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AEC Research and Development Report

A PROBABILITY MODEL
FOR THERMOLUMINESCENT
DOSIMETRY

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A PROBABILITY MODEL FOR THERMOLUMINESCENT
DOSIMETRY

By

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January 1972

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DOSIMETRY

ABSTRACT

The trapped electron description of the thermoluminescence dosimetry mechanism is used to derive a probability model for the dosimetry measurements. Means and variances of measurement distributions can be approximated by linear and quadratic functions of exposure levels. Data from the Hanford Basic Thermoluminescence Dosimetry Process are used to develop precision specifications and audit procedures for that Process.

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A PROBABILITY MODEL FOR THERMOLUMINESCENT DOSIMETRY

Thomas M. Beetle

INTRODUCTION

This report presents the theoretical and experimental results arrived at in a study aimed at the development of an audit procedure for thermoluminescence dosimetry measurements. The experimental data and audit procedure are for the Hanford Basic Thermoluminescence Dosimetry Process. Mathematical descriptions of the relationships between low radiation exposure levels and dosimetry measurements are given in a discussion of bias and precision. A review of the audit procedure and its properties is presented in the final paragraphs.

SUMMARY

The principle of thermoluminescent dosimetry treated here involves the interaction of photons from a radiation source with electrons in a lithium fluoride (LiF) crystal. Some of the electrons become trapped in metastable states at impurity sites or other defects in the crystal. For a fixed time and distance for exposure the conditional probability distribution for the number of trapped electrons in a given crystal is of binomial form with parameters N and $1 - e^{-kM\theta t/(N+1)}$, where

N = number of traps in the LiF crystal

M = number of electrons in the LiF crystal

θt = mean number of photons entering the LiF crystal
during time t

k = proportionality constant.

When the numbers of electrons and traps in the LiF crystals are considered to be random variables, the unconditional mean and variance of the number of trapped electrons during time t are approximated, respectively, by linear and quadratic functions of t .

The trapped electrons are released by heating the crystal, and the resultant light emitted during the process is measured with a photomultiplier tube. After calibration using "known" exposures E , the mean and variance of the dosimetry measurements $\hat{E}$ can be approximated by linear and quadratic functions of E .

The mean of the dosimetry measurements is sensitive to storage time (fading) and annealing procedure. Hence, if calibration and audit or personnel dosimeters differ in the treatments of these factors, biased measurements will result. There is no evidence of potential biases due to differences in angle of radiation incidence, ambient temperatures, humidity, extraneous radiation, or rate of exposure.

Recommended precision specifications and audit procedures were described in an earlier report (9). The probability of declaring a controlled process in control is .996. The probabilities of declaring biased processes out of control vary from .006 for small bias (8% at 50 mR and 2.7% at 1,000 mR) to .982 for large bias (60% at 50 mR and 21% at 1,000 mR). When there is no bias but precision decreases from 80% to 50%, the probabilities of detecting it range from .073 to .751.

DISCUSSION

DESCRIPTION OF PROCESS

The thermoluminescence phenomenon and its applications in dosimetry have been described by several authors (1,3,4,5,7,10,11,13). Ionizing radiation induces stored energy in some materials, which, when heated, release the energy in the form of light. Measurement of the amount of light can then be related to the amount of radiation exposure received by the material. This process is called thermoluminescent dosimetry (TLD).

When a lithium fluoride crystal (LiF) is exposed to a radium source some of the photons emitted by the source interact with electrons in the crystal and release them from the valence band to the conduction band. The freed electrons may recombine with holes left in the valence band or may be trapped in metastable states at impurity sites or other defects in the crystal. Subsequent heating of the crystal will cause the trapped electrons to be released and to combine with the trapped holes. When the electrons and holes combine, visible photons are emitted. The resultant light can be measured by passing it through a photomultiplier tube.

The light emission is a function of the heating rate and the amount of radiation exposure. This function can be depicted as a glow curve on a strip chart. Measurement of the amount of radiation exposure can be accomplished by a calibration of the curve peaks or the area under the curve.

Hanford Basic TLD Process

The Hanford Basic TLD Process is used for non-radiation worker personnel dosimetry. It employs a 3.2 mm square by 0.9 mm thick ^7LiF crystal encased in teflon tape sealed in a plastic card (8).

Readout is at 300°C in a modified Harshaw Model 2000 reader. Light output from the crystal is measured by integrating the glow curve.

Calibration for the process involves data from dosimeters exposed to 0, 50, 100, 200, 400, 600, 800, 1000 and 2000 mR. A least squares linear fit to the data provides estimates of the parameters for use in evaluating radiation exposures.

PROBLEM

The absolute precision of many dosimetry processes decreases as the radiation exposure level increases. That is, the dispersion of measurements of "known" exposures increases with increased exposure levels. It is important to have information on this relationship for judging the quality of the measurements and maintaining control of the process.

In addition to precision, information on potential bias effects is important. Biases may occur if personnel or audit dosimeters are subjected to environmental and treatment conditions that differ from those of the calibration dosimeters. Investigation of sources of bias helps to indicate how the process is to be operated and controlled.

When information on precision and bias is available an audit program can be developed to aid in control of the process. An audit program based on comparisons of dosimeter measurements with "known" exposures can mistakenly indicate that a process is out of control. The natural dispersion

inherent in the audit dosimeter measurements always allows some chance of audit measurements exceeding effective control limits. A problem is to specify limits of measurement dispersion that will detect lack of control with a high frequency and be misleading with a low frequency.

PROBABILITY MODEL

In evaluating the precision of TLD measurements, the experimental procedure usually involves exposing several dosimeters to "known" exposure levels and estimating the variances among the measurements obtained at each level. Since the variance changes with exposure level, it is desirable to have an equation that describes this relationship so that variances at different exposure levels can be estimated from the experimental data. We can derive an equation that describes the relationship between variance and exposure level from a physical description of the process.

The sources of "random" variation inherent in TLD measurements can be classified into four classes:

1. Differences in dosimeters,
2. Differences in exposures,
3. Differences in reader processings and "background", and
4. Differences in successive calibrations.

The model describing these sources of variation in TLD measurements is constructed in three steps: a description of a thermoluminescence measurement for a single dosimeter; an incorporation of that description into a description of the variability manifest in data from several dosimeters; and a description of the effects of that variability on the variability of TLD exposure measurements.

For a single dosimeter exposed to a radium source for a length of time t , the thermoluminescence measurement is assumed to be proportional to the number of trapped electrons. The number of trapped electrons, however, is a random variable because the number of photon-electron interactions and number of subsequent trappings are phenomena dependent upon the Poisson nature of the photon emissions. Hence, we assume the following model as a description of a thermoluminescence measurement for a single dosimeter.

$$(1) \quad TL(t) = \alpha + \beta E(N_t) + \delta_t + \epsilon$$

$TL(t)$ = thermoluminescence measurement for a LiF crystal exposed at a fixed distance from a Ra source for a length of time t

N_t = random variable denoting the number of trapped electrons due to the irradiation

$E(N_t)$ = mean value of the random variable N_t

β = proportionality constant for transforming the number of trapped electrons to $TL(t)$ units

δ_t = random variable with mean 0 and variance $\beta^2 \sigma_{N_t}^2$

$\sigma_{N_t}^2$ = variance of N_t

α = mean value of reader processing and "background" distribution

ϵ = random variable with mean 0 and variance σ^2

σ^2 = variance of reader processing and "background" distribution.

This model includes notation for the sources of variation numbered 2 and 3 on page 5. The thermoluminescence measurement, $TL(t)$, is a random variable characterized by a conditional probability distribution (conditioned in the sense that it is for a single dosimeter) with mean $\alpha + \beta E(N_t)$ and variance $\beta^2 \sigma_{N_t}^2 + \sigma^2$.

The probability distribution for N_t can be derived from a description of TLD as a Pure Birth Process (2). Let

M = number of electrons in the LiF crystal

N = number of traps in the LiF crystal $< M$

$P_n(t) = P(N_t = n)$ = probability that n electrons are trapped during an exposure time interval of length t .

$P_x(t)$ = probability that x photons enter the LiF crystal during a time interval of length t .

$P(y|x,n)$ = probability that y additional electrons are trapped given that n electrons have been trapped previously and that x additional photons have entered the LiF crystal.

Consider time intervals $(0,t)$ and $(t,t+h)$. For $n \geq 1$, there are three mutually exclusive, exhaustive events that describe the manner in which n electrons can be trapped during a time interval $(0,t+h)$:

E_1 : n trapped in $(0,t)$ and 0 trapped in $(t,t+h)$

E_2 : $n-1$ trapped in $(0,t)$ and 1 trapped in $(t,t+h)$

E_3 : $n-y$ trapped in $(0,t)$ and y trapped in $(t,t+h)$ for all y such that $2 \leq y \leq n$.

Hence, letting $P(E_i)$ denote the probability of the event E_i ,

$$(2) \quad P_n(t+h) = \sum_{i=1}^3 P(E_i)$$

We assume that the number of photons entering a LiF crystal during a time interval of length t is not affected by the number of electrons that have been previously trapped. Also, the number of electrons that are trapped when x photons enter a LiF crystal is not dependent on the time t elapsed during the process. So we can write equation (2) as

$$(3) \quad P_n(t+h) = P_n(t) \left\{ \sum_{x=0}^{\infty} P_x(h) P(0|x,n) \right\} + \\ + P_{n-1}(t) \left\{ \sum_{x=0}^{\infty} P_x(h) P(1|x,n-1) \right\} + \\ + \sum_{y=2}^n P_{n-y}(t) \left\{ \sum_{x=0}^{\infty} P_x(h) P(y|x,n-y) \right\}.$$

Because of the Poisson nature of photon emissions from a radium source,

$$P_x(h) = (\theta h)^x e^{-\theta h} / x!.$$

Hence, some of the terms of equation (3) are of smaller order of magnitude than h :

$$\lim_{h \rightarrow 0} \frac{[(\theta h)^x e^{-\theta h} / x!] P(y|x, n)}{h} = 0$$

for $x \geq 2$. So we can write equation (3) as

$$(4) \quad P_n(t+h) = P_n(t) \left\{ \sum_{x=0}^1 P_x(h) P(0|x, n) \right\} + \\ + P_{n-1}(t) \left\{ \sum_{x=0}^1 P_x(h) P(1|x, n-1) \right\} + \\ + \sum_{y=2}^n P_{n-y}(t) \left\{ \sum_{x=0}^1 P_x(h) P(y|x, n-y) \right\} + o(h)$$

From the definitions of the probability symbols it is evident that

$$P(0|0, n) = 1$$

$$P(1|0, n-1) = 0$$

$$P(y|0, n-y) = P(y|1, n-y) = 0 \text{ for } y \geq 2.$$

We assume that the interaction of a photon and electron and the trapping of the electron is dependent only on the number of electrons and traps present in the LiF crystal. That is, we assume that

$$P(1|1, n) = k(M-n)(N-n)/(N+1).$$

An argument that supports this assumption is as follows: the probability that a photon interacts with one of the $(M-n)$ non-trapped electrons is proportional to that number, and the conditional probability that a freed electron is trapped is proportional to the ratio of the number of empty traps to the sum of empty traps plus holes, $(N-n)/(N+1)$; so the probability that both events occur is the product of their probabilities.

Equation (4) can now be written as

$$\begin{aligned}
 (5) \quad P_n(t+h) &= P_n(t) \left\{ e^{-\theta h} + \theta h e^{-\theta h} \left\{ 1 - [k(M-n)(N-n)/(N+1)] \right\} \right\} \\
 &\quad + P_{n-1}(t) \left\{ \theta h e^{-\theta h} [k(M-n+1)(N-n+1)/(N+1)] \right\} + o(h) \\
 &= P_n(t) \left\{ 1 - \left[\sum_{x=2}^{\infty} (\theta h)^x e^{-\theta h} / x! \right] - \theta h e^{-\theta h} [k(M-n)(N-n)/(N+1)] \right\} \\
 &\quad + P_{n-1}(t) \left\{ \theta h e^{-\theta h} [k(M-n+1)(N-n+1)/(N+1)] \right\} + o(h)
 \end{aligned}$$

Hence,

$$\begin{aligned}
 (6) \quad P'_n(t) &= \lim_{h \rightarrow 0} [P_n(t+h) - P_n(t)]/h = \\
 &= -P_n(t) \theta k(M-n)(N-n)/(N+1) + \\
 &\quad + P_{n-1}(t) \theta k(M-n+1)(N-n+1)/(N+1) \\
 &= -P_n(t) \lambda_n + P_{n-1}(t) \lambda_{n-1} .
 \end{aligned}$$

For $n = 0$, event E_1 is sufficient to describe the manner in which n electrons can be trapped during the time interval $(0, t+h)$, so

$$(7) \quad P'_0(t) = -\lambda_0 P_0(t).$$

Equations (6) and (7) constitute a system of differential equations whose solution is simplified by setting

$$\lambda_n = k\theta M(N-n)/(N+1).$$

The solution of $P_n(t)$ under this simplification will be a close approximation for low exposure levels (small n) or for LiF crystals where there are many more electrons than traps, $M \gg N \geq n$. The solution of this system of linear differential equations of the first order is

$$(8) \quad P_n(t) = \binom{N}{n} (e^{-kM\theta t/(N+1)})^{N-n} (1 - e^{-kM\theta t/(N+1)})^n$$

Equation (8) states that the number of trapped electrons for a fixed exposure of time length t (distance from source fixed) has a distribution of binomial form with parameters N and $1 - e^{-kM\theta t/(N+1)}$.

The mean number of photons (or secondary electrons) entering the LiF crystal during the time interval $(0, t)$ is θt . The factor kM is less than or equal to $(N+1)/N$. If the product $kM\theta t$ is small relative to the number of traps N (e.g. at low exposure levels), then the distribution of the number of trapped electrons expressed in equation (8) is approximated by either a Poisson or a normal distribution (2, pp. 142-143, 168-177) with mean and variance equal to $N(1 - e^{-kM\theta t/(N+1)})$. Also, under the same condition, the exponential term in the parentheses can be approximated by

the first two terms of its Taylor Series expansion, so the mean and variance are approximately linear in t :

$$(9) \quad E(N_t) = \sigma_{N_t}^2 = NkM\theta t/(N+1).$$

Equation (9) indicates that under our low exposure assumption the mean and variance of the number of trapped electrons are independent of the number of traps. The mean and variance are most sensitive to the exposure level, θt , and the size of the crystal, M .

The next step in the construction of the model involves the introduction of the variation due to differences in dosimeters. To do this we assume that equation (9) describes a conditional mean and variance given that M and N are fixed, and write

$$(10) \quad E(N_t|M,N) = V(N_t|M,N) = \sigma_{N_t|M,N}^2 = NkM\theta t/(N+1).$$

We also assume that the frequencies of the numbers of electrons and traps in a large group of dosimeters can be described by a probability distribution, and write

$$(11) \quad P(M=\mu, N=\nu) = p(\mu, \nu)$$

$$0 \leq p(\mu, \nu) \leq 1$$

$$\sum_{\mu, \nu} p(\mu, \nu) = 1.$$

Then we can compute the unconditional mean and variance of N_t as follows:

$$\begin{aligned}
 (12) \quad E(N_t) &= \sum_{\mu, \nu} E(N_t | M=\mu, N=\nu) p(\mu, \nu) \\
 &= k\theta t \sum_{\mu, \nu} \mu \nu p(\mu, \nu) / (\nu+1)
 \end{aligned}$$

$$\begin{aligned}
 (13) \quad V(N_t) &= \sigma_{N_t}^2 = \sum_{\mu, \nu} V(N_t | M=\mu, N=\nu) p(\mu, \nu) + \\
 &\quad + \sum_{\mu, \nu} E^2(N_t | M=\mu, N=\nu) p(\mu, \nu) - E^2(N_t) \\
 &= k\theta t \sum_{\mu, \nu} \mu \nu p(\mu, \nu) / (\nu+1) + \\
 &\quad + k^2(\theta t)^2 \sum_{\mu, \nu} \mu^2 \nu^2 p(\mu, \nu) / (\nu+1)^2 - \\
 &\quad - k^2(\theta t)^2 \left[\sum_{\mu, \nu} \mu \nu p(\mu, \nu) / (\nu+1) \right]^2
 \end{aligned}$$

Hence, the mean of the number of trapped electrons for low exposure levels is linear in t , and the variance of the number of trapped electrons is quadratic in t .

Equation (1) can now be written as

$$(1') \quad TL(t) = \alpha' + \beta' E'(N_t) + \delta'_t + \epsilon'$$

with all descriptions of corresponding symbols the same except that they are understood to refer to the probability distributions associated with several dosimeters.

The preceding discussion was concerned with the derivation of a model of the relationships of the mean and variance of thermoluminescence measurements to exposure level. Conversion of these measurements to measurements of exposures by calibration introduces an additional source of variability into exposure measurements: the variability in successive calibration curves.

The calibration procedure for the Basic Thermoluminescence Dosimeter process at Hanford involves data from 9r dosimeters with r exposed to each of the 0, 50, 100, 200, 400, 600, 800, 1000 and 2000 mR exposure levels. The average of the thermoluminescence measurements at exposure level i,

$$\bar{R}_i = \sum_{j=1}^r TL_j(t_i)/r ,$$

is computed for each i, and an unweighted least squares fit is computed to estimate the parameters in the following model:

$$(14) \quad \begin{aligned} \bar{R}_i &= \gamma + \rho E_i + \xi_i \\ E_i &= \text{exposure level} \\ \gamma &= \text{intercept} \\ \rho &= \text{slope} \\ \xi_i &= \text{random variable with mean 0 and variance } \sigma_i^2. \end{aligned}$$

Exposure for a dosimeter with a thermoluminescence measurement R is then computed by

$$(15) \quad \hat{E} = \frac{R - \hat{\gamma}}{\hat{\rho}} , \text{ where}$$

$\hat{\gamma}$, $\hat{\rho}$ are estimates of the parameters in equation (14).

The model expressed by equation (14) is related to the model expressed by equation (1') as follows:

$$\begin{aligned}
 \bar{R}_i &= \sum_{j=1}^r TL_j(t_i)/r = \sum_{j=1}^r [\alpha' + \beta'E'(N_{t_i}) + \delta'_{t_{ij}} + \epsilon'_j]/r \\
 &= \alpha' + \beta'E'(N_{t_i}) + \sum_{j=1}^r \delta'_{t_{ij}}/r + \sum_{j=1}^r \epsilon'_j/r \\
 &= \gamma + \rho E_i + \xi_i
 \end{aligned}$$

$\gamma = \alpha =$ mean of "background" distribution, $\rho E_i = \beta E(N_{t_i}) =$ mean of thermoluminescence measurements due to irradiation at exposure level i , and

$$\xi_i = \sum_{j=1}^r \delta'_{t_{ij}}/r + \sum_{j=1}^r \epsilon'_j/r = \text{random}$$

variable with mean 0 and variance

$$\sigma_i^2 = \beta'^2 \sigma_{N_{t_i}}'^2 / r + \sigma'^2 / r .$$

The equality between ρE_i and $\beta'E'(N_{t_i})$ is a result of E_i being computed by the inverse square law, which is linear in t , and $E(N_{t_i})$ being linear in t as previously shown. Also, since $\sigma_{N_t}'^2$ is quadratic in t , σ_i^2 will be quadratic in t (that is, as t_i varies).

Approximations of the mean and variance of $\hat{E}$ are:

$$(16) \quad E(\hat{E}) \cong E = \text{Dosimeter exposure as computed} \\ \text{by the inverse square law}$$

$$(17) \quad V(\hat{E}) \cong \rho^{-2} [V(R) + V(\hat{\gamma}) + E^2 V(\hat{\rho}) + 2E \text{Cov}(\hat{\gamma}, \hat{\rho})]$$

The variances and covariance in equation (17) are related to exposure levels as follows:

$$(18) \quad V(R) = \beta'^2 \sigma_{N_t}^2 + \sigma'^2 = \beta'^2 (\eta_0 + \eta_1 t + \eta_2 t^2) + \sigma'^2 = \\ = \tau_0 + \tau_1 E + \tau_2 E^2$$

$$(19) \quad V(\hat{\gamma}) = 9^{-2} \sum_{i=1}^9 V(R_i) + \left(\sum_{i=1}^9 E_i / 9 \right)^2 V(\hat{\rho}) - \\ - 2 \left(\sum_{i=1}^9 E_i / 9 \right) \text{Cov} \left[\left(\sum_{i=1}^9 R_i / 9 \right), \hat{\rho} \right] \\ = 9^{-2} r^{-1} \sum_{i=1}^9 (\tau_0 + \tau_1 E_i + \tau_2 E_i^2) + \bar{E}^2 V(\hat{\rho}) - \\ - 2 \bar{E} 9^{-1} r^{-1} \left[\sum_{i=1}^9 (E_i - \bar{E})^2 \right]^{-1} \sum_{i=1}^9 (E_i - \bar{E}) (\tau_0 + \tau_1 E_i + \tau_2 E_i^2)$$

$$(20) \quad V(\hat{\rho}) = r^{-1} \left[\sum_{i=1}^9 (E_i - \bar{E})^2 \right]^{-2} \sum_{i=1}^9 (E_i - \bar{E})^2 (\tau_0 + \tau_1 E_i + \tau_2 E_i^2)$$

$$(21) \quad \text{Cov}(\hat{\gamma}, \hat{\rho}) = r^{-1} g^{-1} \left[\sum_{i=1}^9 (E_i - \bar{E})^2 \right]^{-1} \sum_{i=1}^9 (E_i - \bar{E}) (\tau_0 + \tau_1 E_i + \tau_2 E_i^2) \\ - \bar{E} r^{-1} \left[\sum_{i=1}^9 (E_i - \bar{E})^2 \right]^{-2} \sum_{i=1}^9 (E_i - \bar{E})^2 (\tau_0 + \tau_1 E_i + \tau_2 E_i^2)$$

Equations (19), (20) and (21) are derived in linear regression theory. Substitution of equations (18) through (21) into (17) yields an expression for the variance of an exposure estimate $\hat{E}$. The known quantities are

g = number of calibration exposure levels

r = number of calibration dosimeters exposed at each level

E_i = calibration exposure at level i in mR.

The unknown quantities are

E = exposure as computed by inverse square law

$\tau_i (i = 0, 1, 2)$ = parameters of the quadratic relationship between thermoluminescent measurement variance and exposure in mR.

ρ = regression coefficient parameter (equation (14)) of the linear relationship between thermoluminescent measurement mean and exposure in mR.

Estimation of Parameters

Thermoluminescence measurement data from several dosimeters exposed at each of the nine calibration exposure levels are presented in Table 1 of the Appendix. Estimates of the mean and variance at each exposure level are shown at the bottom of the table as $\bar{R}_i$ and S_i^2 . The sample mean is an unbiased estimate of $\gamma + \rho E_i$ and the sample variance is an unbiased estimate of $\tau_0 + \tau_1 E_i + \tau_2 E_i^2$. Least squares fits were used to estimate the τ 's and ρ .

The linear fit of $\bar{R}_i$ to exposure level yields estimates

$$\hat{\gamma} = .037$$

$$\hat{\rho} = .00121$$

$$\gamma + \rho E_i = .037 + .00121 E_i$$

The coefficient of determination for this fit is $100 r_{R,E}^2 = 99.97\%$, so the fit accounts for more than 99.9% of the total variation in the data. A plot of the fit is shown in Figure 1.

The quadratic fit of S_i^2 to exposure level yields estimates

$$\hat{\tau}_0 = 768 \times 10^{-7}$$

$$\hat{\tau}_1 = 487 \times 10^{-9}$$

$$\hat{\tau}_2 = 15 \times 10^{-9}$$

$$\tau_0 + \tau_1 E_i + \tau_2 E_i^2 = 768 \times 10^{-7} + 487 \times 10^{-9} E_i + 15 \times 10^{-9} E_i^2$$

The coefficient of determination for this fit is $100 R_{S^2,EE}^2 = 99.22\%$. A plot of the fit is shown in Figure 2.

Figure 1. Thermoluminescence Mean vs Exposure Level

Figure 2. Thermoluminescence Variance vs Exposure Level

Substitution of these estimates in equations (17) through (21) gives the following estimate of the relationship between the variance of an exposure measurement and exposure level:

$$(22) \quad V(\hat{E}) = 52.4554 + 966.7509 r^{-1} + \\ + (.332628 - 4.9638 r^{-1})E + \\ + (.010245 + .0082 r^{-1})E^2 .$$

BIAS EFFECTS

Certain operating and environmental conditions (controllable or uncontrollable) of the TLD process might affect the exposure measurements. Some of these conditions are:

Angle of radiation incidence
Temperature
Humidity
Length of time to readout
Annealing
Extraneous Radiation
Rate of exposure.

Biased measurements might occur if the conditions under which calibration dosimeters are produced and evaluated differ from the conditions for personnel and audit dosimeters. Mathematically this can be described by considering equation (14), (15) and (16).

We have seen that

$$(23) \quad E(\hat{E}) = E\left(\frac{R - \hat{\gamma}}{\rho}\right) \approx \frac{\gamma_R + \rho R E - \gamma_C}{\rho_C}$$

$\gamma_R + \rho_R E$ = mean of the TL(t) measurements for
exposure E as computed by the inverse
square law

γ_C and ρ_C parameters estimated by calibration
procedure.

The expression on the right in equation (23) will be equal to E if $\gamma_R = \gamma_C$ and $\rho_R = \rho_C$. That is, if the means of the "background" distributions are equal and $\beta_R E(N_{t_R})/E = \beta_C E(N_{t_C})/E$. This last equality will hold if the proportionality constants (β 's) for transforming the number of trapped electrons to TL(t) units are equal for calibration and personnel (or audit) dosimeters, and the mean numbers of trapped electrons ($E(N_t)$'s) are equal. So, differences in "backgrounds", reader conditions, dosimeters, exposure conditions, or environmental conditions that cause inequalities among these parameters will result in biased measurements.

Angle of Radiation Incidence

In terms of the model presented here, equation (12) states that

$$E(N_t) = Ck\theta t$$

where C is a constant, k is a proportionality constant for the probability that a photon interacts with an electron and the electron is trapped, and θt is the mean number of photons passing through an area at right angle to the radiation source and defined by the projection of the LiF crystal onto the right angle plane. Calibration dosimeters are exposed at right angles to the radiation source. As the angle of incidence decreases, θt will decrease

and one would expect a negative bias to occur. On the other hand, as the angle of incidence decreases, the length of the path of a photon in the crystal increases and one might expect k to increase and offset the negative bias.

Cusimano and Cipperley (14) found that for a lithium fluoride-teflon disc dosimeter there was no appreciable bias down to 45° but that there was about a 20% reduction in response at 0° . Kocher, Kathren and Endres (6) found no effect due to angle of radiation incidence for 8 mm diameter lithium fluoride-teflon disc dosimeters.

Kathren (8) reported on an experiment to test for angular dependence for the Hanford Basic TLD dosimeter. Two dosimeters were exposed to 200 mR of ^{60}Co at each of eleven angles of incidence between 0° and 180° . Comparison of the 90° mean thermoluminescence measurement with the means at each of the other angles using Dunnetts' one-sided procedure (15) did not detect any angle-of-incidence effects. However, a 20% difference in mean thermoluminescence measurements would only be detected half of the time with this size experiment, so some angular dependence might exist for the Hanford Basic TLD dosimeter.

Temperature

Since the principle of TLD involves the emptying of traps by the application of heat, the possibility of a premature emptying of traps at ambient temperatures exists. If the environmental temperature for calibration dosimeters during the time from exposure to reading differs from that for audit or personnel dosimeters, some bias might occur. If any effect does occur, one would expect that dosimeters held at higher than ambient temperatures would have a reduced $E(N_t)$.

Effects due to temperature differences during irradiation were investigated for the Hanford Basic TLD dosimeter. No differences were detected between means of thermoluminescence measurements for 750 mR ^{60}Co exposures at 66°F and 122°F. Effects due to temperature differences over longer periods have not been investigated.

Humidity

Two experiments were conducted to investigate humidity effects on the Hanford Basic TLD dosimeter measurements. Dosimeters exposed in six relative humidity environments from 10% to 95% yielded no significant differences in mean thermoluminescence measurement when exposed to 1R of ^{60}Co (reference (8)). A second experiment involving a ^{60}Co exposure of 750 mR in relative humidities of 10% and 95% resulted in nonsignificance for a test of humidity effects. Long term humidity effects have not been investigated.

Length of Time to Readout

The traps in a LiF crystal are of different types in the sense that they require different temperatures to empty them. Because of this the glow curve for an exposed crystal will have several characteristic peaks. As the period of time between exposure and readout is extended, some of the "low temperature traps" might empty at ambient temperatures and decrease $E(N_t)$. Fading, if it exists, will result in bias if the $E(N_t)$ for calibration dosimeters differ from those for the audit or personnel dosimeters.

Cameron, Suntharalingam and Kenney (1) describe the fading phenomenon in terms of half-lives for characteristic glow curve peaks at ambient temperatures. For a LiF dosimeter, they quantify the fading as less than 5% in a 12 week period. Other authors (5, 7, 12) have reported LiF dosimeter fading as "negligible" to "15% on the first day plus 5% per year".

An experiment on the Hanford Basic TLD dosimeter indicated an average 15% fading effect over a 34 day period for dosimeters exposed to 750 mR of ^{60}Co . Furthermore, the fading effects for two different annealing procedures were different. Analysis details are discussed later in this report under Interrelationship Experiment.

Annealing

Several authors have addressed themselves to the effects that annealing procedures have on TLD processes (1,4,6,7,11,12,13). Reference (1) devotes Chapter 8 to the topic and illustrates the changes in glow curves that occur under various preannealing (before exposure) and post annealing conditions. In terms of the basic TLD theory the preannealing phenomena are not well understood, but various conjectures have been advanced including annealing induced changes in the numbers of traps associated with the various glow curve peaks, changes in the conversion efficiency from trapped electrons to photons, and the formation of aggregates of traps that affects the trapping mechanism.

For the model presented in this report (see equation (9)), a change in the number of traps would not explain annealing effects at low exposure levels. If the conversion efficiency from trapped electrons to photons is changed by annealing procedures, then the model presented here does not offer an explanation since it assumes that all trapped electrons will be released during readout and result in emitted light. The formation of aggregates of traps might invalidate the probability assumption on page 9 : the probability of a freed electron combining with a hole might be different for crystals with

dispersed traps and crystals with aggregated traps.

Whatever the mechanism for preannealing effects might be, a comparison of a reader annealing procedure with an oven annealing procedure indicates a pronounced effect on the thermoluminescence measurements for the Hanford Basic TLD process. Dosimeters annealed at 90°C for 24 hours yield 35% to 40% less light from a subsequent 750 mR exposure (^{60}Co) than dosimeters that are annealed by a readout cycle (300°C for about 20 seconds). This effect, of course, will not bias exposure measurements providing that personnel, audit, and calibration dosimeters are all annealed by the same procedure.

Interrelationship Experiment

Main effects and interactions for the angle-of-incidence, temperature (at time of exposure), humidity (at time of exposure), time-to-readout, and preannealing factors were investigated for the Hanford Basic TLD Process. The "levels" of the factors were:

<u>Factor</u>	<u>Levels</u>	
Angle-of-incidence	10°	90°
Temperature	19°C	50°C
Relative Humidity	10%	95%
Time-to-readout	1 Day	35 Days
Preannealing	20 sec. @ 300°C	24 hrs. @ 80°

Combinations of the first four factors were "applied" to jars containing two dosimeters with different preannealing treatments. All dosimeters were exposed to 750 mR of ^{60}Co . Data are listed in Table 2 of the Appendix.

The data were analyzed in three groups with the following analyses of variance (See Ch. 12 of (15) or Ch. 10 of (16)).

Analysis of Variance 1
(Exposure Temperature = 50°C)

Source	df	Mean Square	F
Angle (A)	1	2.146590	3.455
Humidity (H)	1	.304200	<1
Days-to readout (D)	1	6.913620	11.127
AxH	1	.022472	<1
AxD	1	.017578	<1
HxD	1	.007140	<1
AxHxD	1	.006216	<1
Error (a)	8	.621338	
Preanneal (P)	1	45.821200	350.355**
AxP	1	.008713	<1
HxP	1	.149605	1.144
DxP	1	1.447550	11.068*
AxHxP	1	.703297	5.378
AxDxP	1	.013859	<1
AxHxD	1	.636755	4.869
AxHxDxA	1	.001429	<1
Error (b)	4	.130785	
Total	27		

* Significant at the 5% level.

** Significant at the 1% level.

Analysis of Variance 2
(Angle-of-incidence = 90°C)

Source	df	Mean Square	F
Temperature (T)	1	.037743	<1
Humidity (H)	1	.144319	<1
Days-to-readout (D)	1	6.812660	24.485**
TxH	1	.610789	2.195
TxD	1	.023059	<1
HxD	1	.011288	<1
TxHxD	1	.012521	<1
Error (a)	8	.278238	
Preanneal (P)	1	56.437300	277.591**
TxP	1	.422510	2.078
HxP	1	.492776	2.424
DxP	1	1.669420	8.211
TxHxP	1	.273986	1.348
TxDxP	1	.042706	<1
HxDxP	1	.134550	<1
TxHxDxP	1	.154702	<1
Error (b)	6	.203311	
Total	29		

* Significant at the 5% level.

** Significant at the 1% level.

Analysis of Variance 3

(Angle-of-incidence = 90°, Exposure Temperature = 19°C)

Source	df	Mean Square	F
Humidity (H)	1	.100965	<1
Days-to-readout (D)	1	6.407230	25.250**
HxD	1	.072496	<1
Error (a)	4	.253755	
Preadannealing (P)	1	28.432900	88.201**
HxP	1	.021830	<1
DxP	1	.123728	<1
HxDxP	1	.559877	1.737
Error (b)	4	.322363	
Total	15		

* Significant at the 5% level.

** Significant at the 1% level.

The analyses show that there is a significant fading over a 35 day period for exposures at the 750 mR exposure level, and that readout preannealing results in a significantly different thermoluminescence response than the 80°C for 24 hours preannealing procedure. Also, the fading is different for the two preannealing procedures. These effects are illustrated in Figure 3.

Figure 3. Fading - Annealing Interactions

Over 35 days, readout annealed dosimeters fade an average of 18% and oven annealed dosimeters fade an average of 13%. Oven annealed dosimeters show 39% less response one day after exposure and 35% less response 35 days after exposure.

AUDIT PROCEDURE

A recommended audit procedure for the Hanford Basic TLD Process is described in (9). The precision specification is

Twelve or more of fifteen blind audit exposures, E , between 50 mR and 1,000 mR must be measured, $\hat{E}$, with a precision specified by

$$|\hat{E} - E| \leq 2 (213 - .4947E + .011611E^2)^{\frac{1}{2}} . *$$

The expression in parentheses comes from equation (22) with $r = 6$.

To avoid bias due to the audit procedure (rather than the process) the following restrictions are imposed:

1. Audit exposures should be made at the same time that calibration dosimeters are exposed;
2. Annealing and other handling of the audit dosimeters should be similar to that of the calibration dosimeters; and
3. Exposed audit dosimeters should be stored under conditions similar to the storage of the calibration dosimeters.

The first two restrictions are concerned with the potential biases discussed under the heading Interrelationship Experiment.

*[Subsequent to the publishing of (9), it was decided that the audit should be conducted bimonthly, and that an open audit (dosimeters known to the processor) should be conducted and evaluated prior to the processing of personnel dosimeters. Also, the above inequality is a revised expression.]

Control Probabilities

The spirit of the precision specification limits is to cover approximately 95% of the audit exposure measurements when the process is in control. Calculation of the "exact" probability of declaring a controlled process out of control (and vice versa) is frustrated by the stochastic dependence among the measurements for a single audit period. However, approximations to the probabilities assuming independence of measurements and Normal distributions can be calculated. Under that assumption, the probability of declaring a controlled process out of control (four or more measurements outside of the precision specification limits) is .004. The probabilities of declaring a noncontrolled process in control under various bias and imprecision conditions are presented in the following table.

$\frac{Y_R - Y_C}{\rho_C}$	$\frac{\rho_R}{\rho_C}$	$V(\hat{E})^{\frac{1}{2}}$	% Bias at 50 mR	% Bias at 1,000 mR	Probability of Error in Audit	Probability of Error in "One Measurement Out" Audit
3	1.024	$V(\hat{E})^{\frac{1}{2}}$	8	2.7	.994	.816
5	1.049	$V(\hat{E})^{\frac{1}{2}}$	15	5.4	.989	.698
10	1.097	$V(\hat{E})^{\frac{1}{2}}$	30	10.7	.790	.281
15	1.146	$V(\hat{E})^{\frac{1}{2}}$	45	16.1	.272	.030
20	1.193	$V(\hat{E})^{\frac{1}{2}}$	60	21.0	.018	.0004
0	1	$1.25 V(\hat{E})^{\frac{1}{2}}$	0	0	.927	.498
0	1	$1.50 V(\hat{E})^{\frac{1}{2}}$	0	0	.709	.209
0	1	$1.75 V(\hat{E})^{\frac{1}{2}}$	0	0	.446	.075
0	1	$2.00 V(\hat{E})^{\frac{1}{2}}$	0	0	.249	.026

Columns 1 and 2 refer to equation (23).

Column 3 refers to equation (22).

Column 7 is discussed below.

When precision is in control but there is a bias of .8% to 15% at 50 mR and 2.7% to 5.4% at 1,000 mR, the audit procedure will seldom detect the lack of control. When the bias is 60% at the lower end of the range and 21% at the upper end, the audit procedure will detect it 49 times out of 50. When bias is in control but the standard deviation is at $kV(\hat{E})^{\frac{1}{2}}$, the audit procedure will detect it 1 time in 10 for $k = 1.25$ and 3 times in 4 for $k = 2$.

Future control procedures could be tightened by amending the precision specification to require fourteen or more of the measurements to be within limits. The probabilities for errors in this "One Measurement Out" procedure are shown in column 7. of the above table.

Data Plot

In order to judge the reasonableness of the audit procedure derived in this study, calibration data were used to simulate an audit with the results plotted on the Test Chart displayed below.

Figure 4. Test Chart Plot of Simulated Audit Data

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APPENDIX

Table I. Thermoluminescence Measurements at Nine Exposure Levels

	Exposure Level (mR)								
	0	50	100	200	400	600	800	1000	2000
	.091	.118	.169	.290	.487	.778	1.009	1.264	2.532
	.050	.105	.175	.317	.504	.685	1.077	1.113	2.480
	.037	.087	.168	.271	.628	.701	1.090	1.323	2.417
	.033	.096	.156	.310	.487	.727	1.026	1.007	2.441
	.044	.099	.159	.292	.470	.738	.859	1.304	2.645
	.039	.102	.172	.300	.527	.786	1.136	1.388	2.576
	.042	.100	.168	.250	.512	.783	.980	1.143	2.141
		.094	.157	.336	.547	.725	1.182	1.179	2.611
		.093	.163	.267	.580	.818	1.056	1.362	2.768
		.094	.161	.317	.530	.820	1.151	1.427	2.663
		.086	.166	.304	.615	.785	1.154	1.188	2.753
		.083	.150	.286	.568	.797	.961	1.295	2.647
		.082	.132	.247	.509	.722	.836	1.096	2.365
		.073	.131	.257	.430	.719	.961	1.171	2.261
		.075	.127	.248	.399	.636	.855	1.116	1.840
$\bar{R}_i$	.042	.092	.157	.286	.520	.748	1.022	1.225	2.476
$10^2 S_i^2$	.004	.014	.024	.080	.395	.277	1.285	1.501	6.187

Dark Current $\cong .012$ Light Source $\cong 1.66$

Table II. Thermoluminescence Measurements for an
Interrelationship Experiment

Relative Humidity	Time-to-Readout	Preannealing Temperature	Temperature During Exposure			
			19°C		50°C	
			Angle-of-Incidence		Angle-of-Incidence	
			90°		10°	90°
10%	1 Day	80°C	4.946	5.160	4.945	5.427
		300°C	8.032	8.032	7.519	7.550
		80°C	4.379	4.114	3.372	4.765
		300°C	7.999	7.822	6.181	6.858
	35 Days	80°C	4.309	4.253	3.767	3.969
		300°C	6.537	6.125	5.503	6.213
		80°C	4.202	3.859	3.243	4.504
		300°C	6.541	6.367	6.204	6.016
95%	1 Day	80°C	4.061	4.181	4.797	4.333
		300°C	8.423	6.758	7.760	7.845
		80°C	5.383	5.374	3.860*	5.777*
		300°C	7.528	8.017	6.823	8.255
	35 Days	80°C	4.205	4.641	3.799	3.760
		300°C	6.165	6.456	5.237	5.967
		80°C	3.494	2.919	4.502*	5.099*
		300°C	6.391	6.685	5.940	6.871

[1]

[3]

[1][2]

[2]

* Estimated Values

[1] Data for Analysis of Variance 1

[2] Data for Analysis of Variance 2

[3] Data for Analysis of Variance 3

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