D. RICHTER

# Ancient TL

"The first rule of intelligent tinkering is to save all the parts."  Paul Ralph Ehrlich

## IN THIS ISSUE . . .

## THE SECOND SPECIALIST SEMINAR ON THERMOLUMINESCENCE DATING

The Second Specialist Seminar on Thermoluminescence Dating was held September 1 to 8, 1980, at Oxford, England.  A total of about 75 oral and poster presentations were made which covered virtually all aspects of TL dating.  Topics included:

TL kinetics
TL apparatus
Intensifier imaging
Emission spectrometers
Spectra
Dose-rate dependence
Dose-rate evaluation
Radon and thoron emanation
Series disequilibria

Alpha counting
Gamma spectrometry measurements
Source calibrations
Dating of slag, volcanic rocks, calcite,
    flint, heated rocks, feldspar, loess,
    sediments, stalactites, meteorites
Dating applications
ESR measurements

Papers from the conference (including poster presentations) are expected to be published in the Council of Europe PACT journal series in June, 1981. Inquiries regarding copies of the proceedings should be directed to:  Dr. A. Hackens, President of the Institute, Institut Superieur d'Archaeologie, et d'Histoire de l'Art, 3000 Louvain, 83 Vlamingenstraat, Belgium. A general discussion was held at the conclusion of the meeting during which Vagn Mejdahl offered to host the next seminar at Risø to be held in 1982. His offer was endorsed by the participants.  Watch Ancient TL for details as they become available.

S. R. Sutton

# EMISSION SPECTRUM OF THE 'FACEY' LIGHT SOURCE

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A light source that is suitable as a reference for the TL detection system and based on a design by Facey (J. Sci. Inst., 1966, 43, 658) is manufactured by D.A. Pitman (Weybridge, Surrey, England). It is claimed to have good stability (0.04% $^{o}C^{-1}$), blue/violet emission and low light intensity (nanolumens). The source consists of several microcuries of C-14 uniformly dispersed in a plastic scintillant. The pulse height spectrum is significantly biassed towards double events and so differs from the single photon spectrum obtained for example, with a light source of the type manufactured by Saunders Roe (betalight D08; tritium and phosphor cell) that characterizes TL. However, the betalights that have been used in this laboratory have not been found to have particularly good temperature stability (0.5-1.0% $^{o}C^{-1}$). We have tested a standard light source which is mounted in an aluminium holder.

The emission spectrum has been measured using a spinning interference spectrometer (see Ancient TL, 2, 1978). The spectrum measured with the source at 20ºC, is shown in figure 1 (plot A). It has been corrected for instrument response and normalised on the basis of total photon count. Reproducibility was better than ± 3%. The spectrum has maximum emission in ch5 ($\lambda$ max=420nm). When plotted on an energy scale the spectrum remains asymmetric. This is presumed to be the result of optical absorption in the UV region by the plastic medium in which the scintillant is immersed. The spectrum was also measured after the light source had been stored for 2h at 50ºC. The normalized spectrum shows a slight shift to longer wavelengths (fig.1, plot B). Over the range of the spectrometer a 6 ±2% loss in counts was recorded. There is a spectral dependence to the loss and in the regions chs1-9 and chs10-16 the losses were 5% and 20% respectively. The former interval, containing 94% of the total count, thus showed a loss of 0.17 ± 0.06% $^{o}C^{-1}$. This is higher than the Pitman figure of 0.04% $^{o}C^{-1}$ (below 40ºC), but for TL detection systems biassed towards the blue or shorter wavelengths, the variation in normal use is likely to be less than ±1%.

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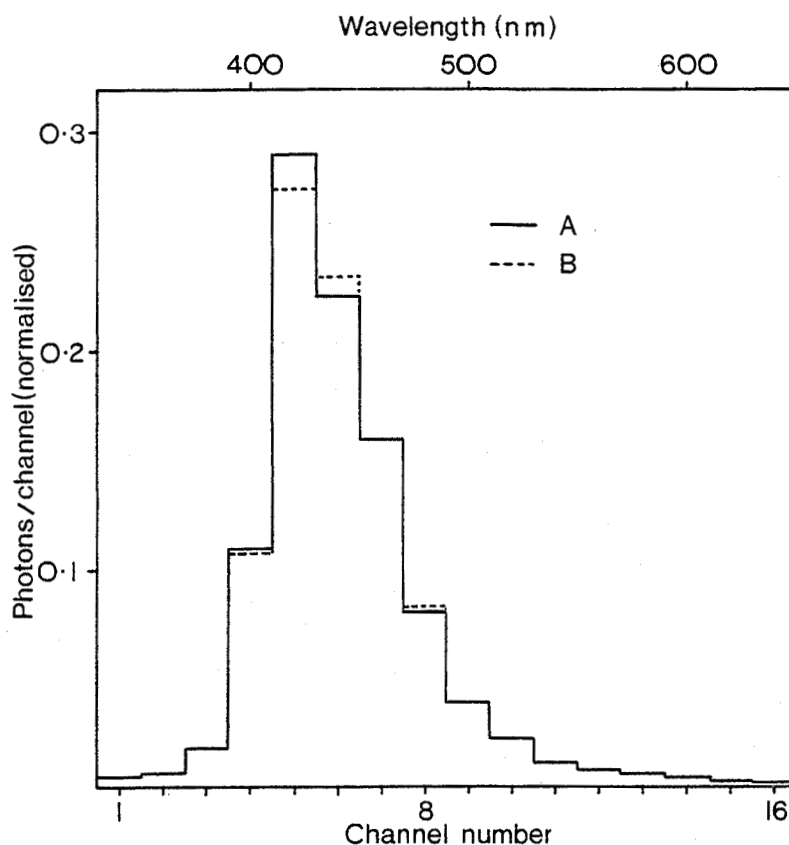
Figure 1          Plot A;   corrected spectrum obtained after
                           a 2 min. count at RT.   Total
                           number of photoelectrons counted
                           was $3.4.10^4$

                 Plot B;   corrected spectrum, measured
                           after storage of light source
                           at 50°C for 2h.


The spectrometer comprises of 16 interference filters each of
which has maximum transmission ($\lambda$max) commencing at 340nm (ch 1)
and progressing in 20nm intervals to 640nm (ch 16).   The scanning
time over the spectral range of the instrument is 125 msec.




## ANCIENT TL WOULD LIKE TO CARRY YOUR CONTRIBUTION !

Space is available in the next issue of Ancient TL (no. 13).  Send your
contributions today to the editor.  The deadline for issue 13 is Dec. 15.

# ALPHA DOSE ATTENUATION
# IN QUARTZ GRAINS FOR THERMOLUMINESCENCE DATING

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Fleming (1969) has calculated the attenuation of the alpha dose from the uranium and thorium series in quartz grains of certain sizes. Aitken and Fleming (1972) used these calculations to predict that the attenuation of the alpha dose in the fine-grain technique of Zimmerman (1971) would be about 6%. As the alpha dose contributes typically only about 40% of the total radiation dose to the fine grains, this implied that there would be a decrease of only about 2.5% in the overall effective dose. It has become quite usual for users of the fine-grain dating method to regard this attenuation factor as negligible when compared to other possible sources of error in the method and hence not to correct for it in the TL age determination (Aitken, 1978). The purpose of this paper is to present the results of the calculation of the alpha dose attenuation in quartz grains in the size range 1 μm - 1 mm using more sophisticated computer techniques than were available to Fleming (1969) and thus give a more exact and detailed description of the attenuation in quartz over a wider grain size range.

## Method of Calculation

The calculation of the alpha dose attenuation was based on the mathematical formalism developed by Charlton and Cormack (1962), which was also the basis for Fleming's (1969) calculations. The calculations essentially involve the development of a geometrical factor, $S_\alpha(x)$, which describes the dose at a point within a non-radioactive sphere, at a distance x from the interface with a surrounding radioactive matrix. The theory makes two simplifying assumptions:

a) the ionising particles are emitted isotropically within the surrounding medium, and

b) the emitted particles travel in straight lines.

Both of these assumptions are valid for the alpha particles emitted by an homogeneous distribution of the uranium and thorium series in a clay matrix. It is further assumed that the quartz grains constitute non-radioactive spheres within the radioactive clay matrix. The actual mathematical expressions for $S_\alpha(x)$ are given by Charlton and Cormack (1962), but Howarth (1965) has reduced these to a more convenient form for numerical evaluation. The values of $S_\alpha(x)$ required here were taken from Howarth's tables for a spherical interface.

In order to assess the mean absorbed dose in a quartz grain, the values of $S_\alpha(x)$ were integrated numerically over the grain volume for diameters from 1 μm up to 1 mm. This integration was performed by a computer programme for numerical integration on a Univac 1100 series computer and the results are shown graphically in Figure 1 for the uranium and thorium series. $\overline{S}_\alpha$ is the mean absorbed dose in a quartz grain of given diameter expressed as a fraction of the dose which would be absorbed by an infinitesimally small grain. The numerical values for $\overline{S}_\alpha$ for each grain diameter are given in Table 1.
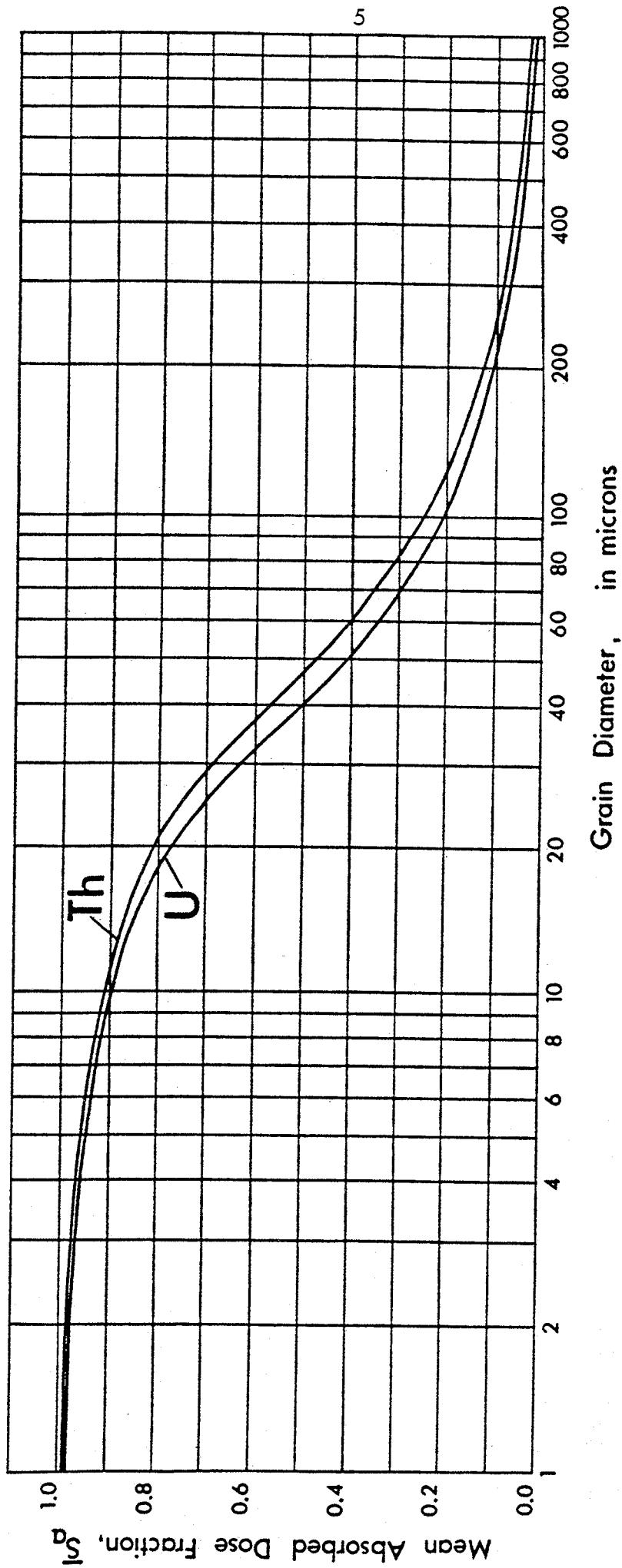
FIGURE 1. The Mean Absorbed Dose Fraction, $\overline{S}_\alpha$, for Alpha Particles from the Uranium (U) and Thorium (Th) Series in Quartz Grains Plotted as a Logarithmic Function of the Grain Diameter from 1 μm up to 1 mm.

## Table 1

Mean Absorbed Dose Fractions, $\overline{S}_\alpha$, in Quartz Grains of Various Diameters for the Uranium and Thorium Radioactive Decay Series.

| Grain Diameter (μm) | Uranium series | Thorium series |
|---|---|---|
| 1.0 | 0.990 | 0.991 |
| 2.0 | 0.979 | 0.982 |
| 3.0 | 0.969 | 0.973 |
| 4.0 | 0.958 | 0.964 |
| 5.0 | 0.947 | 0.955 |
| 6.0 | 0.936 | 0.945 |
| 7.0 | 0.925 | 0.936 |
| 8.0 | 0.914 | 0.926 |
| 9.0 | 0.903 | 0.917 |
| 10 | 0.892 | 0.907 |
| 15 | 0.834 | 0.859 |
| 20 | 0.772 | 0.807 |
| 30 | 0.628 | 0.690 |
| 40 | 0.502 | 0.567 |
| 50 | 0.414 | 0.471 |
| 60 | 0.350 | 0.402 |
| 70 | 0.303 | 0.349 |
| 80 | 0.266 | 0.308 |
| 90 | 0.238 | 0.275 |
| 100 | 0.215 | 0.248 |
| 150 | 0.144 | 0.167 |
| 200 | 0.108 | 0.126 |
| 250 | 0.087 | 0.101 |
| 300 | 0.073 | 0.084 |
| 400 | 0.054 | 0.063 |
| 500 | 0.043 | 0.050 |
| 1000 | 0.022 | 0.025 |

## Use of the Attenuation Factors

It must be stressed that the data given in Table 1 apply only to the case of non-radioactive quartz grains embedded in a clay matrix having an homogeneous distribution of the uranium and thorium series. Singhvi and Zimmerman (1979) have shown, however, that in many fine-grain samples feldspars can be the dominant TL minerals. Therefore the mean absorbed dose fractions for 1-8 μm grains of potassium feldspar were calculated and compared to those for quartz in the same size range. They were found to be less than 1% different from each other. The same calculations were performed for 100 μm grains but here the difference between the quartz and feldspar factors was found to be about 7%, due to the fact that the alpha particles have a greater range in the feldspar which in turn results in a smaller attenuation of the dose. Hence, the data given in Table 1 can safely be applied to most fine-grain samples, but for larger grain sizes the data should only be used for quartz.

There will be certain samples, however, for which the radioactivity content of the TL grains themselves, feldspar or quartz, will not be negligible. In these cases the above data will still be valid for the matrix alpha dose to the grains but account will also have to be taken of the small internal dose from any radioactivity within the grains.

It is worth noting here that there are certain other factors, in addition to the grain size attenuation, which must be included in the determination of the alpha dose to a particular grain size range. The most important of these is the alpha efficiency factor which describes the relatively low efficiency of alpha particles at producing TL compared to that of beta and gamma radiation. This phenomenon has been described in detail elsewhere by Zimmerman (1971, 1972) and by Aitken and Bowman (1975), but it should be remembered that for larger grains at least, the alpha efficiency may vary radially within the grain due to the diffusion of impurities into the outer layers as mentioned by Bell and Zimmerman (1978). Other factors which must also be taken into account in the alpha dose determination are the effect of ground water on the dose-rate (Zimmerman, 1971) and the possible escape of radon from the sample (Zimmerman, 1971; Desai and Aitken, 1974).

Although the data of Table 1 may be used for all grain sizes in the range 1 μm - 1 mm, the two principal TL dating methods employed today utilise grains either in the fine-grain range of 1-8 μm or quartz inclusions of approximately 100 μm diameter. The attenuation of the alpha dose in 100 μm quartz inclusions has already been specifically dealt with by Bell (1979), so let us look here in a little more detail at the fine-grain size range.

Zimmerman (1971) has shown that the separation techniques used in the fine-grain dating method give grains in the size range 1-8 μm and, assuming equal numbers of all sizes in the initial distribution, this results in an average grain diameter of 5 μm. From Table 1 this predicts a mean absorbed dose fraction for fine grains of 0.947 for the uranium series and 0.955 for the thorium series. For typical concentrations of the two series this implies an average attenuation of the alpha dose in fine grains of 5%, in good agreement with the figure of 6% given earlier by Aitken and Fleming (1972). As mentioned above, because of the low efficiency of alpha particles at inducing TL (efficiency factors are of the order of 0.1), the alpha dose contributes typically only about 40% of the total radiation dose, with the remainder coming from the beta, gamma and cosmic ray doses. Hence a 5% attenuation in the alpha dose will normally result in only a 2% decrease in the total dose and thus only a 2% increase in the TL age.

Nevertheless, now that the attenuation factor is accurately known for the fine-grain method and unless any other complications (such as significant internal radioactivity within the grains) are suspected, there appears to be no reason to neglect the correction even though the error in doing so would probably be much smaller than other possible errors in the dating method.

There can, however, be certain circumstances (such as an exceptionally high alpha efficiency factor and/or a very low potassium contribution) which would imply that the alpha dose would contribute greater than 40% of the total dose and hence the alpha dose attenuation would be more significant. This can be illustrated to some degree by the TL dating of a deep-sea sediment core as described by Wintle and Huntley (1979). These authors used fine grains in the size range 4-11 μm and found the grains to consist mostly of clay minerals. Using the data presented here for quartz gives an alpha dose attenuation of 10% for this size range. The potassium content of the core is low and, for the younger samples at least, the radiation dose comes predominantly from precipitated thorium-230. Hence for this particular dating programme, the alpha

contribution to the overall radiation dose is greater than 50% and neglect of the alpha dose attenuation factors will result in TL ages which are more than 5% too young, assuming negligible internal radioactivity within the grains.

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# PREDOSE ANALYSIS OF BRICK SAMPLES:  FEASIBILITY FOR FALLOUT DOSIMETRY

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A number of communities in the regions downwind from the Nevada Test Site received fallout radiation from atomic tests in the 1950's and 1960's, and adverse health effects have recently been suggested (Lyon et. al., 1979). A major uncertainty in evaluating potential health effects is that direct measurements of cumulative exposures to the communities in question have not been made.  Instead, estimates of the cumulative exposures have been calculated based on exposure rate readings taken with field survey instruments shortly following the individual blasts.  The estimates are obtained by assuming that the exposure rate at any time following an atomic detonation may be expressed by the relationship (Way and Wigner, 1948);

$$R_t = R_0 \; t^{-1.2}$$

Where '$R_t$' is the exposure rate at time '$t$', and $R_0$ is the exposure rate at unit time following the blast.  Integrating from the time of deposition of fallout to infinite time yields an estimate of so called "infinite exposure" for the detonation in question.  Summing the estimates for each test which contributed fallout to a given location yields an estimate of "cumulative infinite exposure" for that location (Nagler and Telegadas, 1956).  These calculations indicate that cumulative external gamma-ray exposures of from 1 to 17 roentgens may have been delivered to inhabited regions within 250 miles of the test site (Shleien, 1980).

The relationship above appears to hold for controlled situations where the fallout remains undisturbed on plane surfaces, but the effects of vegetation, uneven terrain, and dispersal or accumulation by wind and water are largely unknown.  Attempts have been made to correct for some of these factors (Dunning, 1959.  Shleien, 1980), but the variety of geographical conditions (from open prairie to well developed community) is considerable and the accuracy of such corrections is not well established.

The predose TL technique (Fleming, 1973) offers the potential for directly measuring the cumulative doses in many locations.  The basic idea is to use TL to determine the dose received by quartz grains in housing bricks of known age.  The equivalent dose from the TL of the quartz is compared with the natural dose calculated from the age of the brick and the natural radioisotope concentrations within the brick.  With sufficient numbers of samples from a given community, average excesses of the measured doses over the calculated doses may be attributed to fallout radiation in the past.

The value of these measurements lies in (1) the verification of estimated cumulative exposures, (2) the examination of intra-community variations in fallout exposures, and (3) the extension of the technique to those communities where exposure measurements with survey instruments were not consistently taken.

The important first question to be answered is, can the expected small equivalent dose of around 10 rads be measured with sufficient accuracy using the predose method? This paper reports results of the analysis of a single brick taken from a 25 year old building at the University of Utah, Salt Lake City. This analysis was undertaken to provide a rough indication of the suitability of the predose technique for determining the expected low doses of radiation. The feasibility of the technique has been suggested by the report of Fleming and Aitken, (1974) which indicated that doses of 10 to 20 rads could be readily detected in bricks from a house in North Berkshire, England. The analyses reported here were performed at Washington University, St. Louis, Missouri.

## Material Investigated

A portion of brick was removed from a height of 7 feet from the exterior west wall of the Radiobiology kennel at the Medical Center of the University of Utah, Salt Lake City. The wall was constructed in 1955 from brick supplied by the Interstate Brick Company of Salt Lake City, the major supplier of brick throughout the state. The exact date of kilning is expected to be within one year of the construction date.

## Sample Preparation

Quartz crystals were obtained by repeatedly vising a small brick portion (34 grams), selecting grain sizes of 74 to 250 microns by sonic sifting, and eliminating ferrous materials with a Frantz magnetic separator. 1.5 g of non-magnetic grains were obtained. 15 mg of unetched crystals were removed for TL analysis while a second portion (0.5g) was etched at room temperature for 50 minutes in 49% HF acid, rinsed in water, soaked for 15 minutes at 50°C in a solution of 20% $AlCl_3$ (Carriveau, 1977) to remove fluoride precipitates, rinsed and finally sieved to remove grains smaller than 74 microns.

## Equivalent Dose Measurements

Glow curves were made on three (3) aliquots of the quartz powder; 1 unetched (7mg) and 2 etched (15mg each). An EMI 9635QB PM tube was used to detect the TL. Laboratory irradiations were made using $^{90}Sr$ beta sources which were calibrated using the Oxford calibration kit (Aitken, 1978). An activation temperature of 550°C was used. The equivalent doses for aliquots 1 and 2 were determined from single added laboratory doses of 26.5 and 17.7 rads, respectively. The test doses of 0.9 rad were significant additions fo the laboratory dose and were included in the equivalent dose calculation. Aliquot 3 was sensitized with three doses of 17.7 rads giving 4 points to which a least squares linear fit could be made (Fig. 1). The equivalent dose for this aliquot was 9.6 ±1.8 rad, with 9.9 rad for the other etched aliquot and 8.7 rad for the unetched.

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FIGURE 1

## Sensitivity Growth Curve



### Growth Curve Data
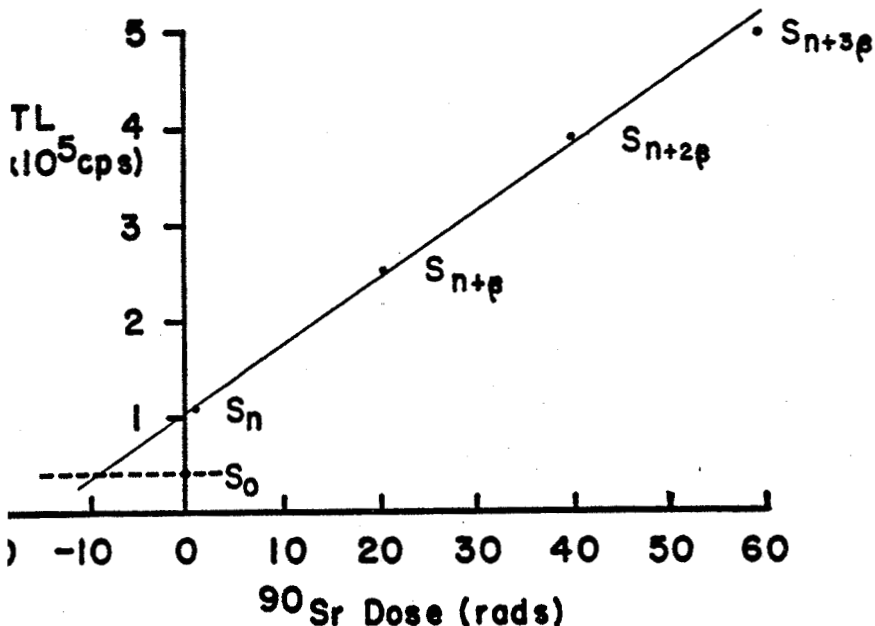
| | Activated $^{90}Sr$ Dose (rads) | TL $(10^5 cps)$ | TL' $(10^5 cps)$ |
|---|---|---|---|
| $S_o$ | 0 | 0.39 | 0.39 |
| $S_n$ | 0.9 | 1.08 | 1.08 |
| $S'_n$ | 0.9 | 1.02 | |
| $S_{n+\beta}$ | 20.5 | 2.43 | 2.49 |
| $S'_{n+\beta}$ | 20.5 | 2.34 | |
| $S_{n+2\beta}$ | 40.1 | 3.72 | 3.87 |
| $S'_{n+2\beta}$ | 40.1 | 3.54 | |
| $S_{n+3\beta}$ | 59.7 | 4.65 | 4.98 |

TL' - corrected for radiation quenching
Test dose = 0.9 rads
$\beta$ dose = 17.7 rads

## Estimation of Dose from Natural Radiation

The expected natural dose was calculated from the approximate age of the brick (25 years), the potassium content of the material (1.0% $K_2O$, determined by atomic absorption spectroscopy), and uranium and thorium contents (2.7 ppm U and 11.0 ppm Th, determined by alpha counting and assuming equal activities of the uranium and thorium series). Table 1 lists the beta and gamma dose-rates calculated using the data of Bell (1979) and the beta attenuation factors of Mejdahl (1979). Several assumptions have been made. First, the brick is assumed to have been dry. Second, radon emanation is assumed to be negligible (gamma-ray spectrometry measurements on a second sample removed from the wall indicated negligible radon loss). Third, because of the unusual geometry and the unknown contribution from the soil, 3 $\pi$ gamma geometry was arbitrarily assumed. Multiplying the estimated annual dose (1/4 rad) by the approximate age of the brick (25 years) gives an expected natural dose of approximately 6 1/4 rads.

DOSE RATES
(mrad/year)

| | BETA | GAMMA | SUBTOTAL |
|---|---|---|---|
| URANIUM (2.7ppm) | 35 | 25 | 60 |
| THORIUM (11.0ppm) | 25 | 45 | 70 |
| POTASSIUM (1.0% $K_2O$) | 65 | 15 | 80 |
| COSMIC RAYS | | | 40 |
| TOTAL | | | 250 |

TABLE 1

## Discussion

The discrepancy between the estimated equivalent dose of approximately 9 1/2 rad and the estimated background dose of 6 1/4 rad could be due to a number of factors unrelated to fallout. For instance, Bell and Mejdahl (1980) have shown that beta source calibration may vary with changes in transparency of quartz crystals due to etching, Fleming (1973) has warned of disruption of the predose sensitivity also due to etching, and changes in $S_O$ over time are occasionally encountered (Ian Bailiff, personal communication). These potentially confounding effects will be explored in future pilot studies as will the errors associated with methods of background dose-rate determination. We are, nevertheless, encouraged in this pilot study by the high signal to noise ratios of the glow curves, the linear growth of sensitivity with increasing dose and the grouping of the equivalent doses of the aliquots analysed. Preliminary analyses (one aliquot per sample) on an additional 9 cores from Utah bricks of various age show similarities in sensitivity and linearity (the mean error on the least squares fit for members of this group was 1.0rad). These preliminary results suggest that the predose technique may provide the sensitivity required to extract estimates of fallout doses on the order of from 5 to 20rads.

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# LETTER TO THE EDITOR

Recently, W.T. Bell (1980 a,b) stated in Ancient TL that the use of
perspex absorbers for gamma irradiations of quartz - as done by Pernicka
and Wagner (1979) - introduces an error of about 5% in the gamma dose cal-
culation to quartz. In principle, we agree with an uncertainty due to
different secondary electron fluences in perspex and quartz. But taking
into account additional error sources such as Compton scattering in the
absorber, gamma attenuation and electron scattering at the quartz/perspex
interface we estimate a smaller uncertainty of less than 2%. In view of the
experimental precision of TL measurement during calibration this uncertain-
ty is barely significant. A more detailed presentation will be published
in the Proceedings of the Second Seminar for TL Specialists (Pernicka and
Wagner, 1980).

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Part 1 and 2: Heat effects, David, M., Sunta, C. M., and Ganguly, A. K., Vol. 15 (1977) pp. 201 and 277.
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(Requests for copies of this article may be sent to S. R. Sutton, Washington University, St. Louis, Missouri, 63130.)

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