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DOSIMETRY TECHNOLOGY STUDIES
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By

Radiological Physics Staff
Radiological Sciences Department
Environmental Health and Engineering Division

September 1969

BATTELLE MEMORIAL INSTITUTE
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DOSIMETRY TECHNOLOGY STUDIES

1968

ACTIVATION OF BODY SODIUM FOLLOWING LOW NEUTRON EXPOSURES

D. McConnon

INTRODUCTION

Upon exposure to a source of neutrons of appropriate energy, a fraction of the stable sodium in the body is activated to form ^{24}Na through the reaction $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$. Sodium-24 is a high energy gamma emitter that decays with a 15-hr half-life. Since the amount of ^{24}Na activation is proportional to the neutron dose, a convenient method for estimating neutron dose, provided the spectrum is known, is by measuring the ^{24}Na content of the body following exposure. This technique is commonly used for estimating personnel doses following accidental exposure to a nuclear critical excursion.⁽¹⁻³⁾

Palmer, et al.,⁽⁴⁾ have shown that ^{24}Na is measurable in human cadavers and phantoms even after exposure to relatively low levels of fast neutrons. This fact suggested the possibility that induced ^{24}Na may even be measurable in certain Battelle-Northwest personnel who receive very low neutron doses during the performance of their normal duties.

EXPERIMENT

A sensitive whole body counter was built and calibrated for ^{24}Na measurements specifically for this study. The counter consists of three NaI(Tl) crystals 9 3/8 in. in diam by 4 in. thick positioned at specified locations over the length of the subject's body. The summed output from the crystals are sorted and stored in a multi-channel analyzer. Calibration of the counter was completed with volunteers who ingested known quantities of ^{24}Na . The counter has a sensitivity of 24 c/m/nCi ^{24}Na in the energy range of 2.54 to 2.84 MeV and a

detection limit at the 99% confidence level of 0.6 nCi ^{24}Na for a 20-min counting period.

The subjects were employees required to enter the process cell of the Plutonium Recycle Test Reactor to perform various functions while the reactor was operating. Measured fast neutron dose equivalent rates in areas entered by personnel ranged from 0.01 to 1.0 rem/hr. In addition to their routine radiation dosimeters, personnel wore fission fragment dosimeters⁽⁵⁾ from which estimates of neutron doses were made. Examinations in the whole-body counters were begun soon after personnel exited from the process cell. A background was obtained for each subject two weeks after exposure to allow complete decay of induced ^{24}Na .

Measurements of induced ^{24}Na activity and estimated neutron doses are shown in Table 1 for five subjects. These five measurements were made prior to reactor shutdown for the remainder of the year for major repairs and maintenance. While the number of measurements are insufficient to indicate any definite correlation between neutron exposure and induced ^{24}Na activity, they do indicate the feasibility of estimating neutron dose equivalents even down to 5 mrem by measuring the induced ^{24}Na activity. This method is rather time consuming for routine use, but may be applicable in instances where appreciable neutron exposures, although suspected, cannot be measured either because dosimeters are not worn or cannot be evaluated.

TABLE 1. *Induced ^{24}Na Activity Following Low Neutron Doses*

Subject No.	Induced ^{24}Na - nCi	Measured Dose Equivalent, mrem
1	0.96 ± 0.09	9
2	1.2 ± 0.10	5
3	1.8 ± 0.12	10
4	3.0 ± 0.20	7
5	3.5 ± 0.22	11

DETERMINATION OF DOSE DISTRIBUTION
FOLLOWING NON-UNIFORM EXPOSURE TO A CRITICAL ASSEMBLY

D. McConnon

INTRODUCTION

The common method of estimating neutron dose received by personnel in a criticality accident is by measuring total body or blood ^{24}Na activation and relating the observed activity to dose. This relationship has been the subject of a number of studies.^(1-3,6)

THEORY

In most circumstances, the neutron dose will not be uniform over the surface of the body because of orientation of the subject to the source and the intervening shielding. The sodium activation method of estimating neutron dose will tend to average the dose because of mixing of ^{24}Na in body fluids. Attempts have been made to estimate non-uniform doses by determining ^{32}P activation in body hair resulting from the $^{32}\text{S}(n,p)^{32}\text{P}$ reaction.⁽⁷⁾ Unfortunately, this reaction has a neutron threshold energy of 2.5 MeV and, therefore, provides a dose estimate involving only a portion of the neutron spectrum. Another method of estimating neutron dose distribution is by means of foil dosimeters attached to a belt or sewn into protective clothing.^(8,9) This method is feasible, but may suffer from problems of a practical nature. Metallic objects carried by the exposed person may also be used for this estimate, but there is no guarantee such objects will be available.

Studies by Palmer, et al.⁽¹⁰⁾ indicate the activation of other body elements besides sodium at relatively low neutron doses. A study was undertaken to determine the feasibility of estimating neutron dose distribution over the body by measuring the gamma activity of induced radionuclides

which remain fixed at the point of formation. The most promising radionuclide for this purpose appeared to be ^{49}Ca formed by the $^{48}\text{Ca}(n,\gamma)^{49}\text{Ca}$ reaction. Since about 99% of the body's calcium is fixed in bone, the induced ^{49}Ca should remain at the point of formation and provide a good indication of dose distribution. However, this radionuclide has such a short half-life (8.8 min) that counting must be done soon after exposure by a relatively simple and rapid method.

EXPERIMENT

A REMAB phantom filled with a tissue equivalent solution and containing a full skeleton was used for the study and the University of Washington cyclotron facility was used as the source of fast neutrons. Following irradiation, the phantom was scanned with a collimated 9-3/8 in. by 4 in. thick NaI(Tl) crystal detector, single channel analyzer, and count-rate meter system. The discriminators of the single channel analyzer were so adjusted that only pulses from the 3.1 MeV photons associated with the decay of ^{49}Ca would be accepted. Fission fragment dosimeters⁽⁵⁾ were placed at various points on the surface of the phantom to estimate the neutron dose distribution.

In order to provide a reference irradiation involving uniform exposure, the phantom was exposed facing and approximately 3 meters distance from the cyclotron target. Dosimeters indicated the dose distribution over the surface of the phantom to be uniform to within $\pm 10\%$. Following irradiation, the phantom was immediately scanned with the results shown in Figure 1. Increases in the relative activity are evident at regions of large bone mass such as the chest, hips, knees and feet. Also shown in Figure 1 is the scan observed following non-uniform irradiation of the phantom. Although the head region is evident in the case of non-uniform irradiation, the increases noted in other areas of the phantom following

FIGURE 1. Relative ^{49}Ca Activation Following Uniform and Non-uniform Irradiation of a REMAB Phantom with Fast Neutrons

uniform irradiation are missing. In the case of non-uniform irradiation, the relative activity is depressed in the lower portions of the body as expected.

These results demonstrate that the dose distribution following non-uniform exposure may be estimated by measuring the relative induced ^{49}Ca activity. However, because of the short half-life of ^{49}Ca , these measurements must be made within about 1 hr following exposure. Because it may not be practical to perform these measurements on such short notice, the search for other induced radionuclides remaining at the point of formation and of relatively long half-life will be continued.

IN VIVO MEASUREMENTS OF GAMMA EMITTERS
WITH A SOLID STATE DETECTOR*

D. McConnon

In vivo measurements of a complex mixture of gamma emitters with conventional whole body or organ counting systems often produce unsatisfactory results because of the poor resolution of the counting system. This problem might be eliminated by the use of a counting system employing a high resolution solid state detector. Such a system was therefore studied to determine its suitability as an organ counter. The response of a dual crystal Ge(Li) detector to gamma emitters contained in various organ compartments of an anthropomorphic phantom was measured. The study indicated that even though its sensitivity is poor relative to that of conventional organ counting systems, the solid state system may usefully supplement in vivo measurements provided the deposition in an organ is sufficiently high and the background and counting geometry are controlled.

*BNWL forthcoming.

A TISSUE EQUIVALENT PROPORTIONAL COUNTER
FOR MEASURING DOSES DUE TO NEUTRON RADIATION

G. W. R. Endres, W. A. Glass, and A. J. Haverfield

INTRODUCTION

Several methods have been used in the past to measure the intensity of neutron radiation fields and relate this information to absorbed dose in tissue or tissue-equivalent material. These techniques (including nuclear track emulsions, moderated BF_3 detectors, and activation foil systems) have, in general, proven inadequate in some phase of operation. A proportional counter developed by Hurst⁽¹⁰⁻¹³⁾ has been used as an absolute neutron dosimeter. This system uses a pulse height discriminator to reject gamma interference.

The approach used in the work to be reported here is one of direct measurement of absorbed dose accomplished with a tissue-equivalent proportional counter (TEPC). The techniques of using the TEPC to measure energy deposited in tissue-like materials have been developed by Rossi⁽¹⁴⁾ and others, and later by Glass⁽¹⁵⁾ at Battelle-Northwest.

THEORY

The technique involved is essentially a study of the manner in which radiation deposits energy in matter. Matter, as used in these experiments, more specifically denotes a spherical site of tissue-like material simulated by the proportional counter. The proportional counter consists of a sphere of tissue-equivalent (TE) plastic surrounding a cavity filled with TE gas. These materials are described in a later section of this report.

The counter produces a spectrum of pulses corresponding to the spectrum of energy losses in the cavity. The cavity can be made to simulate tissue spheres down to a few tenths of a micron in diameter by adjusting gas pressure. At these

low pressures, the differences between energies deposited by gamma radiation secondaries (electrons) and neutron secondaries (protons) can be observed. For some exposure conditions, the absorbed dose due to gamma radiation can be separated from the absorbed dose due to neutron radiation. The maximum rate of energy a secondary electron can lose is 1-5 keV/ μ compared to the maximum of about 100 keV/ μ for secondary protons. In other words, a proton will deposit its energy in a short distance at a relatively high rate while an electron will travel much farther in depositing its energy at a rate as much as 100 times less.

When a cavity is placed in a solid such as TE plastic and filled with TE gas, the cavity itself does not alter the fluence of charged particles if the irradiation is uniform. The spectrum of energy deposited in the cavity by individual events will show the same range of energies with the same relative frequency, as compared to events in a similarly shaped solid, the dimensions of which are reduced by the ratio of the densities of the gas and solid. The absolute frequency of events in the gas cavity is larger by a factor inversely proportional to the square of the density of the gas relative to the solid.⁽¹⁶⁾

The event size Y is defined as the energy laid down in an individual event divided by the sphere diameter. The diameter is determined by the gas pressure inside the counter. In determining the spectrum of energy losses, the counter diameter is assumed small compared to the range of the charged particles. If this condition is met, the total energy deposited in an event is approximately proportional to counter diameter. An event is produced by the passage through the counter of a charged particle and its secondaries. Event size may be expressed in units of keV/ μ , which makes it a quantity nearly independent of counter size. For a good general discussion

of the energy loss techniques, the reader is directed to an excellent review article by Rossi.⁽¹⁷⁾

EXPERIMENTAL EQUIPMENT

Two types of sealed TE proportional counters, as well as a gas flow counter, were used to obtain data in a variety of radiation environments. All three types of counters used as the detector wall a sphere of TE plastic 2 in. in diam and 1/8 in. in thickness. The composition of TE plastic is shown in Table 2, along with the composition of standard man and tissue muscle.⁽¹⁸⁾

TABLE 2. *Composition of Tissue-Like Materials*

Element	Standard Man, wt%	Tissue Muscle, wt%	TE Plastic, wt%
H	10	10	10
C	18	12	76
N	3	3	3
O	65	73	5.2

Although the amounts of carbon and oxygen are essentially reversed in the TE plastic, the dose to the plastic is still very close to the dose to tissue because the carbon neutron cross section is only slightly higher than the oxygen cross section. This difference represents, at the most, a 5% error in tissue dose measurements.

For the data reported here, the gas pressure in the counters was adjusted to give an equivalent sphere size of 2 μ . For a 2-in. sphere, this pressure is 28 Torr absolute. The composition of the gas used is approximately 32.4% CO₂, 3.2% N₂, and 64.4% CH₄.

For one type of sealed counter and the gas flow counter, a single 0.001-in. stainless steel wire was used as the center electrode. This arrangement is similar to a device designed by Benjamin⁽¹⁹⁾ for proton recoil spectrum measurements. The

other type of sealed counter used a single 0.005-in. stainless steel wire as a center electrode and a spiral grid around the center electrode to provide a more constant gas gain along the length of the electrode. The spiral grid is maintained at a potential of 20% of that on the center electrode.

The electronics system used with the counter consisted of a charge sensitive preamplifier, an RC singly differentiated linear amplifier, a high voltage supply, and a multichannel analyzer. Electrical connections between the counter and preamplifier were kept as short as possible to reduce system noise. Integral and differential time constants in the linear amplifier were adjusted for 3.2 μ sec to allow charge collection in the counter. Two different multichannel analyzers were used during the course of this work.

MEASUREMENTS

Experimental measurements of energy loss spectra were conducted in fields of monoenergetic neutrons generated by a 2.0 MeV positive ion Van de Graaff accelerator. This machine is capable of producing monoenergetic neutrons with energies as low as 0.020 MeV to as high as 18.0 MeV. Most of the neutron exposures conducted with the accelerator made use of the tritium(p,n) reaction which yields monoenergetic neutrons with energies from about 1.0 MeV down, depending upon bombarding energy and the target to detector angle. Other reactions used were deuterium(d,n), and beryllium(d,n).

A number of exposures in mixed gamma-neutron radiation fields and gamma only fields were conducted to help define the differences in counter response to gamma and neutron radiations. These radiation fields were produced by a PuF_4 isotopic source, a PuBe isotopic source, and the Plutonium Recycle Test Reactor (PRTR). The PuF_4 source has an average neutron energy of about 0.85 MeV with gamma radiation from plutonium and americium-241.

The source contains 1 kg of PuF_4 with a yield of about 5.0×10^6 n/sec. The PuBe source, containing 80 g of plutonium and 40 g of beryllium, yields 9.5×10^6 n/sec, also mixed with gamma radiation from plutonium and americium. Exposures near the PRTR were conducted in a mixed neutron-gamma field serving essentially as a leakage spectrum from the heavy water moderated reactor.

EXPERIMENTAL RESULTS

A number of exposures to monoenergetic neutrons were conducted to determine the response of the TEPC to neutrons for system calibration and resolution. Calibration is accomplished by noting the very rapid drop in the spectrum at just under 100 keV/ μ . This spectrum drop is called the proton drop and corresponds to the maximum energy loss produced in the counter by proton recoils. The sharpness of the proton drop is related to the resolution of the counter system. A typical energy loss spectrum is shown in Figure 2 for 0.73 MeV monoenergetic neutrons. The pressure in the counter was equivalent to a 2 μ tissue site. The maximum proton energy loss, ~ 96 keV/ μ for this case, corresponds to the rather sharp drop in counts just above Channel 200 in this figure.

To determine the dose distribution in event size, the number of events in each energy interval must be multiplied by the event size of that interval and normalized to unit area. The event size per channel is determined by eye at the midpoint of the sharply descending portion of the curve in the proton drop region. A computer program has been written⁽²⁰⁾ to fit the data in this region and to determine the calibration point from the fitted curve. A typical dose distribution curve as a function of event size is shown in Figure 3 for 0.73 MeV incident neutrons. Reproducibility of

FIGURE 2. Raw Data Spectrum, 0.73 MeV Neutrons

FIGURE 3. Dose Distribution in Event Size - 0.73 MeV Neutrons

the sealed counter with spiral grid and electronics system was checked over a period of three weeks and compared with kerma derived from fluence measurements taken with a long counter and fluence to kerma constants reported by Caswell at NBS. These data are shown in Table 3.

TABLE 3. Comparison of TEPC and Precision Long Counter Measurements

<u>Dose</u> TEPC, mrads	<u>Kerma</u> PLC, mrads	<u>Proton Drop;</u> <u>Channel No.</u>
46.0	51.8	200
45.1	51.8	225
45.1	51.8	220
43.2	41.8	218
32.3	34.6	220
31.6	34.6	223

The proton drop point held constant to within $\pm 5\%$. The counter was evacuated and filled each morning to a pressure of 28 Torr absolute.

To observe the system response as a function of incident neutron energy, exposures were made to neutrons with energies of 0.20 MeV, 0.48 MeV, 1.38 MeV, and 4.0 MeV by again using the Van de Graaff accelerator. Dose distribution curves in event size for these exposures are shown in Figures 4-8. As the neutron energy increases, more events occur at the lower event sizes because proton recoils of high energy (<1 MeV) display a lower rate of energy loss than do those of lower energy (>1 MeV). The exposure at 1.38 MeV included several mR of gamma radiation from the carbon (d,n) reaction used to produce the neutrons. It appears possible from this case to separate the absorbed dose due to neutron interactions and the absorbed dose due to gamma interactions without making a significant error in the neutron dose measurements. With 4.0 MeV incident neutrons, the contribution of heavy ion recoils

FIGURE 4. Dose Distribution in Event Size - 0.20 MeV Neutrons

FIGURE 5. Dose Distribution in Event Size - 0.48 MeV Neutrons

FIGURE 6. Dose Distribution in Event Size - 1.38 MeV Neutrons

FIGURE 7. Dose Distribution in Event Size - 4.0 MeV Neutrons

FIGURE 8. Dose Distribution in Event Size - 4.0 MeV Neutrons

becomes important as indicated by the greater number of events occurring above the proton drop point.

A comparison of the measured spectra taken with two different proportional counters was made using 0.73 MeV neutrons. Resolution of the counter with the center single 0.001-in. wire electrode did not appear as good as the counter with the spiral grid (labeled TEPC-127). The results are shown in Figure 9.

Comparisons of kerma as measured with a long counter, and absorbed dose as measured by a tissue equivalent ionization chamber⁽²¹⁾ and the TEPC were made at energies of 0.25 MeV, 1.0 MeV, and 4.0 MeV. Table 4 shows these results.

TABLE 4. Comparison of Ion Chamber, TEPC, and Long Counter Measurements

Neutron Energy MeV	LC/IC	LC/TEPC
0.25	1.43	1.37
0.1	1.20	1.19
4.0	1.12	1.10

At least for these measurements, the ion chamber and the TEPC demonstrate the same behavior relative to the long counter. The ratios are not equal to unity partly because of the response of the long counter in the particular neutron field produced by the accelerator.⁽²²⁾

The long counter does not respond properly when placed in a non-isotopic neutron field. The response may be high by as much as 30% when used at certain angles with the D(d,n) reaction. Thus, the agreement between the TEPC and the ion chamber is more important than the apparent disagreement with the long counter.

FIGURE 9. Dose Distribution in Event Size - 0.73 MeV Neutrons, Two Types of Counters

Event spectra were measured in a mixed gamma-neutron radiation field generated by a PuF_4 isotopic source. The results are shown in Figure 10. The TEPC with the spiral grid (TEPC-127) provides a wider separation between the pulses produced by gamma secondaries and neutron secondaries than does the counter without spiral grid. As in the previous case of the carbon(d,n) reaction, it appears possible to discriminate against the gamma produced secondaries without making a significant error in neutron dose measurement. The absorbed dose from the PuF_4 source, as measured by the TEPC, is about 50% high when compared with long counter measurements because of the large amount of gamma radiation present. When corrected for the gamma dose, the TEPC and long counter agree within 15%.

Some additional measurements were taken in a radiation field produced by the beryllium(d,n) reaction using 20 MeV deuterons accelerated in the University of Washington cyclotron. The neutron spectrum from the beryllium(d,n) reaction is continuous up to about 24 MeV with a broad maximum at about 8 MeV. As expected, a large number of energy loss events occur above the maximum for proton recoils. In this case, a significant portion of the dose is due to the high energy loss events caused by carbon and oxygen recoils. The dose distribution in event size for this case is shown in Figure 11. Separation between pulses due to gamma and neutron secondaries is not apparent due to a preponderance of high energy proton recoils which have low rates of energy loss.

Measurements taken at the Plutonium Recycle Test Reactor (PRTR) in a mixed radiation field show good separation of gamma and neutron dose even though the neutron dose is rather small. These results are shown in Figure 12.

Three of the measurements of dose distribution in event size were converted to dose distribution in linear energy

FIGURE 10. Dose Distribution in Event Size - PuF_4 Neutrons, Two Types of Counters

FIGURE 11. Dose Distribution in Event Size - Be(D,n) Neutrons

FIGURE 12. Dose Distribution in Event Size - PRTR γ , n Field

transfer (LET) by using a relatively simple formula:

$$D(L) = K[2D(Y) - \frac{YdD(Y)}{dY}]_Y \quad (1)$$

where K is a normalizing constant

$D(L)$ = dose distribution in LET

$D(Y)$ = dose distribution in Y

$\frac{dD(Y)}{dY}$ = derivative of the $D(Y)$ curve

The derivation of this formula assumes localized deposition of energy along the particle track and straight tracks, and that the particle loses a small fraction of its energy in traversing the counter. $D(L)$ curves for 0.73 MeV, and 1.38 MeV neutrons were calculated using formula (1) and compared with theoretical calculations by Boag⁽²³⁾ for water in Figures 13-15.

A sealed counter with the spiral grid was used for the measurements shown at 0.73 MeV. This curve follows the same general form as that calculated by Boag with differences at low values of LET. This form may be due to events caused by proton recoils that start and/or stop inside the counter. The "start and stop" events are not considered in the theoretical treatment.

The average quality factor (QF) for this $D(L)$ spectrum is ~ 14 based on ICRP (1966).⁽²⁴⁾ The $D(L)$ curve for 1.0 MeV neutrons was calculated from data taken with the gas flow counter. This curve compares more favorably with the theoretical curve than does the previous case. The low point in the theoretical LET curve at 80 keV/ μ drops farther down than does the experimental curve. Again, start and stop events are apparent from 1 to 25 keV/ μ in the experimental curve. The average QF for the 1.0 MeV exposure is ~ 12 .

The third $D(L)$ curve was generated from the sealed spiral grid counter exposed to 1.38 MeV neutron. Again, good agreement was demonstrated between theoretical and experimental curves. The average QF for this case was ~ 10 .

FIGURE 13. LET Distribution - 0.73 MeV Neutrons

FIGURE 14. LET Distribution - 0.99 MeV Neutrons

FIGURE 15. LET Distribution - 1.38 MeV Neutrons

It is apparent that the TEPC is operating in a manner which at least approaches what would be expected theoretically. This conversion from D(Y) to D(L) is possible because the assumption that recoiling protons travel across the chamber and lose only a small part of their energy is adequately met. It does not hold for the heavy ions which have very short ranges.

SUMMARY

It has been shown that sealed counters and gas flow counters with single wire electrodes can be made to function satisfactorily for monitoring neutrons in mixed gamma-neutron radiation fields. It should be apparent that, in addition to providing absorbed dose and QF measurements in radiation work zones, there exists a good possibility of developing the TEPC into a standard for absorbed dose measurements.

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The authors wish to acknowledge the work of Mr. Dan Haggard in helping to take and analyze much of the data during the course of this study. The efforts of Mr. Dan McConnon who, at considerable trouble, obtained the data at the PRTR are also gratefully acknowledged.

MATHEMATICAL CONSIDERATIONS ON THE URINARY EXCRETION OF PLUTONIUM DEPOSITED IN THE LUNG

I. C. Nelson

INTRODUCTION

Inhalation of plutonium aerosols continues to present a potential for occupational exposure at the Hanford complex. Because of this potential, a program of surveillance is practiced to reveal possible accumulation of plutonium in workers' bodies. While advances have been made in in vivo counting techniques and in interpretation of fecal analysis results for estimation of body burdens resulting from known exposures, urinalysis continues as the most manageable sampling

technique for surveillance purposes. The basis for local evaluations has been the experimental work of Langham, et al.,⁽²⁵⁾ as extended theoretically by Healy⁽²⁶⁾ in the development of a mathematical model relating urinary excretion to estimates of retention of plutonium in the lung. Uncertainties in the values of urinary excretion of plutonium predicted by this model at times soon after intake have prompted a reexamination of the theoretical development. The result of this work, a mathematical approximation of urinary excretion of plutonium deposited in the lung, differs from the previously mentioned model and appears to be more consistent with the experimental bases.

DISCUSSION

As in the Healy method, let it be assumed that the deposition resulting from the inhalation of a plutonium aerosol constitutes a reservoir of material in the lung from which various quantities pass at various rates to the bloodstream. It can also be assumed that the release of material from the lung to blood is proportional to the quantity in the lung at any time. The quantity in the lung may then be expressed by the exponential function

$$Q = Q_0 e^{-\Lambda t} \quad (1)$$

where Q_0 is the amount initially taken into the lung. The overall elimination constant Λ is both complicated and poorly known due to the variety of removal modes and rates. If it is assumed that the quantity of plutonium to be translocated to blood (q_0) can be differentiated from all other components and pathways, and that the removal of this material with time at a removal rate constant (λ) depends only upon the quantity present to be translocated, the incremental quantity placed in the bloodstream at any time, t , is given by

$$dq = \lambda q_0 e^{-\lambda t} dt \quad (2)$$

The Langham expression for excretion of plutonium in urine may be written as

$$E = BT^{a-1}A \quad (3)$$

where E is the excretion of plutonium in urine collected over one-day periods and in the same units as A, the quantity injected and T is the time from injection to time of urine sampling in days. B and a are empirically obtained constants. Since the urine is collected in one-day periods, the values of T are restricted to the integers.

From Equation (2) the amount of plutonium entering the bloodstream on the first day amounts to

$$q_1 = q_0(1-e^{-\lambda}) \quad (4)$$

If this material is assumed to enter the bloodstream instantaneously at the beginning of the day, the plutonium collected in the first day's urine would be given by

$$E_1 = Bq_0(1-e^{-\lambda})1^{a-1} \quad (5)$$

At the end of the second day, the plutonium excretion from that entering the blood on the first day would be described by

$$E_2 = Bq_0(1-e^{-\lambda})2^{a-1} \text{ plus} \quad (6)$$

the additional amount from the second day, which is

$$Bq_0(e^{-\lambda}-e^{-2\lambda}) = Bq_0(1-e^{-\lambda})e^{-\lambda} \quad (7)$$

with the total excretion being the sum of the two. Similarly, the excretion on the third day would be described by

$$E_3 = Bq_0[(1-e^{-\lambda})3^{a-1} + (1-e^{-\lambda})e^{-\lambda}2^{a-1} + (1-e^{-\lambda})e^{-2\lambda}] \quad (8)$$

In general, the excretion of plutonium on the Rth day after inhalation would be described by

$$E = Bq_0(1-e^{-\lambda}) \sum_{\ell=1}^R \ell^{a-1} e^{-(R-\ell)\lambda} \quad (9)$$

The excretion may be expressed as percent of initial transferable quantity (q_0) and becomes

$$E_u = \frac{100E}{q_0} = B' (1 - e^{-\lambda}) \sum_{\ell=1}^R \ell^{a-1} e^{-(R-\ell)\lambda} \quad (10)$$

In the extended form of Langham's expression, B' is equal to 0.20 and a is equal to 0.26 ($B' = 100B$). For periods up to 138 days, the values of both B' and a are often taken as 0.23. The excretion of plutonium as a function of time as given by (1) is plotted in Figure 16 and tabulated in the Appendix.

By assuming that all of the material translocated from lung to blood during the first day had entered the blood at the beginning of the day, the estimate of urinary excretion of plutonium is maximized. If the time of entry was at the end of the first day, the estimate tends to be minimized and the effect is to move the excretion curve ahead one day in time, with no excretion of plutonium occurring on the first day. The representation of this case is also plotted in Figure 15. It would seem reasonable that the best estimate of plutonium excretion should lie between these maximum and minimum estimates.

The Healy method yields an integral in these same terms as

$$E_u = B' \lambda \int_0^R e^{-\lambda t} (R-t)^{a-1} dt \quad (11)$$

This integral was evaluated using the same previously defined constants. The results are plotted in Figure 16 and tabulated in the Appendix. The Healy expression can be seen to significantly over-estimate the urinary excretion of plutonium by the previous method, particularly at times soon after inhalation. The logic of the Healy method is the same as that developed here, and the discrepancy is concluded to

FIGURE 16. *Plutonium in Urine as Function of Time for Plutonium Entering with Biological 500-Day Half Time*

arise from the combination of a step function (Langham's excretion expression) and the continuous function describing entry of material into the bloodstream.

Just where the time-excretion curve would fall between the maximum and minimum representation is not known. However, the effect is important for only the first 10 days or so after inhalation. An approximation was developed which, although somewhat arbitrary in basis, has the virtue of falling between the maximum and minimum expressions for the first few days, and thereafter following the curves with a difference of about 10%. This function is represented by

$$E_u = B'\lambda \int_0^R e^{-\lambda t} (R+1-t)^{a-1} dt \quad (12)$$

The curve of this function is also plotted in Figure 16 and is tabulated in the Appendix.

While the models of plutonium excretion developed here and presented as Equations (10) and (12) shed no real light on solution of the many problems of diagnosing burdens of plutonium in workers' lungs, they are concluded to offer a more mathematically satisfactory description of the excretion process and will form a more defensible basis for more detailed treatments.⁽²⁷⁾

NEUTRON DOSIMETRY WITH ACTIVATION FILTERS AND TLD

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

INTRODUCTION

The search for a personnel neutron dosimeter for adequate sensitivity and satisfactory energy response has continued for at least the last 10 years. Work conducted at the old Hanford Laboratories, Battelle-Northwest, and at such other AEC installations as Brookhaven, Oak Ridge, and Argonne has led to a number of reasonably accurate active dosimetry systems. As yet, however, no passive device suitably

accurate over the required neutron energy spectrum and of satisfactory sensitivity to gamma radiation is available.

The use of NTA nuclear emulsion continues today at many universities and atomic energy sites where the properties of the emulsion are not detrimental to the accurate measurement of high energy neutron and charged particle radiation. At installations where radiation fields are a mixture of neutron and other types of radiation, the interpretation of nuclear track emulsion is, at best, difficult and, in many instances, impossible due to the interference reaction in the emulsion. In many cases where a degraded fission neutron spectrum in the proximity of a reactor is mixed with gamma radiation, no logical results can be obtained from the NTA system since the low energy cutoff of the emulsion may be as high as 0.8 MeV, which is approximately the most probable energy of the fission neutron spectrum.

The shortcomings of emulsions have led to numerous investigations of various luminescent materials, fissionable materials, TLDs, and elements which may be activated by neutrons of various energies. A particular element (rhodium) has been investigated to determine its characteristics when exposed to mixed radiation fields and is of universal interest. This report includes what is best known at this time about the characteristics of a rhodium-TLD combination dosimeter developed with the intention of replacing nuclear emulsion film.

HISTORY

In early 1963, investigations of the use of various combinations of rhodium, cadmium, tin, and iron as filters in front of ordinary beta-gamma dosimetry film were started (Figure 17). Rhodium was picked as the prime element of the filter system because of its unusually large absorption resonance for neutrons of 1.26 eV. The cross section of 1.26 eV is

FIGURE 17. Activation Neutron Dosimeter

4500 barns compared to the 2200 meter/sec cross section of 150 barns. The basic idea of the rhodium system is then to cause high energy neutrons incident on the dosimeter and human body to be moderated and backscattered into rhodium. Any neutron with energy near the 1.26 eV resonance will be preferentially absorbed causing the rhodium to become activated. For a particular incident neutron energy, the activation of the rhodium should be directly proportional to the neutron exposure or total flux.

The path of an incident fast neutron is as follows:

- Penetration through the dosimeter components into the body near the dosimeter.
- Moderation in the body tissues.
- Backscatter from the body to the neutron dosimeter.
- Capture by an activation foil in the dosimeter

Film was first chosen for a number of reasons as the sensitive material to detect the decay products of the activated foils.⁽²⁸⁾ Film is extremely sensitive to the beta and gamma radiation emitted by rhodium. The energies of this radiation occur at the peak in the unshielded response of the film. The film is identical to film used in the routine beta-gamma dosimeter used for personnel monitoring. Thus, no unusual processing was required for the neutron dosimeter film. TLD materials, upon becoming readily available, were chosen on the basis of their characteristics as the sensitive material.

Several combinations of the above metals were used in various thicknesses to achieve the proper response to beta and gamma radiation. These radiations must be taken into account to achieve uniform response at each filter location. The early work from 1963-1965 consisted largely of working out the proper combinations.

DOSIMETER AND EXPERIMENTAL DESIGN

After many preliminary designs using metal foils of varying thicknesses, a final design was determined by calculation and experimentation in mid-1965. The configuration of the dosimeter is given in Figure 16. The filter materials and thicknesses are as follows:

	γ Filter, in.	Fast Neutron, in.	Thermal Neutron, in.
Badge	Sn-0.040	Cd-0.030	Sn-0.030
Front	SS-0.003	Rh-0.010	Rh-0.010
Badge	Pb-0.010	Pb-0.010	Pb-0.010
Back	Sn-0.010	Rh-0.010	Rh-0.010

With these filter combinations, the TLD response behind the three filters due to gamma radiation is essentially equal for energies lying in the range 0.020 MeV to 3 MeV.

The response behind the tin-iron filter is produced almost entirely by photon radiation. The response behind the cadmium-rhodium filter is produced by gamma radiation and "body moderated" fast neutrons. Some response is also due to prompt gamma radiation after thermal neutron capture in the cadmium. The response at the tin-rhodium filter is produced by gamma radiation, fast neutrons, and thermal neutrons.

With this system, three quantities are measured and related to three types of radiation (photon, thermal neutron, and intermediate to fast neutron) through a set of simultaneous equations. The filters are of sufficient density thickness to yield essentially zero response behind all three filters for beta particles of energies <3 MeV. The response to beta radiation is assumed to be zero in the following derivations. Generally, in a mixed radiation field, the following equations describe mathematically the TLD response behind each filter region:⁽²⁸⁾

$$D_{\gamma A} + D_{N_f A} + D_{N_t A} = D_A \quad (1)$$

$$D_{\gamma B} + D_{N_f B} + D_{N_t B} = D_B \quad (2)$$

$$D_{\gamma C} + D_{N_f C} + D_{N_t C} = D_C \quad (3)$$

where

$D_{\gamma A}$ = Response behind filter due to photon radiation.

$D_{N_f A}$ = Response behind filter A due to intermediate and fast neutron radiation.

$D_{N_t A}$ = Response behind filter A due to thermal neutron radiation.

D_A = Total response behind filter A.

The symbols in Equations (2) and (3) are defined in a similar way for filters B and C.

The photon absorption properties of all three filters is such that the response behind each filter is equal when the system is exposed to photon radiation only.

$$D_{\gamma A} = D_{\gamma B} = D_{\gamma C} \quad (4)$$

Filters B and C are chosen so that equal response is obtained when the system is exposed to intermediate and fast neutrons.

$$D_{N_f B} = D_{N_f C} \quad (5)$$

All three filters provide unequal responses to thermal neutron.

$$D_{N_t A} \neq D_{N_t B} \neq D_{N_t C} \quad (6)$$

Using the condition given in Equation (4) and subtracting Equations (2) and (3), the result is a relation between densities due to thermal neutrons.

$$(D_{N_t C} - D_{N_t B}) = D_C - D_B \quad (7)$$

As long as the assumptions hold, this relationship can be used to measure thermal neutron exposure since the difference $(D_C - D_B)$ is due only to thermal neutrons.

After subtracting Equations (1) and (2) and applying Condition 4, the resulting equations give the relationship of the response due to fast and thermal neutrons.

$$(D_{N_f B} - D_{N_f A}) = D_B - (D_{N_t B} - D_{N_t A}) \quad (8)$$

but

$$(D_{N_t B} - D_{N_t A}) = k(D_{N_t B} - D_{N_t B}) \quad (9)$$

The quantity $(D_{N_t B} - D_{N_t A})$ must be determined by exposures in a "pure" thermal neutron field. The constant k can be determined and Equation (8) modified to:

$$(D_{N_f B} - D_{N_f A}) = D_B - D_A - k(D_{N_t C} - D_{N_t B}) \quad (10)$$

Thus, once k is determined, it is possible to separate the difference due to fast and intermediate neutrons from the difference due to thermal neutrons. These differences are then related to the neutron exposure. For gamma and low energy photon radiation, response reading at the tin-iron filter is calibrated to exposure.

Three quantities of importance can be determined. These are exposure to electromagnetic radiation, to thermal neutrons, and to fast and intermediate neutrons. The basic problem with this type of neutron dosimeter is its nonlinear energy response. The nonlinearity makes necessary a calibration of the dosimeter in a fast neutron field closely duplicating the neutron spectrum to be encountered in the dosimetry application.

To take the place of the human body during design and exposure, a large piece of polyethylene was used. Dosimeters

were attached to the piece of polyethylene and exposed to various radiations from isotopic neutron sources (PuF_4 and PuBe) and a positive ion accelerator. The neutron energy spectrum from PuF_4 is continuous from low energies to approximately 3.0 MeV. The average energy of the PuF_4 neutron spectrum is between 0.8 MeV and 1.0 MeV. The PuBe neutron source produces neutrons in several energy groups from fission neutron energies (~ 0.8 MeV) to approximately 10 MeV.⁽²⁹⁾ Neutrons produced at the positive ion accelerator are very nearly monoenergetic when a thin target is used. The range of neutron energies available is from ~ 10 keV to as high as 18 MeV, though all these possible energies were not used. Dosimeters were exposed to monoenergetic neutrons from about 0.027 MeV to 5.0 MeV to determine the energy response of the dosimeter system.

Thermal neutron exposures were conducted in a calibrated paraffin tank using the PuF_4 and PuBe sources to supply the neutrons. The paraffin tank was calibrated by exposing gold foils in the tank and in a standard graphite pile where the thermal neutron flux is known.

Gamma radiation from radium and cobalt-60 sources and X-rays from 16 keV to 170 keV were used in developing the filter system and measuring the response of the final dosimeter design. A number of K-fluorescent X-ray sources up to 100 keV were available, while above 43 keV, filtered X-rays were used.

EXPOSURES AND RESULTS

The first step in developing the dosimetry system is to achieve gamma equivalence for all three metal filters. This is a process of calculation and trial until a satisfactory compromise is reached. For experimental purposes, the X-ray sources and radium and cobalt-60 sources were used. A large

number of exposures were made to various energies of gamma and X-ray radiation. The gamma energy response was then determined by measuring the relative sensitivity as a function of energy while keeping the exposure constant. The result is shown in Figure 18. The TLD response readings for this study are shown in Table 4.

TABLE 4. Response of Rh-TLD Filter System

Energy, keV	γ Filter	N_f Filter	N_t Filter
16	130	130	140
23	380	370	380
34	480	480	510
43	520	540	540
62	510	490	510
82	420	400	430
100	430	450	420
120	450	440	410
170	470	460	470
Ra- γ	470	480	470
^{60}Co	460	470	470

To determine the thermal neutron response and to obtain a value for K to use in evaluating fast neutron kerma, a set of thermal neutron exposures was made using the PuF_4 source in a large paraffin tank. The amount of paraffin between the source and the badge is adequate to moderate most of the fast neutrons emitted from the source, but a few fast neutrons are still present. Thus, the determination of the constant K is somewhat inaccurate. The results for thermal neutron exposures are shown in Figure 19. For the measurements made, the average value of ke was found to be 1.6.

Exposure response for fast neutrons was measured using a PuF_4 source with varying lengths of exposure time. The response was linear with exposure. The minimum detectable

FIGURE 18. *Gamma Energy Response Characteristics*

FIGURE 19. Thermal Neutron Response

kerma was about 10 mrad for the PuF_4 spectrum. These data are shown on Figure 20. The fast neutron energy response was measured using a positive ion accelerator. Exposures were made to fast and intermediate energy neutrons from 0.027 MeV to 5.0 MeV, and the difference in the cadmium-rhodium and the tin-iron filters determined. This difference is related to the fast neutron kerma as shown by the equations in the design section of this report. While this response is related to fast neutron kerma, the magnitude of the difference for a given kerma of 100 mrad is a function of the incident neutron energy. The observed neutron energy response for the experimental exposures between 0.027 MeV to 5.0 MeV varies by a factor of about 100. No significant thermal neutron contribution occurred in any of the fast neutron exposures. Neutron doses were calculated using the curve in NBS handbook 63. Total neutron fluence needed to accumulate the desired dose was calculated by using known long counter and BF-3 tube responses available from the Radiological Physics group. Errors in this information are not (under usual conditions) greater than $\pm 20\%$.

As can be seen from Figure 21, the energy response varies across the entire energy spectrum. Thus, to be useful, the dosimeter would require calibration in a neutron radiation field quite similar to the field requiring personnel dosimetry. In situations convenient for effecting such calibration, and where no large amounts of moderating material exist close by, the rhodium filter system can perform adequately. These dosimeters could be used to advantage in locations where plutonium is being processed through the PuF_4 stage. Their use, however, need not be limited to these locations. If an exposure to a different spectrum were incurred, say about 0.4 MeV to 2.0 MeV, an error of no greater than 2.0x would be realized.

FIGURE 20. Fast Neutron Response

FIGURE 21. Neutron Energy Response Characteristic

CONCLUSION

In the preceding discussion, the various formulae and experimental measurements leading to a personnel neutron badge have been presented. The particular filter system described has been developed and checked to a logical termination. The rhodium filter badge does provide an excellent improvement over the NTA film badge in its good sensitivity to thermal and intermediate neutrons, as contrasted to the absence of NTA sensitivity at these energies, and improved sensitivity at high energies. As has been stated before, the accuracy of this system is good if the neutron spectrum is known. The use of TLD permits convenient handling of the badge and presents none of the problems of heat fogging and gamma-ray background normally associated with film.

THE RESPONSE OF SELECTED THERMOLUMINESCENT MATERIALS TO FAST NEUTRON EXPOSURES

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

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 time-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 dosimetry have led Battelle-Northwest to study the possibilities of some of the new TLD materials for fast-neutron dose measurements.

Earlier studies of the fast-neutron response of TLD materials^(30,22) 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 frequently 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.

Some nuclear research laboratories are using TLD materials in this manner, notably $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$. To determine the accuracies one would expect from such system, the response of TLD materials was measured as a function of incident-neutron energy and shielding material. By using cadmium and boron-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, recently purchased and used for a portion of the measurements in this series, is shown in Figure 22. Most of the work was accomplished with a Conrad TL analyzer modified to readout through a Dymec analog to digital converter (Figure 23).

The dosimeters used for the study were TLD-100, TLD-200, TLD-600, TLD-700 ribbon in the form of $1/8 \times 1/8 \times 0.35$ -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 in most cases to assure proper readout of the dosimeters, though a few low readings were obtained despite the precautions.

FIGURE 22. Harshaw TLD Reader

FIGURE 23. Conrad TLD Reader

Fast neutron exposures were conducted with 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 24). These neutrons were produced with $T(p,n)^3\text{He}$ reactions. The Hanford precision long-counter was used as the standard for kerma measurements

EXPERIMENT

Within limits of time, convenient exposures to kermas between 20 mrad and 1.0 rads were conducted. The amount of time required depends upon the neutron yield from the targets. With the Van de Graaff of the type at Battelle-Northwest, the yield decreases as the bombarding energy decreases, even though 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 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 admium shielding - Cadmium-moderator.
- Backscatter geometry with B-10 shielding - B-10 moderator.

FIGURE 24. *Experimental Room - Van De Graaff Accelerator*

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, 1-1/4 in. in diam, provided a 1-cm thickness of boron-10 in all directions around the dosimeter placed inside the sphere. The moderator-phantom was a 9-in. diam cylinder of polyethylene with one area cut off to provide a flat surface to face the neutron beam.

All data are reported in terms of net reader counts per rad, with the reader calibrated to 2 units per mR radium γ for TLD-700 ribbon. Exposures of $\text{Li}_2\text{B}_4\text{O}_7\text{:Mn}$ were conducted under the previously described 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 have a positive response 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 25, 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 larger number of cadmium capture gammas present. The response is given in terms of net reader counts per rad. Apparently, 90% or more of the response of $\text{Li}_2\text{B}_4\text{O}_7\text{:Mn}$ 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 increased as the incident neutron energy decreased, as shown in Figure 26. Again, the most sensitive condition of

FIGURE 25. Response of $\text{Li}_2\text{B}_4\text{O}_7$ to Fast Neutrons

FIGURE 26. Response of TLD-100 to Fast Neutrons

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 somewhat 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 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 is extremely sensitive to neutrons, with a very high reading even for the 1.0 MeV exposures (Figure 27). The response in air with no backscatter is higher than for similar measurements for any of the other TLD materials used. TLD-600, composed of 95.62% lithium-6, 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 of 0.20 MeV to 1.0 MeV for the bare-moderator case. For the materials with high neutron cross sections, the bare-moderator condition appears to yield the highest results, with the cadmium shielded condition the next most sensitive. These two conditions make the maximum use of the backscattered neutrons.

The results of exposures using TLD-700 material are shown in Figure 28. TLD-700 is composed of 99.993% lithium-7 and so should be relatively insensitive to the backscattered neutrons and to incident neutrons also. Both response curves obtained using bare dosimeters show a low response.

FIGURE 27. Response of TLD-600 to Fast Neutrons

FIGURE 28. Response of TLD-700 to Fast Neutrons

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 29. 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 in the foregoing, the response of TLD-600 in the bare-moderator condition varies by a factor of 25 over the same energy range for the same exposure condition (see Figure 30). A combination of TLD-600 and TLD-700 to provide a measure of the average incident neutron energy appears 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 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 31.

Glow curves were taken periodically while reading out the dosimeters to assure total readout or rejection of the data if proper readout did not occur. The standard deviations of the data points shown in Figures 4 through 10 are $\pm 10\%$ or less based on the readings. This calculation does not include errors in the long counter measurements, shown by Nichols⁽³¹⁾ to be as much as $\pm 10\%$ for the neutron energies and experimental geometries used here. Even though errors in the experiment may total 20%, the curves give useful information about neutron response of various TLD materials.

FIGURE 29. Response of TLD-200 to Fast Neutrons

FIGURE 30. Response of TLD-100, TLD-600, and TLD-700 on Unshielded Moderator-Phantom

FIGURE 31. Ratio of TLD-100 and TLD-600 Response to TLD-7 Response

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 if 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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APPENDIX

APPENDIX

Value of the functions $E_u(1) = B'(1-e^{-\lambda}) \sum_{\ell=1}^R \ell^{a-1} e^{-(R-\ell)\lambda}$,

$E_u(2) = B'\lambda \int_0^R e^{-\lambda t} (R-t)^{a-1} dt$, and $E_u(3) = B'\lambda \int_0^R e^{-\lambda t} (R+1-t)^{a-1} dt$

as percent of dose are presented below for $B' = 0.20$, $a = 0.26$, and $\lambda = 0.001386 \text{ d}^{-1}$ ($t_{1/2} = 500$ days).

$\frac{R}{\text{Days}}$	$E_u(1)$	$E_u(2)$	$E_u(3)$
1	2.772×10^{-4}	10.65×10^{-4}	2.112×10^{-4}
2	4.428	12.74	3.493
3	5.651	14.14	4.574
4	6.637	15.22	5.461
5	7.470	16.11	6.209
6	8.196	16.88	6.902
7	8.842	17.55	7.484
8	9.424	18.15	8.039
9	9.957	18.69	8.578
10	10.45	19.19	9.145
20	14.01	22.73	12.63
30	16.34	24.98	14.95
40	18.09	26.62	16.69
50	19.48	27.91	18.08
60	20.62	28.94	19.24
70	21.58	29.80	20.21
80	22.42	30.52	21.05
90	23.13	31.13	21.78
100	23.78	31.65	22.42
200	27.14	34.02	25.96
300	27.98	33.99	26.96
400	27.77	32.99	26.86
500	26.99	31.54	26.20
600	25.93	29.88	25.23
700	24.72	28.16	24.11
800	23.45	26.44	22.92
900	22.17	24.78	21.71
1000	20.92	23.19	20.52
2000	11.72	12.28	11.61
3000	7.406	7.545	7.378
4000	5.361	5.393	5.352
5000	4.264	4.271	4.261
6000	3.593	3.593	3.591
7000	3.135	3.134	3.134
8000	2.799	2.798	2.798
9000	2.539	2.537	2.538
10000	2.330	2.328	2.329

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