20 SEP. 1983

TIS-RP/113/CF

TIS DIVISIONAL REPORT

1 September 1983

A COMBINATION OF TLD ALBEDO AND SULPHUR ACTIVATION TECHNIQUES
FOR FAST NEUTRON PERSONNEL DOSIMETRY

J.W.N. Tuyn and A.R. Lakshmanan

ABSTRACT

A combination of a TL albedo dosimeter system consisting of a ${}^6\text{LiF} + {}^7\text{LiF}$ pair and a sulphur pellet has been studied for fast neutron personnel monitoring in high-energy accelerator environments.

P00030835

To be presented at the
7th International Conference on Solid State Dosimetry,
Ottawa, 27 - 30 September 1983

G E N E V A

1983

INTRODUCTION

Thermoluminescent detectors ($^6\text{LiF} + ^7\text{LiF}$) in 12.5 cm $\emptyset \times$ 12.5 cm polyethylene (PE) cylinders are routinely used for environmental gamma and neutron monitoring on the CERN sites ⁽¹⁾. However, for personal neutron dosimetry most of the systems currently used do not perform satisfactorily due to fading or incorrect neutron response ^(2,3). TL albedo dosimeters provide a reasonable response only to neutrons of thermal and intermediate energies. Sulphur activation provides a satisfactory response for neutrons (>2 MeV). Hence, we studied the combination of a TLD albedo system ($^6\text{LiF} + ^7\text{LiF}$) and a sulphur pellet for possible applications in personal neutron dosimetry at CERN. This paper reports the preliminary results of this study.

MATERIALS AND METHODS

For albedo dosimetry the $^6\text{LiF} + ^7\text{LiF}$ TLD pair ($3 \times 3 \times 0.9 \text{ mm}^3$ Harshaw chips) were fixed firmly on a polyethylene (PE) cylinder (25 cm $\emptyset \times$ 25 cm). For calibration a PuBe neutron source having a dose rate of 200 mrem (2 mSv)/h at 1 metre was used. The monitoring sites at CERN covered in this study were certain controlled radiation areas in the vicinity of the 28 GeV Proton Synchrotron (PS) and the 600 MeV Synchro-Cyclotron (SC). The $^6\text{LiF} + ^7\text{LiF}$ TLDs kept inside the PE (25 $\emptyset \times$ 25 cm) cylinders provide a measure of the dose equivalent (H) which was also measured with ionization chambers positioned permanently for routine monitoring at these sites, a neutron rem counter and ^{11}C activation for high-energy particles (>20 MeV).

In order to improve the counting efficiency for beta rays from the ^{32}P radioactivity induced by fast neutrons in the sulphur pellet ($\emptyset$ 51 mm, thickness 6 mm, weight 20 g), the sulphur was sublimated on a steel planchet placed over a hot plate maintained at 450°C in a fumehood⁽⁴⁾. The major advantage of the sublimation technique is that it is possible to burn any number of sulphur pellets on a single planchet (as long as the sulphur does not contain impurities which do not burn) and concentrate the ^{32}P activity thereby eliminating the self-absorption of ^{32}P beta rays in the sulphur pellet(s). For counting the ^{32}P beta rays, a Beckman gas-flow proportional counter having a background counting rate of 1.5 cpm was used.

Since in personal dosimetry the exposure history is normally unknown, a maximum uncertainty in neutron dose estimation due to decay of ^{32}P activity ($T_{1/2} = 14.3$ d) by a factor of 2 is introduced, for a monitoring period of 30 d. This uncertainty will be much less than a factor of 2 if the exposure is at a constant rate or if the exposure occurs close to the middle of the monitoring period, because it is then possible to correct for the decay of ^{32}P activity which occurs during this period. If, however, the major neutron exposure occurs at the extreme of the monitoring period, the uncertainty in dose estimation will be high but could be reduced considerably with some knowledge of the neutron exposure history. For this purpose we studied the differential fading characteristics at room temperature ($\sim 20^\circ\text{C}$) of low and high temperature peaks in the TL albedo dosimeter (^6LiF). As the fading of reader annealed ^6LiF chips to neutrons has been reported to be high⁽⁵⁾, we (pre-irradiation) annealed the ^6LiF and ^7LiF chips twice in the reader after the standard oven annealing treatment (400°C , 1 h + 100°C , 2 h) for this study. A two-temperature

readout (80-170°C and 170-250°C) was used for this purpose. The TLD reader system used was a Teledyne Isotopes (model TLD 7300) for pre-heat, read-out and annealing. A pre-heat of 80°C for 10 s was used to erase the TL contribution from very low temperature peaks. Nitrogen (2 lpm) was flushed during TL measurements. For fading studies, the TL was read during the two temperature intervals, 80-170°C and 170-250°C. For albedo dose measurements the latter readout only was used. For the estimation of time of neutron exposure the ratio of 80-170°C/170-250°C readouts was used.

RESULTS AND DISCUSSIONS

The number of counts after a decay period of 14 days from the ^{32}P activity induced in a sulphur pellet (20 g) by 5 rem (50 mSv) of PuBe neutrons before and after the sublimation of sulphur was found to be 83 ± 18 and 676 ± 32 per 10 minutes respectively, so an enhancement in the counting ratio by a factor of 8.1 could be obtained by sublimation of the sulphur. This enhancement was found to increase proportionally with the number of sulphur pellets (at least up to 10) sublimated in a single planchet. The decay of activity left on the planchet after sublimation of sulphur was found to be exactly the same as that of ^{32}P ($T_{1/2} = 14.3$ d).

If the limit of detection is four times the standard deviation of total counts, the detection threshold works out to be 43 mrem (0.43 mSv) of PuBe neutrons for a counting duration of 100 min. with one 20 g sulphur pellet, with the counting carried out 14 days after irradiation. The threshold detection limit could be lowered by sublimating more sulphur pellets in one planchet or by increasing the counting time (e.g. 14 mrem after 1000 min.).

For PuBe and fission neutrons the average cross-section for ^{32}P production are 173.5 mb and 67 mb, respectively. Hence the detection threshold for fission neutrons will be a factor of 2.6 higher than for PuBe neutrons. Also for neutron fields around high-energy accelerators a higher detection threshold than for PuBe is to be expected, because of the decrease in cross-section at high energies.

Table 1 gives the sensitivity ($\text{counts} \cdot \text{min}^{-1} \cdot \text{rem}^{-1}$) of a sulphur pellet (after sublimation of sulphur) for different neutron exposure conditions. The sensitivity was obtained by assuming exposure in the middle of the month and decay during 17 days. If continuous exposure took place during 30 days followed by two days of waiting before measuring the counts, the sensitivity increases by a factor of 1.11. It is seen from Table 1 that in general as the sensitivity of sulphur decreases, the sensitivity of TL albedo increases, as a result of change in neutron spectrum from hard to soft. Hence, a linear combination of sulphur activation and TL albedo should in principle provide a good personal neutron dosimeter system similar to a combination proposed in the past using ^6LiF and LR115 track detectors⁶⁾. The major advantages of sulphur activation techniques are the complete discrimination of neutrons against accompanying gamma rays, the very wide dynamic range, the independence on angle of incidence and evaluation techniques which are easy to automate using e.g. classical sample changer systems. The number of pellets to be evaluated can be highly reduced using the ^6LiF detector as a "flag" for neutron exposure.

Fig. 1 shows the TL readout ratio (80-170°C/170-250°C) of ^6LiF detector for albedo neutrons (PuBe) and gamma rays as a function of post-irradiation interval. The neutron exposure duration was 11 h and the gamma exposure duration was 10 min. It is seen that the low temperature to high temperature readout ratio is a little higher for neutrons as compared to gamma rays. The readout ratio for neutrons beyond the interval of 2 days could be fitted on a straight line $R = a - b \ln t_w$, in which R = readout ratio, $a = 0.278$, $b = 0.042$ and t_w = post-irradiation waiting time between 2 and 70 days. Thus, in principle if we know the readout ratio R , we can calculate the time of exposure (if the exposure is acute), and thus correct for the decay of ^{32}P . Unfortunately, for $t_w > 6$ days, the value of R decreases at a much lower rate than the activity of ^{32}P as shown in Fig. 1. Since the fading of low temperature TL peaks is relatively fast in the beginning, the effect of fluctuations in R ($\sigma = \pm 10\%$) on the uncertainty in correction factor for the ^{32}P decay increases with post-irradiation interval. This is shown in Fig. 2. A 10% error in readout ratio causes an uncertainty of 14% in the value of ^{32}P decay correction $e^{-\lambda t_w}$, if $R = 0.20$ and of 43% if $R = 0.14$. Table 2 gives the readout ratio R in ^6LiF for neutrons under different exposure conditions at CERN. This represents a practical situation and the exposure was more or less continuous during the monitoring period.

The gamma dose contribution is measured by the ^7LiF detector. For albedo neutron dose measurements the TL readout values of ^7LiF were subtracted from the ^6LiF readout values after taking into account the difference in inherent sensitivity of ^7LiF and ^6LiF , if any, to gamma rays.

For the calculation of R in ^6LiF the same procedure was adopted for both the readouts, i.e. 80-170°C and 170-250°C. The results in Table 2 shows that the decay correction obtained from the readout ratio using Fig. 2 differs still considerably in some cases from the calculated one. It should be mentioned as well that the value of R has been found to vary from batch to batch and from ^6LiF to ^7LiF , thereby increasing the uncertainty in calculating the value of R.

CONCLUSION

It appears that the use of the readout ratio technique described in this paper can in principle reduce the error related to unknown exposure history of activation detectors like sulphur pellets. However, it is shown in this paper that LiF does not have the most suitable fading characteristics for this purpose. Consequently, other neutron-sensitive TL materials should be studied since the technique is promising in view of the good sensitivity of the sulphur pellet technique for personal monitoring which has been overlooked so far.

REFERENCES

1. Tuyn, J.W.N., Radiation Protection Monitoring around High-Energy Proton Accelerators Using Thermoluminescence Dosemeters. Radiat. Prot. Dosim. 2(2) 69-74 (1982).
2. Griffith, R.V., Hankins, D.E., Gammage, R.B., Tommasino, L. and Wheeler, R.V. Recent Developments in Personnel Neutron Dosimeters - A Review. Health Phys. 36, 235-260 (1979).
3. Lakshmanan, A.R. A Review on the Role of Thermoluminescent Dosimeters in Fast Neutron Personnel Dosimetry. Nucl. Tracks 6(2/3) 59-78 (1982).
4. Reinhardt, P.W. and Davis, F.J. Improvements in the Threshold Detector Method of Fast Neutron Dosimetry. Health Phys. 1, 169-175 (1958).
5. Johnson, T.L. and Luersen, R.B. Fading of unannealed LiF (TLD-600) for Thermal Neutrons and γ -Rays. Health Phys. 38(5) 853-856 (1980).
6. Tymons, B.J. and Tuyn, J.W.N. Personnel Neutron Dosimetry by Means of Cellulose Nitrate Film Combined with LiF as Both Radiator and TLD. Health Phys. 32(6), 547-548 (1977).

Irradiation condition	Sensitivity of sulphur (counts/min-rem)	Sensitivity of ^6LiF <u>rad ^{60}Co equivalent</u> rem neutron dose
PuBe	13.5	0.67
PS bridge		
Position 1	4.5 ^{a)} , 8.3 ^{b)}	0.68 ^{a)}
Position 2	4.1 ^{a)} , 7.5 ^{b)}	0.70 ^{a)}
SC	1.3	7.40
PS access door	0.6	7.60

a) Fast neutrons + high-energy particles (>20 MeV) dose equivalent.

b) Fast neutrons only

Table 1 The sensitivity of a sulphur pellet (after sublimation of sulphur) and ^6LiF (after subtracting the gamma contribution) to neutrons for different irradiation conditions.

Irradiation condition	Irradiation duration (d)	Post-irradiation interval (d)	Readout ratio (R)	$\frac{\text{Decay correction using R}}{\text{Decay correction real}}$
PS access door	1	31	0.14	1.27
	6	26	0.16	1.76
	14	18	0.17	1.57
	26	6	0.20	1.78
SC	28	4	0.13	0.43
PS bridge				
Position 1	15	7	0.16	0.86
Position 2	15	7	0.16	0.86

Table 2 Readout ratio R in ^6LiF for neutrons under different exposure conditions.

Fig. 1 TL readout ratio $R[(80-170^\circ\text{C})/(170-250^\circ\text{C})]$ of ^6LiF detector for albedo neutrons ($\bullet$) and gamma rays ($\circ$) as a function of post-irradiation interval t_w in days. The decay factor $e^{-\lambda t_w}$ for ^{32}P activity (∇) is also shown as a function of t_w .

Fig. 2 TL readout ratio R in ${}^6\text{LiF}$ as a function of ${}^{32}\text{P}$ decay factor $e^{-\lambda t_w}$