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ABSTRACTTHE RESPONSE OF SELECTED THERMOLUMINESCENT
MATERIALS TO FAST-NEUTRON EXPOSURES

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The studies described in this paper were undertaken in an attempt to determine the optimum thermoluminescent dosimetry (TLD) material for personnel dosimetry in those facilities where significant fast-neutron fields may be present. Historically, the common methods applied to fast-neutron dosimetry have been the reading of nuclear track emulsions and timed-counting techniques using an active neutron detection instrument. Many studies have shown these methods to be distinctly inadequate for accurate personnel dosimetry.

The rapid advance of solid-state personnel dosimetry using TLD materials has been accompanied by a number of studies^(1,2) examining the response of certain TLD materials exposed to a fast-neutron field. This current study is an extension of the earlier work and is directed toward measuring the fast-neutron response of most of the common TLD materials when placed on a phantom, thus simulating actual personnel dosimetry conditions.

In the current study, the TLD materials used were LiF(TLD-100), LiF(TLD-600), LiF(TLD-700), $\text{Li}_2\text{B}_4\text{O}_7$, CaF_2 , and BeO in either pressed block or teflon matrix form. Exposures to fast neutrons were conducted using a Van de Graaff positive ion accelerator to produce

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monoenergetic neutrons in the energy range 0.020 to 5.0 MeV from the $T(p,n)^3\text{He}$, $D(d,n)^3\text{He}$, and $^{12}\text{C}(d,n)^{13}\text{N}$ reactions. Exposures were made in air with low-scatter conditions, with a polyethylene phantom placed behind the TLD material, and with cadmium and boron-10 shields surrounding the material in an attempt to evaluate the contribution of back-scattered neutrons. The standard used for dose measurement was the Hanford precision long-counter.

The results of the studies are given in the form energy response curves. Since the gamma response of most of the above materials has been investigated by other workers, it is reported only in cases of unusual interest.

At the present stage of study, it appears that the response of LiF(TLD-600) holds the greatest promise for neutron personnel dosimetry. Though most of the TLD-600 response is due to thermalized and backscattered neutrons, it appears possible to design a proper filter system to provide an accurate measure of fast neutron tissue-absorbed dose. The minimum detection limit for neutrons from a PuF_4 neutron source is less than 5 mrad using current readout equipment.

LiF(TLD-100) and $\text{Li}_2\text{B}_4\text{O}_7$ also show a significant response to fast neutrons when placed on a phantom. Both, however, are less sensitive than the TLD-600 material.

Studies are continuing to refine a personnel neutron dosimeter and to provide a gamma-exposure measurement using combinations of the various thermoluminescent materials.

REFERENCES

1. C. L. Wingate, E. Tochlin, and N. Goldstein. "Response of Lithium Fluoride to Neutrons and Charged Particles," Luminescence Dosimetry, USAEC, Oak Ridge, Tennessee, 1967. pp. 421-434.
2. G. W. R. Endres. "Thermoluminescence Dosimetry Studies at Hanford," ibid., pp. 435-443.

THE RESPONSE OF SELECTED THERMOLUMINESCENT MATERIALS TO FAST-NEUTRON EXPOSURES

INTRODUCTION

The problems associated with neutron dosimetry for radiation workers have been apparent for many years. Activation-foil techniques used with sensitive film, nuclear track emulsions, and timed-counting technique using active detectors are some of the systems used in the past to measure neutron dose. The rapid developments in the field of thermoluminescence *documented by R. L. Bittles - Tenthredinid. To study the possibilities* of using some of the new TLD materials for fast-neutron dose measurements.

Earlier studies of the fast-neutron response of TLD materials^(1,2) were limited to one or two materials only and generally were not conducted on suitable phantoms to simulate backscatter from the human body. These studies attempt to extend the earlier work and provide increased understanding of the neutron-energy response of several TLD materials.

THEORY

Generally, the neutron activation cross section of the selected TLD materials decreases as the incident neutron energy increases. To provide efficient detection of neutrons, it is many times necessary to moderate the neutrons before detection. Since the human body provides this function for the purpose of personnel neutron dosimetry, it may be possible to devise a simple system to provide reasonably accurate measurements.

At some nuclear research laboratories, TLD materials are being used in this manner, notably $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$. To determine the accuracies, one can expect from such systems the response of TLD materials was

measured as a function of incident-neutron energy and shielding material. By using cadmium and ⁶rhoron-10 shields, some information can be gained on the relative amounts of activation due to thermal, epithermal, and fast neutrons.

EQUIPMENT

Two thermoluminescence analyzers are available to us at Battelle-Northwest. A Harshaw Model 2000 reader was recently purchased and was used for a portion of the measurements in this series. Most of the work was accomplished using a Conrad TL analyzer modified to readout through a dymec analog to digital converter. ~~The performance of these systems has been generally quite satisfactory. One of our main problems appear to be non-uniform dosimeters.~~

The dosimeters used for the study were TLD-100, TLD-200, TLD-600, TLD-700 ribbon in the form of 1/8 x 1/8 x .035 in. blocks with about 25 mg of phosphor and Li₂B₄O₇:Mn in teflon in the form of 1/2 inch discs. Glow curves were taken in most cases to assure proper readout of the dosimeters, though a few low readings were obtained despite the precautions.

Fast-neutron exposures were conducted using a 2.0 MeV Van de Graaff positive ion accelerator to produce monoenergetic neutrons in the energy range from 0.02 MeV to 5.0 MeV. These neutrons were produced with T(p,n)³He, D(d,n)³He, and ¹²C(d,n)¹³N reactions. The Hanford precision long-counter was used as the standard for dose measurements.

EXPERIMENT

Within limits of time convenient exposures to doses between 20 mrad and 1.0 rads were conducted. The amount of time required depends on the neutron yield from the targets. With a Van de Graaff of the type at Battelle-Northwest, the yield decreases as the bombarding energy decreases; even ^{though} the higher ion current is used. Fortunately, the response of the TLD materials increases as the neutron energy decreases so that a high exposure is not necessary at ^{lower} neutron energies.

The exposures were conducted using sets of three dosimeters to provide a reasonable statistical average of the response. All dosimeters used in a given response study were from the same batch to be sure that the relative response as a function of energy is accurate.

Five different exposure conditions were used in the response studies:

- (1) Air geometry with no shielding - bare air
- (2) Air geometry with B-10 shielding - B-10-air.
- (3) Backscatter geometry with no shielding - Bare-moderator
- (4) Backscatter geometry with cadmium shielding - Cadmium-moderator
- (5) Backscatter geometry with ^BB-10 shielding - B-10-moderator

The cadmium used was 0.020 inches thick and was placed on both sides of the dosimeters as they were exposed in front of the moderator-phantom. The B-10 spheres were 1 1/4" diameter and provided 1 cm thickness of

boron-10 in all directions around the dosimeter placed inside the sphere. The moderator-phantom was a 9 inch diameter cylinder of polyethylene with one area cut off to provide a flat surface to face the neutron beam.

Exposures of $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ were conducted in the conditions given above and read out several days later, so some decay of the TL response may have occurred. All dosimeters were treated in the same way. At reasonable exposure levels, the $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ discs did not have a positive response when exposed in air with no moderator present to provide backscattered neutrons even (for an exposure greater than 1.0 rad at 1.0 MeV.) The response of these discs was positive when placed in front of the moderator-phantom for both bare and cadmium covered cases. No response was evident when using the boron-19 shield in front of the moderator. The response of the $\text{Li}_2\text{B}_4\text{O}_7$ discs exposed in bare geometry was an order of magnitude higher than that of the discs exposed inside the cadmium cover despite the large number of cadmium capture gammas present (as shown in Figure 4.) The response is given in terms of net reader counts per rad. Apparently, 90 per cent or more of the response of $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ is due to backscattered thermal neutrons. Relatively few epithermal neutrons are present, at least for the conditions studied here.

The TLD-100 ribbon showed a response to all exposure levels and conditions studied. The response in all geometries increases as the incident neutron energy decreases, as shown in Figure 5. [All data

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are given in terms of net reader counts per rad with the reader calibrated to 2 units per mR radium γ for TLD-700 ribbon.] Again, the most sensitive condition of exposure is the case with the bare dosimeters placed on the moderator-phantom. The response inside the cadmium shield is about 25% of the bare response, and the response inside the boron-10 sphere is about 10% of the bare response. The gamma response of the TLD material undoubtedly contributes some to the total response of the dosimeters. The gamma yield directly from the target used to produce the neutrons for the exposures is quite small, not more than 10 mR for the high energy exposures. A 10 mR exposure will yield a response of 20 reader units, which is a small fraction of the total response obtained from the TLD materials. So the gamma contribution to the response should be small except for ^{capture} ~~exposure~~ gammas in the cadmium shield and in the moderator. All the response curves have the same general shape for TLD-100.

The TLD-600 material is extremely sensitive to neutrons, with a very high reading even for the 1.0 MeV exposures. See Figure 6. The response in air with no backscatter is higher than for similar measurements for any of the other TLD materials used. TLD-600 is 95.62 per cent lithium-6, which has a high cross section for neutron interactions, mostly (n, α) reactions.

The response of TLD-600 varies by a factor of about 2.5 over the range 0.20 MeV to 1.0 MeV for the bare-moderator case. It appears for

the materials with high neutron cross sections, the bare-moderator condition yields the highest results with the cadmium shielded condition the next most sensitive. These two conditions make maximum use of the backscattered neutrons.

The results of exposures using TLD-700 material are shown in Figure 7. TLD-700 is composed of 99.993 per cent lithium-7 and so should be relatively insensitive to the backscattered neutrons and incident neutrons also. Both response curves obtained using bare dosimeters show a low response. The situation is somewhat reversed over the results obtained using TLD-600. Now the cadmium shielded dosimeters show the highest response, presumably due to the capture gammas in the cadmium.

The results of the exposures using TLD-200 ($\text{CaF}_2:\text{Dy}$) are similar to those using TLD-700 as shown in Figure 8. Again the cadmium shielded condition yields the highest response to fast neutrons. The bare-moderator condition is next in sensitivity. The TLD-200 is about 30 times as sensitive to gamma radiation as is TLD-100 and so should be very sensitive to cadmium capture gamma radiation.

In searching for possible combinations of TLD materials to use for neutron dosimetry, the most logical materials seem to be TLD-100, TLD-600, and TLD-700. As shown above, the response of TLD-600 ^{in the bare-moderator condition} varies by a factor of 2.5 from 1.0 MeV to 0.20 MeV. The response of TLD-700 ^{for the same exposure condition} varies by more than a factor of 25 over the same energy range. (See Figure 9.) It appears possible to use a combination of TLD-600 and -700 to provide a measure of the average incident neutron energy.

To look at the energy variation in response a little differently, the ratios of the TLD-600 to TLD-700 and the TLD-100 to TLD-700 were calculated as a function of energy. The 600 to 700 ratio varies from about 10 at 0.20 MeV to about 100 at 1.0 MeV. The 100 to 700 ratio varies from about 5 at 0.20 MeV to about 23 at 1.0 MeV. These results are shown in Figure 10.

CONCLUSIONS

Although the exposures reported were conducted in an essentially gamma free field, the responses indicate that a satisfactory neutron dosimetry system may be possible, if a proper evaluation of an incident gamma field can be made. TLD-600 is slightly less sensitive to gamma radiation than is TLD-700; this complicates the evaluation somewhat.

FIG 4

$\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ TEFLON

$E_n(\text{MeV}) \rightarrow$

RESPONSE OF $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ TO FAST NEUTRONS

Fig 5

TLD-100

$E_N(\text{MeV}) \longrightarrow$

RESPONSE OF TLD-100 TO FAST NEUTRONS

Fig - C

TLD-G00 - 8078

RESPONSE OF TLD-G00 TO FAST NEUTRONS

Fig 7
TLD-700

CTS
RAD

LOGARITHMIC 46 7400
3 X 3 CYCLES
MADE IN U.S.A.
KEUFFEL & ESSER CO.

RESPONSE OF TLD-700 TO FAST NEUTRONS

Fig 8

TLD-200 - $\text{CaF}_2:\text{Dy}$

CT5
RAD

46 7400
MADE IN U.S.A.

LOGARITHMIC
3 X 3 CYCLES
KEUFFEL & ESSER CO.

RESPONSE OF TLD-200 TO FAST NEUTRONS

FIG 9

ENERGY RESPONSE OF TLD-600, TLD-100 AND TLD-700 ON MODERATORIZ -
PHANTOM - NO SHIELDING.

FIG 10