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Title: MEASUREMENT OF ENVIRONMENTAL GAMMA EXPOSURE BY THERMOLUMINESCENCE DOSIMETRY

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Energy dependence, angular dependence, and linearity of response to exposure levels were studied for the thermoluminescent dosimeter used in an environmental monitoring network. The dosimeter consisted of a manganese activated calcium fluoride phosphor (CaF_2: Mn) bonded to a helical heating element and encapsulated in a gas filled glass tube.

Energy dependence was studied using a K-alpha, K-beta x-ray fluorescence system which produced energies from 6.47 keV to 100.96 keV. The response to ^{137}Cs and ^{60}Co gamma was also tested. The curve generated by the data may be useful to correct dosimeter readings when the energy of the radiation is known.

Angular dependence at 662 keV produced a 20 percent drop in response for radiation parallel to the dosimeter axis compared to the most sensitive incident angle of 90^{o}.

For exposures between 10 mR and 1 R the response of the dosimeter was linear within $\pm$ 5 percent, however for exposures greater than 1 R, there was an over-response which increased with exposure. When the reader was adjusted to read correctly at 100 mR, the response to 3000 R, the highest exposure given, was 32 percent high.

For the conditions usually encountered in environmental monitoring, the dosimeter can be used with no more than a simple calibration correction. If, on the other hand, low energy radiation, very directional beams, or high exposures are encountered, further corrections may be necessary.

Measurement of Environmental Gamma Exposure by Thermoluminescence Dosimetry

by

Charles Kenneth Fitzsimmons

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INTRODUCTION

History and Description of Thermoluminescence

Thermoluminescence (TL) is a phenomenon exhibited by a number of solids, primarily ionic crystals and glasses, whereby stored energy is released and emitted as light when the solid is heated. The energy stored in the crystal is in the form of electrons excited by outside influences into metastable energy states.

The existence of thermoluminescence has been known for a long time. Boyle (5, p. 35) is reported to have observed the thermoluminescence of a diamond in the seventeenth century. In 1928, Lind (14) observed the coloration of glasses when they were exposed to x or γ radiation and the emission of light when the glasses were subsequently heated. Studies of thermoluminescent natural minerals, fluorides, and carbonates were reported in 1925 by Wick (23).

Although TL was known to be related to exposure to x or gamma rays, its application to dosimetry was not attempted until the 1950's. The majority of early research in TL was carried out by Daniels and his students at the University of Wisconsin (5). More recent work at Wisconsin has been done by Cameron (4, p. 4, 197-198) in collaboration with a number of others.

In the course of research at Wisconsin, thousands of rocks and about a hundred pure inorganic crystals were examined for thermoluminescence (5). About half the laboratory crystals exhibited thermoluminescence. It is apparent that thermoluminescence is common to most inorganic crystals which contain suitable amounts and types of impurities. Not all thermoluminescent materials are suitable as ionizing radiation dosimeters, however. The intensity of the luminescence must be relatively high for a given amount of radiation exposure in order to provide sufficient sensitivity. The luminescence must not occur spontaneously at room temperature, or exposure information will not be retained long enough to obtain an accurate reading of the dosimeter.

A number of materials have been discovered or developed which are very good thermoluminescent dosimeters (TLD). Lithium fluoride and calcium fluoride are perhaps the most popular TLD materials. They have a high intensity luminescence which occurs above 200° C, making the dosimeter quite stable in retaining information. Other materials investigated have been $CaSO_3$:Mn, $Li_2B_4O_7$:Mn, and a terbuim activated lithium-aluminosilicate thermoluminescent glass (11). For very high exposures, greater than 10^5 R, carbonates of calcium, barium, strontium, magnesium, and zinc, and thorium dioxide (ThO_2) and barium titanate ($BaTiO_3$) hold promise (2). In addition

to these, Cameron (4, p. 65-66) cites a number of references on other materials including, Al_2O_3, BeO, and SrS:(Eu, Sm).

In addition to the work at Wisconsin, concurrent research on TL was being done at the Naval Research Laboratory (NRL) during the 1950's under the direction of J. H. Schulman (9, 10, 20). A useful dosimeter using CaF_2:Mn was developed out of this work (21, p. 24). A practical dosimeter using CaF_2:Natural was developed in Belgium by Manufacture Belge de Lampes (MBLE) in the early 1960's (13, 19).

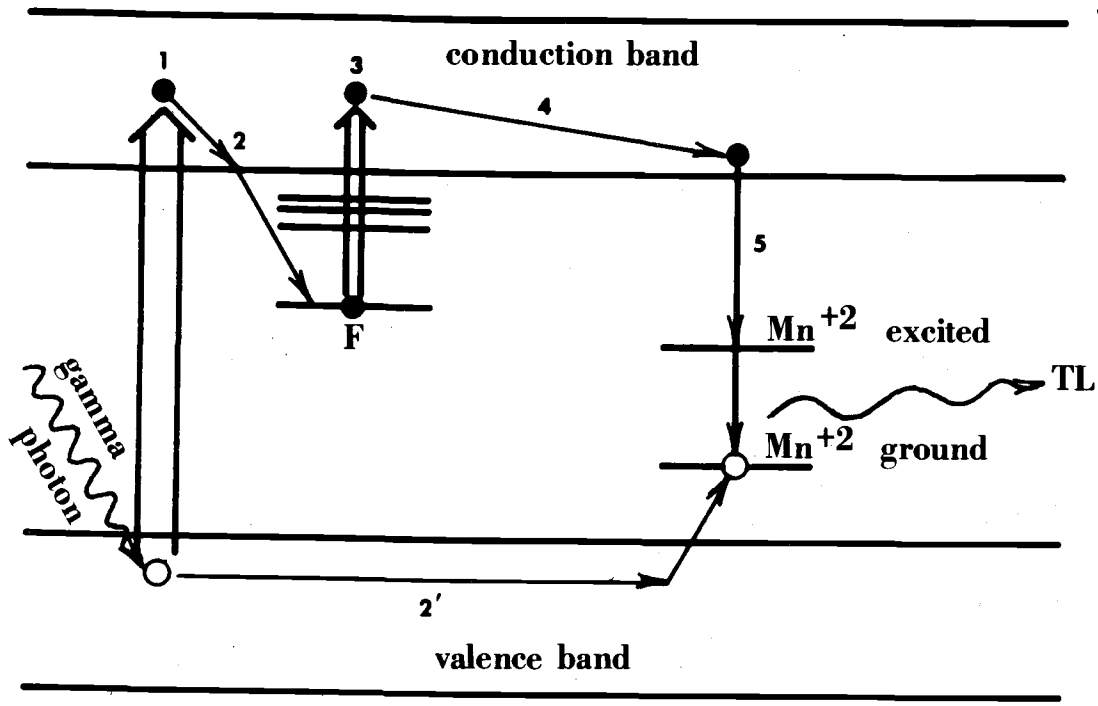
Both the NRL and MBLE dosimeters use a form of CaF_2 phosphor bonded to a heating element which is enclosed in either an evacuated or gas filled glass tube. A similar tube-type dosimeter using CaF_2:Mn was developed for the U. S. Navy by Edgerton, Germeshausen, and Grier, Inc. (EG&G) (18). The dosimeter used for the studies in this thesis is made by EG&G and is a descendent of the dosimeter designed for the Navy. It employs a manganese activated calcium fluoride (CaF_2:Mn) phosphor (17) bonded to a helical heating element enclosed in a gas filled glass tube.

The Physics of Thermoluminescence

The physical theory of thermoluminescence is not well known, but the basic phenomenon is qualitatively understood. Mathematical models for TL are very complicated and not very successful in predicting the observed behavior of phosphors. Solutions to simple

mathematical models are already very unhandy and complicated, but they become hopelessly involved when the parameters of real phosphors are considered. One such model is described by Bräunlich (3).

Thermoluminescence is most often explained (4, p. 5, 17, 21, p. 11) in terms of an energy band diagram such as shown in Figure 1. In step one, energy deposited in the crystal by ionizing radiation activates an electron from the valence band into the conduction band where it is free to wander throughout the crystal. Defects in the crystal structure, usually ion vacancies in the lattice, form local positive charges which trap electrons in metastable energy states shown in step two. An electron trap can be thought of as a type of Bohr atom with its own particular energy levels all different from those which would be available in a perfect crystal. The formation of these traps allows particular wave lengths of light to be absorbed and emitted and thus gives the crystal its color. The electron traps are often referred to as F-centers from the German term Farbzentrum (color center). The probability of an electron in a metastable state of the F-center decaying to the ground state is temperature dependent. If the trap is sufficiently deep (below the conduction band) normal room temperatures will not cause its decay. In the case of CaF_2:Mn, the traps are emptied most efficiently at about 260° C. Application of heat to the exposed phosphor is shown in step three, where the trapped electron is excited into the conduction band. It wanders, step four, until it



● electron

○ hole

Figure 1. Thermoluminescent process in manganese activated calcium fluoride.

recombines in a radiative transition with a trapped hole, step five. The function of the Mn^{+2} doping agent or activator is to trap the holes created in step one. This is depicted by step two prime. The activator insures that the recombination will be a radiative transition. The intensity of the green-orange luminescence of the Mn^{+2} ion is proportional to the energy absorbed by the phosphor and thus can be used to measure the dose received from the original ionizing radiation.

Characteristics of CaF_2:Mn

As the temperature of a thermoluminescent phosphor is increased, luminescence begins at some threshold temperature and increases to a maximum, then falls off again, forming a peak of light intensity. If the phosphor is heated further, a black body incandenscence curve takes over. The plot of luminescence with temperature is called a "glow curve." Each phosphor has its own particular glow curve. Some exhibit more than one glow peak which can be associated with the existence of more than one trap depth in the phosphor.

Figure 2 shows the glow curve drawn by the TLD reader used at the Southwestern Radiological Health Laboratory (SWRHL). The maximum of the main glow peak occurs at approximately 260° C (4, p. 49; 13, p. 741). The heating cycle is terminated shortly after the completion of this peak to prevent thermal damage to the dosimeter. A lower temperature peak shows up in the glow curve for exposures greater than

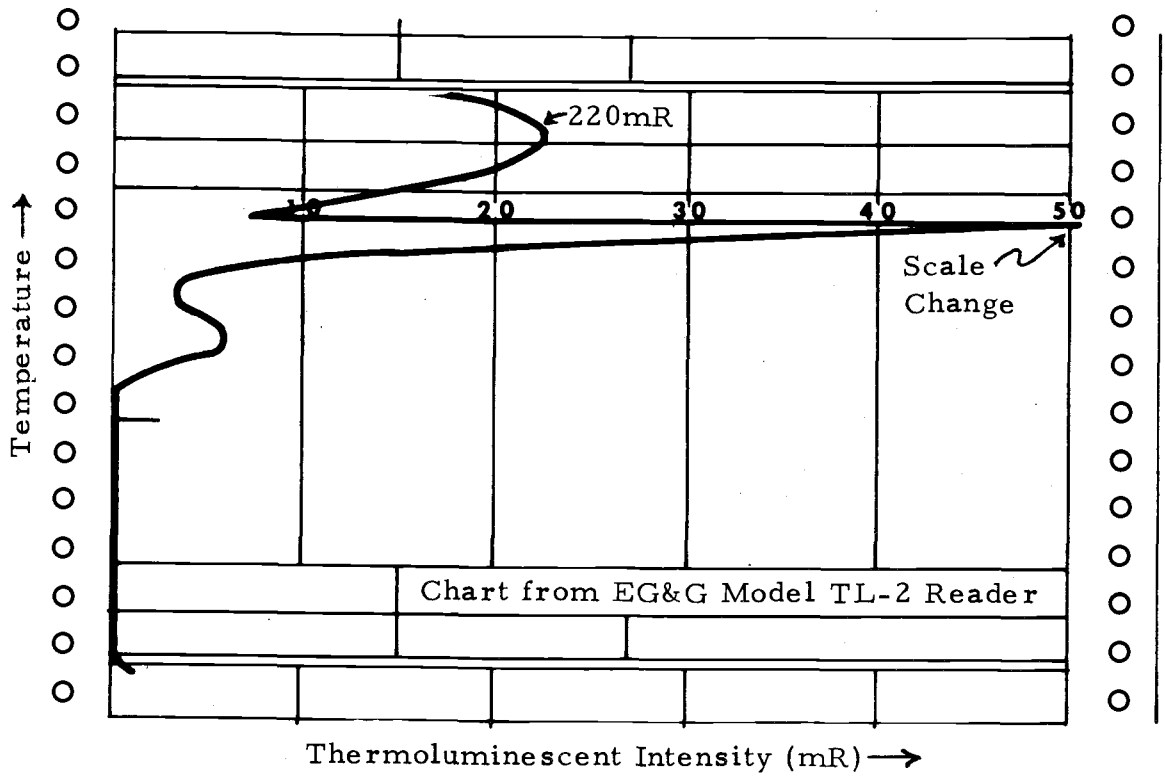


Figure 2. TL glow curve drawn by TLD reader.

about one hundred milliroentgens. This peak may be the one described by Palmer (17) which is associated with the reducing atmosphere used in preparing the phospher. The shape of the glow curve is dependent on the amount and type of activator (Mn^{+2} in this case), the phosphor itself, and the method of preparation as well as the quality and amount of radiation.

If a constant heating rate is used, the height of the glow peak is proportional to the absorbed dose. The area under the curve is also proportional to the dose; therefore, integrating circuits can be used to read the dosimeter. As the peak height becomes smaller, it becomes increasingly more difficult to know where to start and stop integrating. For this reason, measuring the peak height is more accurate for low exposures since it is easier to discriminate against the photomultiplier tube noise and other signals comprising the glow curve. A very stable heating circuit is required to use the peak height method because the peak height is dependent on the heating rate.

The system at SWRHL employs the peak height method of reading. The user must read the value of the exposure from the chart shown in Figure 2. The first full scale deflection represents an exposure of 50 mR. The reader automatically cycles the recorder to the next higher decade if full scale is reached. Thus, the second full scale deflection becomes 500 mR. This is repeated until the highest available scale, 5000 R, is reached. Such an arrangement is necessary to

preserve the reading because the TLD can be read only once for each exposure. The glow curve of Figure 2 has gone over 50 mR and the recorder was cycled to the next scale. The peak height corresponds to an exposure of about 220 mR. The lower temperature peak can be seen on this glow curve as well but it is not quantitated.

The investigators already cited describe thermoluminescent CaF_2:Mn as a phospher with high sensitivity, and a very linear response with respect to exposure levels. Part of the high sensitivity is due to the relatively high density of CaF_2 compared to LiF, for example. The higher density also causes a greater energy dependence below 500 keV. Energy dependence is usually compensated for by special filters around the detectors, but, then, energies below 60 keV can not be measured accurately. No exposure rate dependence has been reported which causes a deviation in response greater than 10 percent.

Non-Radiation Induced Thermoluminescence

Thermoluminescence may be stimulated by other mechanisms than the absorption of ionizing radiation. Such non-radiation induced thermoluminescence (NRI-TL) is referred to as "idiopathic" or "spurious" thermoluminescence (4, p. 123). Investigators of this phenomenon concur that the two major conditions necessary to produce NRI-TL are (a) mechanical movement of the crystals, such as shaking, grinding, or pouring, and (b) the presence of oxygen during read out

(1, 15, 22, 24). The NRI-TL induced by mechanical means is often referred to as triboluminescence. Although the physics of NRI-TL is not well understood, it seems to be a surface effect, since it is affected by ambient gas during the readout and is greater for powdered phosphors than for larger crystals. The TL induced by ionizing radiation, however, is a volume effect. The glow peaks from TL and NRI-TL occur at different temperatures which indicates different electron traps are responsible for the luminescence.

The use of thermoluminescent materials as dosimeters requires that the NRI-TL be minimized. The obvious means is to eliminate the requirements, i.e., immobilize the crystals, and remove the oxygen. In the tube type dosimeters, such as the ones used in this study, the phosphor (CaF_2:Mn) is bonded to the heating element and the whole detector is enclosed in a glass envelope which provides a non-oxidizing atmosphere.

Another effect related to NRI-TL is the sensitivity of some phosphors like CaF_2:Natural and $CaSO_4$:Mn to light (4, p. 33 and p. 103). Depending upon the wave length, light can excite or anneal the phosphor. Light induced glow peaks may not appear at the same temperature as the normal radiation induced TL peak. Dosimeters sensitive to light must be enclosed in light tight containers, but can be handled in normal room light during the short period of readout with no significant error. The dosimeters used in this study are shielded from light by the energy

compensating shield. CaF_2:Mn is not very sensitive to incandescent light; however, sunlight causes a large low temperature peak to appear in the glow curve after a few minutes exposure.

The Dosimetry Network

The work described in this report is part of a continuing effort to understand the behavior of the thermoluminescent dosimetry system used by the U. S. Public Health Service in an environmental monitoring network around the Nevada Test Site. The Southwestern Radiological Health Laboratory (SWRHL) of the U. S. Public Health Service performs off-site radiological monitoring activities in conjunction with nuclear testing under a Memorandum of Understanding (No. SF 54 373) with the U. S. Atomic Energy Commission. In addition to collecting and analyzing environmental samples, including air, water, milk, vegetation, etc., the laboratory maintains a dosimetry network of approximately 100 locations around the Nevada Test Site (NTS). Each of these stations is equipped with three film badges and three thermoluminescent dosimeters (TLD's) which are exchanged monthly.

The dosimetry network serves to document off-site exposures which arise from testing within NTS, and therefore, provides an exposure record which can be referred to when future nuclear tests are being planned. Because it is desirable to document relatively small exposures, of the order of a few milliroentgens, the dosimeters used

must be very sensitive. In fact, the sensitivity required necessarily allows them to measure background exposure. The interference of background with exposure measurements is itself a problem, common to all sensitive systems, and will be discussed later as it pertains to thermoluminescence systems.

The increasing interest in documenting smaller exposures forced the SWRHL Dosimetry Program to seek a more sensitive dosimeter than the film badge which was, at best, only able to measure exposures greater than 30 mR. Perhaps as great a problem as low sensitivity was one of poor reliability of dosimetry film. The film badges used in the dosimetry network suffered an intolerable amount of damage from ambient heat and light. In summer months loss of data due to heat and light damage often ranged as high as 70 percent (7, p. 20).

The search for a better dosimeter brought attention to the capabilities of TL. Manufacturers' claims for TLD performance were typified by the following specifications for the EG&G Model TL-12:[1]

Exposure Range:	0.1 mR to 5 x 10^4 R (Present Model TL-2B Reader. Limits: 5 mR to 5000 R).
Exposure Rate:	Negligible dependence to 10^9 R/sec.
Energy Dependence:	Five percent from 60 keV to 1.25 MeV.
Reusability:	In excess of 100 readings.
Reproducibility:	One percent or 0.4 mR.

[1] Edgerton, Germeshausen, and Grier, Inc., Laboratory Products Division, Data Sheet No. 2.

In addition to these data, it was apparent from numerous articles on thermoluminescence that the state of the art had advanced to a stage which promised to fulfill the needs of a monitoring network, i.e., high sensitivity, good reproducibility, and assured reliability.

In August, 1965, a TLD system[2] was purchased by SWRHL to supplement the existing film badge network. At first, only 20 stations were equipped with TLD's. As procedures were worked out necessary to implement a full scale routine monitoring network, more of the dosimetry stations were supplied with TLD's, until after two years, virtually all the stations had TLD's as well as film. Since then the TLD system has performed so well that film badges are soon to be discontinued in the network (8, p. 27-28).

A great deal of study and experimentation has been done on the TLD system at SWRHL during the last four years. It was necessary, for example, to perfect a calibration technique, and to determine the inherent TLD background before the system could even be practicable (7, p. 4-6). After the system was in operation, other studies were undertaken to refine the results obtainable from the network.

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[2] EG&G Model TL-12 dosimeters and two readers, Model TL-2B and TL-3B.

Purpose of the Study

The purpose for experimenting with the TLD system is to assure that the numbers generated are the best possible estimates of the exposures being measured. When the system was first set up, the manufacturer's specifications were taken for granted. It was assumed that reproducibility was within one percent, that the energy dependence was negligible in the gamma spectrum of interest (fission products), that there was negligible exposure rate dependence, and that the system response was linear over the range of exposures it was capable of measuring. To test these claims earlier would have taken too long at a time when the goal was to expedite a workable TLD system for environmental monitoring.

A number of problems did appear, however, which had to be solved before the system was ready for use. It was found that the dosimeters had to be individually calibrated to obtain results in keeping with the system capability. A procedure was devised (7, p. 4-5) to compare the thermoluminescence of each dosimeter with an ionization chamber reading, thereby establishing correction factors which could be applied to the thermoluminescence from unknown exposures. Accuracy by this method has been $\pm$ five percent and the reproducibility, $\pm$ one percent.

It soon became apparent that the TL-12 dosimeter had an inherent background, independent of the environment. Before low level measurements were possible, the magnitude of this inherent background had to be determined. The internal background, as it is now called, was produced by ^{40}K in the glass envelope and in the material which binds the phosphor to its heating element (16). See Figure 7. To minimize external exposure, dosimeters were placed in a low background chamber for which the exposure rate inside had been carefully measured or calculated. The apparent internal TLD background exposure rate was determined to be 0.7 mR/day $\pm$ 10 percent for all the dosimeters tested.[3]

Once the correction factor and the internal background exposure rate were known, all that remained before environmental measurements could be made was to account for the exposure during transit of the TLD's from the laboratory to the dosimetry station and return. The resulting relationship is written:

$$E_e = X f - a(t_1 + t_2 + t_3) - b(t_1 + t_3)$$

where E_e = External exposure on location (mR)

X = TLD readout (mR)

f = Correction factor

a = Internal background rate (mR/day)

[3] Data collected by William Horn in 1965 and Charles Fitzsimmons in 1969.

b = In-transit background rate (mR/day)

t_1 = Time from annealing to issue (days)

t_2 = Time from issue to collection (days)

t_3 = Time from collection to readout (days)

If the background exposure rate on location is known from previous measurements, a net exposure, E_n, can be calculated by

$$E_n = E_e - ct_2,$$

where c is the on-location background exposure rate. The above expression is used to calculate exposures reported by the present dosimetry program.

The expression, $E_e = Xf - a(t_1 + t_2 + t_3) - b(t_1 + t_3)$ contains no terms which correct for energy dependence, rate dependence, angular dependence, exposure level dependence, or fading, all of which in varying degrees are known to be inherent problems of TLD's. Three of the problem areas were chosen for this study, energy dependence, angular dependence, and exposure level dependence. These three problems are not necessarily the most important but are ones whose solutions might be most readily incorporated into the routine calculations.

The data obtained in this study revealed a classical angular dependence, typical of a cylindrical detector. The determined energy dependence was essentially the same as that published by the manufacturer except that the response to lower energies than those published previously was investigated in this study. The response to

exposures ranging from a few mR to 3000 R was found to be non-linear. Response varied approximately $\pm$ 20 percent of the mean (defined as 100 percent of given exposure) over the range of given exposures.

EXPERIMENTAL PROCEDURES

Energy Dependence Study

The energy dependence of ten TL-12 dosimeters was determined both with and without the energy compensating shield from 6.47 keV to 1,250 keV. Four separate systems were required to provide this range of energies, two x-ray machines, a ^{137}Cs source, and a ^{60}Co source.[4]

The low end of the spectrum was generated by a K-alpha, K-beta x-ray fluorescence system employing a General Electric Model 11DF1 tungsten target x-ray tube, operated continuously at 75 kVp and 36 mA. The 75 kVp machine had 12 fluorescers which produced energies from 6.47 keV (Fe) to 32.99 keV (Ba).

A larger x-ray fluorescence system produced energies from 25.84 keV (Sn) to 100.96 keV (U). This system comprised a Machlett type EG302 x-ray tube powered by a Universal Voltronics Power Supply and was operated continuously at 300 kVp and 15 mA.

The specifications and fluorescers of each system are listed in Table 1. Both systems are identical in principle of operation. As shown schematically in Figure 3, the primary beam from the x-ray tube

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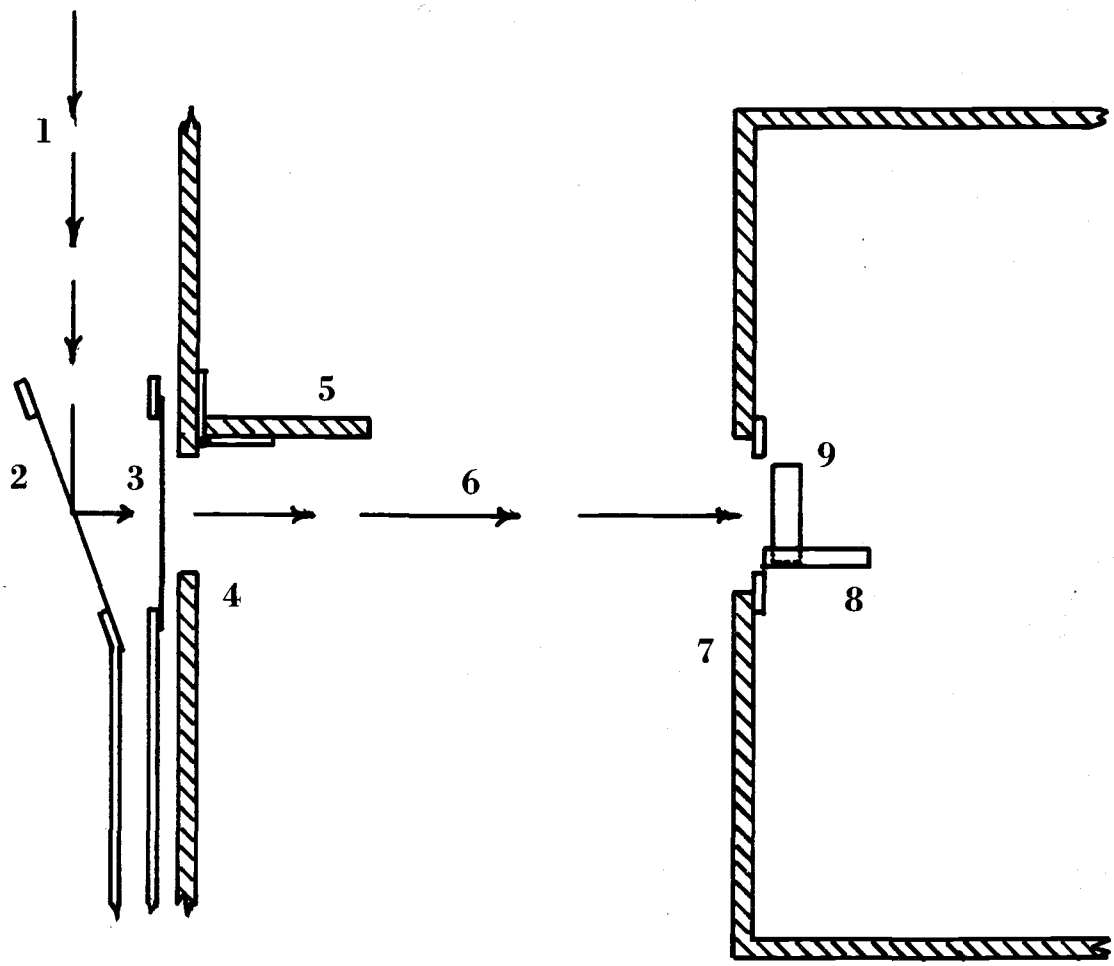
[4] Use of the two x-ray machines and the ^{60}Co source was provided by EG&G, Inc., Las Vegas, Nev. The ^{137}Cs source is operated by SWRHL, USPHS, Las Vegas, Nevada.

Table 1. Energies Available on the Two X-ray Fluorescence Systems.

Energy (keV)	Fluorescer	Filter (mg/cm^2)	
G. E. Model 11 DF 1, W Target, 75 kVp, 36 mA			
6.47	Fe	2	Al
8.14	Cu	2	Al
8.74	Zn	2	Al
11.20	Se	14	Al
15.20	Y	25	Al
16.04	Zr	29	Al
16.90	Nb	29	Al
17.78	Mo	43	Al
22.59	Ag	43	Al
23.63	Cd	58	Al
25.84	Sn	66	Al
32.99	Ba	172.4	Al
Machlett-Universal Voltronics, W Target, 300 kVp, 15 mA			
25.84	Sn[a]	425	Al
38.31	Nd	425	Al
41.16	Sm	425	Al
44.13	Gd	425	Al
52.11	Tm	425	Al
53.80	Yb	425	Al
57.30	Hf	425	Al
60.94	W	425	Al
68.66	Pt	425	Al
77.00	Pb	58	Sn
95.81	Th	175	Sn
100.96	U	175	Sn

[a] Machine operated at 150 kVp, 15 mA for Sn.



1 primary beam
2 fluorescer
3 filter
4 collimator
5 shutter
6 monoenergetic beam
7 scatter shield
8 TLD holder
9 TLD

Figure 3. Schematic diagram of X-ray fluorescence system.

is directed on one of the fluorescers. The characteristic K-alpha and K-beta x-rays of the particular material are emitted, along with a small amount of scattered primary radiation, at 90^{o} to the incident beam. A few centimeters from the fluorescer is a filter whose thickness can be selected for each fluorescer. The resulting beam is about 96 percent K-alpha and K-beta. Figure 4 shows a spectrum of the silver fluorescer on the 75 kVp machine as measured by a solid state detector and a 512 channel analyzer.

The exposures were treated as if produced by a mono-energetic source whose value was the weighted average of the K-alpha and K-beta energies. The TLD's were placed so that the geometric center of the detector was at the 12 inch calibration distance and in the center of the beam. This position was just behind a three inch opening in a steel box which was designed to reduce exposure from scattered radiation.

To accommodate the unshielded dosimeter, a phenolic cylinder was constructed with a diameter equal to that of the energy shield. Two small holes were then drilled into one end of the cylinder to accept the two electrodes of the dosimeter tube. The cylinder was then placed into the TLD holder. The height of the cylinder held the unshielded dosimeter in the same position relative to the x-ray beam as it would have been if in the shielded configuration. All fluorescent lighting was turned off whenever unshielded TLD's were being handled to reduce the chance of visible light dosing.

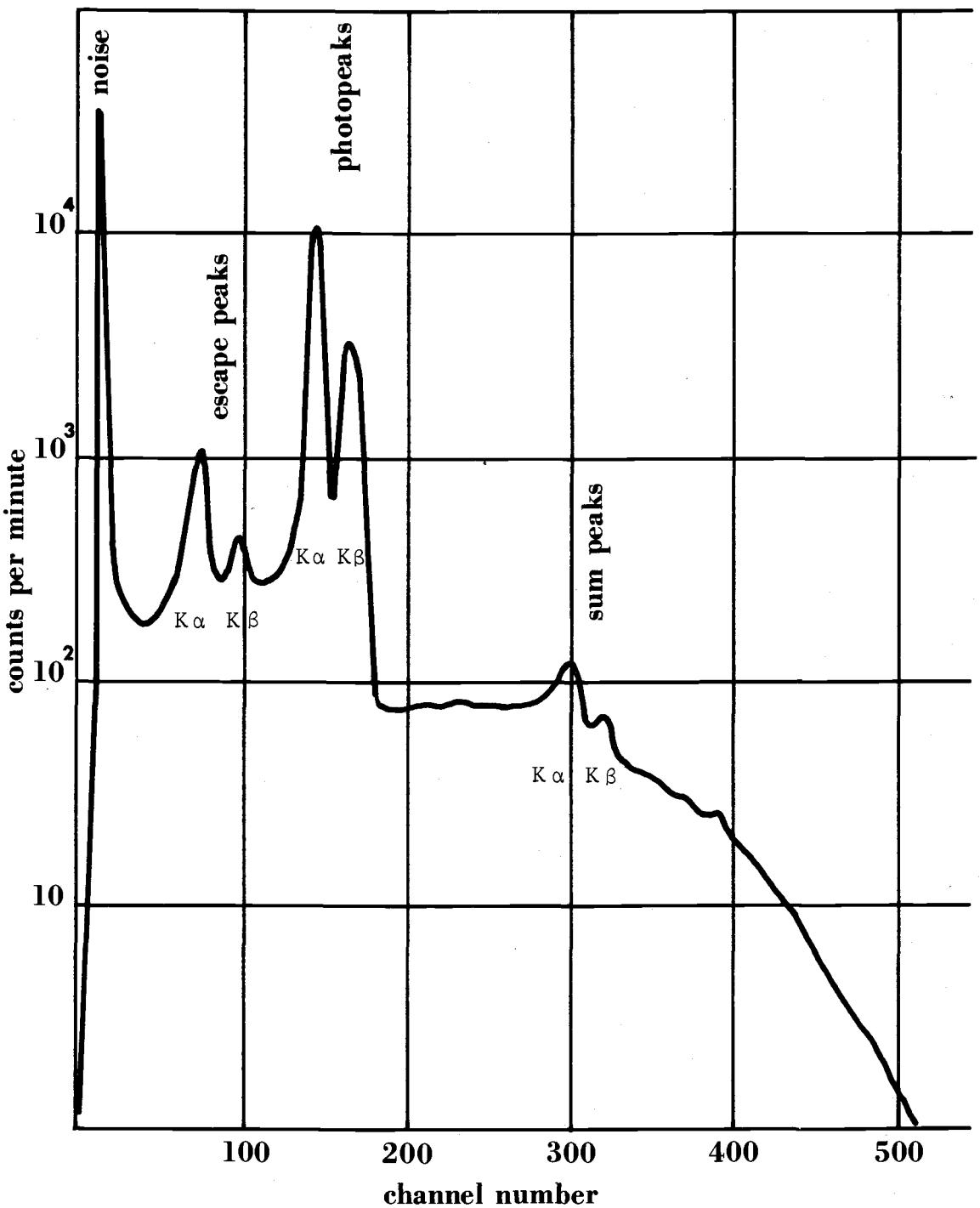


Figure 4. X-ray fluorescence spectrum of silver.

Ten more cylinders were subsequently made of lucite to hold the unshielded dosimeters during the ^{137}Cs and the ^{60}Co exposures, since all ten TLD's could be exposed at once.

Both x-ray machines were calibrated approximately once a week with a free air ionization chamber coupled to a Cary 401 electrometer. Current on the order of 10^{-11} amps was read out on a 10-inch strip chart recorder. Reproducibility from day to day was about $\pm$ one percent.

Exposures were timed with a stop watch and the beam was started and stopped by a lead shutter. Because the 75 kVp machine could be brought up to full voltage in a matter of seconds, each run could be accomplished quickly. The 300 kVp machine, on the other hand, required about five minutes to bring the voltage up in a number of steps to 300 kV. Such a procedure was necessary to prevent dielectric breakdown.

Nearly all exposures on both machines were five minutes long and ranged from 45.6 R for the Fe fluorescer to 2.4 R for the U fluorescer. Timing errors were calculated to be less than 0.3 percent. Eighteen x-ray energies were used in this study.

The TLD response to 662 keV gamma was compared to ionization chamber (R-meter) readings. The dosimeters are normally calibrated by comparing their response to a known ^{137}Cs gamma field on a calibration table built especially for this purpose. See Figure 5. The

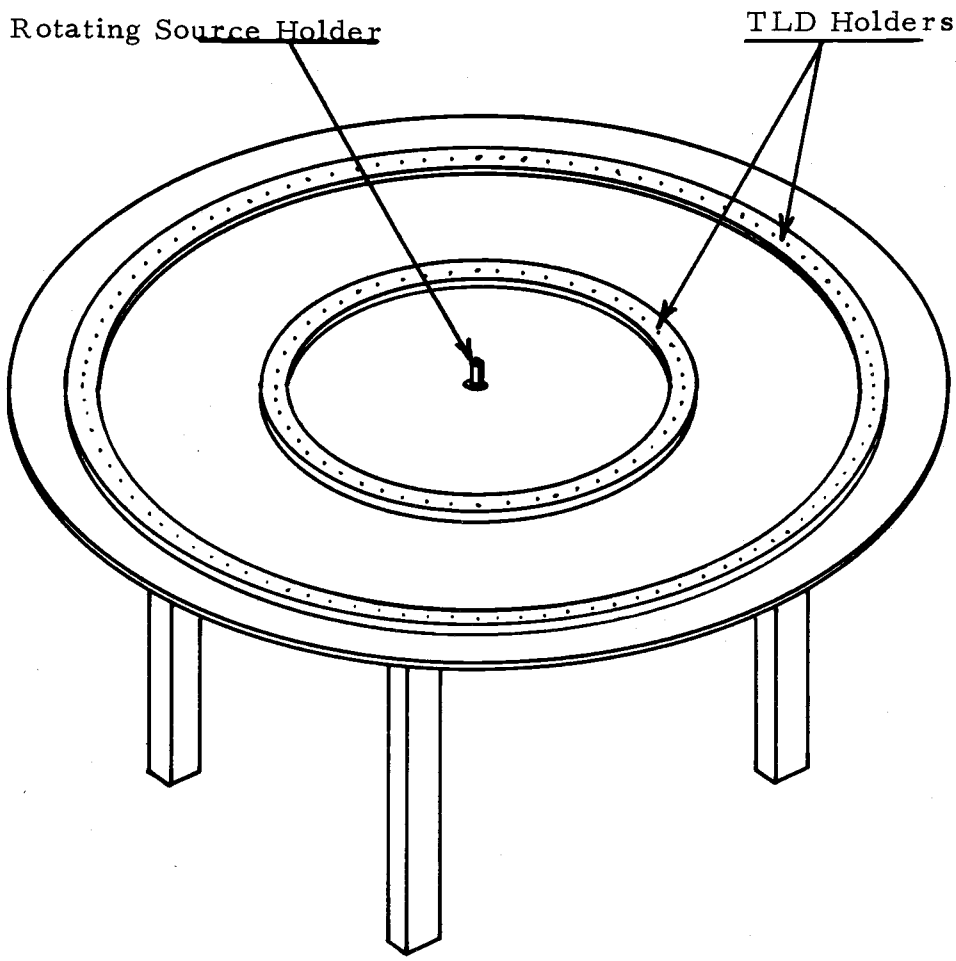


Figure 5. TLD calibration table.

table is a seven foot diameter disk cut from 3/4 inch plywood and supported by four wooden legs. A nylon source holder driven by a one RPM electric motor is mounted in the center of the table. Arranged concentrically around the center are two circles of dosimeter holders, one with a radius of 50 cm, and the other, 100 cm. The dosimeter holders used for normal calibration work are simply holes punched in styrofoam which allow the dosimeters to be placed in vertical positions around the arc.

The ten test dosimeters were placed on the calibration table around the 50 cm arc. The source holder was started in rotation and the source was brought from its pig with a long source handler and quickly placed in the rotating receptacle. At that moment, a stop watch was started. After 30 minutes, the source was returned to the pig. The exposure, calculated from previous calibration of the source, was 322 $\pm$ 16 mR.[5]

The above procedure was to be repeated several times during the linearity study described in the next section, so only one ^{137}Cs exposure was made during the energy response study.

The highest energy source used in the energy response study was ^{60}Co with gamma energies of 1.17 MeV and 1.33 MeV, (average,

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[5] The following calibrations were performed by Sandia Laboratories, Albuquerque, New Mexico, June 18, 1969: ^{137}Cs source No. 122, 1237 $\pm$ 53 mR/hr. at 25 cm (227 mCi), Victoreen chambers, No. 130, X 0.92 and No. 227, X 1.13 $\pm$ 4.5 percent.

1.25 MeV). Ten dosimeters were exposed to 2.00 R at two meters from the ^{60}Co source. The exposure level of 2 R was chosen to be comparable to the higher energy x-ray exposures which had been given, and thus to minimize any exposure level dependence. Holes drilled along a two meter radius arc in a masonite board provided a holder for the dosimeters. The TLD's were positioned so that the geometric center of the detector again intersected the center of the beam as shown in Figure 6.

The ^{60}Co source was remotely operated from a console outside a shielded source room. Exposures were effected by a fast moving shutter in the pig which was controlled by a pre-set timer. Accuracy for the one minute exposure used in this study was claimed by the operator to be better than $\pm$ one percent.

The TLD's used in the energy response study were always read out within an hour after exposure. The thermoluminescent response was compared directly to the calibrated exposure and recorded as a percent of the actual exposure. The same reader was used throughout the study and its gain setting was always set to the same value before reading each batch of TLD's.

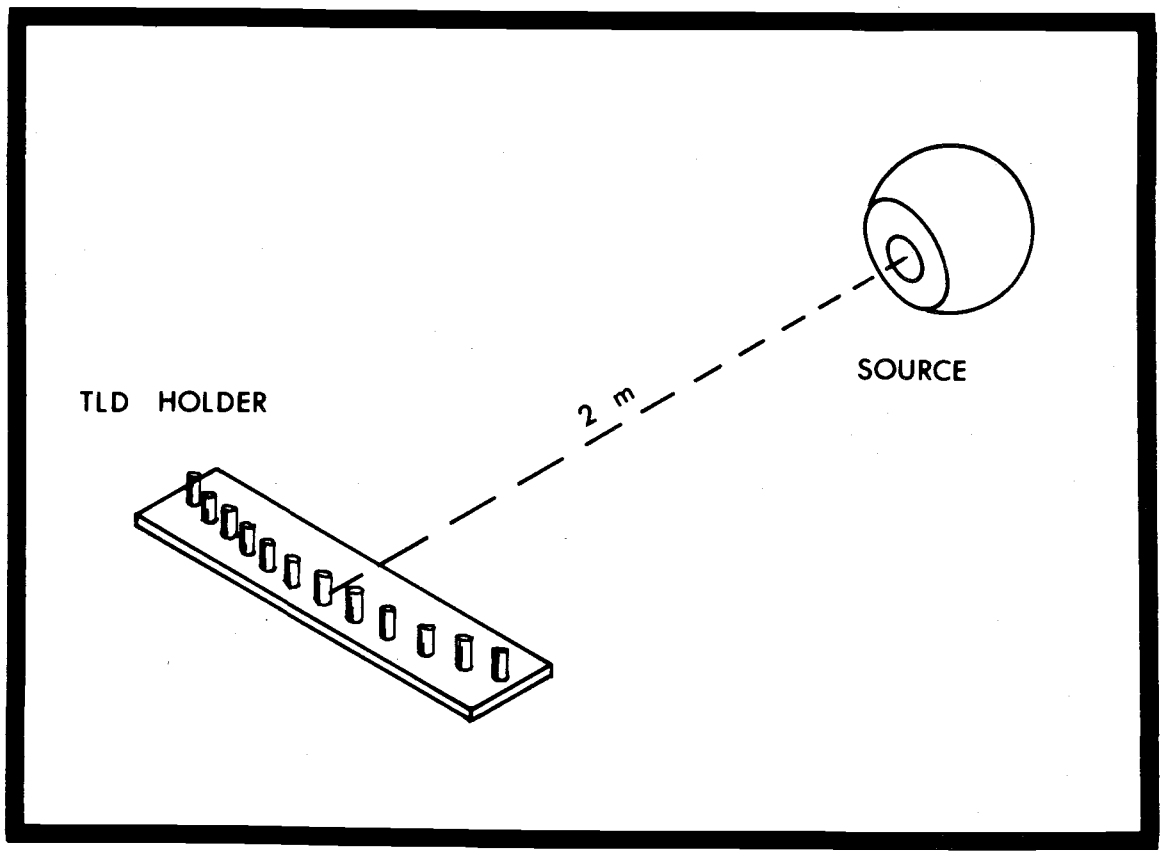

Figure 6. Cobalt-60 exposure facility.

Angular Dependence Study

The angular dependence of the TL-12 response was determined for two energies, 662 keV (^{137}Cs gamma) and 22.6 keV (Ag K-alpha, K-beta x-ray). The response was assumed to be symmetrical around the long axis of the helical detector, but care was taken, nevertheless, to keep the same side facing up for each exposure. The dosimeters were rotated with respect to the source around an axis perpendicular to the axis of the helix. Figure 7 shows the construction of the detector and the energy compensating shield. Rotation was around the geometric center of the shield rather than around the center of the sensitive material which was displaced approximately 1/4 inch from the mid-point of the shield.

The angular response to ^{137}Cs gamma was determined on the circular calibration table. In order to change the orientation of the dosimeter axis with respect to the source direction, a special dosimeter holder was built. The holder was in the form of a shallow trough with a stem underneath which could be inserted into the holes on the table. Figure 8 shows how the TLD holder was designed to keep the center of the sensitive area at the same height as it would be when mounted vertically. This allowed direct comparison of the response in both horizontal and vertical attitudes.

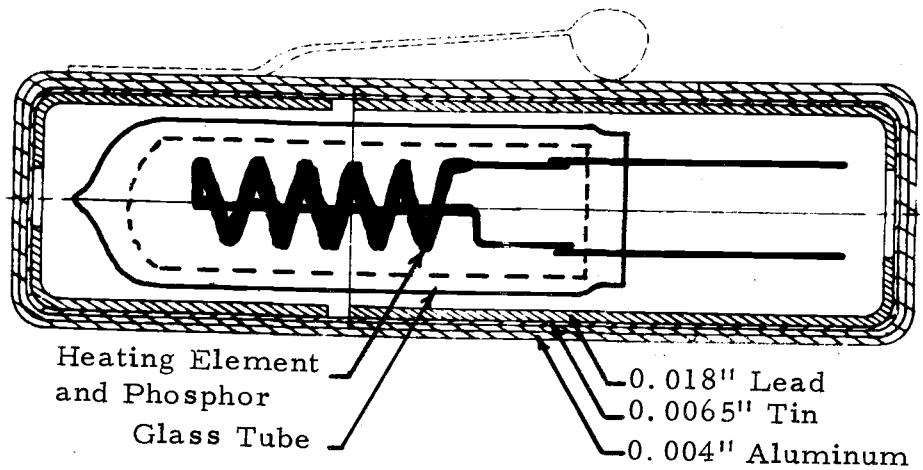

Figure 7. Thermoluminescent dosimeter

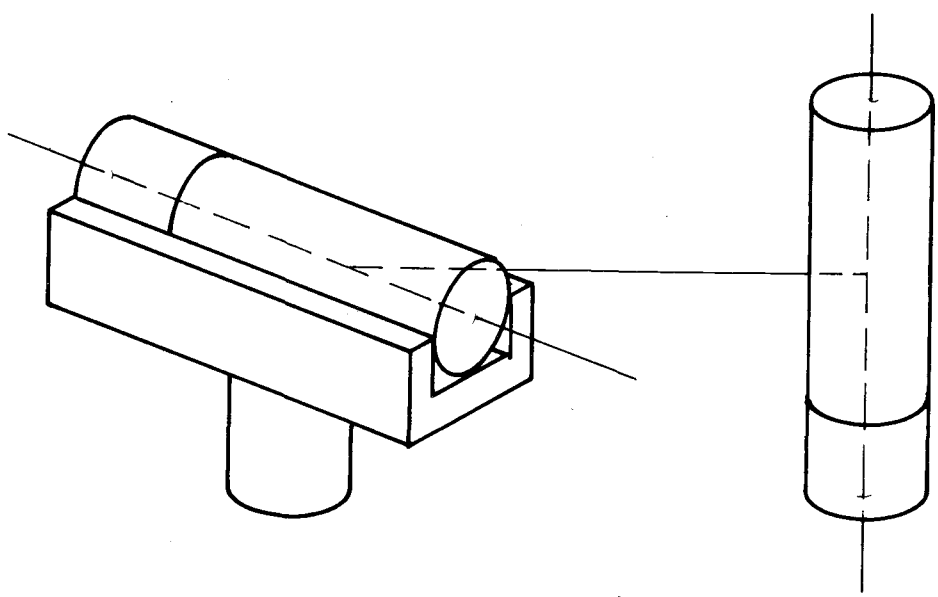

Figure 8. TLD's in holder for angular response and in normal vertical position.

There were 152 equally spaced holes around the 100 centimeter circle and approximately 2.37 degrees between each hole. One hole was chosen arbitrarily to be zero degrees (0^{o}). An array of nine nearly equally spaced positions were then marked off around one-half of the circle. The arrangement can be understood by examining Figure 9. The TLD's in their holders were aligned with a long straight edge so that all the holders were parallel. The straight edge was placed between positions 0 and 76, 9 and 67, 19 and 57, and 28 and 48, whereas positions 38 (90^{o}) and 114 (270^{o}) were aligned by eye. The extra position at 270^{o} was used as a check on the 90^{o} response which in turn was expected to be the same as that of the normal vertical orientation.

Ten dosimeters were used in this study. Every dosimeter was exposed in each of the ten positions which provided ten values to calculate the mean response for each position. Each exposure was timed for 30 minutes in the same manner as described previously. The error in timing was calculated to be less than $\pm$ 0.2 percent. The average exposure for 30 minutes was 32 mR.

Ten additional TLD's were used as controls in this experiment. The position of the controls on the table are indicated by small circles in Figure 9. The controls were placed in the normal vertical orientation. Angular dependence was then calculated by comparing the average response of the TLD's at each test position to the average of the ten controls.

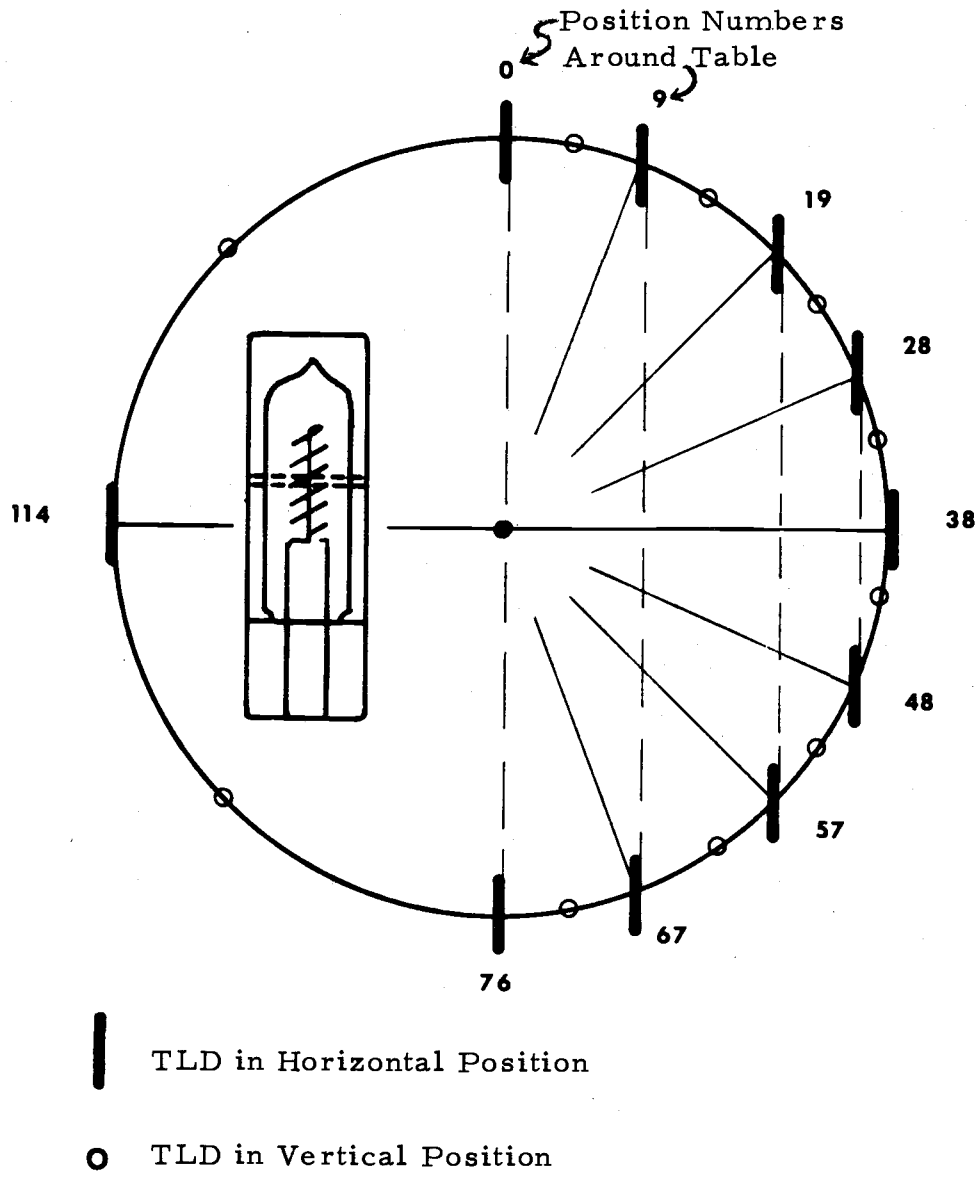

Figure 9. Positions of TLD's on calibration table for angular response study.

To study the angular response to 22.6 keV x-rays, another special TLD holder was constructed which is shown in Figure 10. The holder comprised a vertical sheet of lucite which fitted the standard detector mounting bracket of the x-ray machine and a horizontal plate of lucite upon which was mounted a rotating TLD holder of the type described previously. The horizontal plate was marked off in 22.5^{o} intervals. A center line was scribed in the rotating part so that it could be aligned at any of the eight positions. A TLD was thus positioned so that the center of the x-ray beam intersected the geometric center of the TLD energy shield, and the TLD could be rotated about this point in the horizontal plane.

Since only one TLD at a time could be exposed in this arrangement, no replicate exposures were made. Instead, the angular response of each TLD was compared to the vertical position response to 22.6 keV which had been previously determined for each TLD during the energy response study. Five dosimeters were used in this part of the study. Each exposure was five minutes to Ag K-alpha, K-beta x-radiation from the GE 75 kVp system, producing 5.2 R at the detector distance of 12 inches. The dosimeters were oriented at 0, 22.5, 45.0, 67.5, 78.75, 90.0, 101.25, 112.5, 135.0, 157.5, and 180.0 degrees. Timing errors were calculated to be less than $\pm$ 0.3 percent.

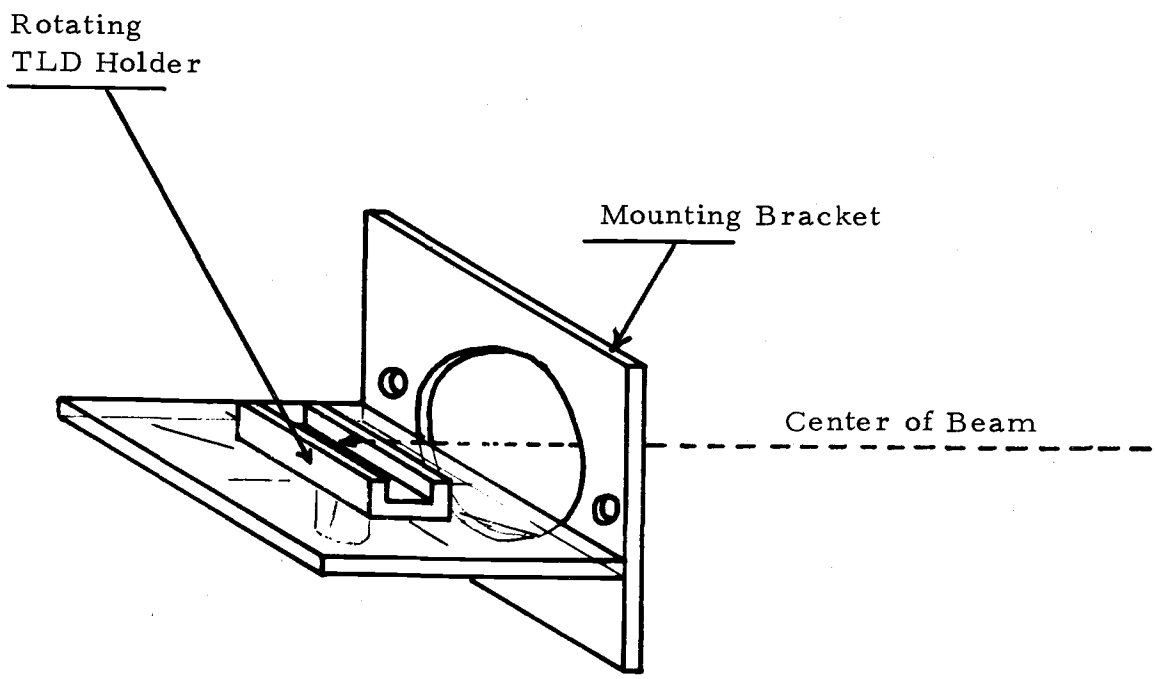

Figure 10. TLD holder for angular response study on X-ray machine.

Exposure Level Dependence Study

Ten dosimeters were given gamma exposures ranging from 3.0 mR to 3000 R. Two facilities already described were necessary to produce the desired range of exposures, the ^{137}Cs source on the calibration table, and the large ^{60}Co source at EG&G.

Using two energies in this study was a possible source of error, but it was known that data obtained in the energy response study could be used, if necessary, to correct for differences in response. As will be shown, the difference in response between 662 keV and 1250 keV was negligible compared to the effects of various exposure levels.

In order to produce a range of exposures from .003 to 3000 R in a reasonable length of time, it was necessary to increase the exposure rate for higher exposures. The possibility of rate-dependence was not tested in this study, although at least one investigation (6) suggests there may be some rate dependence, even in CaF_2:Mn. It was assumed that rate dependence was negligible within the limits of experimental error as found by this and other investigations (6, 18).

A schedule of exposures, shown in Table 2, was prepared which took the ten test dosimeters through the full range of exposures in seven decades, beginning with .003 R and ending with 3000 R. The factor three was chosen because it represented approximately

Table 2. Schedule of Exposures for Linearity Study.

Run	Exposure Given (R)	Historical Exposure[b] (R)
1	.100[a]	.100
2	.003	.103
3	.100	.203
4	.030	.233
5	.100	.333
6	.300	.633
7	.100	.733
8	3.000	3.733
9	.100	3.833
10	30.000	33.833
11	.100	33.933
12	300.000	333.933
13	.100	334.033
14	3000.000	3334.003
15	.100	3334.103

[a] A 100 mR test exposure was given initially and then after each exposure of the linearity study to test for changes in TLD sensitivity.

[b] The Historical Exposure is for the period of the study only, and is the sum of successive exposures including the 100 mR test exposures.

mid-scale on the reader output. To test for the possibility of exposure damage or permanent changes in TLD sensitivity during the experiment, a 100 mR ^{137}Cs test exposure was given after each exposure of the series. The low exposures in the series were also given with the 200 mCi ^{137}Cs source. Exposures above 300 mR had to be given by the ^{60}Co source to expedite the experiment.

To minimize the effect of early fading, the TLD's were read 24 hours after exposure in each case. For example, 24 hours after the 300 mR exposure, the dosimeters were read, then exposed to a 100 mR test exposure, either that day or as soon as possible thereafter. Another 24 hours elapsed before the test exposed TLD's were read. The 3.0 R exposure was given next, and so forth. Since the reproducibility of the 100 mR test exposure was better than $\pm$ one percent, the procedure provided a good check on changes occurring in either the TLD's or the reader.

After the original schedule of exposures was completed, a second, but shorter series was given to complement the data. The exposure range of most interest to the dosimetry program is from a few mR to about 1 R; therefore, the second series of exposures was designed to obtain data in this important range. Ninety TLD's were arranged around the calibration table and the ^{137}Cs source was put in place. The TLD's were then removed from the table in groups of 10 at predetermined time intervals to provide nine exposure levels -

3, 10, 30, 200, 300, 400, 500, 700, and 900 mR. The dosimeters were read 24 hours after exposure, and the response was normalized to the percent of a 100 mR exposure by use of recently determined correction factors.

RESULTS

Energy Dependence Study

During the initial phase of the energy response study, ten dosimeters were exposed, one at a time, to seven of the available energies on the 75 kVp machine, 6.47, 8.74, 15.20, 17.78, 22.59, 25.84, and 32.99 keV. The data revealed that five of the dosimeters were "better behaved" than the other five, i.e., the responses to a given energy were similar, and the magnitudes of the responses remained in the same order with respect to each other. The other dosimeters were more erratic; the range of responses was greater, and the relative positions of the response values changed from time to time.

The original contract for x-ray services called for 1000 minutes[6] of machine time plus 1000 minutes of set-up time. The relatively fast operation of the 75 kVp machine allowed some extra exposures to be made, thus it was possible to expose ten TLD's at first, and then choose the five best behaved dosimeters for the remainder of the study. The more erratic behavior of some of the TLD's indicated a greater variation could be expected from a randomly selected group of dosimeters than from a more carefully selected group.

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[6] 5 TLD's x 20 energies x 5 minutes x 2 runs, shielded and unshielded = 1000 min.

An exposure "run" consisted of either five or ten five-minute exposures at one or more energies. The first few runs used the full compass of energies so that the proper exposure level could be determined. A five minute exposure was adequate for all energies. Mixing energies with runs also reduced the bias which might have been introduced into the results from run to run. Usually only one run was made per day.

Table 3 is a listing of the average energy response for the shielded dosimeters. The average relative response to ^{137}Cs, 662 keV gamma, is shown in the third column. The error term in the fourth column is a one sigma estimate based on the number of replicate exposures for each energy.

Table 4 lists the energy response data for the unshielded dosimeters. There were energies which had fewer than five replicate exposures in portions of the study because several weeks elapsed while the 300 kVp machine was inoperable. Since the variation within the group of five TLD's was small, two or three replicates were considered sufficient.

A graphic display of the energy response data is presented in Figure 11. The response of each TLD was normalized to 662 keV. The average response of the group was then plotted as the percent of total exposure versus energy.

Table 3. Energy Response Data, Shielded Dosimeter.

Energy (keV)	Average Percent Response to Given Exposure	Average Normalized Percent Response	Coefficient of Variation of 5 Dosimeters
6.47	0.0117	0.0150	5.5
8.74	0.0160	0.0206	14.5
15.20	0.232	0.298	39.6
17.78	0.791	1.02	17.5
22.59	4.68	6.02	14.4
25.84[a]	11.3	14.5	8.0
32.99	8.22	10.6	7.1
25.84[a]	11.4	14.7	5.5
38.31	13.9	17.9	4.5
41.16	15.8	20.3	7.5
44.13	19.4	25.0	7.7
52.11	33.7	43.4	8.9
53.80	36.4	46.8	4.5
57.30	45.7	58.8	8.0
60.94	53.2	68.5	3.6
68.66	71.7	92.3	8.2
77.00	84.5	109.0	5.6
95.81	64.1	82.5	8.4
100.96	65.2	83.9	5.1
662.0	77.7	100.0	4.4
1250.0	79.8	103.0	6.6

[a] The Sn fluorescer (25.84 keV) was used on both x-ray systems to provide a calibration crosscheck.

Table 4. Energy Response Data, Unshielded Dosimeter.

Energy (keV)	Average Percent Response to Given Exposure	Average Normalized Percent Response	Coefficient of Variation
6.47	0.684	1.29	4.2
8.74	0.948	1.79	5.2
15.20	82.5	156.0	3.0
17.78	147.0	278.0	6.1
22.59	298.0	563.0	5.2
25.84[a]	353.0	667.0	4.9
32.99	382.0	722.0	4.0
25.84[a]	468.0	885.0	5.3
38.31	508.0	960.0	3.9
41.16	528.0	998.0	7.1
44.13	533.0	1007.0	5.1
52.11	564.0	1066.0	0.6[b]
53.80	537.0	1015.0	1.3[b]
57.30	462.0	873.0	9.6[b]
60.94	458.0	866.0	0.6[b]
68.66	428.0	809.0	9.7[b]
77.00	294.0	555.0	17.6[b]
95.81	294.0	555.0	8.4[b]
100.96	237.0	448.0	3.0[b]
662.0	52.9	100.0	3.7
1250	79.2	150.0	4.3

[a] The Sn fluorescer (25.84 keV) was used on both x-ray systems to provide a calibration crosscheck.

[b] Two replicate exposures, all others had five.

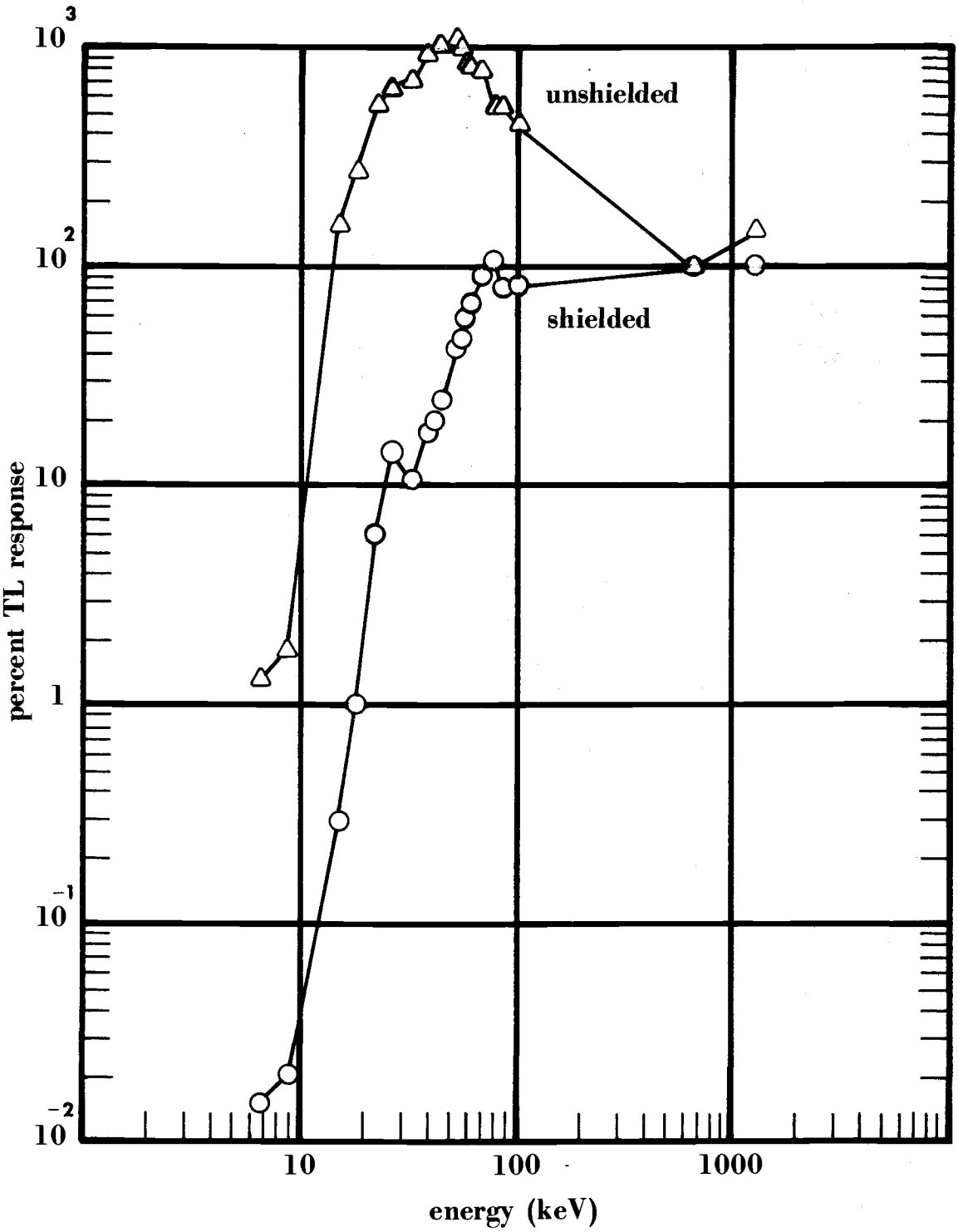

Figure 11. Energy response.

Angular Dependence Study

The angular dependence of the TLD followed a pattern in keeping with the geometry of the detector. The sensitive element of the dosimeter (Figure 7) is a helix approximately 0.2 inches in diameter and 0.6 inches long. The energy compensating shield is a cylinder with an annular window ringing the middle of the detector and a circular window in each end. The dosimeter presents the greatest surface of TL material when the radiation is perpendicular to the long axis. The least surface is presented when the radiation is parallel or end-on.

One would expect the greatest TL response to be at 90° and the least at 0° and 180°. The difference between the two extremes would be expected to increase as the energy of the beam decreased. The data, presented in Table 5, show these assumptions to be correct.

Figure 12 is a polar graph of the relative angular response to 662 keV and 22.6 keV. The response at 90° is considered to be 100 percent in both cases, whereas, in actuality, the TL response to 22.6 keV is only about five percent of the response to 662 keV. It can be seen from the graph that there is little angular dependence at 662 keV, approximately 20 percent between the extremes. It is interesting that the minimum response is not at 0° and 180°, but closer to $\pm 21^{\circ}$ from

Table 5. Angular Response Data.

Angle in Degrees	Average Corrected Percent Response to Given Exposure	Average Relative Percent Response to 90°
662 keV		
0	85.2	83.4
21.3	79.5	77.9
45.0	94.2	92.3
66.3	98.5	96.5
90.0	102.0	100.0
113.7	98.9	96.9
135.0	94.5	92.6
158.7	89.8	88.0
180.0	95.8	93.8
270.0	102.1	100.0
22.59 keV		
0	0.050	1.55
22.5	0.054	1.67
45.0	0.375	11.6
67.5	1.59	49.2
78.75	2.42	74.9
90.0	3.23	100.0
101.25	2.19	67.8
112.5	1.58	48.9
135.0	0.24	7.43
157.5	0.053	1.64
180.0	0.048	1.49

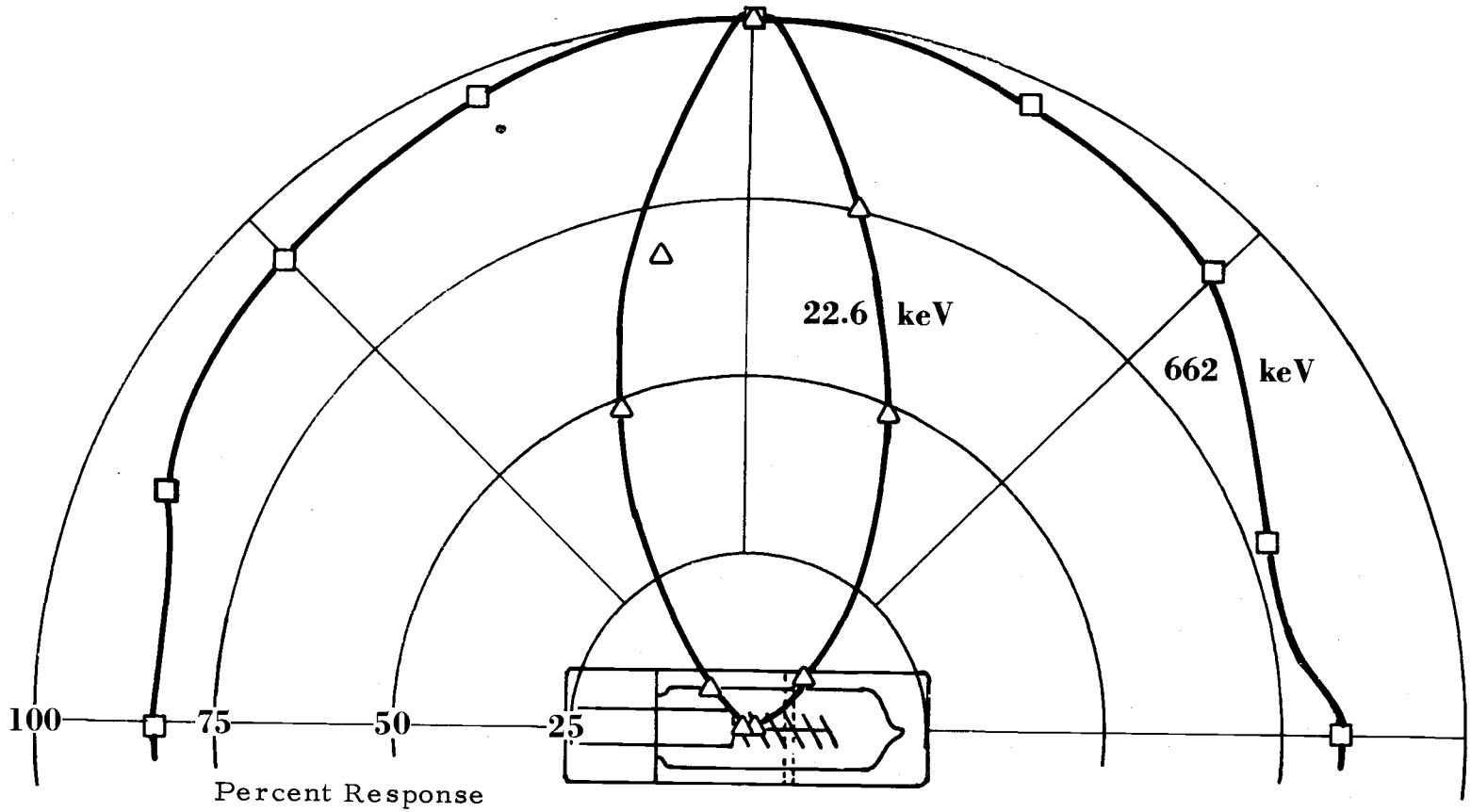

Figure 12. Angular response

these two directions. The effects of the end window and the corners of the shield are most likely being manifested here.

The range of angular dependence for 22.6 keV is about two orders of magnitude wide. The response falls off rapidly from 90^{o} and decreases to about one percent of that value at 0^{o} and 180^{o}. The end window effect was not noticed in the low energy response. It is probable that the effect of self-shielding in the helix is a more important cause of angular dependence than the effect of the energy shield at such low energies.

Exposure Level Dependence Study

Ideally, a good dosimeter should have a linear response to varying exposure levels. This study revealed that there was a non-linear behavior for the TLD system. It was not determined which component, the dosimeter or the reader, if not both, was responsible for the non-linearity. In general, there was an over response at very low exposures and at high exposures. The minimum response occurred between 100 mR and 1.0 R.

Table 6 lists the TL response, the corrected response, and the relative response for all the dosimeters tested in the series. Data from the 100 mR test exposures given after the exposures in the series are listed in Table 7.

Table 6. Exposure Level Response Data.

Exposure (R)	Average Corrected Percent Response to Given Exposure	Coefficient of Variation of 10 Dosimeters
First Series		
0.003	89.0	8.8
0.030	92.5	8.2
0.100	100.0	7.0
0.200	95.5	1.4
0.300	93.6	2.6
3.00	105.0	1.4
30.0	104.0	3.3
300.0	123.0	6.2
3000.0	132.0	4.2
Second Series		
0.003	106.0	6.2
0.010	99.6	1.4
0.030	98.3	1.6
0.100	100.0	1.4
0.200	99.8	2.4
0.300	98.5	1.3
0.400	101.0	2.2
0.500	98.3	2.7
0.700	102.0	4.0
0.900	95.3	1.0

Table 7. One Hundred mR Test Exposure Data.

Cumulative Exposure (R)	Average Percent Response to 100 mR Test Exposure	Coefficient of Variation of 10 Dosimeters
0.100	80.6	7.0
0.103	a	
0.133	a	
0.533	81.9	7.4
3.633	85.3	9.1
33.733	82.7	9.4
333.833	80.8	7.8
3333.933	81.0	6.5

[a] No 100 mR test exposure was given after the 3 and 30 mR exposures.

The results of this study are best summarized by Figures 13, 14, and 15. The linearity of a dosimeter is usually depicted as in Figure 13, TL response versus exposure. A straight line with a slope of 45 degrees seems to fit these points quite well, although, upon close observation, there appears to be a slight supralinearity to the data. Figure 14 exaggerates this non-linearity by plotting the percent response versus exposure. The ideal dosimeter would display a horizontal line at 100 percent throughout the range of exposures. The greatest departure from the ideal response occurred at exposures above one roentgen.

The data which were collected in the second part of the experiment are plotted as triangles. It was hoped that these points would confirm a trend which appeared during the first part of the experiment (circles). The response did decrease again from 100 mR to 300 mR, but then became very erratic from 400 mR to 900 mR. The variation is small, however, and the TLD response seems quite flat between 10 and 700 mR.

The true nature of the response to varying exposures is left undertermined, although it is obvious that the system is not as linear as was once hoped. Results from the energy response study should rule out any effects of changing from ^{137}Cs to ^{60}Co during the linearity experiment. The effects of rate dependence, on the other hand, cannot be ruled out at this time.

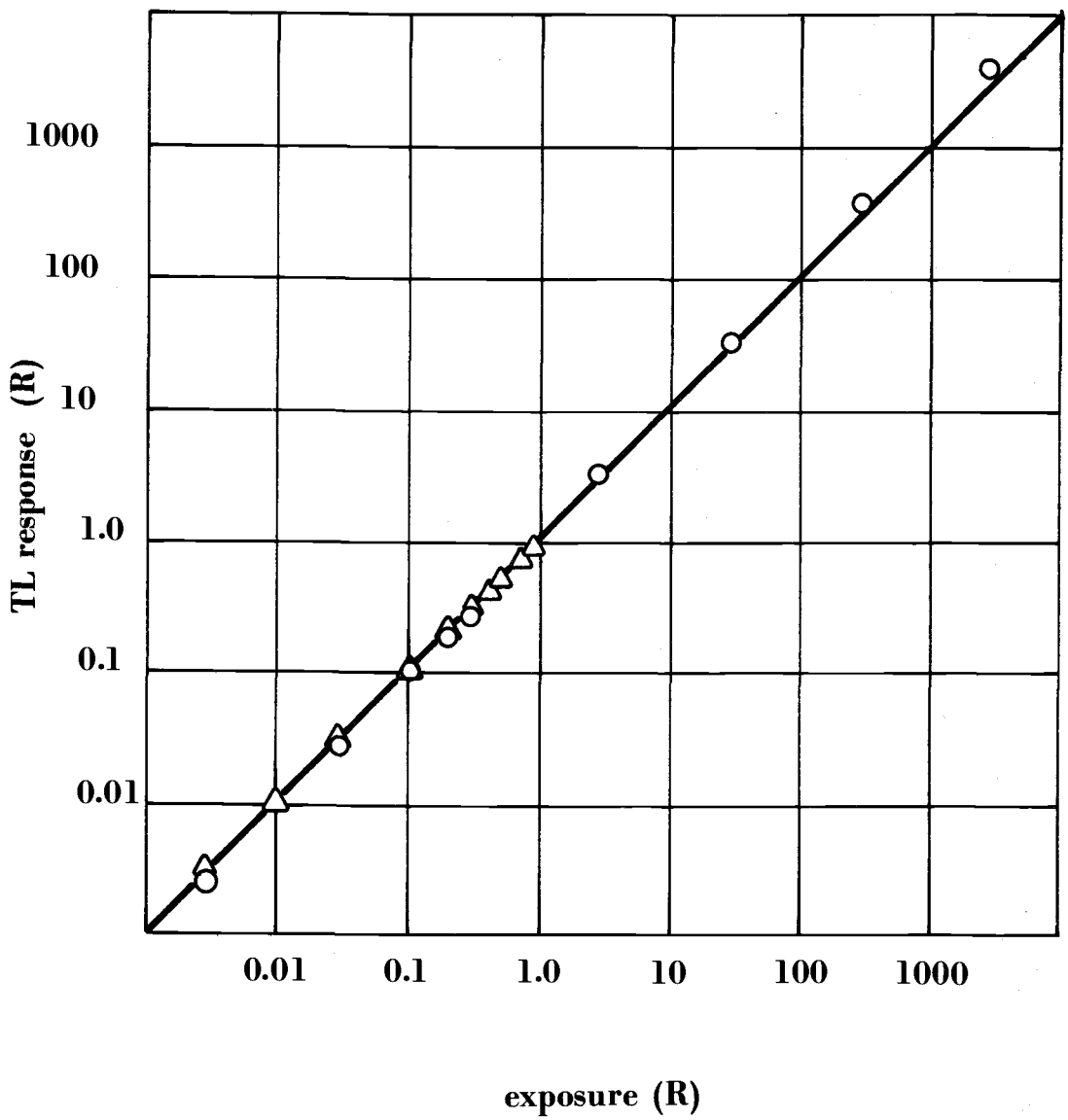

Figure 13. TL response versus exposure.

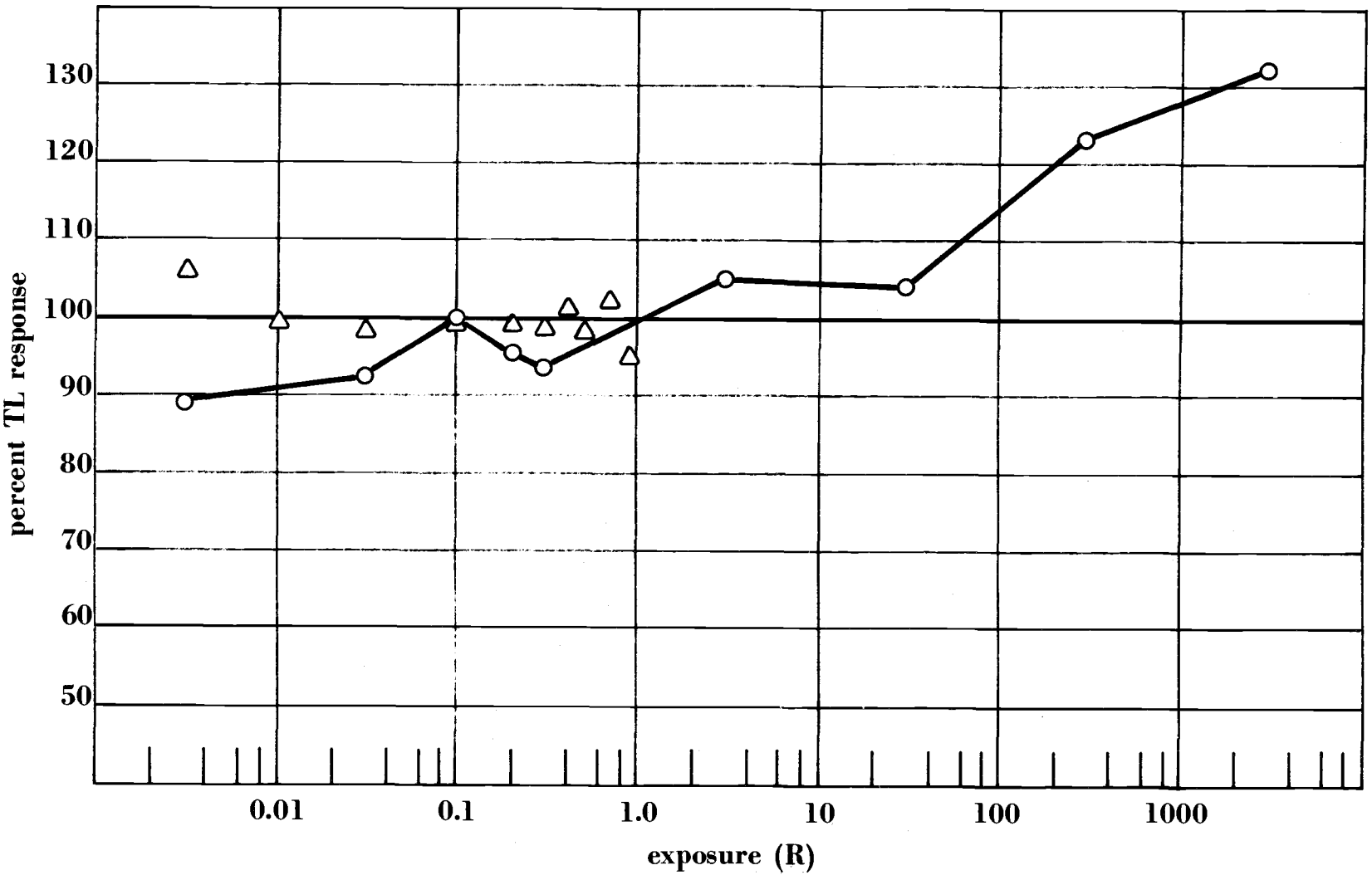

Figure 14. Percent TL response versus exposure.

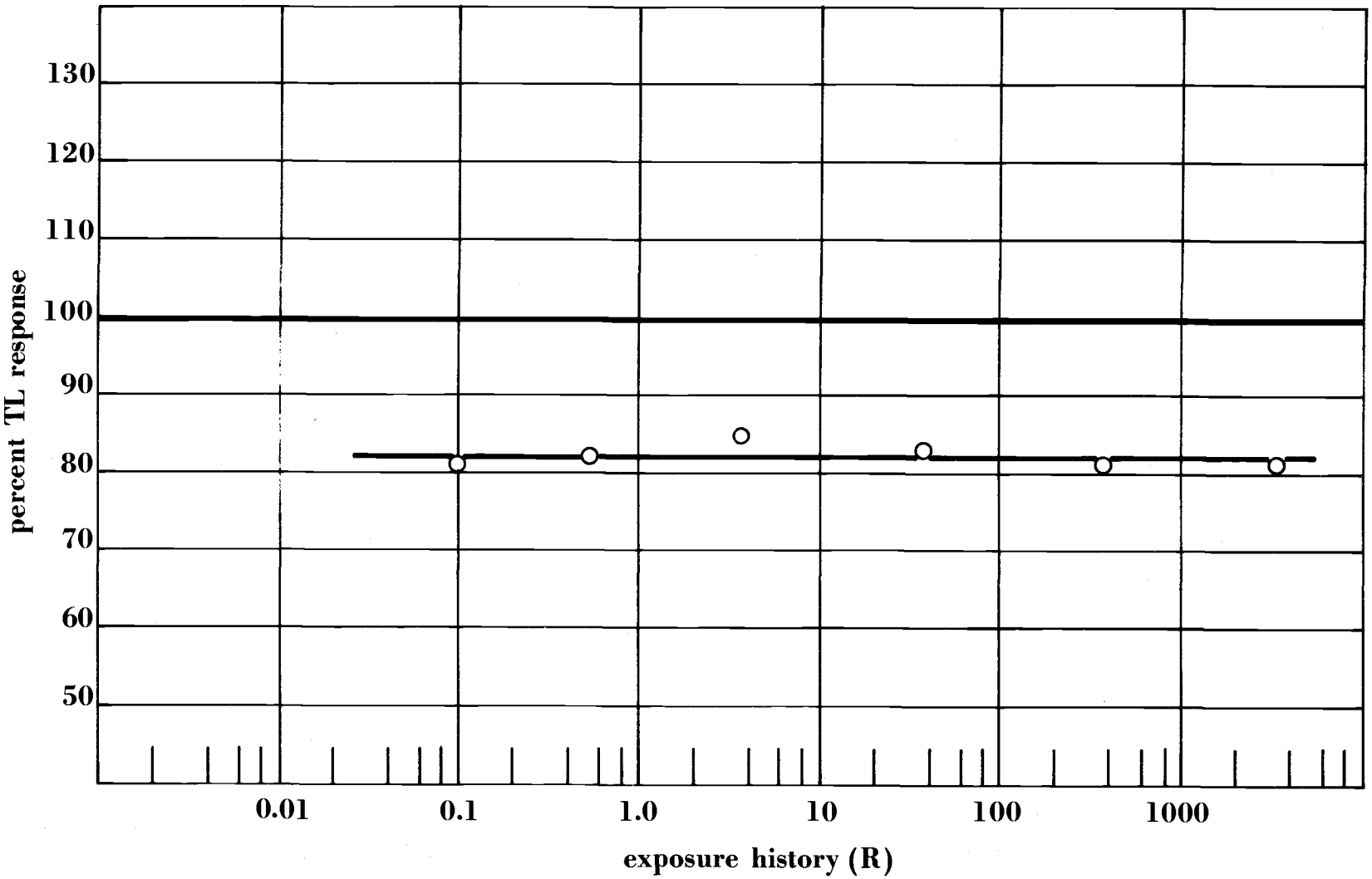

Figure 15. Percent TL response to 100mR test exposures.

Most of the error associated with exposures below 100 mR probably can be attributed to uncertainties in the internal background value and in the estimate of a 24 hour natural background exposure. The sum of the two backgrounds is approximately one milli-roentgen per day.

The response to 200 mR has habitually been less that the response to 100 mR ever since the system was put into use. Routine TLD calibrations were performed at both 100 and 200 mR for several years. The gain of the reader always had to be increased to make the standard TLD's direct reading, but the increase was not always constant. The amount of data on hand is sufficient to point out a problem but is still too limited to permit a systematic explanation.

Trends in reader response over the period of the experiment are ruled out because of the data presented in Figure 15, which plots the percent response to the 100 mR test exposure versus the exposure history of the dosimeters. No significant change in the system response was apparent during the course of the experiment.

DISCUSSION AND CONCLUSIONS

The shielded TL-12 dosimeter has a rather sharp response cut-off below about 70 keV (See Figure 11). The response drops nearly four orders of magnitude from 70 keV to 6.5 keV. The "flat" portion of the response curve ranges from 70 keV where the response is down eight percent to 1250 keV where it is up three percent. In general, the variation in response is bounded by $\pm$ 10 percent of the response to 662 keV gammas over the range from 70 keV to 1250 keV.

The K-absorption edges of tin and lead occur at 29.190 keV and 88.001 keV respectively. The effect of these two metals in the dosimeter shields can be seen in Figure 11. The absorption edges have an overall smoothing effect on the response curve, although they produce rather sharp changes in response near the K-absorption energies. The unshielded response curve shows no K-absorption edges, because the characteristic absorption energies of the glass which is the only absorber present are much lower than the range of energies investigated. The 50 percent over-response of the unshielded dosimeter at 1250 keV was unexpected.

The energy response curves presented in Figure 11 should be representative of all the TL-12 dosimeters and therefore can be used to correct the TL response if the energy of the measured radiation is known. The curves are most complete and therefore most useful for

energies below 100 keV. The large gap in data points from 100 keV to 662 keV makes this region of the curve nearly useless, especially in the unshielded case. Since the dosimeter is always used in the shielded configuration, at least for higher energies, the lack of data does not present much of a problem. The energy response for the shielded dosimeter above 100 keV is reasonably flat.

Knowing the energy response of the unshielded dosimeter well suggests a possible extension of the use of the device. The dosimeter may be useful to measure low energy radiation, such as from medical x-ray installations, if a suitable light shield were devised which would not attenuate the x-rays appreciably. Such a light shield could easily be made of black opaque plastic in the form of a small envelope. The only, but very important, requirement for using the dosimeters in this manner would be to know the effective energy of the radiation to be measured. The over-response of CaF_2:Mn to low energy radiation could be an asset since the sensitivity of the dosimeter would be enhanced about ten fold.

The angular dependence of the TL-12 was tested at the normal calibration energy, 662 keV, and at a low x-ray energy, 22.6 keV. The results show that if the radiation field to be monitored has a known preferred direction, then for best results the TLD must be oriented so that its long axis is perpendicular to the incident radiation. Only then will the calibration and energy response corrections be valid.

It can be seen in Figure 12, that for low energies, the angle is critical, whereas for the energy of ^{137}Cs, it is not so critical. The worst situation for ^{137}Cs produces a 20 percent drop in response.

Most environmental monitoring involves either a nearly isotropic source with the detector immersed in it, or a large or infinite plane source such as a cloud overhead or fallout on the ground. In these cases, there is a range of angles of incidence and the TLD response is an integral over a solid angle. Since the calibration factors are derived for the highest response situation, i.e. 90^{o}, the response to other than a point source would be expected to underestimate the exposure. In the case of ^{137}Cs, the response curve of Figure 12 can be approximated to an ellipse with the semi-major axis equal to 1.0 and the semi-minor axis equal to 0.8. The ratio of the area of the ellipse to a unit circle is 0.81. Since the response integral is proportional to the area of the response curve, the dosimeter could be expected to underestimate an isotropic field of 662 keV gamma by 19 percent.

If the radiation field is well defined for both energy and direction, then it may be appropriate to correct for the angular response. In most cases, however, the field is not well defined and the 90^{o} calibration figure is the best estimate, and any other adjustments in the exposure value would be unwarranted. The angular dependence

changes slowly near 90^{o} for the higher energies, and if some care is taken to orient the dosimeter for its best response, no correction will be necessary for most applications.

The dosimeter was found to be linear within $\pm$ five percent over the exposure range of most interest to environmental monitoring, i.e., from a few milliroentgens to about one roentgen. In Figure 14, the solid curve connects only the data points of the first linearity experiment to show the trend more clearly. The data of the second experiment, plotted as triangles, had less variation than the first. The only significant departure from linearity occurred at 300 and 3000 R, where there was a 23 percent and 32 percent over-response, respectively.

A trend of decreasing response between 100 and 200 mR was seen but the nature of the trend was not confirmed. If it does exist, the degree of departure from 100 percent is about the same order of magnitude as the inherent error in the system, i.e. five percent. Since all exposure results depend on the calibration factors derived at 100 mR, it is essential for the sake of accuracy that the response at other exposure levels be known if the response is indeed not the same.

Although the curve generated by this study is not yet well founded enough to be used for exposure corrections, such a curve is being developed by further work. The plan is to develop a curve (and its

function if possible) which can be applied to all the TL-12 dosimeters. First the read-out would be corrected by the 100 mR calibration factor, and then corrected further by an exposure dependent factor obtained from the curve.

It is possible that each batch of dosimeters, if not each dosimeter, will have a unique curve of its own. If such is the case, it may be more practical to simply calibrate each dosimeter at several points in the range of interest. It was such a practice that suggested that there was an exposure dependence in the first place. The dosimeters were calibrated at 100 and 200 mR for some time and it was noted that the relative response to 200 mR was consistently down a few percent compared to the response to 100 mR. Typical monthly background exposures measured in the environment with the TL-12 (including the internal background) are about 30 mR; therefore, 30 mR would be one obvious choice for calibration should it be necessary.

Although the problems of monitoring the environment for low levels of radiation exposure are many and varied, reasonably good data can be obtained by using a suitable dosimeter. Any dosimeter can be used which has the required sensitivity and good reproducibility. The problems of accuracy, energy dependence , angular dependence, non-linearity, and other response characteristics peculiar to the particular dosimeter can be overcome. The characteristics

of the chosen dosimeter must be documented in detail in order to get the most out of the device, that is, to obtain from it the best possible estimate of the exposure being measured.

The data collected during the course of this study have augmented the description of the TL-12 dosimeter. The results will now be used to refine the exposure estimates being reported in the off-site Dosimetry Network. It is hoped that the information will be helpful to other users of this dosimeter.

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