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PUBLICAÇÃO IEA N.º 372
Janeiro — 1975

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SÃO PAULO — BRASIL

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ABSTRACT

The thermal neutron monitoring system used with ordinary thermoluminescence dosimeters has been studied. In this monitoring system we used the capture gamma rays from the reaction between cadmium and thermal neutrons. The effects of the back-scatter and building-up of radiation have been studied for approaching the n/γ ratio of the system to that of the mixed field. It has been found that the ratio of a system using these effects is greater than that of one without these effects and that the minimum measurable fluence is about 10^{6} n/cm^{2}. This method can be used for all kinds of thermoluminescence dosimeter phosphors.

INTRODUCTION

The thermoluminescence dosimeter (TLD) has been widely used for radiation dosimetry because of high sensitivity to the radiation dose, its convenience, and the freer selection of TLD phosphors it makes possible. Many kinds of phosphors have been developed and their properties studied by a number of investigators. These phosphors may be classified into two groups. One comprises phosphors for γ, β, α and X-rays, and the other, phosphors for neutron dosimetry, such as ^{6}LiF and ^{7}LiF. On the other hand, recently, the dose measurement of each radiation in a mixed field of neutron and γ-rays has become importat in the field of radiation dosimetry.

The sensitivity of TLD in treating neutrons has been mainly investigated on LiF, CaF_2 and $Li_2B_4O_7$ for thermal neutrons and fast neutrons[1-4]. For example, the difference in sensitivity between ^{6}LiF and ^{7}LiF has been used for thermal neutrons. Watanabe *et al*, have reported a self activation method[5]. This method has one of the same merits as TLD for measuring thermal-neutron and fast-neutron fluences; namely, it is designed specifically for measuring neutrons. However, the self activation method has a low sensitivity. For example, the minimum detectable fluence is about 10^{10} n/cm^{2} of thermal neutrons using natural CaF_2 TLD[5]; this is because the activation crossection of ^{44}Ca on thermal neutrons is about 0.72 barn. On the other hand, in the case of the LiF method of neutron dosimetry, isotope separation from the natural Li element must be carried out. Therefore, the cost of these phosphors is more than ordinary. Therefore, we have proposed another monitoring system by which it is possible to detect the neutrons with all phosphors; however, n/γ ratio of the detector system is lower than that of the radiation mixed field used. The method of improving the n/γ ratio in the monitoring system has not been studied.

The present study was undertaken in order to ascertain the effect of the back-scatter and building-up of radiation on the n/γ ratio of the monitoring system; this paper will report the results.

EXPERIMENT

In the present experiment, TLD-100 LiF and TLD-600 (purchased from the Harshaw Chemical Co.) $CaSO_4$(Tm) and BeO(Na) purchased from Matsushita Electric Co.), Mg_2SiO_4(Tb) (purchased from the Dai Nippon Toryo Co.), NTL-50p LiF (purchased from the Nemoto Tokushu Kagaku Co.), and natural CaF_2 (from Brazil) were used. These phosphors were in powdered form, and Mg_2SiO_4(Tb), $CaSO_4$(Tm), BeO(Na) and natural CaF_2 enclosed in small glass tubes about 2 mm or 0.8 mm in diameter and 13 mm or 10 mm long were used as thermoluminescence dosimeters. The TLD-600 LiF was a hot pressed crystal, while the TLD-100 LiF and NTL-50p LiF phosphors were powder.

Van de Graaff generator, purchased from the High Voltage Co., was used as neutron generator from which neutron fluence was produced from the reactions of both Be(d, n) and T(d, n). The mean energies of these fast neutrons were about 2.5 and 14 MeV respectively. The thermal neutrons were obtained from thermalized fast neutrons of the Be(d, n) reaction with a paraffin block, as is shown in Fig. 1 (a). On the other hand, the neutrons from the T(d, n) reaction were thermalized with the block, as is shown in Fig. 1 (b).

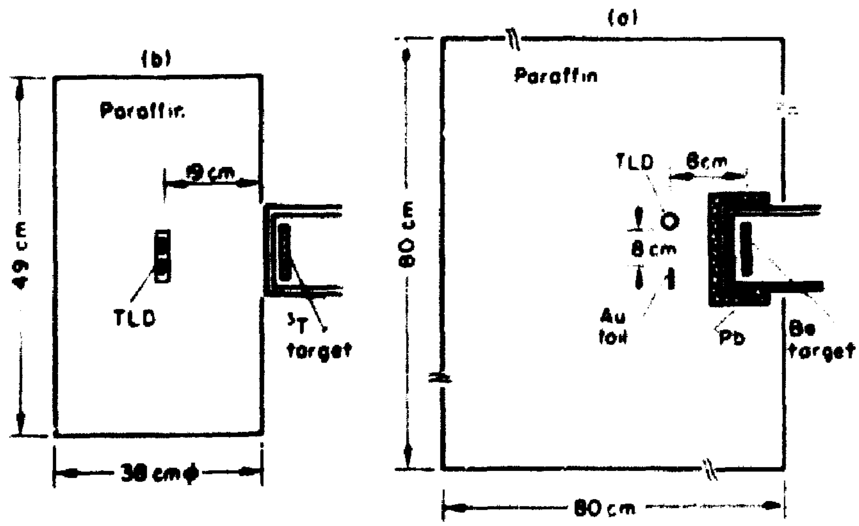

Fig. 1

Diagrams for obtaining thermal neutrons from fast neutron. (a) is for fast neutrons from reaction Be(d, n), and (b) for the fast neutrons from T(d, n) reaction.

The TLD phosphors were irradiated 8 or 20 cm in front of a target of the generator with thermal neutrons, the fluence of which was measured by means of the activation method of 10 x 10 mm² gold foil; the fluence was varied from 10^7 to 10^{11} n./cm². In the case of reading the thermoluminescence intensity, the phosphor was heated from room temperature up to about 360°C using a Dai Nippon Toryo or Harshaw TLD reader.

Figure 2 shows a diagram of the TLD holder for thermal-neutron monitoring. In such a holder, a tin or lead filter is used for correcting the energy dependence of the TLD phosphor on γ rays from the neutron generator, while a cadmium filter is used for obtaining the capture gamma rays due to the reaction of Cd(n, γ) between thermal neutrons and the cadmium element. A lead filter inside or outside of the cadmium filter was used for ascertaining the building up of the secondary electrons and the back-scatter of the gamma rays.

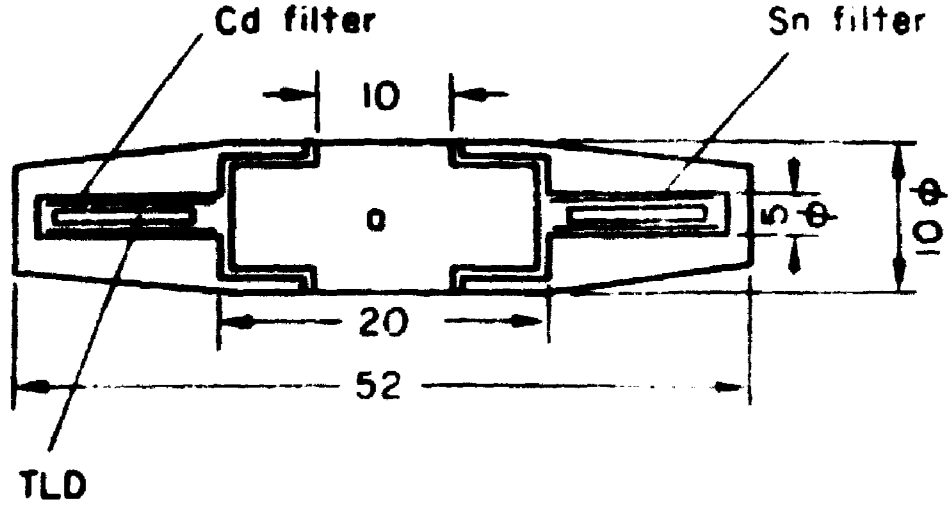

Fig 2

Thermoluminescence dosimeter holder for the thermal neutron monitor

RESULT AND DISCUSSION

The termoluminescence intensity of each filtered phosphor in the mixed radiation field may be represented as follows:

$$TL = \alpha D + gD'$$
$$TL_{Cd} = \alpha D + \delta D_{Cd} \tag{1}$$

where TL and TL_{Cd} are the thermoluminescence intensities of a non-cadmium-covered and a cadmium-covered thermoluminescence dosimeter respectively; D is the radiation dose due to gamma-rays in the mixed radiation field, and D' and D_{Cd}, the doses due to the radiation caused by a reaction between thermal neutrons and filtered materials, and by cadmium, respectively. The values of α, β and δ are proportional constants related to the thermoluminescence emission.

The amount of D' and D_{Cd} are proportional to σnt and $\sigma_{Cd}nt$, where σ and σ_{Cd} are the crossections of the filter material and cadmium to thermal neutrons; nt is the fluence of the thermal neutrons. In equation (1), D_{Cd} is much greater than D'; D' will be neglected if σ_{Cd} is much greater than the filter material. Therefore, the difference between TL_{Cd} and TL approximately represents tne value proportional to the neutron fluence.

It is well known that when a material is irradiated with gamma-rays the concentration of the secondary electrons reaches its maximum at a suitable thickness of the irradiated material. Then, if the phosphor is enclosed with some material the thickness of which corresponds to that of the maximum building-up of the capture gamma-rays, the n/γ ratio of the monitoring system will be improved and will approach the n/γ ratio of the mixed field.

On the other hand, it is well known that when gamma-rays are scattered with some material, the energy of the scattered radiation comes to be decreased. The thermoluminescence intensities or energy dependence of $CaSO_4$, Mg_2SiO_4 and CaF_2, which have larger atomic numbers than air, generally increase with a decrease in the photon energy until about 30 keV. If the phosphor is irradiated by back-scattered radiation, its thermoluminescence sensitivity will increase, but an increase in the thermoluminescence sensitivity is dependent on the ratio between initial radiation and the back scattered radiation intensities.

First, the phosphor usually lapped with cadmium foil 0.5 mm thick and successively enclosed with lead foil of various thicknesses and other phosphors for detecting the reaction gamma rays in the mixed radiation field was only lapped with lead foil of a thickness equal to that of the lead foil for the neutron detector. Second, the phosphor was enclosed with the lead foil of various thicknesses and successively lapped with cadmium foil 0.5 mm thick.

Table 1 gives the back-scatter and building-up effects of the radiation on the n/γ ratio of the detector system.

Table 1

Back scatter and building up effects on the n/γ ratio of the monitoring system ($(n/\gamma)_0$ is the n/γ ratio of the monitoring system without these effects and thermal neutron was obtained from thermalized fast neutron of *T*(d,n) reactions)

	Back scatter effect					Building up effect				
Pb(mm)	0.05	0.2	0.3	0.6	1.2	0.2	0.4	0.5	1.0	1.5
$(n/\gamma)/(n/\gamma)_0$	1.12	0.96	1.33	1.07	1.23	1.11	1.24	1.50	1.18	1.07

When the phosphor was enclosed with the filter of the lead foil to obtain either the back scatter or building-up of the radiation in the phosphor, the n/γ ratio varied as compared with that of an unfiltered phosphor. Each ratio in the monitoring system was normalized with that of the unfiltered one. In the case of an unfiltered system with cadmium foil 0.2 mm thick, the n/γ ratio was about 1.2, but, as is given in Table 1, the n/γ ratio in the filtered system has a maximum of 1.5 at 0.5 mm and a minimum of 0.96 at 0.2 mm, and was improved. It is found that the n/γ ratio in the monitoring system is improved by using these effects. Therefore, the feasibility of these effects for improving the ratio in the monitoring system is established.

The correlation between the thermal-neutron fluence and the thermoluminescence intensity of the Mg_2SiO_4(Tb)-TLD put into the holder is shown in Fig. 3. In this figure, the vertical axis is the difference in the thermoluminescence intensities between the cadmium-filtered and non-filtered TLD elements, calibrated with gamma-rays of ^{60}Co. As is shown in Fig. 3, the difference between the thermoluminescence intensities is nearly proportional to the thermal-neutron fluence. The linearity of the thermoluminescence response due to the dose of capture gamma-rays is maintained in the thermal-neutron-fluence range from 10^7 to 10^{11} n/cm^2 used in the present experiment.

On the other hand, Ayyangar *et al.*[6] have reported that the thermoluminescence response of the ^{6}LiF TLD crystal to the thermal neutrons was decreased in the fluence range of 10^{12} n/cm^2 or more. In this case, it may be inferred that the ^{6}LiF crystal contains many latice defects caused by the reaction of $^6Li(n,\alpha)^3H$. Accordingly, in the case of re-using the phosphor, the thermoluminescence intensity of the phosphor is affected by the lattice deffects. On the other hand, in the case of the application of the capture gamma-rays for the dosimetry of thermal neutrons, the radiation damage is less than that due to a direct reaction between thermal neutrons and the phosphor. Therefore, with regard to re-using the phosphor, the application of the capture gamma-rays to the neutron monitoring is better than that of the direct reaction between the neutrons and the phosphor, and the linearity of the thermoluminescence response is maintained until the same dose range as the case of the irradiation of ^{60}Co gamma-rays, because of the irradiation of the gamma-rays. It is considered that, when the response is extrapolated from high fluence experimental results, the minimum measurable fluence is about 10^6 n/cm^2 or less. Therefore, it may be concluded that this monitoring system can be used for monitoring the thermal neutrons from the point of view of health physics.

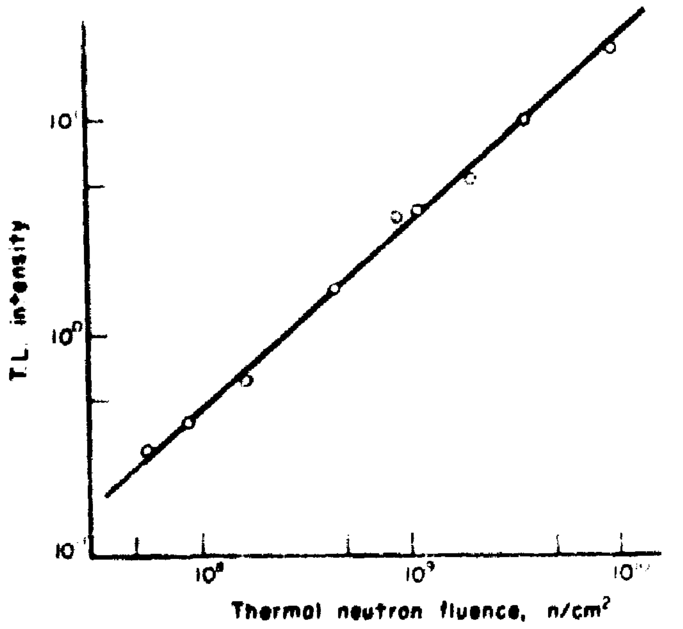

Fig. 3

Linearity of Thermoluminescence response of the monitor system on thermal neutrons (cadmium cylinder of 0.2 mm thick was used for the experiment)

The ratio between the thermoluminescence intensities of the phosphors in the monitoring system caused by the capture and reaction of gamma-rays was about 1.2 when the phosphor was enclosed with a 0.2 mm thick cadmium foil and when it was irradiated with the thermal neutrons obtained by thermalizing the fast neutrons of 2.5 MeV, but it has been reported that, when they are thermalized in paraffin 5 cm thick, the fast neutrons of 2.5 MeV used in the present experiment are about 40 per cent thermalized and that the tissue dose due to the fast collision of the fast neutrons is about 5.9 times the gamma-ray dose of this Van de Graaff generator. Therefore, it may be considered or inferred that the n/γ ratio in the mixed radiation field used in the present experiment is about 2.4. This value is larger than that obtained from the TLD monitoring system used in the experiment. Accordingly, when the thickness of the cadmium foil for enclosing the phosphor is varied, the optimum thickness of the cadmium foil with regard to the n/γ ratio of the monitoring system may be found.

Table 2 gives the n/γ ratio of the system based on thermal neutrons from the thermalized fast neutrons of 2.5 MeV. The n/γ ratio in the system was dependent on the thickness of the cadmium foil and increased with the thickness of the cadmium foil.

Table 2

The n/γ ratio of the monitoring system on various cadmium thickness (thermal neutron was obtained thermalizing fast neutron from *T*(d,n) reaction)

Cd(mm)	0.1	0.2	0.4	0.5	0.8	1.0
$(n/\gamma)/(n/\gamma)_0$	1.0	1.27	1.44	1.42	1.51	1.72

There are several elements with crossection larger than that of cadmium for example, Gd, Sm and Eu. When 4 g of Gd_2O_3 powder were used instead of Cd Foil, the ratio in the monitoring system was about 0.1, smaller than that of Cd foil. When cadmium is used, the thermoluminescence dosimeter can detect thermal neutrons which have energies below the effective cadmium cut off, which is near 0.5 eV, and the cost of the cadmium is much less than that of rare earth elements. Accordingly, in practice, the use of cadmium covered TLD is useful in the personnel monitoring system of thermal neutrons.

The influence of such phosphors as ^{6}LiF, LiF, ^{n}LiF, natural CaF_2, BeO(Na), $CaSO_4$(Tm) and Mg_2SiO_4(Tb) on the n/γ ratio of the monitoring system is given in Table 3. In this experiment, thermal neutrons were obtained from thermalized fast neutrons of the *T*(d, n) reaction. The n/γ ratios of all these phosphors except the LiF phosphors were larger than zero and were nearly equal to each other. This phenomenon will be discussed as the cause of the negative values of the n/γ ratio. The irradiation effect on the thermoluminescence emission due to α particles from the ^{6}Li(n, α) ^{3}H reaction in the LiF TLD phosphor without cadmium foil is larger than due to gamma rays from the Cd(n, γ) Cd reaction, because the LET of the α particles is much larger than that of the capture gamma rays.

Table 3

The n/γ ratio of phosphors in the monitoring system to thermal neutron and crossection (thermal neutron was obtained from thermalized fast neutron of *T*(d,n) reaction)

	^{6}LiF	^{n}LiF	BeO	CaF_2	$CaSO_4$ (Tb)	Mg_2SiO_4 (Tm)
n/γ	-0.93	-0.15	0.77	0.84	0.88	0.96
σ	880 barn	71 barn	94 mb	2153 mb	279 mb	143 mb

The thermoluminescence sensitivites of $CaSO_4$(Tm) and natural CaF_2 were about 1.0 and 0.3 times that of Mg_2SiO_4(Tb) respectively. Therefore, it may be concluded that these two phosphors can, like the Mg_2SiO_4(Tb) phosphor, be used for this monitoring system of thermal neutron detection. However, the thermoluminescence sensitivity of BeO(Na) is about one percent of that of the Mg_2SiO_4(Tb) phosphor. Accordingly, BeO(Na) is not suitable for use in the monitoring system for detecting a low fluence of thermal neutrons.

The holder shown in Fig. 2 was used as the TLD holder in this experiment. A part of "a" in Figure 2, which was named the holder space, is made of plastic material. It may be inferred that the thermoluminescence intensity of the TLD used to detect the gamma rays in the mixed field is disturbed by the capture gamma-rays from the cadmium foil. Therefore, the shielding effects of the holder space materials, such as lead, iron, brass and plastic, on the capture gamma-rays were studied.

The shielding effect of each material on the capture gamma-rays is given in Table 4. In this experiment, the n/γ ratios were compared with each holder space material. According to those results, the shielding effect was hardly dependent at all on the holder space materials. Therefore, it may be concluded that the plastic material is used as the holder space.

Table 4

Effect of holder space material on the n/γ ratio of the monitoring system (thermal neutron was obtained from thermalized fast neutron of the T(d, n) reaction)

	Plastic	Iron	Brass	Lead
n/γ	0.77	0.87	0.97	0.92

Acknowledgements

The authors wish to express their thanks to Prof. Dr. R. R. Pieroni, Director of the Institute of Atomic Energy, São Paulo, Brazil, and to Dr. T. Hashizume, Chief of the Physics Division, National Institute of Radiological Sciences, Chibashi, Japan, for their advice and encouragement. A part of the work was supported gy grants of CNEN and FAPESP, Brazil.

Resumo

Um sistema de monitoração de neutrons térmicos usado com dosímetros comuns de termoluminescência foi investigado. Neste sistema de monitoração, nós usamos raios gama de captura da reação entre o Cd e neutrons térmicos. Os efeitos de retro-espalhamento e "building-up" de radiação foram estudados, aproximando a razão n/γ do sistema ao do campo mixto. O resultado observado mostra que a razão de um sistema usando estes efeitos é maior do que a do sistema sem esses efeitos, e que a fluência mínima mensurável é cerca de 10^6 n/cm^2. Este método pode ser usado para todos os tipos de dosímetros termoluminescentes.

Résumé

Un système contrôleur des neutrons thérmiques utilisé avec dosimètres ordinaires de thermoluminescence est étudié. Pour ce système, on a utilisé les rayonnements gamma de la capture dans la reaction entre le Cd e les neutrons thérmiques. Les effets du "back scatter" e du "building up" du rayonnement sont examinés en mettant la raison n/γ du système auprès de celle du champ mixte. Le resultat observé montre que la raison d'un système en utilisant ces effets est plus grand que celle d'un système sans ces effets, et que la fluence minimum qui peut être mésurée est environ 10^6 n/cm^2. Cette méthode peut être utilisée pour tous les types des dosimètres thermoluminescents.

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