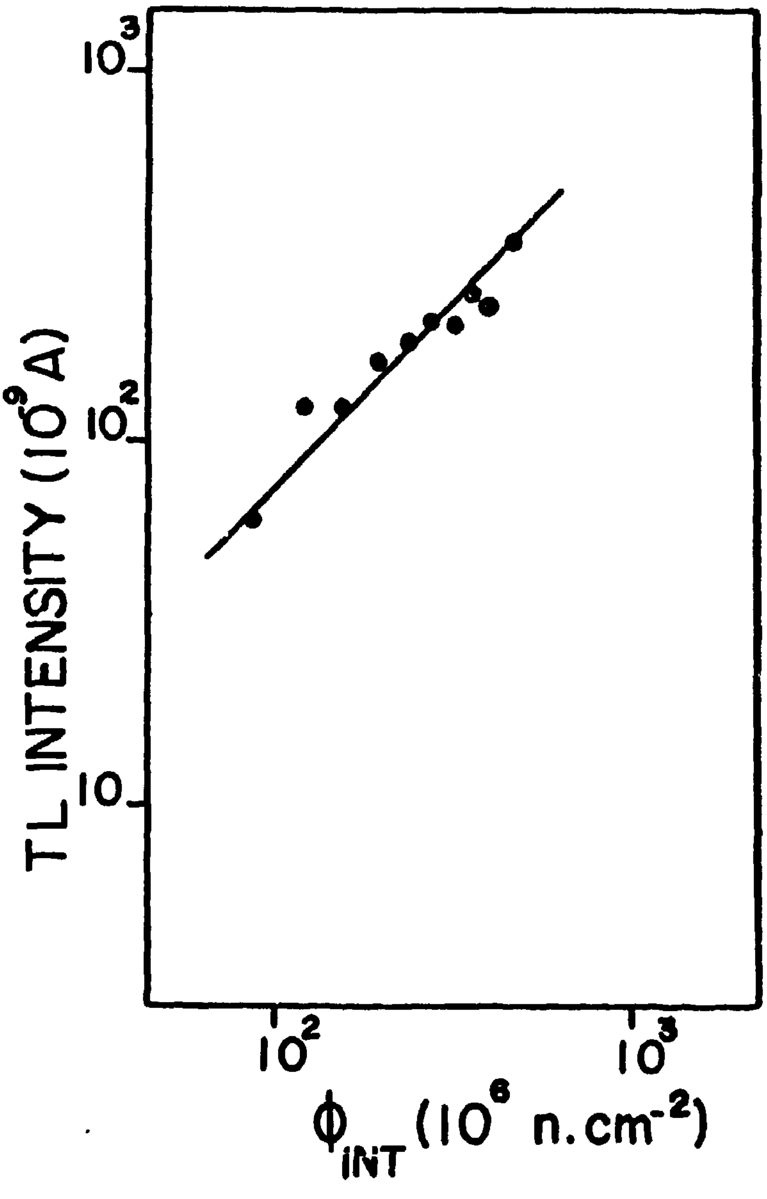

Figure 4 – Self-Induced TL Intensity as a Function of Intermediate Neutron Fluence of a ^{252}Cf Source; Phosphor: 60 mg $CaSO_4$: 0.1% Dy + 30 mg Dy_2O_3 + 90 mg KCl

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