

Alpha dose to a thin layer

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With increasing success in ESR dating of shells and teeth it becomes topical to consider the alpha dose within thin layers. This is done here using the approximation of constant effect (TL or ESR) per unit length of track, which is the basis of the three current systems of assessing alpha dose: *a*-value (Aitken and Bowman, 1975), *b*-value (Bowman and Huntley, 1984) and omnidirectional flux (Guérin and Valladas, 1980). The dose in terms of energy deposited increases as the particle slows down but because of a compensatory decrease in TL effectiveness it turns out that constant TL per unit length of track is a good approximation; it has now been shown that the same is true for ESR (see accompanying note by Lyons).

Dose in inert medium

First we consider the effective dose delivered by a radioactive medium to an inert medium, the media being separated by a plane boundary of infinite extent; see Fig. 1. Let the alpha particles be emitted with initial range R_0 , the same in both mediums. Then the residual range carried by a particle across a plane at depth x in the inert medium is $(R_0 - R)$ where,

$$R = (h+x) / \cos\theta$$

and h is the distance of the point of emission from the boundary, with θ being the angle to the vertical.

If N is the number of particles emitted per unit volume then the total residual range carried across the plane by particles emitted from unit volume at distance h from the boundary with paths lying between θ and $\theta+d\theta$ is given by

$$\frac{2\pi r dr \cos\theta (R_0 - R) N}{4\pi R^2} \quad \dots (1)$$

which may be written as

$$\frac{N (h+x)(R_0 - R) dR}{2R^2} \quad \dots (2)$$

since $\cos\theta = \frac{(h+x)}{R}$ and $r dr = R dR$

The total residual range carried across the plane by particles emitted from the unit volume is obtained by integration, between the limits of $R = (h+x)$ and $R = R_0$, as,

$$\frac{N}{2} \{ R_0 - (h+x) - (h+x) \ln R_0 + (h+x) \ln(h+x) \} \quad \dots (3)$$

as long as $(h+x) \leq R_0$.

We now integrate over the radioactive medium and obtain the residual range delivered across the plane by particles emitted from a cylinder of area A and height $(R_0 - x)$ as

$$\frac{NA}{8} (R_0^2 - 4xR_0 + 3x^2 + 2x^2 \ln R_0 - 2x^2 \ln x) \quad \dots (4)$$

with the restrictions that $x \leq R_0$, and $A \gg R_0^2$. From this expression we may derive the effective alpha dose in the inert medium for various situations.

Case A: Inert medium of thickness $t > R_0$.

Substituting $x = 0$ into (4), the tracklength entering the inert medium is obtained as $NA R_0^2 / 8$ and hence the average effective dose, ie the track length per unit volume, is

$$\frac{M R_0}{8 t} \quad \dots (5)$$

where $M = N R_0$ is the effective dose within the radioactive medium for $h > R_0$, ie the infinite matrix dose.

Case B: Inert medium of thickness $t \leq R_0$

The track length that exits across the plane at $x = t$ is obtained by substitution into (4), whence the average effective dose between $x = 0$ and $x = t$ is

$$\frac{M}{2} \left\{ 1 - \frac{3t}{4R_0} - \frac{t}{2R_0} \ln \left(\frac{R_0}{t} \right) \right\} \quad \dots (6)$$

Case C: Inert medium of thickness t from which a surface layer I is removed before measurement.

By subtracting the track length exiting across $x = t$ from that entering across $x = I$ the average dose is obtained as

$$\frac{M}{2} \left\{ 1 - \frac{3(t+I)}{4R_0} - \frac{(t+I) \ln R_0}{2R_0} + \frac{(t^2 \ln t - I^2 \ln I)}{2R_0(t-I)} \right\} \quad \dots (7)$$

with the restriction that $t \leq R_0$. If $t \geq R_0$ then there is no exiting of track length and the average dose is

$$\frac{M}{2} \left\{ \frac{R_0}{4(t-I)} - \frac{I}{(t-I)} + \left[\frac{I^2}{4R_0(t-I)} (3 + 2 \ln \frac{R_0}{I}) \right] \right\} \quad \dots (8)$$

The effective dose at depth x , obtained through differentiation of (4) is

$$\frac{M}{2} \left\{ 1 - \frac{x}{R_0} - \frac{x}{R_0} \ln \left(\frac{R_0}{x} \right) \right\} \quad \dots (9)$$

This is illustrated in Fig. 2 as curve (a); curve (b) shows the average dose, illustrating Cases A and B. In table 1 numerical values are given for all three cases.

In application the expressions should be summed over the individual alpha particle emissions of the thorium and uranium chains (see accompanying note by Grün). An approximate result can be obtained by using the average value of R_0 for each chain, viz. 0.026 mm for the thorium series and 0.021 mm for the uranium series, both in equilibrium; these values assume, for both media, a density of 2.7 g/cm³ and a stopping power equal to that of pottery as given by Bowman (1982) who also indicates that the stopping power of calcium carbonate is close to that of pottery. In practical application the values of R_0 are liable to be different between the two media on account of differing densities; the best approximation is then given by using the value corresponding to the inert medium.

Dose in radioactive medium

The residual range exiting from each surface of a layer of thickness $H (\leq R_0)$ of radioactive medium sandwiched between inert media is obtained by putting $x = 0$ in expression (3) and integrating from $h = 0$ to $h = H$, ie

$$\frac{NR_0 H}{2} \left\{ 1 - \frac{3H}{4R_0} - \frac{H}{2R_0} \ln \left(\frac{R_0}{H} \right) \right\} \quad \dots (10)$$

By doubling this and subtracting from the residual range generated in the layer the average effective dose is obtained as

$$M \left\{ \frac{3H}{4R_0} + \frac{H}{2R_0} \ln \left(\frac{R_0}{H} \right) \right\} \quad \dots (11)$$

For the case when $H > R_0$ the residual range exiting from each surface is obtained by substituting R_0 for H in (10), ie $(NR_0^2/8)$. Hence the average effective dose is

$$M \left\{ 1 - \frac{R_0}{4H} \right\} \quad \dots (12)$$

Because, for alpha particles, backscattering effects are negligible the dose in the radioactive medium is not influenced by the stopping power or density of the inert medium and the assumption that R_0 is the same in both media.

Table 1.
Effective alpha doses (expressed as fraction of infinite matrix dose)

(x/R_0)	Average dose for $t=x$	Dose at depth x	Percentage removed
0.01	0.485	0.472	3.9
0.02	0.473	0.451	7.6
0.03	0.463	0.433	11
0.04	0.453	0.416	15
0.05	0.444	0.400	18
0.1	0.405	0.335	32
0.15	0.373	0.283	45
0.2	0.345	0.239	55
0.25	0.320	0.202	64
0.3	0.297	0.170	71
0.4	0.259	0.117	83
0.5	0.226	0.077	90
0.6	0.199	0.047	95
0.7	0.175	0.025	98
0.8	0.156	0.011	99
0.9	0.139	0.003	
1.0	0.125	0	
1.5	0.083		
2	0.063		
2.5	0.050		
3	0.042		
4	0.031		
5	0.025		

Note: 'Percentage removed' gives the percentage decrease in the average dose in a layer of thickness $t = R_0$ due to removal of $I = x$.

Acknowledgement

The need for these calculations was brought to my attention by Rainer Grün, who in the accompanying paper, reports values based on the individual ranges. I am grateful to Dr Barnaby Smith for checking my calculations.

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Figure 1

Alpha track crossing boundary between radioactive medium and inert medium; the initial range R_0 is assumed to be the same in both media. The residual range crossing an imaginary plane at depth x is $R_0 - R$.

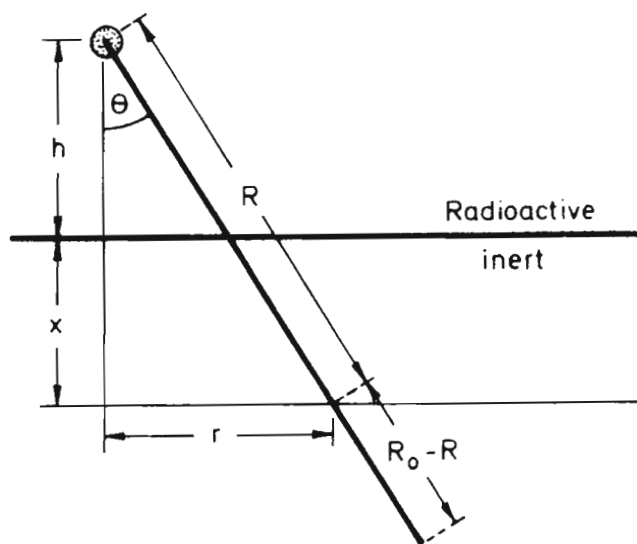
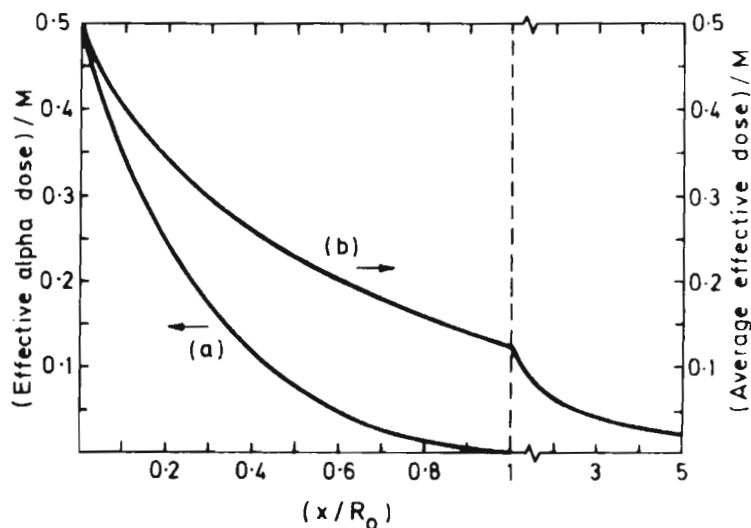


Figure.2

Effective alpha dose in surface layer of inert medium close to its plane boundary with radioactive medium in which the infinite matrix effective alpha dose is M ; the distance into the inert medium is x and the initial range is R_0 . Note change of horizontal scale at $(x/R_0) = 1$. Curve (a) represents the dose at distance x ; curve (b) represents the average dose between $x = 0$ and x .



Alpha effectiveness in ESR dating: a preliminary note on energy dependence

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A small research accelerator has been used as a source of alpha particles of selected well-defined energies to investigate the relationship between alpha effectiveness and the energy of the incident alpha particles. A general description of the accelerator and the irradiation procedure is given in Lyons et al (1985), and further details of the method will be given in due course.

The experimental method differed conceptually from the standard method in TL dating, as the sample thickness chosen was sufficiently great to allow all the energy of the incident alpha particles to be absorbed in the sample, thus simulating an infinite medium: the effectiveness of alphas of different energies could therefore be compared directly without further correction.

A range of energies from 1.5 - 8.8 MeV was used. Not all results are to hand as yet, but preliminary results strongly suggest that alpha effectiveness is dependent on the energy of the incident alpha particles (Fig.1): the total number of defects, as measured by ESR, is not simply a function of total dose but also depends on alpha energy. This can be seen clearly by the non-linearity of the ESR/particle vs MeV plot (Fig.2). When the same data is plotted vs range of the incident alpha, a straight line provides a reasonable fit (Fig.3). Thus alpha effectiveness appears to be linearly related to track length rather than to alpha energy. This is consistent with a model of local saturation along the track of the alpha particle, so that the increase in the rate of energy deposition when the alpha particle energy falls below 2 MeV does not cause a corresponding increase in the rate of defect production.

This concept underlies the a value of Aitken and Bowman (1975) and the b value of Bowman and Huntley (1984), but it is not always appreciated that direct use of the dose-rate tables of Bell (1979) implicitly assumes that alpha effectiveness is proportional to dose rather than range; consequently care needs to be taken in converting radionuclide concentration into effective dose-rate. The size of the correction depends on the reference energy used in evaluating k , as defined by Zimmerman (1971). For example, if a reference energy of 3.7 MeV is used, the effective dose derived for the ^{238}U chain in secular equilibrium on the assumption that the k -value is independent of particle energy is approximately 20%

lower than the value calculated on the basis indicated as being correct by the results given above. A fuller report is in preparation.

Acknowledgement

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PI Reviewers Comments (M.J. Aitken)

It is reassuring to find that these measurements, carried without influence from knowledge of the TL situation, indicate that the concept of constant ESR per unit length of track is valid. This is in keeping with the basic model of alpha effectiveness that stems from David Zimmerman's work.

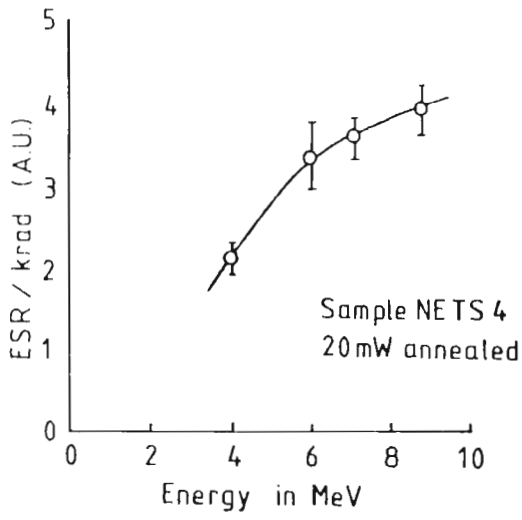


Figure 1. Alpha effectiveness (ESR/krad) as a function of energy.

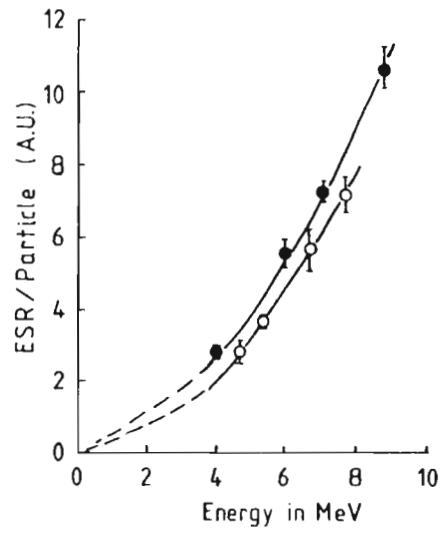


Figure 2. ESR/particle vs. energy. The two lines arise from the different storage times of the sample sets before measurement and are due to interference from an unstable peak.

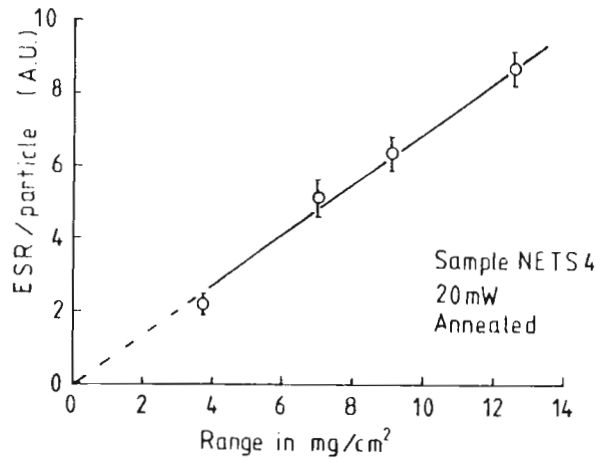


Figure 3. ESR/particle vs. range.

Alpha dose attenuation in thin layers

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When dating thin walled samples like foraminifera, terrestrial snails, or thin walled mollusc shells by TL or ESR it is sometimes not possible to remove the outer 20 μm in order to eliminate most of the volume being penetrated by α -rays from the matrix. Bell (1980) calculated α -dose attenuation curves for quartz grains based on formulas given by Charlton & Cormack (1962) which were tabulated by Howarth (1965).

The absorbed α -dose D_α at a depth x into a non-radioactive layer being irradiated by a radioactive infinite matrix (from one side = 2π -geometry) can be expressed by (Howarth, 1965):

$$D_\alpha = \frac{n_0 E_0}{m^s} \cdot G_\alpha \left[\frac{x}{R_0} \right]$$

where n_0 is the number of α -particles with initial energy E_0 and maximum range R_0 . $G_\alpha(x/R_0)$ is a geometrical function which depends on the configuration of the interface of radioactive/non-radioactive bodies and the energy-range relation for α -particles; m^s is the ratio of the mass stopping powers of matrix over sample (in the following calculation = 1).

Howarth (1965) tabulated the values for P_α (the geometrical function for a plane interface) in steps of $1/100 R_0$. In this paper, these values were numerically integrated for the α -decays occurring within the U- and Th-decay chains using maximum α -ranges from Aitken (1985: Tab. J1-J3) yielding average α -doses received by a volume of thickness x subjected to an α -emitter up to a thickness of 50 μm which corresponds to the maximum range occurring; that of ^{212}Po with an initial energy of about 8.8 MeV. The calculated values are shown in Table 1. For thicknesses above 50 μm a linear extrapolation can be carried out using the values for 50 μm .

However, as shown by Zimmerman (1971) although the energy loss of an α -particle increases as it slows down, the α -efficiency decreases. These effects led to the development of the a -value system (summarized by Aitken, 1985) in which it is assumed that the TL (and ESR) intensity along unit length of α -track is more or less constant. This system was

experimentally confirmed for ESR by Lyons (1987). Therefore, it seems appropriate to apply the formulae from Aitken (1987).

Table 2 can be used to determine the average effective α -dose for TL or ESR age calculations, given a matrix with a homogeneous distribution of alpha-emitters such as clay or loess.

The values of the average α -doses in Table 2 are slightly lower than in Table 1. This raises the question about the validity of α -attenuation curves for grains as estimated by Bell (1980), who did not consider an a -value system.

In the case of terrestrial snails, which have typical shell thicknesses of about 45 μm the α -contribution of an average clay (3 ppm U; 9ppm Th; 1% K; $k = 0.1$; 4 π -geometry) is about 7.5% of the total external dose. With thicker samples it seems to be sufficient to remove the outer 20 μm in order to eliminate the influence of external α -irradiation, since this treatment removes the volume which contains 96% (^{232}Th -decay chain) to 100% (^{238}U to ^{230}Th) of the total external α -dose. As can be seen from Table 2 the α -attenuation within various the U-decay chains (^{235}U ; ^{238}U to ^{230}Th ; ^{230}Th to ^{206}Pb) is slightly different, which should be taken into account when considering U-series disequilibria. However, the uncertainties in the determination of the effective alpha efficiency (especially in ESR dating) and estimation of the precise thickness of a sample is normally much larger than these differences in α -attenuation.

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Table 1

Average α -doses (as percentage of the infinite matrix dose) for calcite samples with thickness x irradiated from one side (2π geometry) and infinite matrix doses (imd) for 1ppm U and Th (Nambi and Aitken, 1986). Density of calcite = 2.7 gm/cm³

Thickness x (μm)	²³² Th-decay chain	U-series	²³⁵ U-decay chain	²³⁸ U - ²³⁰ Th	²³⁰ Th - ²⁰⁶ Pb
imd [$\mu\text{Gy/a}$]	738	2781	124	555	2103
1	46.8	46.2	46.6	45.4	46.7
2	44.1	43.0	44.0	41.4	43.5
3	41.8	40.5	41.6	38.4	41.1
4	39.7	38.1	39.3	35.4	38.9
5	37.7	36.0	37.5	33.0	36.8
6	35.9	34.0	35.7	30.5	34.9
7	34.2	32.3	33.9	28.7	33.3
8	32.7	30.5	32.4	26.6	31.6
9	31.3	28.9	31.0	24.7	30.0
10	29.9	27.4	29.6	23.1	28.5
15	24.1	21.3	23.6	16.7	22.4
20	19.7	16.9	19.2	12.7	17.9
25	16.4	13.9	15.9	10.1	14.8
30	14.0	11.7	13.4	8.4	12.5
35	12.1	10.1	11.6	7.2	10.7
40	10.6	8.8	10.1	6.3	9.4
45	9.5	7.8	9.0	5.6	8.4
50	8.5	7.0	8.1	5.1	7.5

Table 2

Average effective α -doses (EAD) in a layer with thickness x , effective α -doses (ED) at depth x (as percentages of the infinite matrix dose) for calcite samples irradiated from one side (2π geometry) and percentage of total α -dose (PR) which is eliminated by removing x from a layer with thickness $>$ maximum α -range. Density of calcite = 2.7 gm/cm³.

Thickness x [μm]	²³² Th decay chain			U-series			²³⁵ U-decay series			²³⁸ U - ²³⁰ Th			²³⁰ Th - ²⁰⁶ Pb		
	EAD	ED	PR	EAD	ED	PR	EAD	ED	PR	EAD	ED	PR	EAD	ED	PR
1	45.1	41.3	13	44.5	40.2	16	45.2	41.3	14	43.4	38.3	21	44.9	40.8	15
2	41.7	35.4	26	40.7	33.8	30	41.7	35.5	26	38.9	30.9	38	41.3	34.7	28
3	38.8	30.7	36	37.5	28.7	41	38.9	30.9	37	35.2	25.1	52	38.2	30.0	39
4	36.3	26.7	45	34.7	24.4	51	36.4	26.9	46	32.1	20.4	63	35.6	25.7	49
5	34.0	23.2	53	32.3	20.8	59	34.1	23.5	54	29.3	16.4	72	33.2	22.1	57
6	32.0	20.3	59	30.1	17.6	66	32.1	20.4	61	26.9	13.1	79	31.1	19.0	64
7	30.1	17.7	65	28.1	14.9	72	30.2	17.8	67	24.7	10.3	85	29.2	16.3	70
8	28.4	15.3	70	26.3	12.5	77	28.5	15.4	72	22.8	7.9	89	27.4	13.9	75
9	26.8	13.3	75	24.7	10.4	82	26.9	13.3	76	21.0	6.0	93	25.8	11.8	79
10	25.4	11.5	79	23.1	8.6	85	25.5	11.5	80	19.4	4.3	95	24.3	9.9	83
15	19.6	5.3	91	17.2	3.0	95	19.6	5.0	93	13.6	0.4	100	18.3	3.7	94
20	15.6	2.1	96	13.4	1.0	98	15.5	1.9	98	10.2	0		14.3	1.2	98
25	12.7	0.8	99	10.8	0.4	99	12.6	0.5	99	8.1			11.6	0.5	99
30	10.7	0.4		9.0	0.1		10.6	0.1		6.8			9.7	0.2	
35	9.2	0.2		7.8	0		9.1	0		5.8			8.3	0	
40	8.1	0.05		6.8			7.9			5.1			7.3		
45	7.2	0		6.0			7.1			4.5			6.5		
50	6.5			5.4			6.3			4.2			5.8		

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PR Reviewer's Comments (M J Aitken)

We are indebted to Dr Grün for these useful reference data. Note that for material other than calcite the thickness given should be multiplied by that ratio of the densities.

6th INTERNATIONAL SPECIALIST SEMINAR ON THERMOLUMINESCENCE AND ELECTRON SPIN RESONANCE DATING

Early July 1990
Clermont-Ferrand, France

The 6th *Specialist Seminar on TL and ESR Dating* will be held at
Clermont-Ferrand, early July 1990.

Clermont-Ferrand is situated in the Massif Central, 380 km south of Paris. The weather is usually fine in July (20-30°C). Clermont Airport is linked by daily flights to Paris and weekly flights to London. The Railway Station is linked by several daily trains to Paris and other main directions.

The meeting will be held in the same spirit as the previous ones, including both oral and poster communications concerning recent developments in the fields of TL and ESR applied to dating in archaeology and geology. Special emphasis will be given to review papers.

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Experimental TL techniques for the Inclusion method

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Our TL data-gathering is based on weight-normalized procedures using a modified Littlemore 711 TL system. Laboratory gamma doses are administered separately in a Co-60 facility for the additive dose method. The TL data are collected in a clock-driven multichannel scaler (MCS), which necessitates a strictly linear temperature-time relation. The Littlemore TL system compares a chromel-alumel thermocouple output voltage responding to the temperature of the heating strip, with an electronically developed voltage ramp rising linearly with time. The difference signal drives the heater current source in the feedback loop. However, the chromel-alumel junction voltage output is not linear, and this not only misidentifies the temperature to be associated with a given channel in the clock-driven MCS recording of the glow curve but also produces a systematic error in the temperature interval corresponding to the channel width. This latter error is the more serious of the two and has been measured to be as large as 14% in the range 100°C to 500°C.

The necessary linearity is achieved by using a Fluke thermocouple reader with automatic ice point correction and a microprocessor circuit to remove the chromel-alumel thermocouple nonlinearity. The temperature read out is accurate to $\pm 0.1^\circ\text{C}$. Attempts to use the analog readout of the Fluke directly as the input to the Littlemore temperature feedback loop failed because of the long time constant involved in the sampling time of 3 msec in the Fluke. This problem was overcome by using two identical thermocouples welded to the same spot on the heating strip. One fed the Fluke and the other was used as a fast responding signal capacity coupled to an adding circuit is the input to the Littlemore temperature feedback loop. A capacitor was adjusted to give critical damping. The resulting slope, dT/dt (temperature/time), was constant to within the limits of measurement i.e. approximately 1 or 2%.

The MCS has a relatively long deadtime ($\tau \sim 6 \mu\text{s}$). To avoid excessive correction for high counting rates, the PM pulses were fed, after appropriate discrimination, to very fast stacked decade prescalers ($\tau \sim 30 \text{ ns}$). The effect of the prescalers is not only to reduce the effective counting rate seen by the MCS but also to de-randomize the pulse sequence so that pulse overlap is minimized. Thus the only deadtime

correction we have applied is that for the input to the fast prescaler.

At the end of a run the contents of the MCS, which have had the oven glow subtracted, are dumped to an Apple IIe Computer. The deadtime corrections are made, weight normalization performed and the data stored on disk for subsequent processing. The temperature-channel relation is also computed and stored with the data.

Aliquots of the sample of about 1 mg in weight are contained in a small bowl set in a hemispherical depression in the Mo heating strip. To minimize thermal gradients the bowl is made of copper, but the surface must be stabilized (Templer, 1986). Oxidizing the surface by boiling 2 hours in H_2O_2 produced an adherent black surface of low reflectivity, minimizing the effect of changes in the surface properties. This worked well with quartz but proved to be unstable with calcite. Electroplating first nickel and then gold on the copper produced a very stable surface that has worked with calcite as well as with quartz, and produces no spurious TL.

The measured temperature is that indicated by chromel-alumel thermocouples spot-welded to the underside of the hemispherical well in the heating strip. With the aliquot in the copper bowl nestled in the well, there is thermal resistance between the material of the strip and the bowl and between the material of the strip and the bowl and the grains of TL material. The desired temperature within these grains can lag seriously behind the recorded temperature of the thermocouple. Elementary calculations indicate that the effect of this lag is to shift the glow response by a constant temperature.

The presence of this lag can be reduced by increasing the thermal conductance of the contact. Wintle and Dijkmans (1985) suggest the use of thermal contact substances. However, we have been concerned with the possible condensation of volatile substances on the photomultiplier tube window and have avoided this expedient. The use of helium as an atmosphere and the use of the lowest possible heating rate help minimize the thermal lag. We use about 1°C/s , somewhat lower than customary in the TL dating field.

There are actually two thermal barriers of interest in the system, between the heating strip and the copper bowl and between the bowl and the grains of the specimen. We cannot eliminate the latter, but because only a simple linear shift in temperature is involved we can reduce or eliminate the former by calibration. Some characteristic point on the glow curve is chosen, such as the point on either the low or the high temperature side at which the intensity is some fixed fraction of the peak intensity (we usually use half). Each experimental run is corrected in the computer by sliding the data along the temperature axis so that this characteristic point for the experimental run coincides with that for the temperature calibration run. Shifts ranging up to 25°C are required. Figure 1 illustrates the technique, showing two runs with TLD 400, one in the copper bowl and one with the dosimeter powder directly in the well in the heating strip. The data are normalized to the same area. Fig. 1a. shows the data as taken, fig. 1b the same data after a downward shift of 22°C for that taken with the copper bowl. The shifts we observe are similar to shifts reported by Wintle and Dijkmans (1985).

With the normalized glow data and the corresponding shifted temperatures stored in the computer repeat runs can be averaged channel by channel to produce mean glow curves with standard deviations at each channel. Manipulations such as calculating plateau's are then readily performed.

In the additive dose method of obtaining the Estimated Dose it is necessary to extrapolate the TL data to the dose axis. Provided the glow growth curve is linear the extrapolation is relatively simple using some form of linear regression. However, non-linearity is often present. Mejdahl (1985) has shown that in a number of cases a "saturating exponential" involving the added dose x fits the data quite well:

$$y = y_{\infty} \{ 1 - \exp [- \beta(x + x_0)] \}$$

A generalized non-linear least squares program can be used to produce a best-fit of eqn. 1 to the data. We have found a simpler technique that produces fits almost identical to the generalized result. By rearranging eqn. 1 and taking natural logarithms of both sides we produce

$$\ln (y_{\infty} - y) = A - \beta \cdot x$$

with $A = \ln y_{\infty} - \beta \cdot x_0$

A simple one-parameter least squares program then searches for the value of y_{∞} producing the minimum variance of the experimental data about the calculated line, obtaining values for β and x_0 for each value of

y_{∞} examined by the linear regression implied by eqn. 2. Invariably the generalized method produces a lower variance but the difference in parameters is usually small. At the very least, the simplified technique gives a good starting point for the generalized method.

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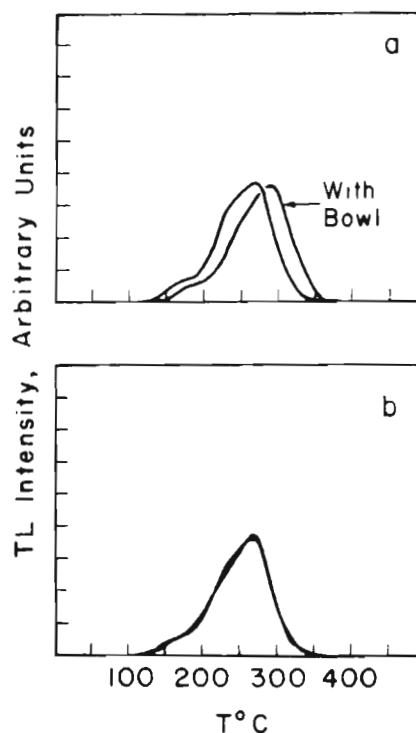


Figure 1
a. Glow curves of TLD 400, with sample in the copper bowl, and without the bowl (sample directly in the well of the heating strip).
b. Glow curves as in 1a, after temperature - shifting.

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