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A model for mid-term fading in TL dating

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Experimental evidence indicates that identical radiation doses acquired by archaeological materials during burial give smaller induced luminescent signals than those resulting from laboratory calibration dosing, thus leading to an underestimation of archaeological ages. A theoretical model is proposed that accounts for this behaviour based on the thermal release of electrons trapped at centres possessing a distribution of activation energies. The predicted degree of age underestimation is found to be sensitive to the ambient burial temperature. In the presence of several independent recombination centres this underestimation will depend on the centre selected by the employment of appropriate colour filters for detection.

Introduction

Suitable archaeological materials may often be used as dosemeters to reveal the palaeodose of radiation received during the specimen's burial. This information is generally obtained by the use of thermoluminescence (TL) techniques. It is essential in such determinations to use TL signals that are produced by stable charge trapping states capable of giving a response based on the total absorbed dose. However, when dating archaeological samples with ages in the range of many thousands of years, it has been found (Wintle 1973) in some cases that samples that are expected to be stable over this time range apparently lose part of their TL signal as anomalous fading. This means that the TL dependence on dose given in the laboratory is not the same as that imparted during burial. Clark and Templer (1988) explained this as being due to thermally assisted tunnelling, an effect previously discovered by Visocekas (1979) for calcite. In a series of papers, Mejdahl (1988a, 1988b; 1989) pointed out that when the ages to be determined are in the 10^5-10^6 year range, the loss of TL that occurs may very well be due to a long-term thermal decay rather than the above mentioned anomalous fading. Thus, in a study of Tertiary sands, he found TL ages in the region of 300-500 ka rather than 700-2000 ka predicted from the stratigraphy. Recent measurements with feldspars have also indicated a systematic underestimation of TL ages when compared to expected geological ages, however, the extent of the underestimation appeared to be dependent on the wave lengths selected by the optical filter used to observe the luminescence (Balescu, et al. 1991 and 1992).

In a recent paper, Xie and Aitken (1991) describe a model which explains this 'mid-term' thermal fading during burial periods the order of 10^5-10^6 years. They assume that the dating signal consists of a stable component and an unstable one, and that while the lifetime of the stable component is long compared to the oldest sample to be dated, that of the other is 'mid-term', namely, long compared to laboratory times but short compared to the ancient ages. They assumed that the unstable component has the same growth characteristics as the stable one, both exhibiting saturating exponential functions of the dose, and that the fraction of the signal which is not stable is constant for all samples. Using these assumptions they showed that for relatively old samples, ages may be appreciably under-estimated. In an example a sample known to be 730 ka old appeared to be only 127 ka old based on TL evidence, presumably as a result of the mid-term fading effect.

Model

Xie and Aitken do not specify the nature of the fast and slow fading components. A plausible explanation is that a distribution of trapping states exists in which the deeper bound states are more stable than the shallower bound ones. The idea of a distribution of activation energies of trapping levels is as old as the theory of TL itself as first suggested by Randall and Wilkins (1945) who also gave evidence that such distributions exist in calcites and dolomites. Kikuchi (1958) showed that distributions of trapping level energies play an important role in the TL of glasses. Medlin (1961) suggests that discrete trapping levels may be broadened into a continuous band of levels as the result of local distortions in the crystal fields due to dislocations, vacancies and impurities. Since both the local distortions and the trapping centres may be randomly distributed throughout the crystal, a logical form for the distribution of activation energies would be a Gaussian, namely, in obvious notation,

$$\rho(E) = N\sqrt{a/\pi}\exp[-a(E-E_o)^2]$$

The actual filling of these trapping states may or may not result in Gaussian charge distribution, depending on the circumstances. The effect of such a distribution on TL characteristics has been studied in detail (Hornyak and Franklin 1988).

Very likely, a distribution of closely spaced discrete or sharp trapping states having an occupation capacity following a normal distribution about some preferred activation energy would produce a very similar situation. In this connection it should be noted that a thermal or time sweep across an activation energy distribution possesses a substantial 'window' width thus making such detailed model distinctions difficult.

Hornyak et al. (1992) have recently shown that the TL characteristics of the 375 °C glow peak in quartz can be successfully explained using a model with such a Gaussian distribution for trapping states and two recombination centres yielding green and uv emissions. In the present study, the difference between dosing a sample in the laboratory times and in burial times is investigated when such a Gaussian distribution for traps is present. The differential equations governing the filling of traps and centres during long burial times are numerically solved and the results are compared to those found when the same dose is imparted during laboratory times (say of the order of minutes). The distribution of trap activation energies given in equation (1) is simulated with a 96 segment histogram following the procedure used in the above cited work. To avoid convergence problems and to achieve reasonably short computer running times the earlier VAX program had to be altered from a continuous dosing rate to that of doses administered in steps. For dosing during archaeological burial the probability of thermal release per unit time for a given trapped electron given by:

$$\alpha = se^{-E/kT}$$

is used while for laboratory dosing is set equal to zero. The results for this latter calculation were confirmed by using the standard computer program with a total dose achieved in a 10 second period and an appropriately much higher continuous dose rate.

In order to present the relevant behaviour in its simplest form a hypothetical model for charge trafficking during the dosing and subsequent glow periods is adopted that consists of a single type of active electron trap and a single type of recombination centre. The physical parameters employed are taken from the above cited study using only the so called green member of the pair of recombination centres. These parameters are: E = 1.450 eV for the central activation energy, $s=5.1\cdot10^{11}$/s for the frequency factor, a ratio of retrapping to radiative recombination cross sections of 2.7, and $N=10^6$/mg for the total available trap states (this corresponds to $N=10^{14}/cm^3$ when corrected for counting efficiency). An incomplete sun bleach is assumed at the beginning of the palaeodose period with $n_o/N=0.135$ for the relative trapped electron population density. The presence of a deep saturated thermally disconnected electron trap with a relative population density of p/N = 1.197 is also assumed.

The computer program assumes doses administered in steps of 10^7 s duration, delivering 1.54×10^{-4} Gy and generating the appearance of 0.96 electron-hole pairs in the conduction and valence bands. These values are again based on the above cited study; note that the actual e/h pairs created when corrected for counting efficiency yield $10/cm^3$ per s). The calculation for a given total dose results in the number and distribution of trapped electrons remaining at the end of the dosing. This is then the starting point for calculation of TL glow curves or isothermal decay curves.

To generate a point in a plot of true age vs. apparent or laboratory age the dosing calculation is done twice, each time resulting in the same number of remaining trapped electrons and therefore (in the absence of thermal quenching) the same area under the glow curve assuming that this signal is used in the dating procedure. One calculation is done with $\alpha = 0$, representing laboratory dosing administered so rapidly that no thermal loss of trapped electrons occurs, and the time to reach the required number of trapped electrons is the apparent age. The other calculation is done with given by eqn (2) with T appropriate for the burial condition, thus turning on the thermal decay and allowing for the loss of trapped electrons. The time required to reach the same number of trapped electrons in spite of this loss is then the true age to associate with the laboratory dosing signal or the resulting apparent age.

Results

The quantity Δ (= true age - apparent or laboratory age) is found to be very strongly dependent on the burial temperature in addition to the obvious dependence on the width parameter a = 2.77 $(FWHM)^{-2}$ appearing in Eq (1). Figures 1 and 2 show these results for as a function of apparent age for some typical cases of burial temperature and FWHM values.

Figure 3 displays these same results showing the TL signal (area under the glow curve) in units of n/N as a function of total dose both under burial and laboratory dosing conditions. As an example the indicated TL signal corresponding to n/N = 0.732 would suggest a sample age of 300 ka based on laboratory dosing whereas the true burial age would be 90 ka older. Contrary to the model employed by Xie and Aitken the ratio of the TL signal generated by a laboratory dose to that developed under burial conditions by the same dose is not a constant, rather the ratio increases sharply with increasing dose. For the case illustrated, materials less than 10 ka in age would involve negligible values of Δ.

Figure 4 shows the distributions of trapped electrons for the same value of n/N selected as the illustration in figure 3. The electron density is per mg·eV (uncorrected for detection efficiency). Under laboratory dosing the distribution has the same Gaussian shape as that for the trap activation energies. Under burial conditions for the same value of n/N the low energy electrons are severely reduced while the higher energy electrons are correspondingly increased as a consequence of retrapping. The areas under the two curves are equal. Note that the acquired dose is not the same in these two cases. For the same dose the TL signal for burial dosing would give n/N = 0.661, the loss of Δ(n/N) = 0.071 representing the thermal decay during burial.

Figure 5 shows the glow curves corresponding to the value of n/N previously selected as an illustration in figure 3 and displayed as electron distributions in figure 4. Again, as required, the area under the glow curves are equal. Although the peak position and FWHM of the two curves are almost the same, the effect of the reduced

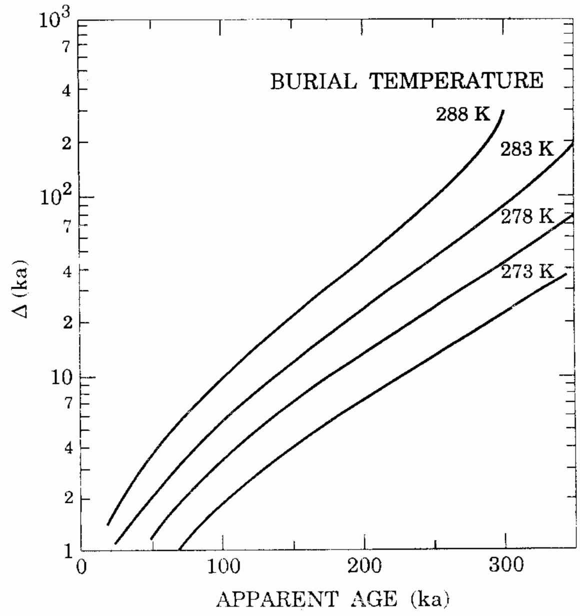

Figure 1.
The quantity Δ (= true age - apparent age) as a function of the burial temperature. The FWHM of the activation energy distribution is 0.186 eV and the central energy E_o = 1.450 eV.

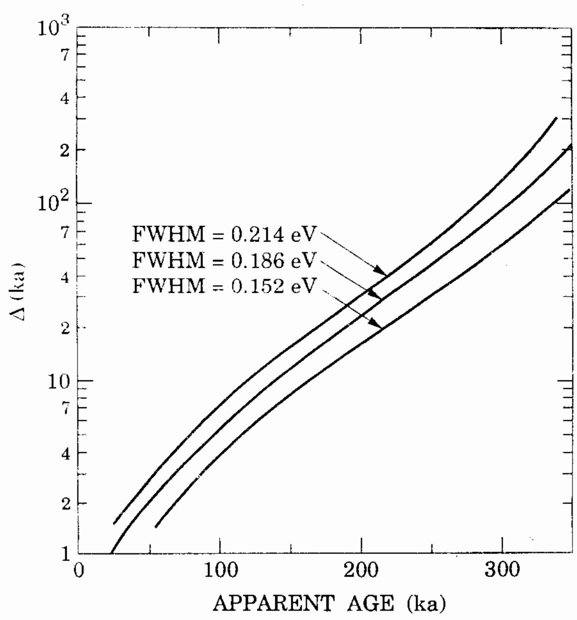

Figure 2.
The quantity Δ (= true age - apparent age) as a function of the width (FWHM) of the activation energy distribution. In all cases the central energy of the distribution E_o = 1.450 eV. The burial temperature is taken to be 283 K.

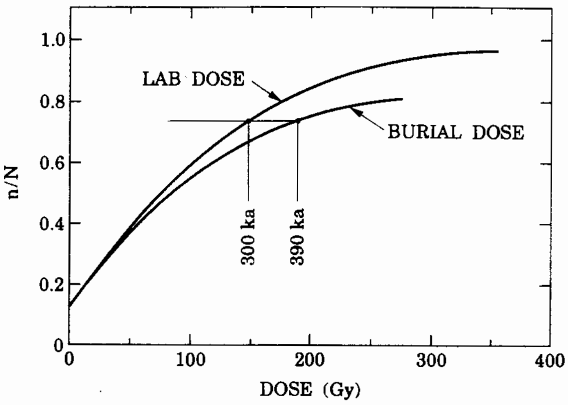

Figure 3.
The relative trapped electron population density, n/N as a function of applied dose for both burial and laboratory dosing situations. The burial temperature is 283 K and the FWHM = 0.186 eV with E_o = 1.450 eV. For the selected value n/N = 0.732, the corresponding TL signal (area under glow curve) would indicate an apparent age of 300 ka while the true burial age would be 390 ka for the same TL signal.

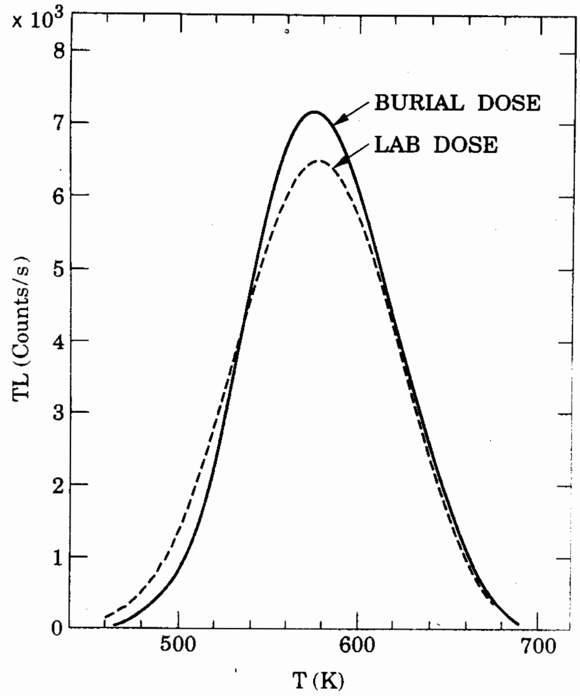

Figure 5.
The two glow curves corresponding to the electron distributions shown in fig. 4.

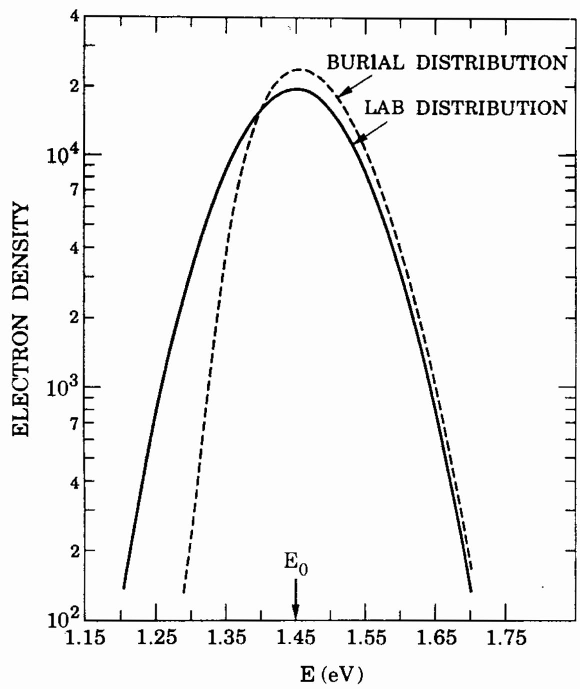

Figure 4.
The distribution of trapped electrons for the same value of n/N or TL signal selected as an illustration in fig. 3 contrasting the laboratory and burial dosing. The units for electron density refer to a total number of detectable electrons = 7.32 x 10^5/mg.

low energy electron population is clearly evident in the steeper initial rise for the burial situation. A standard plateau test comparing the two glow curves should also reveal the shift of the electron population to higher temperatures for the buried sample.

All the above described calculations were also repeated for the two recombination centre model for quartz reported in the earlier investigation (Hornyak 1992). This study demonstrated quite conclusively that the 375 °C peak in quartz involved a distribution of activation energies for the electron traps involved. Results very much like those shown in figures 1-5 followed. One novel feature worth reporting is the substantially larger mid-term effect noted for the uv emission compared to that in the green. This behaviour is attributed to the fact that the glow peak observed in the uv appears at a lower temperature (about 20 °C) than the glow peak observed in the green and hence is more sensitive to the loss of electrons with low activation energies under burial conditions.

Conclusions

The existence of a distribution of activation energies for an electron trap site may result in the appearance of a mid-term thermal fading phenomenon. It may readily happen that supposedly long-lived states will possess a significant number of low activation energy components with intermediate or mid-term life times, long compared to laboratory dosing times but short compared to burial ages. In that event thermal losses that occur during the palaeodose period will not be evidenced in laboratory dosing experiments. Thus, the same absorbed dose will yield a smaller TL signal if acquired during archaeological burial than if administered under laboratory conditions. The usual techniques for age determination will yield palaeodose values that result in underestimating the archaeological age of these sample. To be precise even the various values of TL observed for different added laboratory doses will arise from electron distributions of mixed origins, those combining the original 'natural' population with the added laboratory population in some nonlinear superposition retrapping process.

A complicating phenomenon not considered in this investigation involves the possibility of defect migration. This behaviour can be simulated by allowing thermally driven transitions between the various portions of the activation energy distribution of N itself (Piters and Box ,1991). While this effect by itself would keep the glow area a constant, in connection with the presently described effect it might, if present, alter the escape rate of trapped charge under long burial conditions.

It would appear to be advisable to obtain glow curve data at various ramp heating rates and isothermal decay data on the natural sample to establish the activation energy distribution width and central energy. Such information would assist in sorting out the expected correction to the age underestimation. It would be ideal to use a glow peak occurring at a high enough temperature, of sufficiently narrow temperature width, and behaving with a more or less first order character in order to reduce the need for any important mid-term correction.

However, in general it is rather unfortunate that the amount of palaeodose underestimation due to mid-term thermal fading is very sensitive to the ambient burial temperature, particularly for samples of very old age. This effect will require the archaeologist to not only determine the present temperature of the burial site but to also involve some geological estimation of the temperature history of the site, a more daunting process.

A very favourable result of the present investigation is that for specimens not older than about 10,000 years no significant mid-term correction is called for if dating were based on using this glow peak. After ages over 20,000 years the ratio of observed TL under laboratory dosing to that under burial dosing rapidly increases with increasing dose. Underestimation of archaeological age will certainly occur.

Finally, the results obtained with the two recombination centre model suggests that when TL emission occurs at several wave lengths it may be advisable to examine each emission band separately for its dosing response. One may very well result in a smaller age underestimation, as apparently is the case for feldspar reported by Balescu and Lamothe. These investigators also report a much larger underestimation of age when observing the TL at shorter wavelengths.

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PR Reviewer's Comments (S.W.S. McKeever)

This is a very interesting paper and presents a feasible model for 'mid-term' fading. I tthink it should be stressed that a distribution of states is probably not necessary for the model to work; closely overlapping discrete states should produce the same effect. One may wonder what a plateau test looks like for the case described.