BNW-S4-4355
CONF-721203--9

RECEIVED BY TIC FEB 9 1973

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A THERMOLUMINESCENT DOSIMETRY SYSTEM FOR GAMMA,
BETA AND NEUTRON PERSONNEL EXPOSURE*

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INTRODUCTION

The phenomenon of thermoluminescence and its application to personnel dosimetry has been studied in great detail by Kocher and Endres at Hanford and other people throughout the world since 1964.^[1,2] In the two or three years preceeding this date, efforts at the University of Wisconsin under the direction of John Cameron solved the problems of doping lithium fluoride in a manner which provided sufficient sensitivity to radiation so that the material could be used in personnel dosimetry.

The thermoluminescent properties of LiF as measured by Endres^[3] are nearly independent of the incident photon energy, and this property makes it suitable as a replacement for the health physics film badge. The sensitivity of LiF relative to soft tissue is constant within 10 percent for photons from

* This paper is based on work performed under U. S. Atomic Energy Commission Contract AT(45-1)-1830.

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10 keV to 10 MeV^[4] so that a multielement array of LiF blocks with suitable filters can act as a simple spectrometer as well as an accurate dosimeter.

As part of the continuing effort to improve the health physics program at Hanford, a new personnel dosimeter utilizing LiF has been developed and placed in service. The main criteria for the development of a personnel dosimeter to replace the Hanford film badge are: that the badge is to be used at all work locations to estimate radiation dose to persons from exposure to various combinations of neutrons, photons and beta particles; and, that the badge be designed so that it can be automatically processed and returned to service in less than one month.

DESCRIPTION OF DOSIMETER

The multipurpose card (Figure 1), which houses the TL blocks measures 7.8 cm x 3.8 cm x 0.1 cm. The LiF TL blocks (0.32 cm x 0.32 cm x 0.089 cm) are contained in Teflon TFE skived tape that is 0.005 cm (12.2 mg/cm²) thick on the front side of the block and 0.009 cm (16.0 mg/cm²) thick on the back and positioned at the center of five 0.95 cm diameter holes in the dosimeter card. The tape on the back side of the card is coated on one side with pressure sensitive silicone polymer adhesive. The Teflon TFE tape on the front side is not coated with an adhesive for reasons discussed by Shipler.^[5] This semitransparent tape allows the thermoluminescent blocks to be read out without removing them from the dosimeter card. The

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dosimeter card is injection molded from a thermoplastic resin which has a heat deflection of 130°C .

The dosimeter holder is injection molded from Noryl plastic and is designed to hold the dosimeter card, the Hanford security credential and appropriate filters for the blocks as shown in Figure 1. The holder measures 8 cm x 4.5 cm x 0.6 cm. The 1.27 cm diameter filters are retained in cavities in the holder with an adhesive and covered with mylar tape. When the dosimeter card is inserted into the holder, a snug friction fit is obtained and each TL block is centered between the appropriate filter materials.

The security credential is 0.076 cm thick and has a density thickness of 80 mg/cm^2 . The Hanford Multipurpose Dosimeter was conceived after it was determined that LiF could be read out at the normal reader temperature ($300\text{--}310^{\circ}\text{C}$) and held for one month at room temperature, maintaining a constant sensitivity. Subsequent studies have shown that a standard readout with a 24-hour 60°C oven anneal with a five-day room temperature holding period will also anneal the multipurpose dosimeter card for a dose of 10 rads or less.^[6] We have adopted this latter procedure because it will allow the dosimeter cards to be returned to field use faster and the statistical variations are less than the variations observed with a standard readout and 30 day hold at room temperature.

BETA AND PHOTON DOSIMETRY

The multipurpose dosimeter card has two ^7LiF TL blocks that are isotopically enriched in ^7Li to 99.993 percent for beta and photon dosimetry. The positions of the two blocks (#1 and #2) in the dosimeter card are shown in Figure 1. When the dosimeter card is inserted into the holder, a snug friction fit is obtained and the block in position #1 is located in the open window where it is filtered by only its associated Teflon TFE tape and the security credential. The TLD in position #2 is shielded by an additional 0.089 cm of plastic and 0.064 cm of aluminum. The two blocks serve as a simple two-element photon-beta particle spectrometer. The 0.089 cm block thickness was chosen to provide adequate sensitivity and to eliminate breakage problems encountered with thinner blocks.

The thermoluminescent response of block #2 is interpreted as penetrating dose and its shield was designed to simulate one centimeter of soft tissue shielding for photon energies from 10 keV to 10 MeV. For beta dosimetry, this shield (including the security credential) is about 365 mg/cm² thick.

If the photomultiplier tube current is integrated during the readout process, then the dose interpretation is accomplished with the following equations:

$$\text{Penetrating dose} = \frac{R_2}{k_1} \text{ mrad} \quad (1)$$

$$\text{Derma dose} = \text{penetrating dose} + \frac{R_1 - k_2 R_2}{k_3}$$

where R_1 and R_2 are the net integrated phototube currents from the #1 and #2 TL blocks and k_1 , k_2 and k_3 are the calibration constants determined by exposures to selected sources.

The sources used are selected to provide spectra of photon and beta radiation which are representative of radiation fields at various Hanford work areas.

The energy response function of #1 TL block within the Teflon envelope and shielded by 0.076 cm of plastic is essentially flat from 100 keV to 1.25 MeV. There is a small region around 40 keV where the shield is less effective than one cm of tissue; but this makes the interpretation in a mixed energy field conservative. The energy response curves for the filter TL block combinations are also given by Kocher et al. [6]

An experiment was conducted to study the variation in angular response of the dosimeter to radiation from a ^{60}Co source placed twenty centimeters from the center of the chest of a Remab phantom.

For angles near 90° to the right, the effective shielding of plastic in the card and holder is only slightly larger than the shielding of the filter system and the response remains relatively flat. When the source is to the left of center, the effective shielding of the badge and filter system continually increase through 90° , and the response drops off by about 40%.

It is imperative that the dosimeter be capable of accurate dose interpretation of the various kinds and spectra of radiation to which it may be exposed. To study this property of the dosimeter, eleven combinations of 16-keV x-ray, uranium beta, radium gamma and PuF_4 (neutron source) exposures were arranged and two TLD badges were exposed to each combination. It is evident from the data in Table I that the dosimeter does meet the criteria described above.

When ^{90}Sr radiation is used to evaluate k_3 of equation (2), the value of k_3 is about twice as large as it is when a uranium beta source is used. The photon-beta contributions for all Hanford sources of exposure interpret well with the ^{90}Sr calibration.

Several field studies were conducted by placing TL dosimeters on Hanford personnel along with their regular film badges and, in some cases, pencil dosimeters. The data from this study is reported in another paper,^[7] but in summary, the multipurpose dosimeter results were more consistent and reliable than results obtained from the film dosimeter. With our current readout system we consider the low level detection limits to be 20 mrad of penetrating radiation and 10 mrads of nonpenetrating radiation.

NEUTRON DOSIMETRY

The slowing down of neutrons in a human body is due primarily to elastic scattering with hydrogen nuclei and it is the

subsequent recoil protons which are responsible for most of the neutron dose delivered to the body. For neutron energies below 10 MeV, the elastic scattering is nearly isotropic in the laboratory system and, because of the amount of hydrogenous material in the body, the leakage spectrum of neutrons from the body is primarily a thermal spectrum. By monitoring this leakage spectrum of neutrons, a first order measurement can be made of the fast neutron dose delivered to the body.

The neutron dosimetry portion of the multipurpose badge consists of three LiF blocks. Two of the blocks are isotopically enriched to about 95 percent ^6Li and the third block is enriched to about 99.993 percent ^7Li . The positions of the three elements in the dosimeter card are shown in Figure 1. Positions #3 and #4 are ^6LiF blocks and position #5 is the ^7LiF blocks. Thermal neutrons deposit energy in the ^6LiF block by the nuclear reaction

The alpha and triton particles share the excess energy from this reaction and deposit most of this energy in the TL crystals since the dimensions of the TL blocks are large compared to the range of these particles in LiF.

The bare ^6LiF block responds to the backscattered neutrons from the body as well as to incident neutron flux. The incident neutron flux can be thought of as consisting of just slow and fast neutrons, ϕ_s and ϕ_f , respectively. The $^6\text{Li}(n,\alpha)$ cross section is about 1000 barns for thermal neutrons whereas it is only 0.3 barns for a 1.0 MeV neutron. Therefore, the ^6LiF TL

block is a relatively poor detector for fast neutrons and for practical applications, the contribution to its thermoluminescence due to fast neutron capture can be ignored. The ^6LiF TL block in position #4 is shielded from incident slow neutrons by placing 0.051 cm of cadmium in front of it. The cadmium absorbs essentially all of the incident neutrons of energies below 0.4 ev.

The backscattered neutrons that are captured in the ^6LiF blocks are backscattered slow neutrons (ϕ'_s) and tissue moderated fast neutrons (ϕ'_f). The magnitude of backscattered neutron flux density per unit incident fast flux has been calculated by P. S. Nagarajan and D. Krishnan^[8] and experimentally measured by J. A. Dennis, J. W. Smith and S. J. Boot.^[9] If we ignore photon response, the above discussion can be summarized by writing equations for the relative response of TL blocks #3 and #4.

$$R_3 = n\sigma(E) [\phi_s + \phi'_s + \phi'_f] \quad (3)$$

$$R_4 = n\sigma(E) [\phi'_s + \phi'_f] \quad (4)$$

where R_3 and R_4 are the relative thermoluminescent response of TL blocks #3 and #4, n is a proportionality constant, and $\sigma(E)$ is the $^6\text{Li}(n,\alpha)$ H cross section. The difference between R_3 and R_4 ($n\sigma(E)\phi_s$) can be related to the incident thermal neutron dose:

$$\text{Incident slow neutron dose} = \frac{R_3 - R_4}{k_4} \text{ mrad} \quad (5)$$

where k_4 is a calibration constant. By exposing the badge to a source of thermal neutrons, ϕ_f and ϕ'_f , are zero so that equations (3) and (4) become

$$R_3 = n\sigma(E) [\phi_s + \phi'_s] \quad (6)$$

$$R_4 = n\sigma(E) \phi'_s \quad (7)$$

The ratio of backscattered thermal to incident thermal neutrons can be defined as a constant:

$$k = \frac{\phi'_s}{\phi_s} = \frac{R_4}{R_3 - R_4} \quad (8)$$

If we subtract that portion of the backscattered neutrons which is due to incident slow neutrons ($k_5\phi_s$) from the bracketed term in Equation (4), then the response of R_4 is proportional to ϕ_f . This can then be related to fast neutron dose.

The ^6LiF blocks are also photon sensitive and it is impractical to adequately shield them from all photon radiation. Therefore, a third block of ^7LiF enriched TL material (insensitive to neutrons) is placed in position #5 of the dosimeter card and shielded front and back by 0.102 cm of tin to prevent beta particles and low energy photons from interacting in the TL blocks and possibly causing statistical problems in low neutron dose interpretations. The TL block in position #3 is similarly shielded and the block in position #4 has 0.102 cm of tin on the back and 0.051 cm of tin on the front between the cadmium and the block. This arrangement makes all three shields photon equivalent and helps

reduce the photon contribution from the activated cadmium in position #4. The fast neutron dose is then interpreted using the following equation:

$$\text{Fast Neutron Dose} = \frac{R_4 - k_6 R_5 - k_5 (R_3 - R_4)}{k_7} \text{ mrad} \quad (9)$$

where k_6 is a correction factor for the difference in photon sensitivity between the ^6LiF and ^7LiF TL blocks, and k_7 is a calibration constant.

The constant k_4 is evaluated by solving equation (5) after known exposures of the badge to thermal neutrons in the Hanford sigma pile. The constant k_5 is evaluated by solving equation (8) after setting the fast neutron dose equal to zero and evaluating the responses for the same exposures as described above for evaluating k_4 . The constant k_6 is evaluated by solving

$$k_6 = \frac{R_4}{R_5}$$

after exposing the badges to a penetrating photon calibration source. For fast neutron calibration, the badges are attached to a polyethylene phantom. The constant k_7 is then evaluated by solving equation (9) after exposing the badges to neutrons from a plutonium fluoride neutron calibration source.

Spectral considerations and associated quality factors were studied at a variety of work locations at Hanford. The measurements were made with a tissue equivalent proportional counter (TEPC) which measures absorbed dose directly. An average quality factor ($\overline{QF}$) can also be obtained from the TEPC data.

These data can then be compared for variations in many of the neutron spectra at Hanford. The $\overline{QF}$ range is from 8.5 for a bare ^{252}Cf source to 12.0 for a heavily shielded plutonium storage vault. The average of all these $\overline{QFs}$ is 10.1. The PuF_4 neutron source used for fast neutron calibration has a $\overline{QF}$ of 10.0. The accuracy of these measurements is about ± 1 $\overline{QF}$ unit so it would seem that the use of PuF_4 as a fast neutron calibration source is justified.

When the TLD neutron badge is worn by a person, the badge is very seldom in contact with the body. Since the fast neutron response depends on backscattered neutrons, a study of the relationship between response and badge-to-phantom distance was made. Sets of three badges each were positioned in front of a chest phantom and exposed to the standard PuF_4 neutron calibration source. Each set was positioned at a different distance from the surface of the phantom. In this study, the phantom was set at 35 cm from the source, and therefore, the badges were at various distances from the source. The response, when the badge is close to the phantom surface, is smaller than one would expect compared to the response at 2 cm due to the effect of the relatively large cadmium shield in the badge (see Figure 2). The response increases out to about one centimeter as the effective shield size decreases. At 3-6 cm, the response drops off in a manner similar to a R^{-1} curve. A datum point from 0.56 MeV neutrons produced by our Van de Graaff accelerator and one from a Harwell study^[9] using 0.120 MeV neutrons fall on this curve.

To study the response of the dosimeters as a function of weaving geometry, several dosimeters were taped at various locations on a water phantom and exposed to fast neutrons. Monoenergetic neutrons of 4.5, 1.0 and 0.35 MeV were used for the study. The results from dosimeters placed in symmetrical positions relative to the neutron sources were averaged and corrected for background. These results are shown in Figure 3.

As stated previously, a dosimeter under field conditions is very seldom exposed to a single source of radiation. It is therefore necessary that the TLD badge be capable of accurate neutron dose interpretation when exposed to a variety of different kinds of radiation. To study this property, eleven combinations of 16 keV x-ray, uranium beta, radium gamma and PuF_4 neutron exposures were made using two TLD badges for each combination. These data, shown in Table II, show that the multipurpose dosimeter provides reasonable dose interpretation for neutrons when mixed combinations of radiations are present.

The lower limit of fast neutron interpretations in high photon radiation fields is a matter of statistics and is about 10 mrad $\pm$ 100 percent for fast neutrons in a mixed exposure with about 3000 mrad of photons. Statistical analysis of our data indicates that the TL neutron dosimeter can consistently detect 0.5 mrad of thermal neutrons and 5 mrad of fast neutrons within $\pm$ 50 percent when the photon exposure is 150 mR or less.

Exposures were conducted at the Health Physics Research Reactor (HPRR) at Oak Ridge National Laboratory to simulate a nuclear criticality event. Measurements were made for three bursts of the fast burst reactor: one with no shielding, one with a lucite shield between the reactor and the dosimeters, and one with an iron shield between the reactor and the dosimeter. The dosimeter performed satisfactorily in radiation fields with dose rates as high as 1.1×10^7 rads/sec neutron plus gamma. The results for the burst with no shielding are shown in Table III. For the shielded burst the dosimeter results were high as expected due to an increase in scattered intermediate energy neutrons.

SYSTEM OPERATION

The total system has been in operation since January of 1972 and several problems have become apparent. Some of these difficulties have been corrected and others have not. The dark current plus background signal was rather large (about equivalent to 100 mR gamma exposure) when the system was first put into service. By experimenting with various light filters this signal was reduced to about 20 mR equivalent, half of which is phototube dark current. Some of this signal may be due to thermal shock from the heater at 300°C contacting the cold thermoluminescent chip but it is a relatively small part of the background. The filter eliminated a background due to luminescence of the teflon tape and adhesive.

It is highly recommended that all thermoluminescent chips be purchased from the same batch of material to assure uniform sensitivity.

All multipurpose dosimeters are processed in a computer controlled automatic reader. The computer automatically controls the readout of each dosimeter and then calculates the photon and neutron dose for each dosimeter. Such parameters as heater temperature during readout, phototube dark current, several important circuit voltages, phototube gain and positive identification of employee number, are checked automatically. Control exposures of 1000 mR and 0 mR are read periodically and must fall within prescribed limits. In general, the automatic system required a minimum amount of operator attention.

CONCLUSIONS

A multipurpose thermoluminescent personnel dosimeter has been developed to replace the Hanford film dosimeter. The dosimeter can be used repeatedly with proper annealing and maintain a constant sensitivity. Its photon energy response and shielding provide quite accurate penetrating dose and total derma dose interpretations over the wide range of radiation sources and spectra found at Hanford. The calibration and interpretation procedures are simple and do not leave gaps or unreasonable limits for any one kind of mixture of radiation. The dosimeter has a reasonable independence from body wearing position and angle of radiation incidence. The ^7LiF blocks are relatively insensitive to neutrons of any energy so no

neutron corrections are necessary for beta-photon dose interpretations.

Our experimental evidence and that of other authors shows that 300°C is an adequate preexposure annealing temperature for precision dosimetry. At this temperature, we are able to adequately read out and pre-anneal the blocks in the same process. Data indicates that an additional 24-hour 69°C oven anneal with a five-day holding period at room temperature restores the dosimeters to a uniform sensitivity. This process can be repeated indefinitely.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the valuable assistance of W. H. Bischoff, W. A. Crook and D. L. Haggard for their work in processing the tens of thousands of TL blocks associated with the experiments, prototypes and final dosimeters. The authors are also indebted to J. P. Corley, K. R. Heid, R. L. Kathren and H. V. Larson of the Battelle-Northwest Occupational and Environmental Safety Department for their encouragement, advice and support, and D. H. Denham and his staff of the Radiological Calibrations Laboratory for their dependable service and technical assistance.

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TABLE I
MIXED DOSE COMPARISONS (mrads)

<u>Run</u>	<u>Dose Given</u>		<u>Dose Interpreted From TLD</u>	
	<u>Penetrating</u>	<u>Derma</u>	<u>Penetrating</u>	<u>Derma</u>
1	380	710	380	640
2	250	450	270	430
3	120	350	120	250
4	240	340	260	390
5	1140	1310	1020	1020
6	240	410	300	412 ⁰
7	140	600	160	420
8	240	600	210	720
9	220	350	190	290
10	240	350	190	350
11	70	400	30	350

TABLE II
MIXED DOSE COMPARISONS

<u>Run</u>	<u>Dose Given (mrad)</u>				<u>TLD mrads</u>	
	<u>16 keV</u>	<u>Uβ</u>	<u>PuF₄ Neutron Source</u>	<u>Ray</u>	<u>n_s</u>	<u>n_f</u>
1	200	200	100	200	3	74
2	0	200	50	200	2	43
3	50	200	50	50	2	43
4	100	50	100	100	3	91
5	100	100	100	1000	2	124
6	100	100	100	100	2	122
7	100	400	0	100	0	0
8	400	100	0	100	0	0
9	50	100	0	200	0	0
10	100	50	0	200	0	0
11	200	200	0	0	0	0

TABLE III

GAMMA AND FAST NEUTRON DOSE RESULTS

Burst #1

 7.7×10^{16} Fissions

Bare Reactor

<u>Distance (Meters)</u>	<u>Gamma Exposure (R)</u>	<u>Fast Neutron Dose (rads)</u>	<u>Relative Interpretation</u>		
			<u>Photon</u>	<u>Neutron</u>	<u>1/R²</u>
3	91.0	452	1.0	1.0	1.0
4	52.1	262	0.57	0.57	0.56
5	35.8	166	0.39	0.37	0.36
6	24.8	149	0.27	0.26	0.25
9	11.6	61	0.13	0.13	0.11
15	3.4	21.5	0.04	0.05	0.04

FIGURE CAPTIONS

1. View of dosimeter holder, security credential and dosimeter card.
2. Fast neutron response as a function of the distance between the multipurpose badge and the surface of a polyethylene phantom.
3. Relative monoenergetic fast neutron response as a function of the position of the badge on a phantom. Data have been normalized to front center position.

Figure 1.

VIEW OF DOSIMETER HOLDER, SECURITY CREDENTIAL AND DOSIMETER CARD

Holder-Front view

Holder with card
and Credential

Credential

Holder back
interior view

Card assembly

Holder back Front
interior view

FAST NEUTRON RESPONSE AS A FUNCTION OF THE
DISTANCE BETWEEN THE MULTIPURPOSE BADGE AND
THE SURFACE OF A POLYETHYLENE PHANTOM

Fig 2

Figure 2

RELATIVE MONOENERGETIC FAST NEUTRON
RESPONSE AS A FUNCTION OF THE POSITION
OF THE BADGE ON A PHANTOM. ~~DATA HAS
BEEN NORMALIZED TO FRONT CENTER POSITION~~