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A correction procedure for ambient activation in pre-dose dating

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Introduction

The initial TL sensitivity of the 100°C peak, S_o, used in predose dating is often non-negligible compared to the sensitivity measured after activation of the palaeodose, S_n. The question then arises as to whether the measured initial has increased during burial due to "ambient activation" of the palaeodose. Described here is a procedure which can be used to estimate the degree of ambient activation experienced by such a quartz sample. The basic idea is that the thermal activation characteristic (TAC) for an ambiently activated sample will lie at higher temperatures than if the sample had not experienced this effect. In principle, the difference between natural and artificial TAC's can be used to quantify the magnitude of the activation.

Basic Principles

Consider some arbitrary TAC defined in relative terms using the nomenclature of Bailiff.

$$\int_T = \frac{S_T - S_o}{S_{max} - S_o} \qquad (1)$$

If an identical aliquot of this sample were ambiently activated by some fraction f of the total possible $(S_{max}-S_o)$, the measured initial sensitivity, S_o^* would be greater and the measured TAC will instead be $\int_T{}^*$

$$\int_T{}^* = \frac{S_T - S_o{}^*}{S_{max} - S_o{}^*} \quad \text{for } S_T \geq S_o{}^* \qquad (2)$$

$$= 0 \qquad \text{for } S_T < S_o{}^*$$

where

$$S_0^* = S_0 + f(S_{max} - S_0) \text{ and } 0 < f < 1$$

The two-part solution is required by the condition that the new TAC cannot be negative. Substituting in the definition of S_0^* we get

$$\int_T^* = \frac{\int_T - f}{1 - f} \text{ for } \int_T > f \tag{3}$$

$$= 0 \qquad \text{for } \int_T \leq f$$

The difference $\;_T$ between the original and ambiently activated TAC's is

$$\Delta_T = \int_T - \int_T^*$$

$$= \frac{\{f\}}{\{1-f\}}\{1-\int_T\} \qquad \text{for } \int_T > f \tag{4}$$

$$= \int_T \qquad \text{for } \int_T \leq f \text{ (since } \int_T^* = 0)$$

Thus, at increasing glow curve temperature, Δ_T increases as $\int_T$ until the temperature at which $\int_T = f$. From this temperature onward, Δ_T follows $(f/1-f)(1-\int_T)$. Since this function decreases with increasing temperature, it can be seen that Δ_T peaks at the temperature at which $\int_T = f$ and it has a maximum value of f. The maximum value of the difference between the two TAC's, therefore, gives the degree of ambient activation. It can also be seen that f is always less than or equal to S_0^*/S_{max} since

$$\frac{S_0^*/S_{max}}{\Delta_T(max)} = 1 + \frac{(1-f)S_0}{fS_{max}} \tag{5}$$

which is always ≥ 1.

Application to Natural Samples

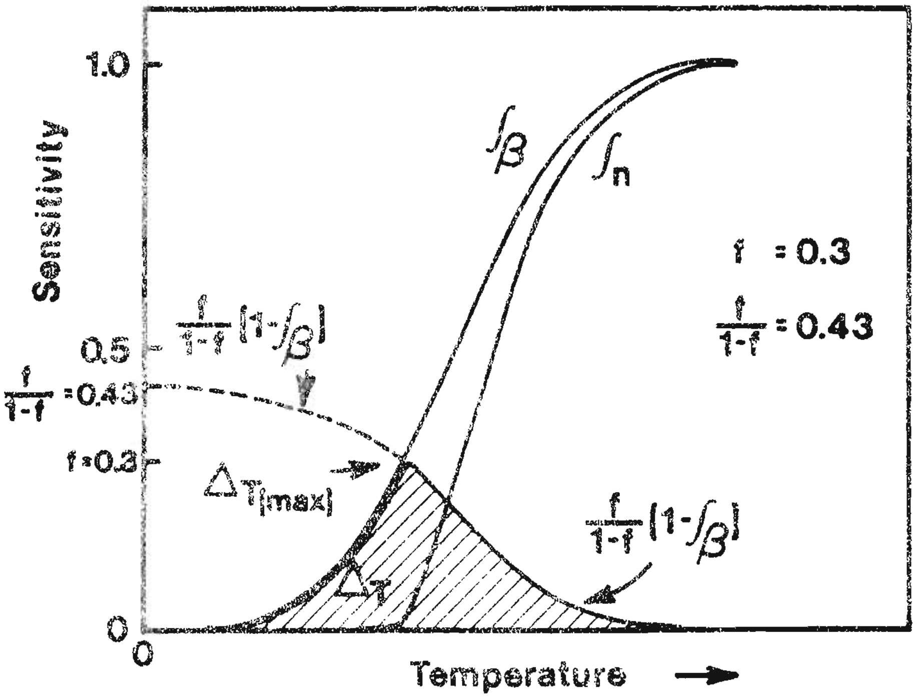
The ambient activation correction is obtained from two TAC's - one for the natural dose $\int_{n,T}$ and one for a laboratory administered dose $\int_{\beta,T}$. The latter is actually from a "natural + artificial" TAC $\int_{n+\beta,T}$, rather than by measuring a second glow activation, to avoid modifications in the activation characteristic which can occur during the first heating. The "$n+\beta$" TAC is a weighted convolution of the "n" and "β" TAC's where the weighting factor is determined by the fraction of TL contributed by each. That is,

$$\int_{\beta,T} = \frac{\int_{n+\beta,T} - \alpha\int_{n,T}}{1 - \alpha} \tag{6}$$

and α = natural dose/(natural dose + lab dose). When a laboratory dose roughly equal to the natural dose is used, $\int_{\beta,T}$ is not very sensitive to α so only approximate knowledge of the natural dose is required. For highest accuracy an iterative approach must be taken.

In this case, Δ_T is taken as $\mathcal{S}_{\beta,T} - \mathcal{S}_{n,T}$ so that equations 4 are used after substituting $\mathcal{S}_{\beta,T}$ for $\mathcal{S}^{\dagger}$. A hypothetical case for "f=0.3" is shown in figure 1. Δ_T peaks at the temperature where $\mathcal{S}_{\beta,T} = f$. The function "$(f/1-f)(1-\mathcal{S}_{\beta,T})$" intercepts the sensitivity axis at $f/(1-f)$ in the limit that $\mathcal{S}_{\beta,T} = 0$.

Fig.1: An Hypothetical Example



<u>Correction of Natural Dose Determinations</u>

The fractional error in the uncorrected natural dose determination D_n^* due to ambient activation is equivalent to the fractional error in the measured sensitivity enhancement. The true natural dose D_n, that obtained after correction for ambient activation, is then

$$D_n = \frac{\{S_{n,max} - S_0\}}{\{S_{n,max} - S_0^*\}} D_n^* = \frac{D_n^*}{1 - \;_{T(max)}} \qquad (7)$$

<u>Simulation Experiment</u>

A simulation experiment was performed to test this method. Two aliquots of pottery quartz (WU-116D, S. Missouri) were given add-on beta doses equivalent to the uncorrected natural dose (using the full value S_0^*) of 100 rads. For the first aliquot, $\mathcal{S}_{n+\beta,T}$ was measured at 50°C intervals up to 450°C. $\mathcal{S}_{\beta,T}$ was then computed as described above. From this TAC, it could be seen that the laboratory dose was activated to 40% of maximum by an activation to 312°C. Accordingly, the second aliquot was pre-activated to 312°C to simulate an ambient activation, and $\mathcal{S}_{\beta,T}$ determined as for the first aliquot. Δ_T computed from these two TAC's peaked at 0.4 in agreement with the prediction.

Ambient Activation Corrections for Some Pottery Quartz

This correction method has been applied to ceramic artifacts from site 23BU10 in the Ozark National Scenic Riverway. The site correlates with the Powers Phase which has 30 associated C14 dates between 630 and 705 years. TL dating results are shown in table 1. Measured ambient activation correction factors ranged from 0.06 to 0.38 and figure 2 shows representative TAC's. In general, the corrected dates are in better agreement with C14 than the uncorrected dates.

Two caveats should be pointed out. First, it is assumed that the TAC's are identical except for ambient activation effects. Second, high accuracy is difficult to attain in practice. This is because high precision temperature control is essential and values are determined by subtracting sensitivities of similar magnitude.

Fig. 2: 116D Quartz TAC's

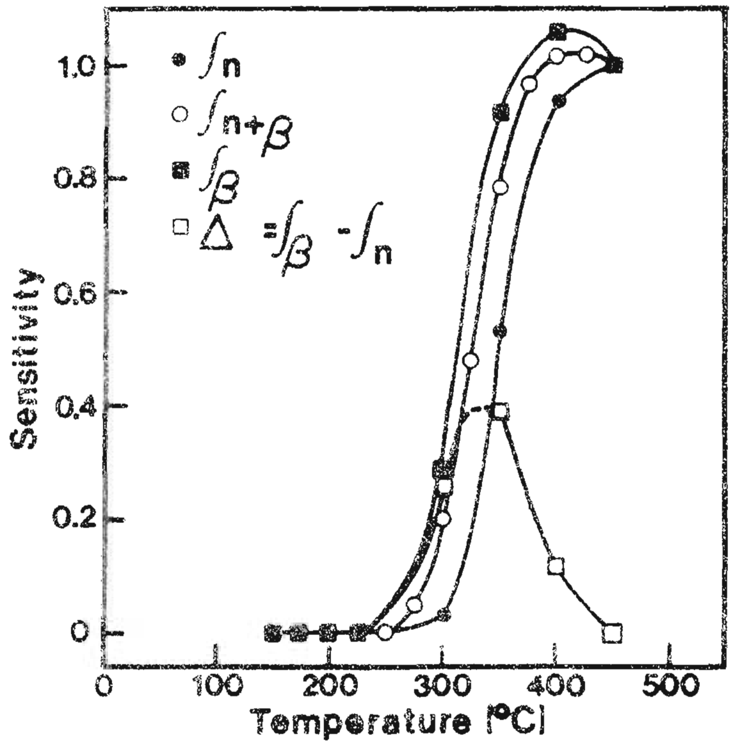


Table 1

Sample	D_n^* (rads)	S_o^*/S_n	Age using S_o^* (years)	Δ_T(max)	Corrected D_n (rads)	Age using correction (years)
116A	125	0.30	465	0.12	140	530
116B	145	0.20	515	0.20	180	650
116C	150	0.12	540	0.10	165	600
116D	100	0.42	495	0.38	160	800
116E	95	0.14	405	0.14	110	470
116F	210	0.12	680	0.06	225	720
116J	180	0.47	615	0.30	255	880

C14 = 630-705 years B.p.

Reviewer's Comments (IKB)

The paper provides a good exposition of the quantitative use of TACs to approach the problem of activation of the accrued dose at ambient temperatures during antiquity. High initial sensitivity is one of the remaining stumbling blocks of the technique that can give rise to significant uncertainty in the evaluation of the accrued dose. Consequently, any procedure that can be used to reduce such error is of importance. However, from our experience in this laboratory we would question its practicability for reasons which are mentioned as caveats by the authors. It is commonly the case that TAC form alters significantly with dose and annealing treatments and these effects will complicate the evaluation of the true initial sensitivity, in addition to the problem of obtaining high precision. The authors do not discuss the use of UV reversal and it would be interesting to test whether; (i) all samples except 116 D, F, and J, where it appears the true initial sensitivity is negligible, are fully UV reversible and (ii) the remaining samples, where ambient activation accounts for part of the measured values of initial sensitivity, are similarly partial UV reversible.

For archaeological samples the possibility of secondary heating should be considered. If it can be excluded there remains the question of whether the measured extent of activation is to be expected at ambient temperatures.

Author's Response

Clearly, additional work is required to test the general applicability of the proposed correction procedure. Comparisons of UV reversibility with activation correction factors would indeed be informative. Errors due to alteration of TAC's by annealing treatment are avoided by using "first glows" as described; however, alteration with dose remains a potentially serious complication. In principle, the correction procedure properly evaluates the integrated effect of any post-firing thermal activation of the initial sensitivity including minor secondary heatings.

Problems with linear regression as applied to TL data

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The purpose of this note is to compare various techniques that are commonly used by workers within TL to fit the 'best' straight line to a set of data points. Despite the fact that the use of some of these techniques has become routine, a comparison is instructive, revealing differences of up to 6% in the intercept on the dose axis.

Discussion will be confined to the 'best fit' rather than to the error terms, and concerns fitting a regression to TL signals vs radiation dose for the simplest of cases i.e. Nat and Nat + beta, in order to determine the intercept on the dose axis when signal = zero or I_0. The nature of the TL data places certain constraints on the regression techniques. First, the pairs of values of signal and dose are not drawn at random, the dose values are selected in the laboratory, usually at regular intervals, whereas the values of TL signal are allowed to vary. Second, the data points may be clustered, with several values of signal for each dose value. Lastly, although both variables will be subject to errors, the errors in signal will tend to be much larger than those for dose.

In order to fit the equation:

$$y = a + bx$$

values of b (slope) and a (intercept on y axis) need to be determined. Owing to the apparently trivial nature of the problem, one is tempted to use a standard regression package without considering its suitability. Details of 5 different techniques for determining the slope of the regression line are given below, in all cases except equation 4, the intercept, a, is given by:

$a = \overline{Y} - b\overline{X}$ where: $\overline{Y} = \Sigma y/n \quad \overline{X} = \Sigma x/n$
n = no. pairs data points

Details of equations.

Equation 1 : least squares regression of y (signal) on (x) dose

The sum of the squares of the vertical distances from the regression line is a minimum (Davis, 1973, pp. 192-200).

$$b = \frac{\Sigma xy - \{(\Sigma x . \Sigma y)/n\}}{\Sigma(x^2) - [(\Sigma x)^2/n\}}$$

Equation 2 : least squares regression of x (dose) on y (signal)

The sum of the squares of the horizontal distances from the regression line is a minimum. Since errors in dose are required for ED estimates this technique may seem the obvious choice. Problems stem from the fact that dose values are not selected at random and Williams (1985) considers that in such a case "the regression of x on y may be so greatly changed as to be meaningless".

$$b = \frac{\Sigma(y^2) - \{(\Sigma y)^2/n\}}{\Sigma xy - \{(\Sigma x \ \Sigma y)/n\}}$$

Equation 3 : major axis regression

The sum of squares of the perpendicular distances from the points to the regression line is a minimum (York, 1966, p. 1079, eqn. 1). This technique represents an attempt to take account of the fact that both the dependent and independent variables are subject to error.

$$b = \frac{\Sigma V^2 - \Sigma U^2 + \sqrt{\{(\Sigma V^2 - \Sigma U^2)^2 + 4(\Sigma UV)^2\}}}{2 \ \Sigma UV}$$

$$U = x - \overline{x}, \qquad V = y - \overline{y}.$$

Equation 4 : weighted least squares regression of y (signal) on x (dose)

This approach is used by Debenham (pers. comm.) and employs a weighting term based onthe assumption that the errors in signal are a fixed percentage of the signal (5% in this case).

$$b = \frac{\Sigma w \ \Sigma wxy - \Sigma wx \ \Sigma wy}{\Sigma w \ \Sigma wx^2 - \Sigma wx \ \Sigma wx}$$

$$w = 1/\ (\sqrt{0.0025\ y^2})^2 \qquad a = \overline{Y} - b\overline{X} \quad \overline{Y} = \frac{\Sigma wy}{\Sigma w} \quad \overline{X} = \frac{\Sigma wx}{\Sigma w}$$

Equation 5 : least squares cubic fit of x (signal) on y (dose)

This weighted regression of x on y is employed by Berger (Wintle pers. comm.) and uses a special solution of the least squares cubic given by York (1966, p. 1082) that assumes no error in y, but that x is subject to error. Unconventionally, Berger treats dose as his dependent, rather than independent variable.

$$b = \frac{\Sigma(wV^2)}{\Sigma(wUV)}$$

$$U = \Sigma\{x - \frac{\Sigma(wx)}{\Sigma w}\} \qquad V = \Sigma\ \{y - \frac{\Sigma(wy)}{\Sigma w}\}$$

$$w = 1/x^2$$

In order to compare these equations a real data set of N, N+beta values (n=8) was used. The data have a correlation coefficient of +0.983. The results are given in Table 1. The raw data are given in the appendix.

Table 1 : Results for different linear regression techniques

Equation	b (slope)	a (intercept)	'ED' dose intercept (y = 0)
1	0.467	5.435	11.639
2	0.483	5.316	11.005
3	0.470	5.413	11.517
4	0.482	5.283	10.956
5*	n/a	n/a	10.992

* slope, intercept not comparable with 1-4

A comparison between equations 1 and 2 using an artificial data set (with r = +0.991) gave values for dose intercept differing by 2.9% (for n=10) and 1.3% (for n=20). The extent of disparity between the different techniques will vary as a function of the correlation coefficient and the number of points in the data set.

Discussion

It is apparent from the example given above that there is a significant variation in intercept values depending on which regression strategy is used. The use of weighting procedures allows the investigator to take account of known analytical errors of the co-ordinates of data points, and such a technique appears particularly valuable in the case of isochron data (Faure, 1977, pp. 89-90). However, if we can assume that the errors in the TL signal are large compared with those for dose, the simplest technique is to use Equation 1 and regress y on x minimising the squares of the vertical deviation about the regression line. Alternatively, if we can assume that the errors in signal are a fixed percentage of the signal then a weighting procedure may be more appropriate. Comments from other workers as to which techniques they use, and why, will be welcomed.

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Reviewed by S. G. E. Bowman, N. C. Debenham and M. Leese.

Appendix: Regression Data

TL signal x 10^3		Dose (min)
4.922	5,2	0
5.536		0
7.569	[illegible]	5
8.608		5
10.279	10	10
9.888		10
12.973	12,3	15
11.729		15

TL studies of Quaternary sediments at the University of Gdansk

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Introduction

A TL laboratory for the dating of Quaternary sediments came into being in our Department in 1982 as the result of increasing interest being shown by scientists working on chrono-stratigraphy and Quaternary geology. The laboratory, run by the authors (the first is a physicist and the second a geomorphologist), is housed in four rooms providing chemical, electronic, analytical and measurement facilities. In this paper we discuss methods for dating Quaternary sediments and the measurement apparatus used in the laboratory.

The laboratory equipment

The apparatus used for estimating the dose-rate comprises (see figure):

1. An AZAR gamma spectrometer with cosmic radiation shield (manuf. by ZZUJ "Polon", Warsaw) which is linked to an RC-82 digital recorder.

2. A ZSG-81 single channel gamma spectrometer (manuf. ZZUJ).

3. A ZR-16 radiometric unit with bisectional proportional counter and including an anticoincidence shield for the measurement of alpha and beta radiation.

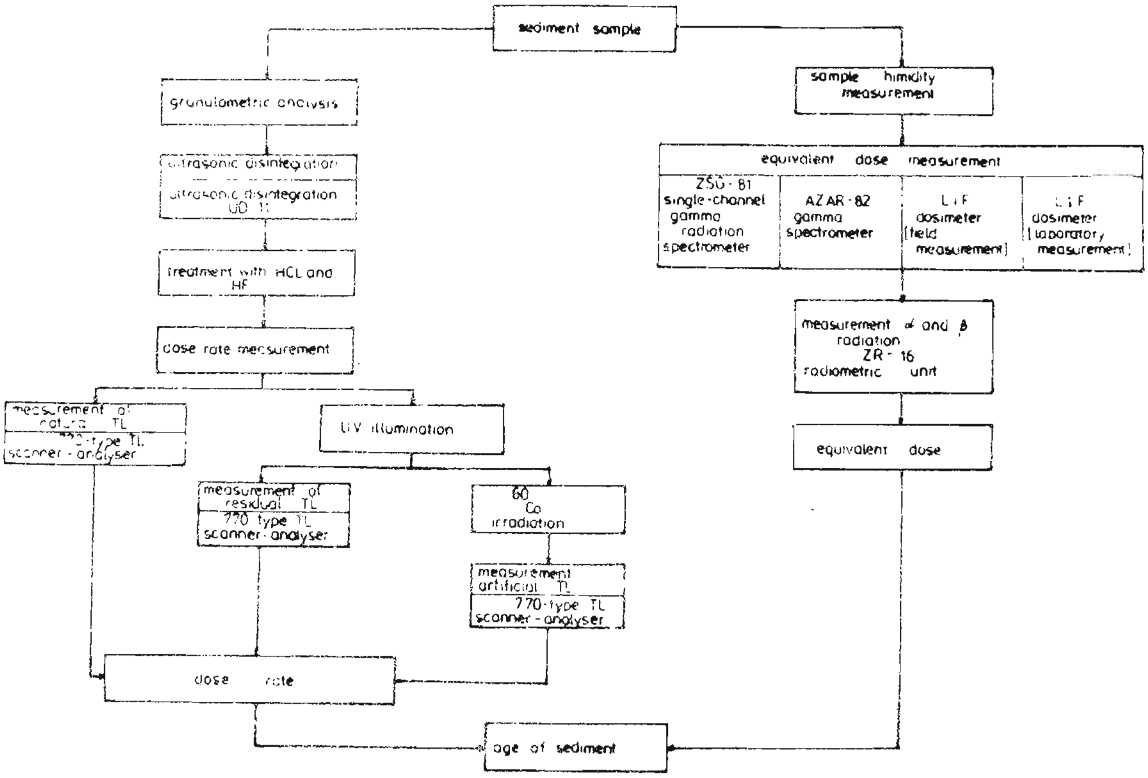
4. A TL scanner - analyser (type 770, manuf. Inst. of Nuclear Physics in Cracow).

The equipment required for equivalent dose measurements comprises:

1. A THYR-2 vibrator with a set of sieves of mesh size from 40 µ to 6.3 mm (made in GDR).
2. A UD-11 ultrasonic disintegrator (manuf. ELPAN, Poland).
3. A TL scanner, as above, coupled to an NE-240 X-Y recorder (manuf. EMG, Hungary).
4. A Co-60 irradiator (located in the Radiological Inst. of the Gdansk Medical Academy) which is capable of administering doses with an accuracy of better than 1.5%.

Methodology of measurements

THE SEQUENCE OF OPERATIONS IN THE TL LABORATORY
OF THE UNIWERSITY OF GDAŃSK IN GDYNIA



Dose-rate

After removal of a sediment sample from the field to the laboratory, it is tested for humidity (%), then dried at 20-50°C. The dose rate, as indicated in the figure, is measured in three ways.

A. Gamma Spectrometry:

A container with 1.5 x 10^{-3} m^3 of sample is placed in the AZAR-82 spectrometer and each measurement, which takes 2000s, is repeated 20 times. The dose-rate is obtained by determination of the concentrations of K-40, Ra-226 and Th-232. The error associated with the gamma dose-rate determination is estimated to be approximately 3%. The single channel ZSG-81 spectrometer is also used and enables the concentration of uranium and thorium to be determined with a measurement error of about 4%.

B. Beta and Alpha Spectrometry:

Samples are placed directly in the active volume of the ZR16's counter and alpha and beta dose-rates are determined with an error of about 3%.

C. TLD:

LiF dosemeters in perspex containers are placed directly in the sample container and stored in the laboratory for a period of at least three months. The equivalent dose, evaluated using the TL scanner, is obtained with a measurement error of approximately ±15%. If field conditions are suitable the dosemeters can also be placed in the sampled deposit.

Determination of Equivalent Dose

We use the reproductive method (N+SL+γ) of Wintle and Huntley (1982). The dried sample is subjected to granulometric analysis. A fraction weighing about 2g is selected from the 800-1000 μm fraction for further analysis (Hutt 1977; Fleming, 1970). The outer surfaces of the quartz grains are cleaned in distilled water for 20 mins in the UD-11 ultrasonic disintegrator operating with an ultrathermostat. The sample is then treated with 5% HCl for 1h and then with 5% HF for a similar time. After each acid treatment, the fraction is rinsed with distilled water and then dried at 20-25°C.

One part of the sample prepared in this way is used for measuring the natural TL and the remainder is illuminated with a sunlamp (uv). The thermal radiation is measured and the sample is then given an accurately controlled dose of radiation from the Co-60 irradiator. The artificial TL is then measured. The TL signals, measured using a heating rate of 1.7 $^{o}C\ s^{-1}$, are analysed in the 270-320°C temperature range and their growth investigated as a function of dose.

The Calculation of Age

The age of a sediment is calculated on a Sinclair microcomputer using a Basic programme. The measurement error is computed by the complete differentiation method (Szydtowski, 1981) and we are presently able to obtain an accuracy in the region of 15%.

Conclusion

So far, our TL laboratory has dated some 300 sediments from all over Poland, including till, glaciomarine and marine deposits. Our laboratory works with and carries out comparative studies for the other Polish laboratories in Gliwice, Lublin and Warsaw, and the Soviet laboratories in Kiev and Tallin. Recently we have been sounding out the possibilities of similar co-operation with laboratories in Denmark, Italy, Great Britain and Australia.

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Reviewed by Galina Hutt and accepted after revisions by the Editor.

REVIEWER'S COMMENTS (Dr. Galina Hutt)

This is a paper worthy of publication in Ancient TL because it describes the research effort and progress of the new TL dating centre in Poland. The apparatus is modern and suitably comprehensive for the present requirements of TL dating.

The comments I would like to make are concerned with the methodology described by the authors:

1. I am doubtful that the authors are employing the quartz inclusion technique. The use of heavy liquid separation (2.6-2.64 g/cm^3) followed by acid treatment of 5% HCl and 1 h and 5% HF for 1h is not, in my opinion, sufficient to obtain pure quartz samples where the alpha-dosed outer layer of grains has been removed.

2. It is advisable to obtain both first and second dose response curves to check for any change in sensitivity after bleaching.

3. The zeroing technique described gives the ideal zero (i.e. where the maximum bleachable TL has been removed). I believe that most of the samples will not have been zeroed to this level at the time of deposition.

Nonetheless, the good dating results achieved by the laboratory suggest that the above problems were not of great importance for the geological region that they investigated.

Cleaning chert with HF -a note

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Carriveau (1977, 1979) has suggested cleaning quartz grains in concentrated HF prior to measurement of their TL spectra. While this procedure has proven successful with quartz grains from pottery, caution should be used in extending it to chert.

During the recent dating of a heat-altered Florida chert tool, a sample of the tool was viced to < 63µm and etched in 50% HF for 15 minutes. On addition of the acid, the chert immediately foamed into an opaque sludge with marked evolution of heat. Testing of the sludge showed that its TL signal had been destroyed. A repeat test showed that the foaming reaction was rapid, going to completion in 10-15 seconds. Weaker HF solutions were then tried. Tests of 1% (60 seconds and 10 minutes) and 2% (10 and 20 minutes) solutions showed neither caused foaming or heat evolution. Examination of the etched grains with a petrographic microscope confirmed that the 50% HF etched grains were completely destroyed while the 1% or 2% etched grains appeared unaffected.

This exothermic sludge may be similar to the "spectacular" failure observed by Huxtable (1982) in a flint from the La Cotte (France) site when she tested it with 40% HF. In both cases, there is the possibility of calcite contamination. Calcite alone, though, would not fully account for the effects observed as the calcite-HF reaction does not markedly evolve heat.

A clue to a possible origin of the effect may lie in a report by Donna Ruhl (Florida State Museum, pers. comm.) that a similar severe foaming and evolution to heat was observed in several North Florida clay samples when she added 40% HF to them. Her solution was to treat the clay samples three times with 10% HCl, with centrifugation and decantation of the supernatant plus a deionized water rinse after each addition, before using the HF. With our chert samples, we use a 3 minute, 38% HCl bath followed by a 20 minute, 4% HF etch.

In conclusion, chert/flint samples which foam when etched with 50% HF may be successfully etched in weaker HF solutions (1-4%) with or without a prior HCl bath.

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Reviewers Comments (S. Sutton)

The advantage of etching ground chert is unclear. As pointed out by Fleming, the purpose in etching quartz grains (from pottery, for example) is twofold; first, to increase transparency by removing material contaminated by impurity diffusion during weathering and, second, to remove part of the alpha-irradiated material. Chert "grains" are not obviously analogous. These "grains" would be expected to have more or less uniform transparency since they are composed of microcrystalline quartz and their surfaces have experienced no weathering. Also, the uranium and thorium distribution in the chert and in the "grains" is likely to be more uniform. It is stated that "grains" etched in dilute HF "appeared unaffected" under petrographic examination. How then was it determined that they were "successfully etched?"

Author's reply

Because chert frequently contains both a glassy phase and trace mineral impurities as well as quartz grains (Clark and Purdy, 'Electron microprobe analysis of weathered Florida chert' American Antiquity 44(3):517-524), the HF etch, preceeded by a HCl etch, is used to try to remove the former while leaving most of the quartz grains behind. Secondly, "successfully etched" was imprecisely worded. The intended meaning was "did not foam into sludge".

Sensitization of TL signal by exposure to light

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Abstract

The regeneration method of obtaining the equivalent dose for polymineral loessic deposits has been shown to give rise to apparent TL ages which are far too young compared with known geological age over the period 50-700 ka. Debenham (1985) proposed that loss of luminescence centres over that time period was the cause of the underestimation. This conclusion was reached on the basis of the apparent exponential nature of the discrepancy between the TL dates (by regeneration) and the geologically estimated ages. This paper presents a different model which could give rise to a similar discrepancy in the dates. It proposes dose-dependent sensitivity changes occurring when the sample is exposed to light. It predicts a hyperbolic, rather than an exponential, discrepancy but it is difficult to differentiate between these two functions.

Introduction

Debenham (1985) has reported that when equivalent doses (EDs) are obtained by regeneration of the TL signal following exposure to a solar simulator, the TL ages obtained for loess from north-western Europe reach a limit of about 100 ka. He concludes that this effect is due to decay of the TL signal through time with a mean life, τ, of about 100 ka; hence the TL age, T, is obtained by $T = \tau\ (1 - \exp[-t/\tau]$ where t is the true geological age of the sample. Wintle et al. (1984) found ages in the range 75-139 ka for the loess and soil complex below the hiatus at 1.8 m at Saint Romain. They concluded that these represented the terrestrial climatic record equivalent to oxygen isotope stages 5e to 5a in the marine record in agreement with the geological interpretation (Lautridou, 1982). However, a later study by Wintle (1985) on soils deeper in the section did not show any increase of TL with depth. This thus confirmed the flattening off of TL age versus real age reported by Debenham and suggests that the interglacial soil at Saint Romain is pre-Eemian. Similar underestimates have been obtained for well-developed interglacial soils in Poland when this technique is employed (Proszynska-Bordas, 1985).

These results can be explained by the slow decay of luminescence centres over the period of interest as shown by Debenham. Fig. 1 shows the effect of such a decay (τ = 150 ka) on the measurement of TL ages. Loss of luminescence centres would affect not just the regeneration method of ED determination, but also the other methods, the total bleach ($N + \beta - I_0$) method and the partial bleach method ($R-\Gamma$) method, to the same extent. On the other hand, several authors (e.g. Buraczynski and Butrym, 1984) have obtained TL dates older than 150 ka using these methods. However, it should be emphasised that their dates could have been overestimated (thus fortuitously cancelling any loss of luminescence centres if the latter is occurring) because of non-linearity of the TL response as the dose is increased.

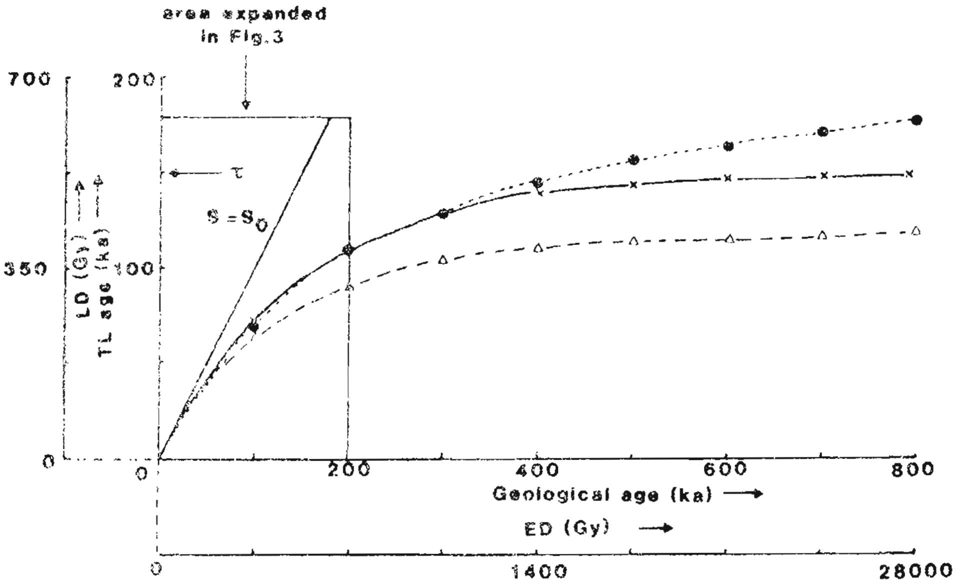

Figure 1: *LD plotted versus ED (also converted to TL age versus geological age using a dose rate of 3.5 Gy/ka). x----x for loss of luminescence centres with τ = 150 ka, ●....● for linear growth and sensitivity change △---△ for exponential growth with a sensitivity change which depends upon the fraction of filled traps.*

Effect of dose-dependent sensitivity change on regenerated growth curves

Another phenomenon which could give rise to an apparent flattening off of the TL age versus geological age plot is now considered. It concerns the effect of a sensitivity change produced by the laboratory light exposure. It is constrained by the experimental knowledge that for young European loesses the sensitivity after bleaching, S, differs negligibly from that for an unbleached sample, S_0. Let us assume that the sensitivity after bleaching depends upon the radiation dose that the sample has received since it was last exposed to sunlight, in a way which is analagous to the pre-dose sensitisation found for quartz and feldspars from pottery when they are heated in the laboratory (e.g. Mejdahl, 1985).

(a) Linear growth curve

In the simplest case, where the growth curves are linear let us assume that the sensitivity after bleaching is given by the equation $S = S_0\ (1 + \dot{D}t/B)$ where t is the geological age, $\dot{D}$ is the annual dose rate, and B is a constant, taken as 800 Gy. The effect on laboratory growth curves is shown in Fig. 2 for samples of various EDs. When the natural TL intensity produced by ED is matched to that induced by laboratory dose LD, we can write

$$S_0 \ .\ ED = S \ .\ LD$$

and hence $S_0\ .\ ED = S_0\ (1 + \dot{D}t/B).LD = S_0\ (1 + ED/B).LD$.

The unique points obtained when the light levels are matched fall on the hyperbola LD = ED (1 + ED/B). This function is also plotted in Fig. 1, with $\dot{D}$ maintained as 3.5 Gy/ka, a fairly typical value for loess. It can be seen that for samples up to 400 ka there is negligible difference between this function and the exponential proposed for decay of luminescence centres with τ = 150 ka. Above 400 ka the curves deviate as the hyperbolic nature of the above equation becomes more pronounced. For the dose region below 200 Gy the responses of these two curves can be seen better in the expanded plot in Fig. 3.

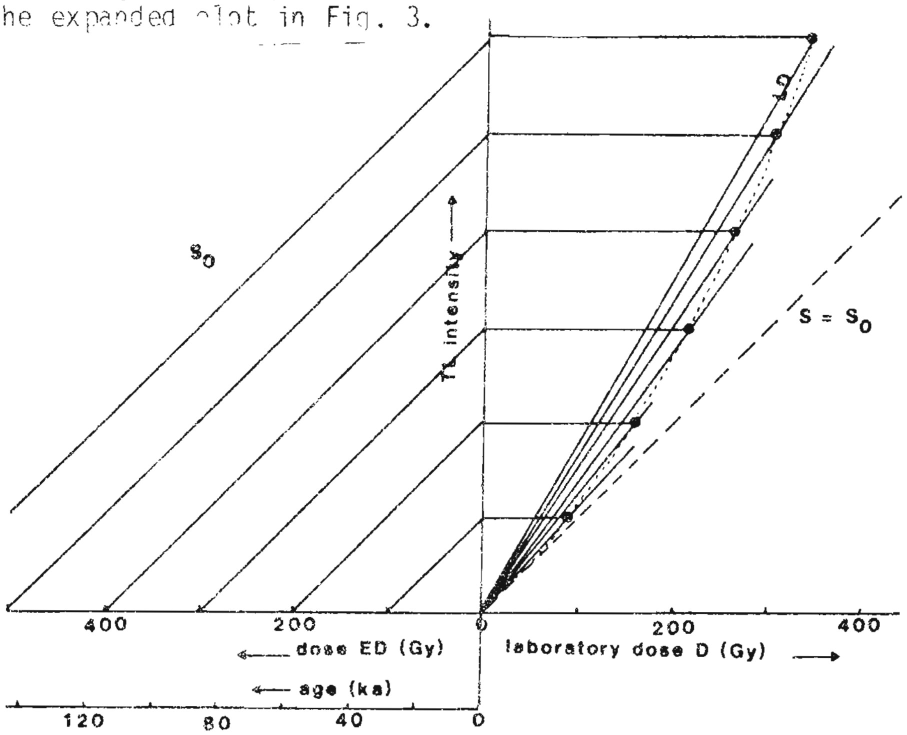

Figure 2: TL intensity plotted as a function of dose for a theoretical sample having linear growth but for which the sensitivity after laboratory bleaching depends upon the equivalent dose ED. This results in an underestimate of the TL age, since the laboratory dose LD which matches the natural TL intensity is systematically smaller than the ED as shown by ● .... ● .... ●

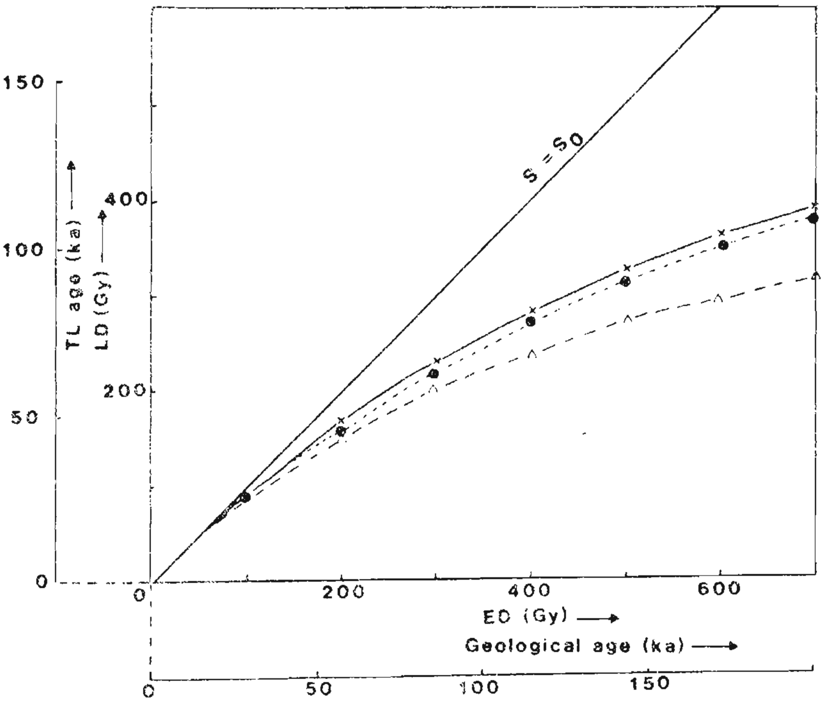

Figure 3: Expansion of the box in Figure 1.

(b) Exponential growth curve

The above linear growth curve model may be insufficient since it has been demonstrated that the growth curve for fine grain loess after bleaching becomes noticably non-linear for doses greater than 200 Gy (Debenham, 1985). Fig. 4 shows a composite regenerated growth curve obtained by matching data for two samples from Saint Romain and two much older ones from Paks and Stranzendorf (H. Proszynska-Bordas, personal communication 1985); normalisation of the data was performed by matching the light level at particular doses assuming that only the sensitivity and not the shape is dose dependent. These data can be fitted reasonably well by a saturating exponential of the form $I/I_0 = (1\text{-}\exp[-D/D_0]$ with $D_0 = 600$ Gy. An equation of this form is preferred since it is that predicted as the finite number of electron traps becomes filled.

Assuming the same exponential growth for the natural TL and for the post-bleaching TL measurements (i.e. no change in growth curve shape caused by bleaching), we can examine the effect of a sensitivity change which depends upon the degree of trap filling i.e. $S/S_0 = 1 + [1 - \exp -ED/D_0]$. The effect of this sensitivity change is shown in Fig. 3, where the relationship between LD and ED is obtained by matching the TL level. This results in

$$(1 - \exp\text{-}ED/D_0) = [1 + (1 - \exp\text{-}ED/D_0)]\ [1 - \exp\text{-}LD/D_0].$$

However, this model predicts that LD will not reach a value greater than about 410 Gy and it also produces a sensitivity enhancement of $S_R / S_N = 2.3$ for a regeneration dose of 270 Gy when the slope of the regenerated curve (S_R) and the N + β curve (S_N) are compared at the same light levels.

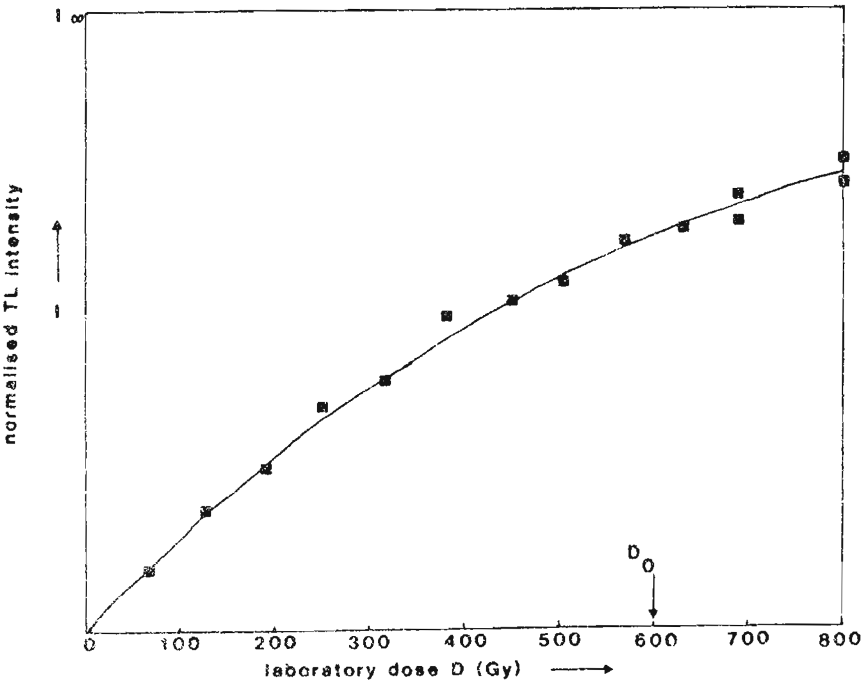

Figure 4: Composite, normalised growth curve data obtained after 450 min. unfiltered sunlamp exposure showing exponential growth for 300-310°C TL signal.

Discussion

Unlike Debenham's hypothesis, the model described above can be tested in the laboratory. Debenham's data implied that S_R / S_N does not increase above 1.6 for regenerated doses up to 400 Gy in his study of a number of geological samples. However, the best test of the model will be a series of experiments of a young loess.

Sensitivity changes after bleaching have been reported in a bleaching study on polymineral fine grains by Rendell et al. (1983) for loess from Pakistan. In their work they found the gradient of the regenerated TL (i.e. S) varied in a non-linear way with the length of bleaching time. If this is a general phenomenon then it adds an additional variable to the evaluation of existing data and to proposed experimental procedures.

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Reviewers' comments:

The virtue of the model presented in this paper is that experiments may be devised to test it. Nevertheless the two problems that can never be resolved are (i) the possibility of dose rate effects and (ii) defect migration either by normal thermal diffusion or by optically enhanced diffusion, because the laboratory to natural dose rates vary by some 10^6:1, and diffusion times of 100 ka could allow defects to migrate throughout the silica or silicate grains of the loess. The familiar demonstration of these problems occurs in the photographic process, and there is no justicication in assuming that the loess presents us with a simpler problem. We should add that in addition to the paper cited (Rendell et al., 1983), we have more recent experimental data indicating that post-bleaching sensitivity changes are extremely complex, at least as far as the Pakistan loess is concerned.

Finally, contrasting TL and geological age estimates is really only helpful if the geological age estimates are related to absolute age determinations - unfortunately geological age estimates often have a flimsier base than those of TL.

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Alpha particle effectiveness: numerical relationship between systems

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There are now four systems: the original k-value system formalized by Zimmerman (1971), the a-value system of Aitken and Bowman (1975), the b-value system of Bowman and Huntley (1984), and the omnidirectional flux system in use at Gif-sur-Yvette (Guerin and Valladas 1980; Valladas and Valladas, 1982; Guerin 1982; Valladas 1985); the latter system deals also with response of coarse grains to an external alpha flux. The purpose of this note is to collate, for convenience, numerical relationships between the various coefficients.

In the k-value system (though Zimmerman used the symbol ε in place of k) the basic definition is:

$$k_{3.7} = \frac{\text{TL/gray for 3.7-MeV alpha particles}}{\text{TL/gray for beta irradiation}}$$

The alpha particles received during antiquity have a spectrum that spreads from 8.8 MeV to zero and to make allowance for this Zimmerman introduced k_{eff}, which for a sample having equal thorium and uranium activities is equal to 0.83 $k_{3.7}$. The drawback to this system is that the value obtained is strongly dependent on particle energy, as also is the absorbed dose. The other three systems are based on the approximation that the TL per unit length of alpha particle is independent of particle energy. This was implicit in the work of Zimmerman and was demonstrated explicitly by Bowman (1976) for a variety of TL phosphors.

The first of these systems was based on the definition

$$a = x'/1300S \qquad (1)$$

where x' rads is the beta (or gamma) dose necessary to induce the same level of TL as 1 minute's irradiation by an alpha source of strength S micron^{-2} minute^{-1}; this latter represents the rate at which track length is delivered to unit volume of the sample. If grays are used instead rads then the definition becomes

$$a = x/13S \tag{2}$$

The numerical factor is equal to the energy loss per micron for a 3.7 MeV alpha particle in quartz divided by the density, i.e. (13S) is the dose-rate for an alpha source delivering 3.7 MeV alpha particles to a thin sample of quartz. Hence a, like k, is dimensionless and the value obtained for quartz, measured on the basis of equation (2), is by definition equal to $k_{3.7}$; the value obtained for a is only weakly dependent on particle energy (to the extent that the TL per unit length of track does have some dependence on particle energy) whereas to measure (or use) k the particle energy must be specified. For a sample having an alpha stopping power ratio r relative to quartz,

$$a = r\,k_{3.7} = r.k_{eff}/0.83 \tag{3}$$

The b-value system avoids any numerical factor in the definition of b, which is,

$$b = \frac{\text{TL per unit alpha track length per unit volume}}{\text{TL per unit absorbed beta dose}} \tag{4}$$

Unlike a, b is not dimensionless and if it is expressed in terms of (gray micron^2),

$$b = 13\,a \tag{5}$$

The Gif system if similar to the b-value system except that the units used are (rad cm^2), so that

$$S_\alpha = 10^{-6}b \tag{6}$$

where S_α represents the beta rads necessary to match the TL level due to a perpendicular alpha flux of 1 particle per cm^2.

Coarse grains

In the quartz inclusion technique (Fleming, 1970) the skin of material irradiated by alpha particles from the clay matrix is removed by etching with hydrofluoric acid. An alternative approach, stimulated by uncertainties in the etching process (Valladas and Valladas, 1982), is to dispense with etching and to make direct experimental measurement of the alpha response of the grains. An important 'theorem' here is that the irradiation situation for a spherical grain exposed to an omnidirectional flux, Φ is identical to that of the grain when it is exposed to a parallel flux of the same value, both fluxes haing the same energy distribution (loc. cit., p. 176); the omnidirectional flux corresponds to the alpha dose received by the grain during antiquity and the parallel flux to laboratory irradiation (the grains are placed on a vibrating pan during measurement so as to minimize error due to grains which are non-spherical). The value of the omnidirectional flux for a sample is equal to four times the particles incident on unit area exposed to 2π steradians of the sample, i.e. the flux rate equals four times the alpha count rate (for zero threshold) per unit area.

It may be useful to note incidentally (see Aitken, 1985, p. 318) that the 'true' effectiveness as obtained by irradiation of fine grains is related to the 'apparent' alpha effectiveness as obtained by irradiation of coarse spherical grains by the ratio

$$\frac{2D}{3\eta' R} \tag{7}$$

where D is the grain diameter, R is the alpha range and η' is the ratio, for the alpha energy used, between the average TL per unit track length for total absorption and that for a thin layer of fine grains. Guerin (1982) reports an experimental value of 6.11 ± 0.55 for this ratio using 3.5 MeV particles and grains of diameter 100 microns. This is consistent with the value of 6.3 obtained for the above ratio on substituting $R = 13$ microns and $\eta' = 0.81$ as appropriate to 3.5 MeV, the value for η' being obtained using the representative energy dependence of TL per unit track length given by Aitken and Bowman (1975); this latter consists of a constant level for energies above 2 MeV with a linear rise between 0 and 2 MeV from 25% of that constant level.

Reviewed by G. Valladas

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An automated beta irradiator using a Sr^{90} foil source

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Introduction

A new off plate beta irradiator has been constructed for the Paisley TL laboratory based on a 4 cm^2 extended area 40 mCi Sr^{90} foil source with a 50μm silver window (type SIQ 9, Amersham International PLC). The design requirements were to maintain external dose rates at below 0.75 mR hr^{-1}, to improve the irradiation geometry and stability compared with previous designs used in this laboratory and to provide an accurate automatic delivery and recording system.

The irradiator features a shutterless irraditation cavity, a linear transport mechanism ensuring uniform distribution of transit dose, and a simple control system based on a cheap home computer with a minimum of external components.

Description

The layout of the irradiator and its central cross-section are shown in figures 1 and 2.

Up to 12 samples on 1 cm discs are mounted on a brass carrier bar which passes through the cylindrical irradiator on a linear track. The central part of the irradiator consists of two interlocking stainless steel sections with a cylindrical cavity on the central axis above which the source is located in a steel capsule.

Stainless steel tubing welded in the central sections encloses a set of lead inserts to provide shielding. We had originally intended to convolute lead into the outer part of the central sections, however to simplify machining an external lead collar was used to sheild this area.

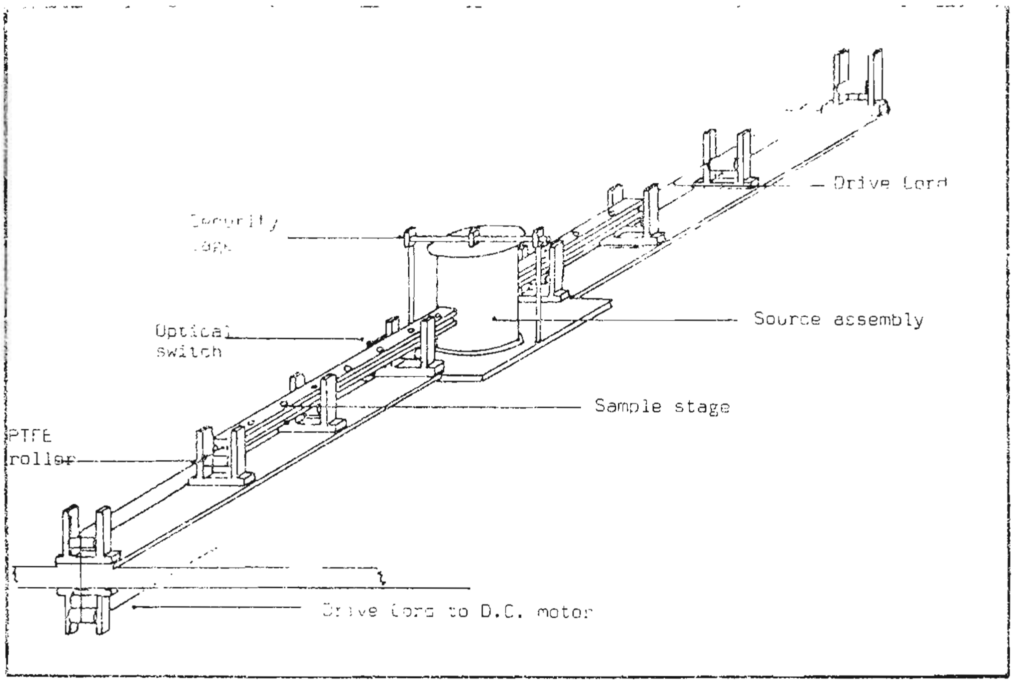

Figure 1: Layout of the beta irradiator.

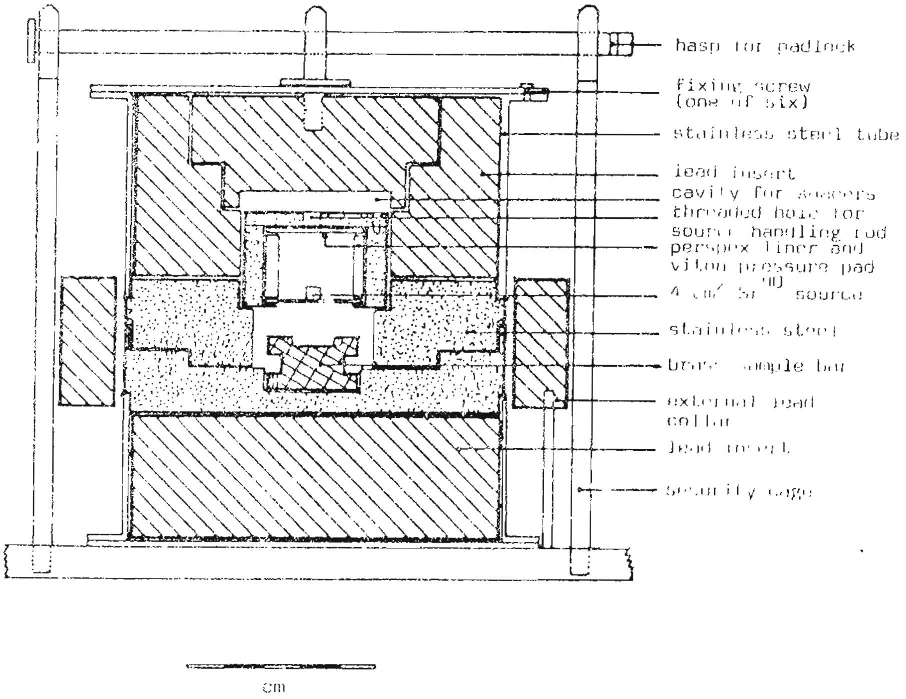

Figure 2: Central cross-section of the source assembly.

The source itself can be loaded and removed on a threaded rod with minimal handling time and a security cage with a padlock prevents unauthorised access to it. Spacer rings can be inserted to alter the source-sample separation.

The sample carrier itself is located by keys on the outside edge of the lower stainless steel section and supported along its track by a series of PTFE rollers whose height is adjusted by shims. A drive cord joins both ends of the carrier to a 12 V reversible d.c. motor mounted underneath the source holder. A series of indexed holes along one edge of the bar are used in conjunction with an optical switch to control the sample position for exposure.

All the central components and the sample carrier were machined to high precision such that the source-sample distance varies by less than 25μm across all 12 positions. Materials in these critical areas were chosen with this in mind and to be self lubricating.

The surface dose rates around the irradiator have been monitored using a calibrated Berthold meter and fall within the 0.75 mR hr^{-1} limit.

Control

A simple, and cost effective controller has been built using a Sinclair ZX Spectrum home computer with an 'Interspec' interface (DCP Microdevelopments Ltd., 2 Station Close, Lingwood, Norwich NR13 4AX) and a CPI ROM card (CML Products, 1 Milton Road, Cambridge, England). The only external electrical components are a 12 V power supply and d.c. motor, a cross-over relay and a Schmidt triggering switch.

The control program is stored in ROM and provides facilities for fixed, incremental exposure sequences accounting for the transit dose to each sample as part of the exposure. A dot matrix printer keeps a permanent log of each exposure which would be particularly useful if an overnight sequence were interrupted by a power cut.

Calibration and Performance

The source has been calibrated using a series of gamma irradiated quartz and CaF_2 samples exposed to an accurately calibrated Co^{60} source at Harwell, and also by intercomparison on two separate occasions with beta sources in the Oxford laboratory. The smallest dose which can be delivered is the transit dose received by a sample passing directly through the irradiation chamber. We have also measured the vertical and lateral fall offs of dose rate from the centre of a sample disc using a similar technique to that described by Bailiff (1980).

These results and the calibrated dose rates to 100 μm quartz grains on 0.25 mm stainless steel discs, with their respective random errors, are tabulated below. We believe that the overall error in the calibration is within ± 5%.

Source-Sample Separation (mm)	Transit Dose/m Gy	Static Dose Rate/m Gy s^{-1}	Vertical Fall off %/mm	Lateral Fall off (at edge of 1 cm disc)
9.75	290 ± 5	32.6 ± 0.4	7%	-3%
14.75	212 ± 4	23.9 ± 0.3	4%	-

Using the SIP type Sr^{90} source most commonly used for TL dating, Bailiff reports a lateral fall off of 10% when comparing dose rates 5 mm off axis and a vertical fall off of some 14% per mm with a source-sample separation of 14.82 mm. The larger area source reported here gives a modest improvement to both parameters.

Conclusions

The larger source has provided an improvement in the beta irradiation geogmetry compared with our previous sources and with those which we believe are most commonly used elsewehre. The external dose levels around the unit are below the 0.75 mR hr^{-1} level allowing access to non-designated persons. While a linear delivery system is less compact than a turntable design it is easy to machine to high precision and when combined with type of control system described is both cost effective and reliable.

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