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The Hanford Environmental CaF_2:Mn Thermoluminescent Dosimeter

by
J. J. Fix
M. L. Miller

March 1978

Pacific Northwest Laboratory
Richland, Washington 99352
Operated for the
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INTRODUCTION

Beginning during January 1977, CaF_2:Mn (TLD-400[a]) thermoluminescent dosimeters were introduced into the Hanford Environmental Surveillance Program to measure ambient penetrating radiation attributable to cosmic and terrestrial radiation as well as any contribution from Hanford facilities. This report describes the routine procedures used to anneal, package, and read out the chips as well as methods used to interpret the data in terms of dose. Additional information pertaining to the energy dependence of the lead-tantalum field capsule, directional dependence of the capsule, and methods used to optimize the readout procedure is also given.

[a] Harshaw Chemical Company.

SUMMARY

The TLD-400 chips combined with the Pb-Ta field capsule provide a sensitive method of measuring penetrating ambient radiation in the environment. The method is best used for field deployments of about 1 month or less to minimize problems associated with fading. A correction factor of about 10% is necessary for the readings obtained for a 28-day field deployment and a 1-day wait before readout. This factor is only an approximation; the use of any adjustment is dependent on the calibration methods employed.

Integration of reader output from 150°C to 280°C provides a good signal-to-noise ratio for TLD-400 chips exposed to 5 mR for the reader and planchet described herein. Visual inspection of the glow curves is recommended during startup of any new program or following any major instrument repair. The glow curves can be easily drawn with an X-Y recorder.

Because of the large energy dependence of bare TLD-400 chips, an energy-flattening filter is necessary to allow a direct conversion from a reference exposure to observed field exposures. The field capsule used, consisting of 10 mil of tantalum and 2 mil of lead, provides an approximate uniform energy response above 70 keV. Below 70 keV, the response decreases rapidly because of the shielding.

Experiments conducted have not shown the TLD-400 chips to be sensitive to the extremes of summer temperature (~50°C) occasionally encountered at Hanford. Although the field dosimeter exhibits a directional dependence, this is of primary concern during calibration, in which the axis of the dosimeter should be normal to the photon beam. The procedures used to interpret the TLD chip reader output in terms of dose are fully described in the text.

TLD-400 ENVIRONMENTAL DOSIMETER

The Environmental Surveillance Program TLD System consists of CaF_2:Mn chips, an energy-flattening field capsule, and a TLD reader. The reader is currently a Harshaw 2000 Model B manufactured by the Harshaw Chemical Company.[a] During readout, the heating chamber is purged with dry nitrogen gas at ∿7.5 ft^3/hr (3.5 ℓ/min).

Each field dosimeter consists of three to five Harshaw TLD-400 chips placed in a slitted polystyrene foam plug which is then fitted into an energy-response-flattening capsule. The capsule itself consists of a tube of 10-mil (0.254-mm) tantalum + 2-mil (0.051-mm) lead with an appropriate end cap held together with heat-shrinkable, opaque vinyl tubing (see Figure 1). The vinyl tubing serves to hold the energy-flattening filter securely in place and to provide a light-tight exterior.

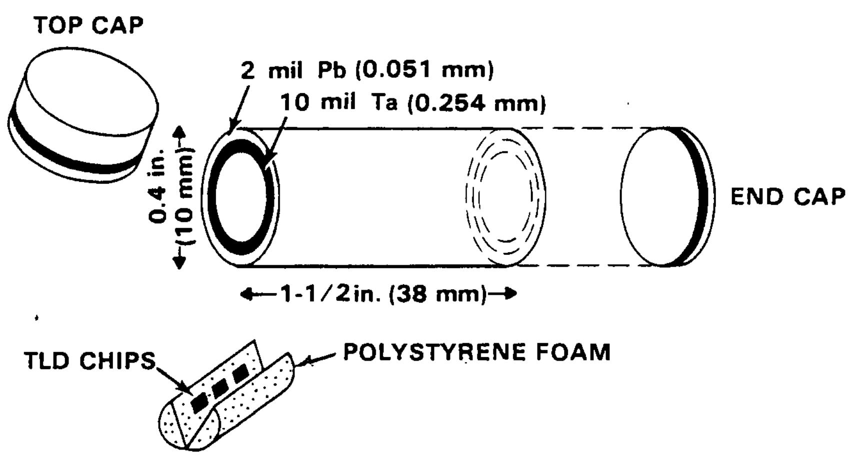

FIGURE 1. Energy-Response-Flattening Filter

(a) Reference to a company or product name does not imply approval or recommendation of the product by the Department of Energy to the exclusion of other suitable products.

ENERGY DEPENDENCE

CaF_2:Mn exhibits a pronounced energy dependence, primarily because of the atomic number of Ca (20). Although the atomic number of Mn (25) is also relatively high, the effect of Mn is insignificant because of its extremely low concentration (∿ppm). Empirical energy dependence data for both the bare chips and the chips in the energy-compensating filter are shown in Figure 2 in which the data were normalized to the response obtained from ^{60}Co (∿1250 keV). The maximum over-response observed occurred at 40 keV and is a factor of 11 greater than a unit response. This over-response is similar to the results obtained by Denham et al. for CaF_2:Dy chips.(1)

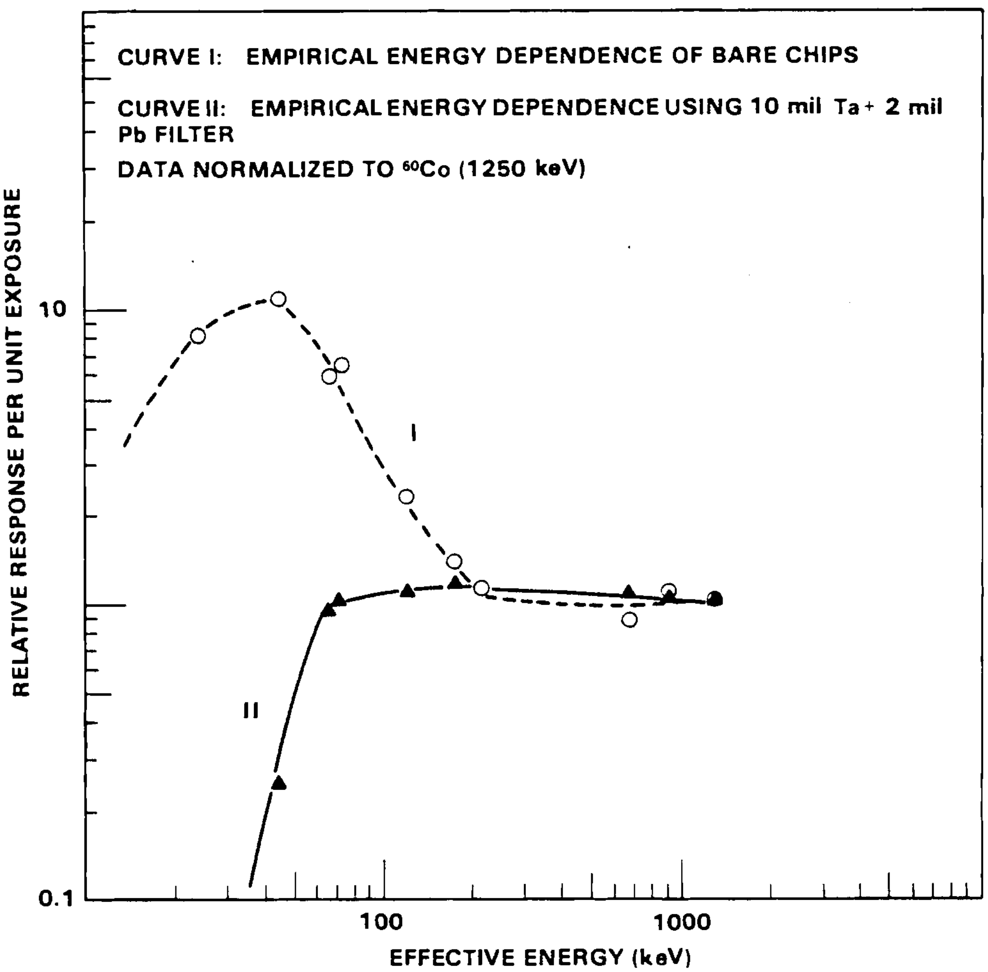

FIGURE 2. Energy Dependence of CaF_2:Mn Chips

DIRECTIONAL DEPENDENCE

In most calibration work, the dosimeters are exposed to X or gamma radiation incident perpendicular to the plane of the TLD chips. The information obtained from the calibrated chips is used to convert the light output of the field chips to units of radiation dose. Since any appreciable amount of directional dependence could affect the accuracy of the dosimeter, an experiment was performed to determine if the dosimeter was directionally dependent.

The experimental setup, depicted in Figure 3, was composed of eight individual field capsules containing five TLD chips each, oriented at different angles relative to the ^{137}Cs source beam. The experiment was performed on a large, styrofoam-covered, aluminium-frame, low-scatter table. Care was taken to insure that each capsule's center was the same distance from the source.

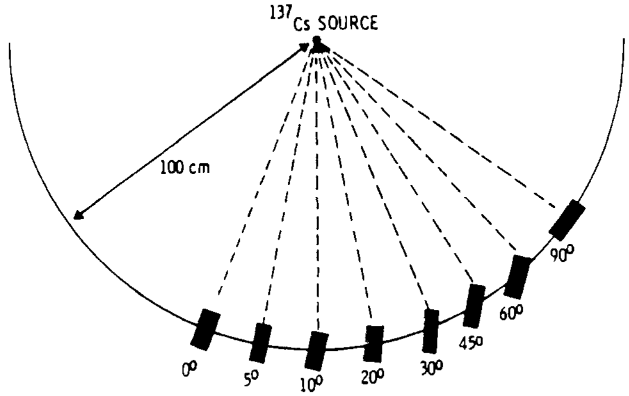

FIGURE 3. Experimental Setup

The results of the experiment are shown graphically in Figure 4. Only the left "half" of the graph shows data actually observed. The right "half" in dashed lines was added to indicate the assumed symmetry in response. The dose recorded for those capsules whose axis was ≤10° from parallel to the beam (0°, 5°, 10° in Figure 3) was significantly less than the dose recorded for the same exposure made more nearly perpendicular to the axis of the capsule (20°-90° in Figure 3).

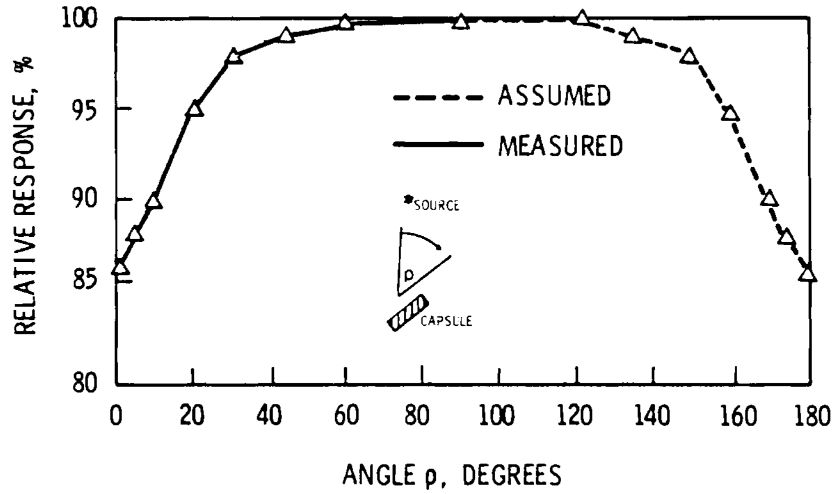

FIGURE 4. Angular Response of Field Capsule

Theoretically, the decrease in response at orientations other than normal to the incident beam might be attributed to increasing attenuation of the beam as it "sees" increasing wall thickness with decreasing angular orientation. This is not, however, a satisfactory explanation for the response at 0°, which should have been the same as at 90° since the thickness and composition of the end caps are identical to those of the capsule's sidewalls.

Another possible cause of the decreasing response is a decrease in flux density per cross sectional area of the TLD chip as its orientation to the beam changes. To determine if this was indeed a factor, the experiment as described above was repeated except that the TLD chips were placed in a thin cardboard "exposure card" instead of the field capsule. The results of this test were identical, within experimental error, to the first set of results. It was concluded, therefore, that the observed directional dependence was due not to the field capsule, but to the angular orientation of the TLD chip relative to the source beam.

The field capsules are mounted parallel to the ground. In this situation, the radiation incident on a capsule is essentially uniform from all directions, and some under-estimation of the field exposure could result.

TEMPERATURE DEPENDENCE

Two separate experiments were run to test the temperature dependence of the TLD-400 chips. Both experiments consisted of exposing a group of chips, waiting 1 day to read out 10 chips and then dividing the remaining chips into two equal groups. The two groups were placed into two Thermolyne ovens, one at room temperature and the other at an elevated temperature. The chips were left in the ovens from 3 to 5 days. By comparing the results of the two groups of chips, any additional fading caused by the increased temperature should be apparent. Also, during readout, chips from each group were read alternately to eliminate any possible reader effects.

Experiment #1

Thirty chips were exposed to approximately 10 mR of ^{137}Cs gamma. Ten chips were read after 24 hours. The other 20 chips were divided and placed in one oven at room temperature or another oven at approximately 100°C. The chips were left in the ovens for about 5 days. The mean and 2-sigma standard deviation for two groups of chips were 0.476 ± 0.071 (room temperature) and 0.451 ± 0.074 (100°C), respectively. There is no apparent difference between the two groups of numbers; this observation was substantiated by a t-test of the data at the 95% confidence level.

Experiment #2

Sixty chips were exposed to approximately 50 mR of ^{137}Cs gamma. Twenty chips were read after 24 hours. The other 40 chips were divided and placed in one oven at room temperature or another at 60°C. The chips were left in the ovens for 3 days. The mean and 2-sigma standard deviation for the two groups of chips were 2.10 ± 0.58 (room temperature) and 2.06 ± 0.36 (60°C), respectively. Again, there was no apparent difference between the groups of numbers and a t-test of the data at the 95% confidence level resulted in the same conclusion.

READOUT OPTIMIZATION

Glow curves resulting from heating TLD-400 chips were plotted using zn X-Y recorder. An example of one of these glow curves is shown in

Figure 5. The chip was esposed to 5 mR of ^{137}Cs gamma several days prior to readout; this exposure is about the same as the cumulative exposure received by the field chips. As seen in Figure 5, the integration cycle was chosen from 150°C to 280°C based on a comparison of the 5 mR output signal with the increasing infrared signal above about 290°C. A platinum planchet with 7.5-ft^3 hr dry nitrogen flow was used throughout these experiments.

In actual practice, the heating cycle lasts 20 seconds, with a rapid ramp (1-2 seconds) to 150°C and a gradual increase (18-19 seconds) to 325°C. The reader output is integrated from 150°C to 280°C. As shown in Figure 5, a small residual signal remains after the first readout. However, the residual signal is a small fraction of the signal observed during the first readout, and from a practical standpoint does not cause any problems. Sequential readout of numerous chips including the glow curve plotting has shown the readout procedure to provide excellent reproducibility.

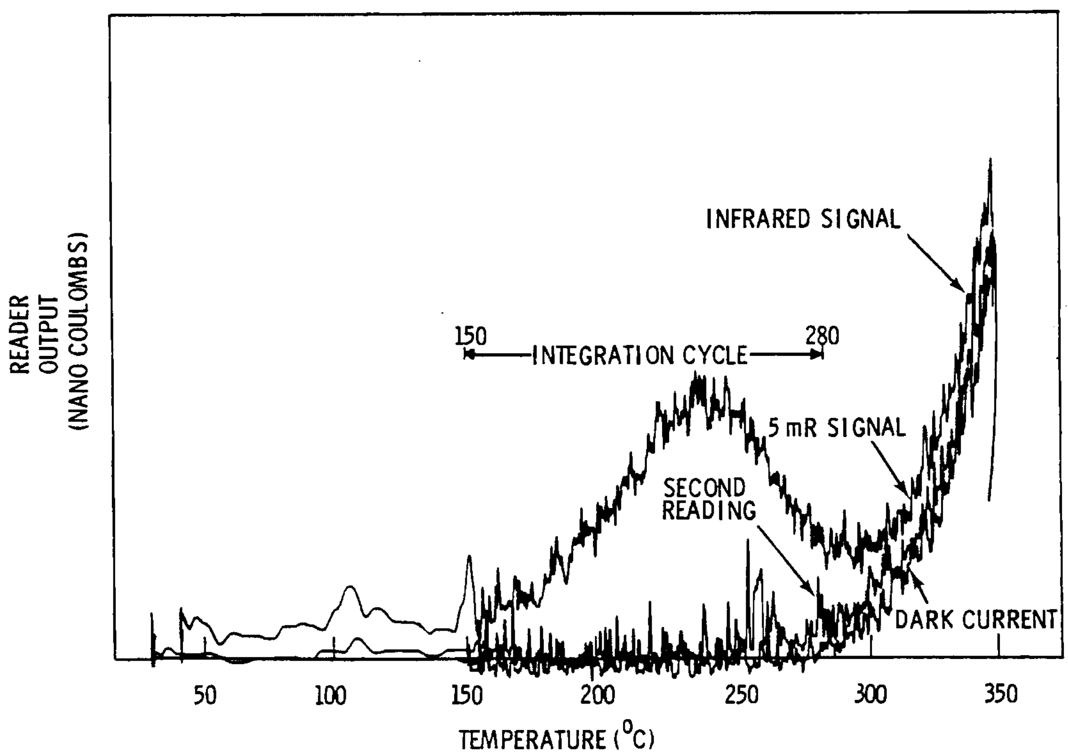

FIGURE 5. Glow Curves Resulting from 5-mR ^{137}Cs Gamma Exposure of TLD-400 Chips, Second Readout of Chips, and Dark Current with No Chips

FADE CORRECTION

An experiment was conducted to measure the loss of signal from the TLD-400 chips as a function of time after exposure. Sixty TLD-400 chips were exposed to 50 mR of ^{137}Cs gamma and read out in groups of 10 at 1 hr, 16 hr, 24 hr, 96 hr, 264 hr, and 432 hr postexposure. The results are shown in Figure 6 as the mean and one standard deviation for each group of 10 chips. It is apparent that fading does occur and a correction factor of about 10% (corresponding to 15 days postexposure) is necessary for the results obtained for a 28-day field deployment plus a 1-day wait before readout. This factor is only an approximate correction but provides a more realistic measure of ambient exposure rates than using no correction at all. The use of any adjustment is dependent on the calibration procedures employed.

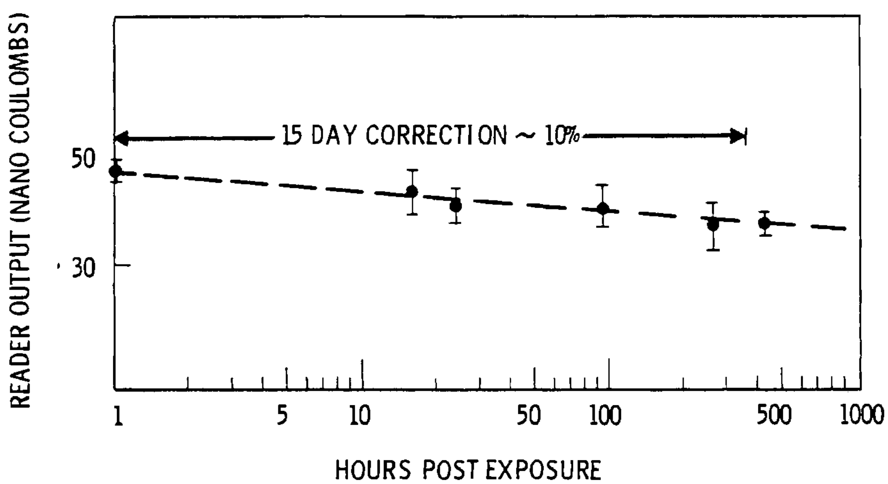

FIGURE 6. Fade Characteristics of TLD-400 Chips Postexposure

DOSE CALIBRATION

Conversion of the reader output for the field chips is done by using a calibration curve. The calibration curve for the environmental dosimeters is determined by exposing sets of five chips to 1.5, 5, 10, 15, 20, 50, and 100 mR of ^{137}Cs gamma. Another set of five chips is not exposed and the mean reader output from these chips is subtracted from the mean reader output from each of the other sets of chips. A typical calibration curve is shown in Figure 7. The ^{137}Cs gamma exposure is considered equivalent to the dose absorbed by the chips during calibration. Similarly, the field dosimeter results are interpreted in terms of dose because only penetrating gamma radiation can penetrate the shielding provided by the field capsule.

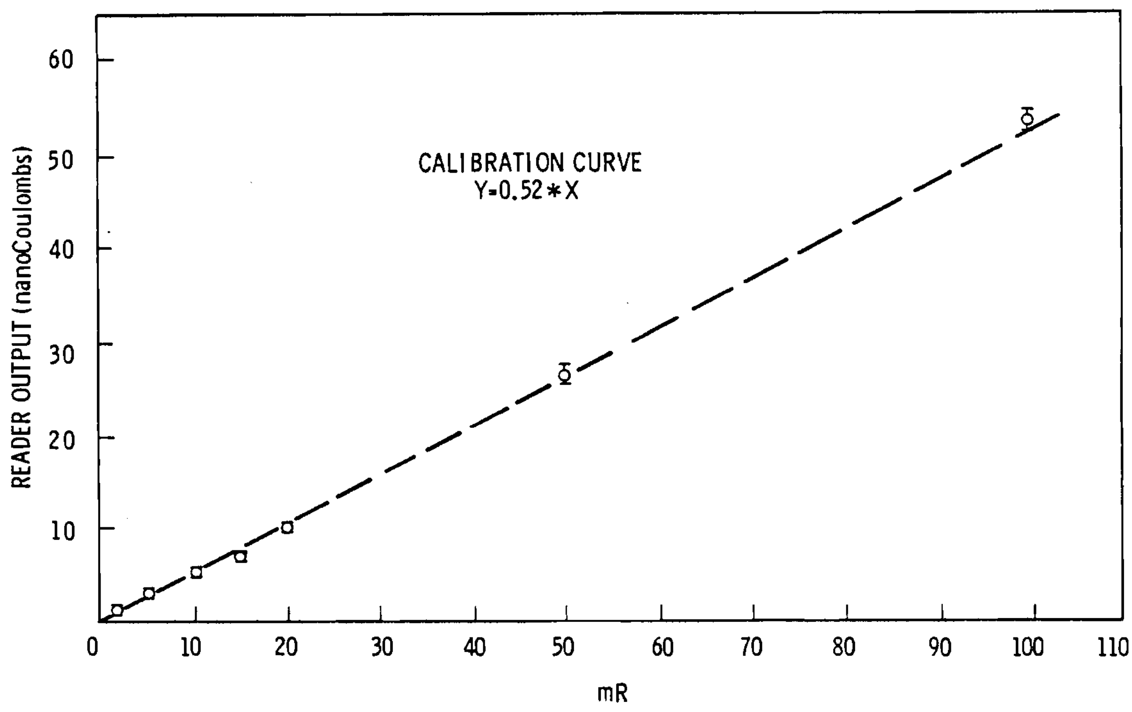

FIGURE 7. Typical Calibration Curve for TLD-400 Chips

CHIP PRECISION

The precision of the entire group (∿1000) of TLD-400 chips used to measure ambient penetrating radiation is generally less than 10% variability between individual chips within the group. Typical variability observed is shown in Figure 8, in which the results from 300 chips exposed to 50 mR of ^{137}Cs gamma are plotted. Before accepting a group of chips for the program, all of the chips are screened to remove any defective chips and to determine the overall variability of the group. The determination of field doses generally has less variability because the mean of 5 chips is used to estimate the dose received.

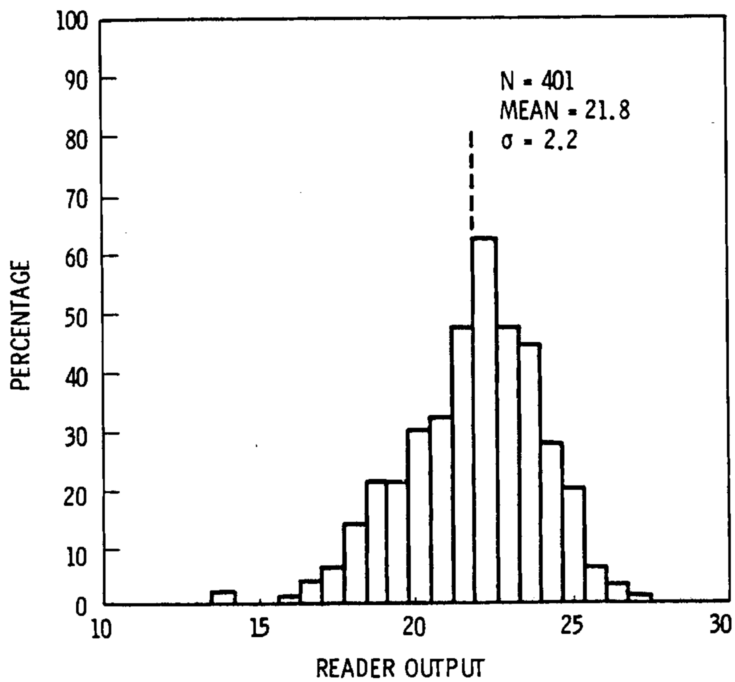

FIGURE 8. Precision of TLD-400 Chips

ROUTINE PROCEDURES[a]

PRE-EXPOSURE ANNEALING

The annealing procedure consists of a 1-hour 400°C anneal plus a 2-hour 100°C step-down anneal. The procedure is as follows:

1) Clean TLD chips if dirty with ethanol and thoroughly dry.
2) Discard any chips that are chipped, cracked, etc.
3) Place TLD chips on clean, flat-bottom, annealing plates making sure that no chips are lying on top of another chip.
4) Place annealing plates in a 400°C furnace for 1 hour. Remove and place in a 100°C oven for 2 hours.
5) Remove from oven and allow to cool to room temperature for 20 minutes. To minimize ultraviolet exposure, work under minimum incandescent lighting only being especially careful to avoid flourescent lights and/or sunlight when the chips are hot.
6) The chips are now ready for use.

FIELD DEPLOYMENT

1) Load the chips into the Pb-Ta capsules and place in the lead pig until needed.
2) Record the dosimeter numbers upon field deployment.
3) After collection from the field, the dosimeters are placed in the lead pig for 24 hours. This compensates for the rapid fading which occurs within 24 hours after exposure.
4) The chips are ready to be read.

READOUT

1) Working in subdued light, remove chips from the Pb-Ta capsules.
2) Start nitrogen (99.997% pure) flow through reader at approximately 7.5 ft^3/hr.
3) Obtain the internal light source reading by pulling out the drawer and pushing the read button. The light source reading should be obtained intermittently during readout of the chips to check reader stability.

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[a] These procedures have been in use since November 1977. Refinements to these procedures are expected.

4) Warm up the reader by running it through three successive read cycles. A blank chip should also be read.
5) The Harshaw 2000 read cycle is set for 20 seconds. The heating cycle is from room temperature to about 325°C, with a very rapid ramp to 150°C. Reader counts are integrated between 150°C and 280°C.
6) Successive TLD chips should be placed on the planchet when the planchet is less than 60°C.
7) A teflon-coated forcep should be used to handle the chips.

CHIP CALIBRATION

1) Anneal 40 chips and separate into groups of five chips each.
2) Expose Pb-Ta capsules containing five chips each (total of 35 chips) to 1.5, 5, 10, 15, 20, 50, and 100 mR of ^{137}Cs gamma.
3) Subtract the mean of the five chips not exposed from the mean of each of the exposed groups.
4) Plot the net reader output of each group versus exposure.
5) Check the plot for linearity.

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1. D. H. Denham, R. L. Kathren, and J. P. Corley, A CaF_2:Dy Thermoluminescent Dosimeter for Environmental Monitoring. BNWL-SA-4191, Battelle, Pacific Northwest Laboratories, Richland, WA, August 1972.

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