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THERMAL NEUTRON DETECTION BY ACTIVATION OF $CaSO_4$:Dy + KBr THERMOLUMINESCENT PHOSPHORS*

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ABSTRACT

Thermoluminescence (TL) studies to detect thermal neutrons were performed in cold-pressed $CaSO_4$:0,1%Dy + KBr samples. The detection is based on the self-irradiation of the $CaSO_4$:Dy TL phosphor by the Br isotopes activated by exposure to a mixed neutron-gamma field.

In 1970 a new method for the detection of thermal neutrons by the Thermoluminescence (TL) auto-activation technique was proposed[3]. It consists in the measurement of the TL output of a phosphor previously exposed to a neutron-gamma field, thermally annealed to bleach the radiation effects, and stored to undergo self-irradiation due to the decay of the thremal neutron activation products. The TL output can then be related to the activation of the phosphor and consequently to the thermal neutron fluence. The main advantage of the method is based on the ability to detect thermal neutrons without gamma contributions. In ref (3) the self-irradiation was provided by ^{45}Ca, which is a beta-emitter, resulting from the activation of ^{44}Ca in natural CaF_2 phosphor. The low ^{44}Ca natural abundance (2.08%) as well as its relative low thermal neutron cross section (0.7 b) lead to a minimum fluence detection of the order of 10^9 neutrons/cm^2. Later on CaF_2:Dy (TLD-200) commercial phosphors were used allowing an increase of the sensitivity of the method by more than two orders of magnitude[2]. An obvious extension of the method to the detection of fast neutrons has been proposed[1,4,5].

In this work, improvements in the self-activation method are reported by combining a highly beta-sensitive TL phosphor with a highly effective activator, i.e., by mixing a TL phosphor with a material with high thermal neutron cross sections. 0.1% Dy-doped $CaSO_4$ was choosen as the phosphor due to its high sensitivity to ionizing radiation as well as to the ease with which it is prepared in the laboratory[7]. KBr was choosen as the activator due to relatively high thermal neutron activation cross sections for the n-gamma reactions: ^{79}Br to ^{80m}Br(8.5 b; half-life: 18 min), ^{79}Br to ^{80}Br(2.9 b; 4.6 h), and ^{81}Br to ^{82}Br(3.3 b; 35.9 h).

Detailed experiments to find out the optimized phosphor activator proportion, the TL response to thermal neutrons, and the nuclides responsible for the self-irradiation will be described. An estimate of the minimum detectable fluence will also be made.

0.1% Dy-doped $CaSO_4$ crystals were grown following the procedure described by Yamashita and cols[6]. Before usage the powders were selected for grain sizes from 85 to 185 microns and annealed at 600 C/2 h in air. Reagent grade KBr powder was selected as above, annealed at 400 C/1 h, and then added to $CaSO_4$:Dy to be cold-pressed (1200 lb/in^2) to discs of 1 mm thickness. All samples were

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evaluated for TL peak heights on a commercial 2000 A/B Harshaw thermoluminescent reader. Pre-selection of the pellets were done by weighing and by glow curve analysis of samples exposed to ^{60}Co gamma rays.

The selected samples were exposed to n-gamma fields of a swimming pool Reactor at this Institute operating at 2 Mw producing fast fission neutrons plus the order of 10^8 R/h of photons and about 10^{11} n/cm^2.s to 10^{12} n/cm^2.s at our irradiation positions. A ^{252}Cf source immersed in a water tank with about 1.7×10^6 n/cm^2.s at a choosen irradiation position was also used for low exposures. The conventional activation of gold foils was the method used to determine the thermal neutron flux densities.

The experimental procedure leading to the evaluation of the thermal neutron induced thermoluminescence was as follows:

1 – $CaSO_4$:Dy + KBr samples are exposed to the mixed n-gamma field during the irradiation time t_{irr};

2 – the irradiated sample is thermally annealed at 600 C/15 min to eliminate the defects produced by the gamma and neutron prompt dose;

3 – the sample is stored at room temperature for a period of 24 h to undergo self-irradiation;

4 – the TL glow curve is then obtained giving a TL signal prpportional to the thermal neutron flux. The proportionality constant is determined by following the above sequence but exposing the specimen to a known thermal neutron flux density.

Figure 1 shows the TL output of $CaSO_4$:Dy + KBr pellets, obtained according to the experimental procedure described above, for various relative proportions of phosphor and activator. The total mass of each pellet was 180 mg. The thermal neutron flux in the irradiation position was determined by the gold foil activation technique to be 6.66×10^{11} thermal neutrons/cm^2.s; the exposure time was 20 s. As the TL outputs of the different pellets was within less than 25%, the optimized proportion was choosen according to the one providing easiness of hendling of the pellets: 100 mg of $CaSO_4$:Dy and 80 mg of KBr. The TL glow curve for that pellet is shown in the insert.

Henceforth all the results are shown for the optimized pellets, and the TL signal is taken as the height of the higher amplitude TL peak.

In Figure 2 the thermoluminescence response of $CaSO_4$:Dy + KBr to different thermal neutron fluences is shown. For this work another irradiation position at the reactor was used, with a thermal neutron flux density of 5×10^{12} neutrons/cm^2.s. The upper curve was obtained for samples allowed to undergo self-irradiation for a period of 24 h. The bottom curve represents the second TL reading, namely, after obtaining the TL output resulting from the 24 h self-irradiation, the sample is stored for 1 h and its TL analysis is made again. These results show that even though the TL analysis is a destructive one, further measurements can be made.

In Figure 3 similar results are shown for pellets exposed to the radiation field of a 400 micrograms ^{252}Cf source immersed in water. The fluence range was from 10^{10} to 10^{11} thermal n/cm^2, determined by gold foil activation techniques. Every irradiation has been done using one bare specimen and another wrapped in 1 mm Cd foil in order to deduce the contribution due to intermediate neutrons. The minimum detectable fluence was estimated to be of the order of 10^6 thermal neutrons/cm^2.

Successive TL readings at one hour intervals were carried out in a $CaSO_4$:Dy + KBr pellet exposed to a mixed radiation field in the Reactor Station. The results summarized in Figure 4 show a

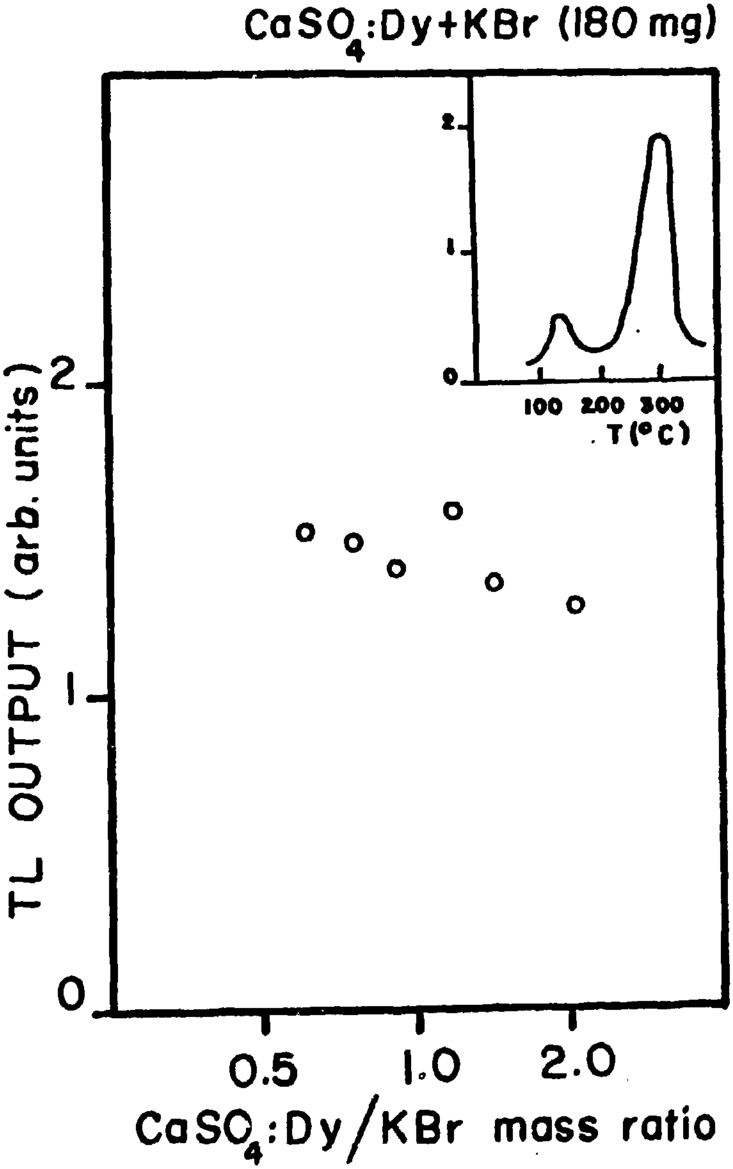

Figure 1 – Self-induced thermoluminescence output of 180 mg pellets of mixed $CaSO_4$:Dy (phosphor) and KBr (activator) for varying the relative amounts of phosphor and activator in the mixture. Thermal neutron flux density: 6.66 x 10^{11} n/cm^2.s; irradiation time: 20s; storage time: 24 h. Insert: typical glow curve for a pellet with 1.25 mass ratio

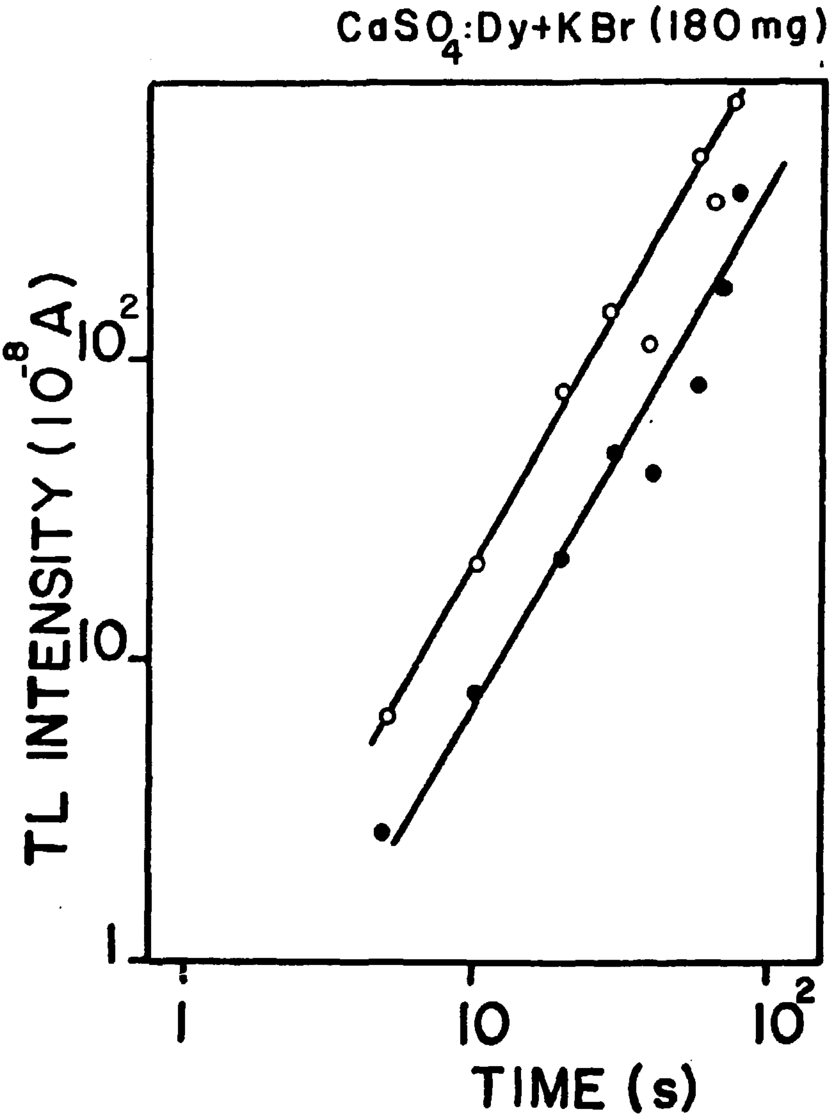

Figure 2 – Self-induced TL output of 180 mg pellets of $CaSO_4$:Dy + KBr as a function of irradiation time. Thermal neutron flux density: 6.66 x 10^{11} $n/cm^2.s$; self-irradiation times: 24 h (upper curve) and 1 h after the first reading (bottom curve)

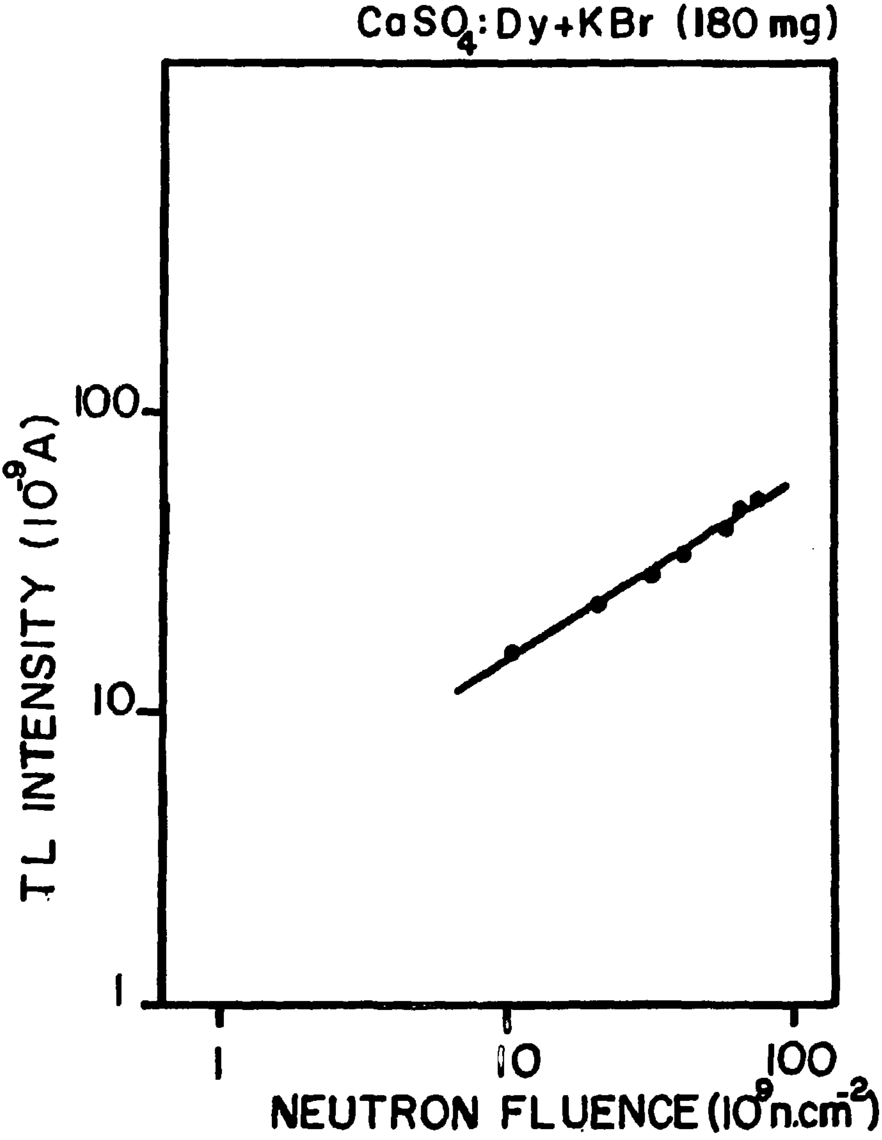

Figure 3 – Self-induced TL output of 180 mg pellets of $CaSO_4$:Dy + KBr as a function of thermal neutron fluence. Thermal neutron flux density: 6.66×10^{11} n/cm^2.s; storage time: 24 h

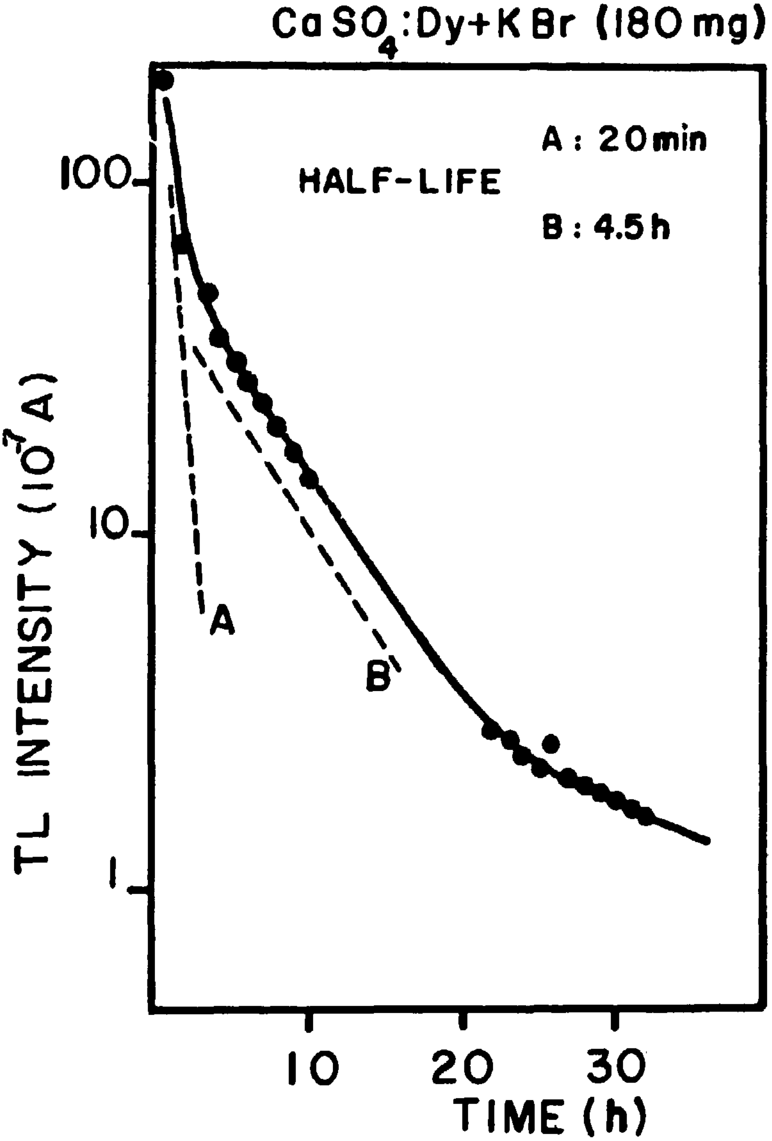

Figure 4 – Hourly decay of the self-induced TL in 180 mg pellets of $CaSO_4$:Dy + KBr

decrease which was fitted to a sum of exponential decays. Only two half-lives could de evaluated with a fair accuracy: 20 min and 4.5 h, which were assigned to ^{80m}Br and ^{80}Br, respectively. The same identification process was used previously in other materials[1,2,3].

The main results suggest then the use of $CaSO_4$:0.1%Dy + KBr pellets and thermoluminescence as a complimentary technique in thermal neutron detection.

RESUMO

Foram feitos estudos de Termoluminescência (TL) para a detecção de nêutrons térmicos em amostras de $CaSO_4$:0,1% Dy + KBr compactadas a frio. A detecção baseia-se na auto-irradiação de fósforos de $CaSO_4$:Dy pelos isótopos de bromo ativados pela exposição em campos mistos nêutron-gama.

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