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A Study on the Neutron Dosimetry with LiF Thermoluminescent Dosimeters

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Abstract

A study was made on the neutron dosimetry in a mixed gamma-neutron field with LiF thermoluminescent dosimeter. In order to estimate the neutron dose in a mixed field, ^{6}LiF and ^{7}LiF dosimeters were used for fast and thermal neutron doses. The over-all conversion factors for the effects of dosimeter positions were derived for personnel monitoring and the glow curves of the LiF dosimeters for neutron and gamma-ray doses were also analyzed.

요 약

혼합 방사선장에서 LiF 열형광 선량계에 의한 중성자 선량측정법에 관해 연구하였다. 혼합 방사선장에서 중성자선량을 선택적으로 측정하고 평가하기 위해서 ^{6}LiF 와 ^{7}LiF 선량계를 속중성자 선량과 열중성자 선량측정에 이용하였다.

개인 방사선 피폭 선량측정에 사용키 위한 보정상수를 유도하였고 중성자와 감마선 선량측정을 위한 그로우곡선을 분석하였다.

1. Introduction

Personnel monitoring for radiation workers is required by international recommendation and national regulation for the purpose of controlling their radiation exposure. At present, the nuclear track emulsions, Kodak personal neutron monitoring film, are widely used for determining neutron exposures. The neutron monitoring by means of nuclear emulsions is known as an inadequate method for neutron energies less than 0.5 MeV or for mixed field exceeding 500 mR of gamma-rays. The biological shielding for protection of radiation degrades the neutron energy so that the energy of a large fraction of neutrons falls below the response threshold of the film. Therfore, an adequate personnel neutron dosimeter is required to detect neutrons in this low energy range. According to the development of thermoluminescent dosimeter, particularly the lithium fluoride, many investigators are concerned themselves in the application to the fields of the personnel neutron dosimetry.

LiF enriched in ^{7}Li responds primarily to gamma-ray, and LiF enriched in ^{6}LiF responds to both gamma-ray and neutrons of thermal and resonance energies. The radiation characteristics of these materials have been invetigated. Preston1) used ^{6}LiF and ^{7}LiF powder to detect albedo neutrons and Enders[2)] attempted the fast neutron dosimety using a pair of ^{7}LiF (TLD-700). Simpson[3)] described the possibility of measuring both thermal neutron and gamma-ray dose in mixed field by the combined use of TLD-100 (natural-abandance ^{7}Li in LiF) and TLD-700. Several related studies were carried out by researchers.[4-8)]

The purpose of the present study is to establish the methods of ^{6}LiF and ^{7}LiF dosimety for fast and thermal neutrons in order to use thermoluminescent dosimeters for personnel monitoring in neutron-gamma mixed fields and to obtain glow curves to get an imfornation about the contribution of neutrons to the TLD. The effects of incident direction of neutron and the distance of dosimeter from the phantonm on the response of the dosimeters are also studied.

2. Neutron dosimetry with TLD

Since the thermoluminescence of LiF dosimeter is based on the (n, α) reaction in the ^{6}Li, the ^{6}LiF dosimeter is more sensitive to thermal neutrons than to fast neutrons. The $^6\mathrm{Li}(n, \alpha)^3\mathrm{H}$ cross section is 945 barns for the thermal neutrons whereas it is only 0.3 barns for a 1 MeV neutron. Thermal neutrons absorbed energy in the ^{6}LiF dosimeter by the following reaction;

$$^6_3\mathrm{Li}+^1_0n \longrightarrow ^4_2\mathrm{He}+^3_1\mathrm{H}+4.78\,\mathrm{MeV}.$$

The alpha and triton particles share the excess energy from this endothermic reaction and absorb most of this energy in the ^{6}LiF dosimeters.

The neutron dosimetry system consists of three parts of LiF dosimeters of which two are shielded with tin (No. 1 and No. 3) and the others with $Cd+Sn$ filters (No. 2). The position of the three elements are shown in Fig. 1. The cadmium filter in position No. 2 absorbs all the incident neutron of energies below 0.4 eV. Since the cadmium absorbs neutrons by the (n, γ) reaction, the resulting photons are shielded by the tin between the cadmium and the ^{6}LiF dosimeter.

Regarding to the backscattered neutrons from the phantom, the relative response of No. 1 and No. 2 dosimeters are descrided as,

$$D_1=n\sigma(E)\ (\phi_s+\phi_s'+\phi_f') \quad (1)$$

$$D_2=n\sigma(E)\ (\phi_s'+\phi_f') \quad (2)$$

where ϕ_s is the incident thermal neutrpn flux, and ϕ_s' and ϕ_f' are the flux of the back-scattered thermal neutron and tissue moderated fast neutrons respectively, n is a proportional constant, $\sigma(E)$ is the $^6\mathrm{Li}(n, \alpha)^3\mathrm{H}$ cross section. The difference between D_1 and D_2 can be related to the incident thermal neutron dose as follows,

$$\text{Thermal Neutron Dose}=\frac{D_1-D_2}{f_1} \quad (3)$$

Where f_1 is a calibration factor for thermal neutron.

If the badge were exposed to a source of thermal neutrons, fast neutron component are zero, therefore, the ratio of backscattered thermal neutron to incident thermal neutrons can be defined as follows,

$$f_2=\frac{\phi_s'}{\phi_s}=\frac{D_2}{D_1-D_2} \quad (4)$$

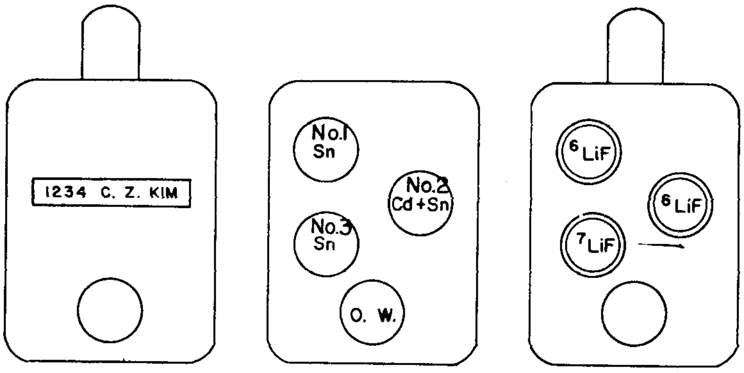
Since the ^{6}LiF dosimeter responds to both neutron and gamma-ray, the ^{7}LiF dosimeter is used to substract the gamma response

$$f_3=\frac{D_2}{D_3} \quad (5)$$

where f_3 is the fraction of the contribution by gamma-ray in the ^{6}LiF and ^{7}LiF dosimeter.

Considering the gamma-ray response, the fast neutron dose can be expressed as follows,

$$\text{Fast Neutron Dose}=\frac{D_2-f_3D_3}{f_4} \quad (6)$$



Front cover Filter holder Back plate

Fig. 1. **Thermoluminescent Dosimeter Badge**

where f_4 is a calibration factor. To calibrate fast neutron, the badges are attached to a polyethylene phantom.

3. Experimental

(1) Description of dosimeter

The TLD badge is consisted of thermoluminescent dosimeter discs, cadmium and tin shields and plastic case. The thermoluminescent materials used are LiF-teflon disc of ^{6}LiF(^{6}Li;95%) and ^{7}LiF(^{7}Li;99.99%) each of which has been uniformly incorporated in the teflon (polytetrafluoroethylene) with a careful mixing and forming technique by Teledyne Isotope Co. The dimensions of the disc are 12.0 mm(^{6}LiF), 12.7 mm(^{7}LiF) in diameter and 0.4mm in thickness. The discs contain about 30 mg of the thermoluminescent materials.

Fig. 1 shows the construction of the TLD badge which was developed by Teledyne for usage by personnel who are occupationally exposed to radiation. This badges are consisted of three main components. The front cover contains the filters to distinguish the quality of incident radiations. The final layer contains compartments for the LiF-teflon disc. Since the ordinary filters in the badge are made for gamma-ray dose measurement, the filters are chosen for the neutron dosimetry as follows; the ^{6}LiF dosimeter in position No. 1 with a 1.0 mm of tin filter, another ^{6}LiF dosimeter in position No. 2 with a Cd(0.5mm)+Sn(0.5mm) filter and ^{7}LiF dosimeter in position No. 3, for the correction of gamma-ray response, with the same filters in position No. 1.

(2) Irradiation

The responses of ^{6}LiF and ^{7}LiF dosimeters to the gamma-ray are measured with the ^{60}Co (18Ci) and ^{226}Ra(13.7mg) gamma-ray sources. For thermal neutron the response of ^{6}LiF is obtained at Bulk Shielding Experimental Tank of TRICA Mark-Ⅱ Reactor. The sample, placed inside a 2.5 in. diameter by 12 ft long aluminum pipe, is subject to irradiation at a given location along the horizontal center line of the thermalizing column door in the BSET. The thermal neutron flux[9)] has been reported to be $2.3\times10^6\,n/\text{cm}^2$-sec at the position 16 cm inside the reactor shield. The dose conversion factor[10)] of 2.1×10^{-10} rem/n is chosen for thermal neutron dose coversions. Since the fast neutrons are incorporated in the BSET facility[11] it is unsuitable as a thermal neutron sources. Fast neutron irradiations are made with a Pu-Be ($8.81\times10^5 n$/sec) neutron source and dose conversion factor[12] is 3.55×10^{-8} rems/n/cm^2. In this tests a TLD badge is attached to a phantom.[13]

The LiF dosimeters are annealed for 5 hr at 300°C followed by 24 hr at 80°C. The thermoluminescence of a sample was measured and the thermoluminescent glow curve was recorded with TLD-7100 and its associated dual-channel recorder of Teledyne Isotope Co.

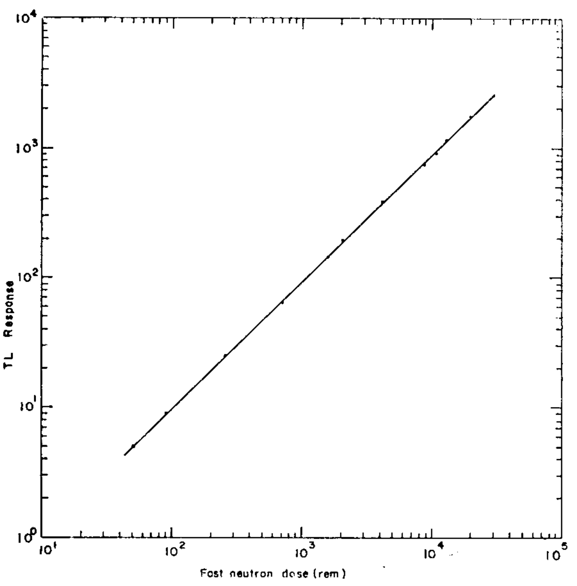


Fig. 2 TL Response as a function of neutron dose for ^{6}LiF

4. Results and Discussion

(1) Neutron dose measurement

The calibration factor, f_1, for thermal neutron dose estimation using tow ^{6}LiF dosimeters which were filtered by Sn and Sn+Cd filter respectively was 3.63 ± 0.69. The standard deviation of the value was about ± 13.7%. The errors in the experiments were mainly due to the influence of the fast neutrons and gamma-rays in the BSET facility. The ratio of the backscattered thermal neutron to incident thermal neutrons, f_2, was 3.5 ± 0.8. Since ^{6}LiF dosimeter is sensitive to gamma-ray as well as neutron, it is necessary to correct for the gamma-ray response in mixed neutron-gamma fields. This correction was done by the use of ^{7}LiF dosimeter. The response characteristics of ^{6}LiF dosimeter to gamma-ray is similar to that of ^{7}LiF dosimeter. The correction factor, f_3, obtained using ^{226}Ra gamma-ray was 1.021 ± 0.002. The calibration factor, f_4, for fast neutron was 0.393 ± 0.064, which was evaluated by equation (6) after exposing the badge to Pu-Be neutron source. These correction factors were obtained under the fixed condition of the thermoluminescent meterial, TLD badge, and TLD Reader. If the experimental conditions were changed the correction factors have to be reexamined.

The response of ^{6}LiF dosimeter was measured as a function of fast neutron dose from 50 mrad to 10^4 rad. The results are shown in Fig. 2. For ^{6}LiF dosimeter, the response versus dose curve was linear at least up to 2×10^4 rad. The sensitivity of readout instrument of LiF dosimeter for gamma-ray dose was 1.06 with a standard deviation of 0.8%. The room-temperature fading of the dosimeter was determined to be less than 0.5% over a period of 3 months and also dose rate dependence was negligible for Pu-Be neutron exposure in the range of mrem/hr.

(2) Effects of dosimeter wearing position

When the TLD badge is worn by a radiation worker, the badge is closely in contact with the body. The fast neutron response depends on backscattered neutron, therefore, the relationships are checked with dosimeter placed at distances up to 8 cm from

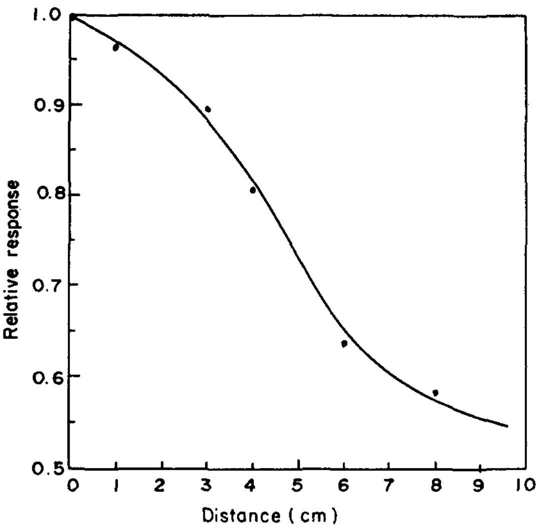


Fig. 3 Fast neutron response as a function of the distance between the TLD badge and the surface of a phantom

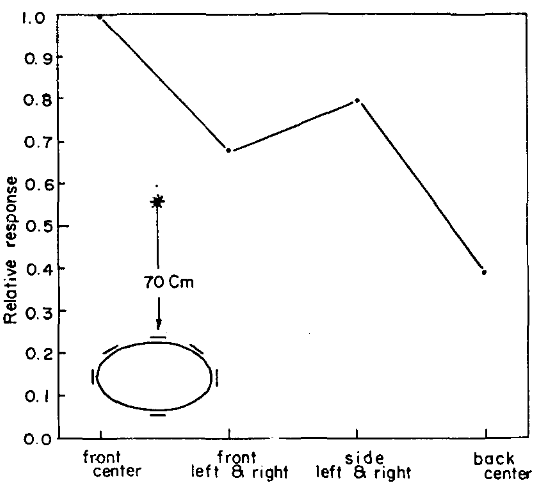


Fig. 4 Fast neutron response with the position of the TLD badge on a phantom

the front surface of the phantom. In this study, the polyethylene phantom is set at 30 cm from the Pu-Be neutron source.

As shown in Fig. 3, the results are normalized to unity as the response of TLD badge in contact with surface of the phantom. The response suddenly decreases out to about 6 cm as the badge-to-phantom distance increases, i, e, the backscattering effect is smaller than

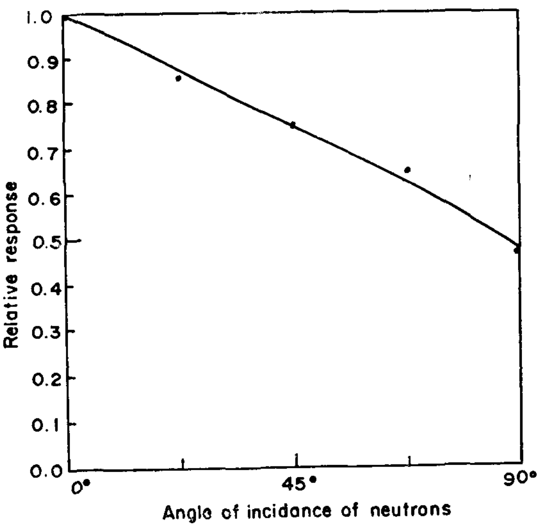


Fig. 5 Variation in response with angle of incidenc.

as one would expect. When the distances are 1 cm and 3 cm, the response of the LiF dosimeter falls by 3% and 11%, respectively. These ranges are typical distances of the dosimeter from the body when being worn on the clothing. When the neutron dose are estimated the error due to the effect of badge-to-body distance will be as much as 10%.

To study the response of the TLD badge in given wearing position of a radiation worker, TLD badge was attached to the middle-trunk, each shirt pocket positions, two lateral positions, and across the back of the phantom, as shown in Fig. 4. The fast neutron response was maximum when the TLD badge was on the center chest position of the phantom. The 30% divergency in maximum due to the change of the wearing position of the TLD badge was included in case of dose estimation.

Also the varying response of the badge to the different direction of the incident neutrons was studied because of the TLD badge on the person may not always be perpendicular to the neutron beams. A Pu-Be neutron source was placed 30 cm from a phantom and the angle of incident direction to TLD badge was adjusted from 0° to 90°. The fast neutron response under these conditions depends primarily on the scattering geometry of the source and phantom. The neutron response dropped off slowly with decreasing angle as shown in Fig. 5. A lot of difference due to the angular variation of incidence to the TLD badge were appeared. This results would indicate that the directional effect must be considered for neutron dose interpretation.

(3) Glow curve of the LiF dosimeter.

Morehead and Daniels[14] have studied the glow curves of LiF dosimeter with various types and dose of irradiation and found serveral peaks present. Wingate[15] also reported the characteristics the glow curve of LiF dosimeter induced by the ^{60}Co gamma-ray and that induced by thermal neutron. Furuta[6] has pointed out that the differences could be arisen by the different manners of excitation in thermoluminescent materials for the kinds of radiation when the irradiation dose is not so high.

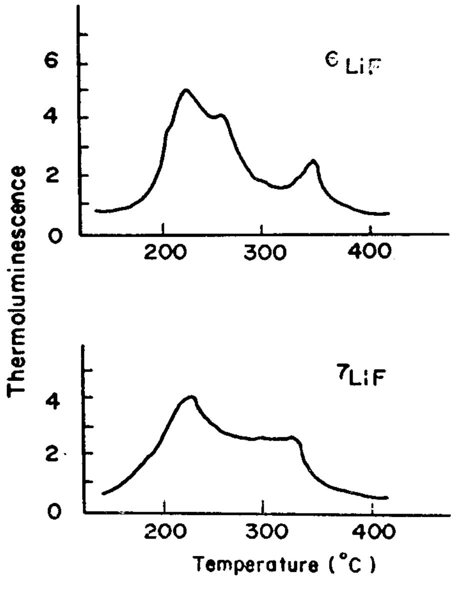


Fig. 6 TL glow curves of ^{6}LiF and ^{7}LiF irradiated by ^{60}Co gamma-rays.

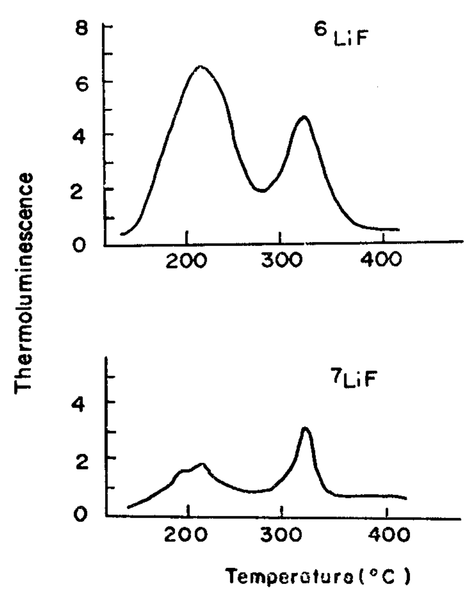


Fig. 7 TL glow curves of ^{6}LiF and ^{7}LiF irradiated by Pu-Be neutron source.

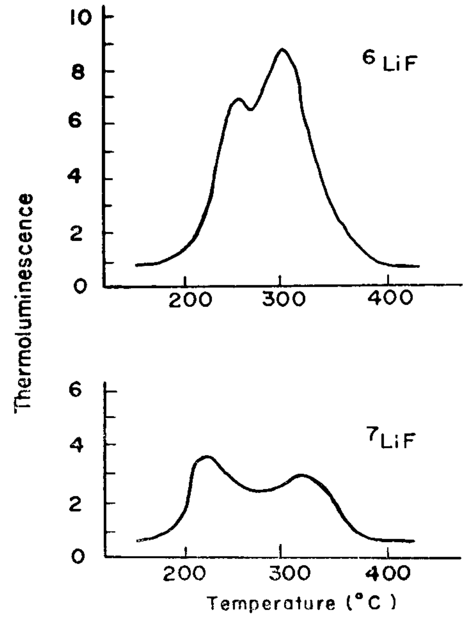


Fig. 8 TL glow curves of ^{6}LiF and ^{7}LiF irradiated by thermal neutrons.

In the case of neutron irradiation, the

thermoluminescence in LiF dosimeter is induced by triton and alpha particles or recoiled nuclei. The difference in glow curves of ^{6}LiF and ^{7}LiF dosimeters irradiated by neutron may be caused by the $^6Li(n,\alpha)^3H$ reaction in LiF dosimeter. The glow curves of the ^{7}LiF and ^{6}LiF dosimeter irradiated with 500 mR of ^{60}Co gamma-rays are shown in Fig. 6. The heating rate of readout system was 10°C/sec. The remarkable difference between the two glow curves was not observed. The main peak in the glow curve was found at about 220°C and it nearly corresponded with the reported data. The peak also has a tendency shifting to higher temperature region according to the irradiated dose of ^{60}Co gamma-ray. The glow curves of ^{6}LiF dosimeter show two distinguished peaks at 260°C, and 345°C, and one peak appears at 325°C in case of ^{7}LiF. All the data were taken 24 hr after irradiation with the fixed instrumental conditions.

Fig. 7 shows the glow curves of ^{7}LiF and ^{6}LiF dosimeter irradiated by Pu-Be neutrons (average energe; 4.2 MeV) of 8.6 rem. In the former the glow peak at 320°C was predominant and at 220°C in the latter. Fig, 8 shows the glow curves of LiF dosimeter irradiated by the thermal neutron in the BSET thermal column. As seen in this figure, the remarkable difference between two glow cuves was observed. The glow curve of ^{7}LiF dosimeter was nearly the same as that of Fig. 6. It is supposed this kind of similarity comes from the fact that the former glow curve is mainly due to the gamma-ray in the BSET facility. The glow peaks of ^{6}LiF dosimeter were appeared at about 250°C and 300°C, and the latter was more predominant than the former. This may be attributed to the fact that the glow curve is induced by the $^6Li(n,\alpha)^3H$ reaction in the ^{6}LiF dosimeter.

5. Conclusion

A thermoluminescent neutron badge using ^{6}LiF and ^{7}LiF dosimeter has been developed to replace the neutron film badge for the further application to the routine personnel monitoring. The TLD badge has the approximate size of film badge and contains a pair of ^{6}LiF dosimeter and a ^{7}LiF dosimeter. The LiF dosimeter can be used repeatedly with proper annealing and maintains a constant reading conditions. The fast neutron response of LiF dosimeter is widely linear range with the neutron dose. The smallest neutron dose necessary to measure is of the order of 10 mrem. The dosimetry problem at this low dose range is left. LiF disimeter may be used for the dosimetry of mixed neutron and gamma-ray fields by utilizing cadmium and tin filters. Neutron and gamma-ray exposure in the mixed fields can be seperated. The response of ^{6}LiF and ^{7}LiF dosimeter to gamma-ray are nearly the same. Since LiF dosimeter are comparatively insensitive to neutron, the correction for response of neutron are unnecessary for gamma-ray dose estimations. The response of TLD badge has some dependence on the incident angle of radiation and wearing position of the badge.

The glow curve of ^{7}LiF dosimeter irradiated by neutron is slightly different from the case of ^{6}LiF dosimeter and that is not much different from the curve of ^{7}LiF by gamma-rays. Further experiments are in progress to determine the variation response with neutron spectral shapes.

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