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THE RESPONSE OF SELECTED
THERMOLUMINESCENT MATERIALS
TO FAST NEUTRON EXPOSURES

July 1969

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THE RESPONSE OF SELECTED THERMOLUMINESCENT
MATERIALS TO FAST NEUTRON EXPOSURES

by

G. W. R. Endres and L. F. Kocher

Radiological Sciences Department
Environmental and Life Sciences Division

July 1969

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ABSTRACT

This current study, an extension of the earlier work, is directed toward measuring the fast neutron response of most of the common TLD materials when placed on a polyethylene phantom to simulate actual personnel dosimetry conditions. The TLD materials used were LiF(TLD-100), LiF(TLD-600), LiF(TLD-700), $\text{Li}_2\text{B}_4\text{O}_7$, and CaF_2 in either pressed block or teflon matrix form. Exposures to fast neutrons were conducted using a Van de Graaff positive ion accelerator to produce monoenergetic neutrons in the energy range 0.2 to 1.0 MeV from the $\text{T(p,n)}^3\text{He}$ reactions. Exposures were made in air with low-scatter conditions. A phantom was placed behind the TLD material, and cadmium and boron-10 shields surrounded it for the purpose of evaluating the contribution of backscattered neutrons. The standard used for dose measurement was the Hanford precision long-counter. LiF(TLD-600) presently appears to hold the greatest promise for neutron personnel dosimetry. Though most of the TLD-600 response is due to thermalized and backscattered neutrons, design of a proper filter system to provide an accurate measure of fast neutron tissue-absorbed dose is considered possible. 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.

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INTRODUCTION

The problems associated with neutron dosimetry for personnel exposed to radiation environments have been apparent for many years. Activation-foil techniques used with sensitive film, nuclear track emulsions, and timed-counting techniques using active detectors are among systems already employed in measuring neutron dose. Rapid developments in the field of thermoluminescence (TLD) dosimetry have prompted the study of new TLD materials for possible use in measuring fast neutron doses.

Earlier studies of the fast-neutron response of TLD materials^(1,2,3) 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, moderation of the neutrons before detection often becomes necessary. Since the human body provides moderation of neutrons before detection, 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, the response of TLD materials was measured as a function of incident-neutron energy and shielding material. The use of cadmium and

boron-10 shields assisted in obtaining information on the amounts of activation due to thermal, epithermal, and fast neutrons.

EQUIPMENT

Two thermoluminescence analyzers are available for use in the laboratory investigation. A Harshaw Model 2000 Reader, recently purchased and used for a portion of the measurements in this series, is shown in Figure 1. Most of the work was accomplished with a Conrad TL Analyzer, modified to readout through a dymec analog to digital converter (see Figure 2).

The dosimeters used for the study were TLD-100, TLD-200, TLD-600, and TLD-700 ribbon in the form of $1/8 \times 1/8 \times 0.035$ in. blocks, with about 25 mg of phosphor and $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ in teflon in the form of $1/2$ in. discs. Glow curves were taken for all readings as a precautionary measure to assure proper readout of the dosimeters.

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 0.2 MeV to 1.0 MeV (see Figure 3). These neutrons were produced with $\text{T}(\text{p},\text{n})^3\text{He}$ reactions. The Hanford precision long-counter was used as the standard.

EXPERIMENT

Within limits of time, convenient exposures to kerma between 20 mrad and 1.0 rad were conducted. The amount of time required was dependent upon the neutron yield from the targets. With a Van de Graaff of the type at Battelle-Northwest, the yield decreased as the bombarding energy decreased despite an increase in ion current. Fortunately, because the response of the TLD materials increased as the neutron energy decreased, a high exposure was 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

FIGURE 1. Harshaw Reader System

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FIGURE 2. Conrad Reader System

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FIGURE 3. Van de Graaff Accelerator

dosimeters used in a given response study were obtained from the same batch to assure accuracy of the relative response as a function of energy. Neutrons in the energy range from 0.20 MeV to 1.0 MeV were used for exposures.

Five different exposure conditions were used in the response studies:

- Air geometry with no shielding, bare air.
- Air geometry with B-10 shielding, B-10 air.
- Backscatter geometry with no shielding, bare moderator.
- Backscatter geometry with cadmium shielding, cadmium moderator.
- Backscatter geometry with B-10 shielding, B-10 moderator.

The cadmium used was 0.020 in. thick and was placed on both sides of the dosimeters as they were exposed in front of the moderator-phantom. The B-10 spheres, 1.25 in. in diameter, provided 1 cm thickness of boron-10 in all directions around the dosimeter placed inside the sphere. The moderator-phantom was a 9-in. diameter cylinder of polyethylene with one area cut off to provide a flat surface to face the neutron beam.

All data reported were obtained in terms of net reader counts per rad, with the reader calibrated to 2 units per mR radium γ for TLD-700 ribbon. $\text{Li}_2\text{B}_4\text{O}_7\text{:Mn}$ was exposed under these conditions and read out several days later, so some decay of the TL response may have occurred. All dosimeters were treated in the same way. The $\text{Li}_2\text{B}_4\text{O}_7\text{:Mn}$ discs did not respond positively for an exposure greater than 1.0 rad at 1.0 MeV when exposed in air with no moderator present to provide backscattered neutrons. 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-10 shield in front of the moderator. As shown in Figure 4, 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

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FIGURE 4. Response of $\text{Li}_2\text{B}_4\text{O}_7$

the large number of cadmium capture gammas present. The response was indicated in terms of net reader counts per rad. Apparently, 90% or more of the response of $\text{Li}_2\text{B}_4\text{O}_7\text{:Mn}$ was due to backscattered thermal neutrons. Relatively few epithermal neutrons were 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 increased as the incident neutron energy decreased, as shown in Figure 5. The most sensitive condition of exposure was obtained by placing the bare dosimeters on the moderator-phantom. The response inside the cadmium shield was about 25% of the bare response, and the response inside the boron-10 sphere was about 10% of the bare response. The gamma response of the TLD material undoubtedly contributed some to the total response of the dosimeters. The gamma yield directly from the target, used to produce the neutrons for the exposures was quite small, not more than 10 mR total 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. The gamma contribution to the response, therefore, should be small except for capture 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, extremely sensitive to neutrons, displayed very high readings even for the 1.0 MeV exposures (Figure 6). The response in air with no backscatter was higher than for similar measurements of the other TLD materials used. TLD-600 is 95.62% lithium-6, which has a high cross section for neutron interactions, mostly (n,α) reactions.

The response of TLD-600 varied by a factor of about 2.5 over the range 0.20 MeV to 1.0 MeV for the bare-moderator case. For the materials with high neutron cross sections, the bare-moderator condition appeared to yield the highest results, with

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FIGURE 5. Response of TLD-100

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FIGURE 6. Response of TLD-600

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% lithium-7 and so should be relatively insensitive to the backscattered neutrons and incident neutrons also. Both response curves obtained using bare dosimeters showed a low response. The situation was somewhat reversed over the results obtained using TLD-600. The highest response was shown by the cadmium shielded dosimeters, presumably due to the capture gammas in the cadmium.

The results of the exposures using TLD-200 ($\text{CaF}_2\text{:Dy}$), as shown in Figure 8, were similar to those using TLD-700. Again the cadmium-shielded condition yielded the highest response to fast neutrons. The bare-moderator condition was next in sensitivity. The TLD-200 was about 30 times as sensitive to gamma radiation as 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 appeared to be TLD-100, TLD-600, and TLD-700. As previously reported, the response of TLD-600 in the bare-moderator condition varied by a factor of 2.5 from 1.0 MeV to 0.20 MeV. The response of TLD-700 varied by more than a factor of 25 over the same energy range for the same exposure condition (Figure 9). A combination of TLD-600 and -700 to provide a measure of the average incident neutron energy was indicated to be possible.

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 varied from about 10 at 0.20 MeV to about 100 at 1.0 MeV. The 100 to 700 ratio varied from about 5 at 0.20 MeV to about 23 at 1.0 MeV. These results are shown in Figure 10.

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FIGURE 7. Response of TLD-700

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FIGURE 8. Response of TLD-200

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FIGURE 9. Response of TLD-100, TLD-600, and TLD-700 in Bare-Moderator Condition

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FIGURE 10. Variation in Ratios TLD-600/TLD-700, and
TLD-100/TLD-700 as Function of Neutron Energy

Glow curves were taken continuously while reading out the dosimeters to assure total readout or rejection of the data where proper readout was not obtained. The standard deviations of the data points shown in Figures 4 through 10 were $\pm 10\%$ or less, based on the readings. This calculation did not include errors in the long counter measurements, shown by Nichols⁽⁴⁾ to be as much as $\pm 10\%$ for the neutron energies and experimental geometries used here. Even though the total errors in the experiment may be 20%, the curves provided useful information about the neutron response of the various TLD materials.

CONCLUSIONS

Although the exposures reported were conducted in an essentially gamma-free field, the response indicates that a satisfactory neutron dosimetry system may be possible where an incident gamma field can be properly evaluated. The fact that TLD-600 is slightly less sensitive to gamma radiation than is TLD-700 complicates the evaluation somewhat. This work is continuing with emphasis on extending the neutron energy range of the measurements and on developing a suitable gamma filter system.

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