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The validity of the laboratory reconstruction of palaeodose

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Introduction

The problem of laboratory reconstruction of palaeodose is common to all the palaeodosimetry dating methods (TL, OSL, ESR). It is known that the quartz TL dose response curve, obtained using β and γ laboratory sources, can be represented by a multi-stage process, as shown in figure 1:

> Stage I may be explained by competition between traps with different effective cross sections which are empty at the initial moment of irradiation;
> Stage II is characterised by the filling of existing defects (traps) by charge carriers;
> Stage III corresponds to saturation, where all the existing traps are filled;
> Stage IV represents the generation and filling of new defects.

The fourth stage is simultaneous with the second one, but due to its lower efficiency it becomes apparent only after the stage of saturation. The accumulated dose at which the fourth stage is observed has not been fixed since it depends on the number of traps in the palaeodosemeter, i.e. on the second and third stages.

However, in nature the dose rate is several orders of magnitude lower than that used in the laboratory and the validity of the assumption that the growth characteristics are similar for the two situations has been considered previously (eg Aitken, 1985, p.1412). The essence of the problem is that in case of low natural dose rates saturation may be reached, not as a result of a shortage of traps, but due to thermodynamic equilibrium between the input and output of trapped charge carriers. In this case the values of laboratory and natural saturation levels may be different if the lifetime of charge carriers in traps is not sufficiently long. The implication of such a difference is, in principle, that the saturation value (the second stage) will depend on dose rate.

Experiments with CaF_2 (Aitken, 1985, p141) have shown that saturation TL values do not change if the laboratory dose varies from 0.3 mGy/min to 3 Gy/min. However, changes may have taken place at considerably lower dose rates where it is impossible to model experimentally a situation close to the natural one. Consequently the level of dose at which the dependence on dose-rate for traps with trapped charge mean lifetimes comparable with dating timescales becomes significant may be within the regime of accrued palaeodose for dating samples.

The first results indicating a possible difference between natural palaeodose formation and its laboratory reconstruction were obtained from the TL dating of loess sections. In spite of the absence of saturation under laboratory conditions, no ages greater than 100 ka were obtained (Debenham, 1985). In previous work (by G.H.) on loess of ages ranging from the contemporary to 1.5 Ma, it had been found that TL ages greater than 30 ka were systematically younger than reliable geological estimates. These results suggest that the apparent limits of the TL method may vary according to geographic location and that these limits are considerably lower than those predicted by the laboratory dose response results. The accuracy of the dates obtained remains topical as well.

Shljukov and Shakovets (1986), after attempting to reconstruct palaeodose by means of X-ray sources of radiation with a dose rate 30 Gy/min. concluded that laboratory calibration was not a valid procedure. They also assumed the natural saturation value for quartz to be constant for all geological regions.

Model for palaeodose formation and age correction

Vagn Mejdahl (1988) discovered a difference between natural and laboratory saturation for alkali feldspars from one of the regions and took it to be a common phenomenon related to long term fading. He suggested the correction of dates in view of the discrepancy between the values of natural and laboratory saturation.

In this paper a further method for the correction of dates is presented, based on a consideration of differences between natural and laboratory saturation values. We leave aside the agents accounting for this discrepancy: in complex many-impurity system losses may take place during such a long term period due to non-radiative transmission, redistribution of charge carriers, diffusion etc. In the case of fine grains (4-11 µm), competition between surface and volume defects (Kortov, 1986) may partly account for this effect.

The proposed model permits the evaluation of the critical dose rate, i.e. the minimal dose rate at which the above mentioned dependence is negligible, but also the effective lifetime of change carriers in the traps. The correction formula is based, not on formal rules, but on actual processes of palaeodose formation which enables experimental parameters to be used. Only stages II and III of the dose response curve (figure 1) are considered.

Palaeodose formation is a monomolecular process, and, unlike the optical bleaching process, the elucidation of its order of kinetics requires special experiments. The main kinetic formula of palaeodose formation is as follows:

$$\frac{dn}{dt} = \beta(N - n) - \alpha n \qquad (1)$$

where,

$\frac{dn}{dt}$ is the number of electrons stored per unit time
N - the total number of traps of the type considered
n - number of traps occupied at time t
α - probability for carriers to escape from traps
β - probability of capture for electrons from the conduction band

$$\frac{1}{\tau} = \alpha = P_o\, e^{-E/kT} \qquad (2)$$

where,

τ - is the life time of carriers in the trap
E - trap thermal activation energy
P_o - frequency factor
k - Boltzmann's coefficient
T - storage temperature

Solving the differential equation (1),

$$dt = \frac{\alpha n}{\beta N - n(\alpha+\beta)} \qquad (3)$$

$$\frac{-1}{\alpha+\beta} \ln\,[\beta N - n(\alpha+\beta)] = t + \ln C \qquad (4)$$

$$\ln C = \frac{-1}{\alpha+\beta} \ln \beta N \qquad (5)$$

$$t = \frac{-1}{\alpha+\beta} \ln \frac{\beta N}{\beta N - (\alpha+\beta) n} \qquad (6)$$

For saturation, $n \rightarrow n_\infty$, $\frac{dn}{dt} = 0$

Equation (1) yields

$$\beta N - n_\infty(\alpha + \beta) = 0 \qquad (7)$$

$$\alpha + \beta = \frac{\beta N}{n_\infty} \qquad (8)$$

Substituting (8) in (6),

$$t = \frac{n_\infty}{\beta N} \ln \frac{\beta N}{\beta N - \frac{\beta N}{n_\infty} n} \qquad (9)$$

$$t = \frac{n_\infty}{\beta N} \ln \frac{1}{1 - \frac{n}{n_\infty}} \qquad (10)$$

Introducing βN, the probability of capture with unoccupied traps, and γ, the dose sensitivity at the linear stage of the dose response curve, the following formula may be defined,

$$\beta N = \gamma P \qquad (11)$$

where P is the dose rate, $\frac{dD}{dt}$.

From equations (11) and (10),

$$t = \frac{n_\infty}{\gamma\ P} \ln \frac{1}{1 - \frac{n}{n_\infty}} \qquad (12)$$

Initially ($N \gg n_0$) there are many more unoccupied traps than charge carriers and the change in the number of charge carriers in the traps is determined by the dose sensitivity.
Putting γ, the change in the number of trapped charge carriers per unit laboratory dose, into (12),

$$t = \frac{n_\infty dD}{dn_o P} \ln \frac{1}{1 - \frac{n}{n_\infty}} \qquad (13)$$

n_∞ may be realised both at the natural (n_{∞_N}) and the laboratory dose rate (n_{∞_L}).
Assuming,

t_1 - age corresponding to the palaeodose induced by the natural radiation
t_2 - age corresponding to the palaeodose reconstructed by laboratory irradiation

we get from (10),

$$t_1 = t_2 \frac{n_{\infty_N}}{n_{\infty_L}} \left(\ln \frac{1}{1 - \frac{n}{n_{\infty_N}}} \right) \cdot \left(\ln \frac{1}{1 - \frac{n}{n_{\infty_L}}} \right)^{-1} \qquad (14)$$

For correction, the natural saturation level, n_{∞_N}, must be known i.e. using samples with ages beyond the limits of the method.

Dependence of saturation value on dose rate
As was shown above, the model enables the dependence of saturation value on dose rate to be established for traps with different trapped charge carrier life times. Using formula (8) it is possible to determine the number of trapped charge carriers in the saturation stage.

$$n_\infty = \frac{\beta N}{\alpha + \beta} \qquad (15)$$

It is worth noting here that β is the probability for electrons located in the conduction band to be captured in vacant traps and consequently must depend on dose-rate. From (11) and (15),

$$n_\infty = \frac{P\gamma}{\alpha + \beta} \qquad (16)$$

If there are two dose rates, P_1 and P_2, corresponding to two different saturation values $n_{1\infty}$ and $n_{2\infty}$ respectively, and assuming γ is constant, we obtain

$$\frac{n_{1\infty}}{n_{2\infty}} = \frac{P_1(\alpha + \beta_2)}{P_2(\alpha + \beta_1)} \qquad (17)$$

If storage takes place in shallow traps (ie $\alpha \gg \beta$) and neglecting β, (17) may be written as,

$$\frac{n_{1\infty}}{n_{2\infty}} = \frac{P_1}{P_2} \qquad (18)$$

which shows that for shallow traps (τ is small) the saturation value is proportional to dose-rate. For deep traps ($\alpha \ll \beta$) with sufficiently long charge carrier lifetimes, substituting in (19) the expression for β obtained from (11) gives,

$$\frac{n_{1\infty}}{n_{2\infty}} = \frac{P_1 \frac{\gamma P_2}{N}}{P_2 \frac{\gamma P_1}{N}} = 1 \qquad (19)$$

Dose rate dependence of n_∞ is not observed with deep traps. The proposed model enables the estimation of the values of dose rate at which the probabilities of trapping and eviction of charge carriers from a particular trap are of the same magnitude (potential equilibrium conditions).

If we assume $\alpha = \beta$, and as in the laboratory saturation stage all the vacant traps are filled, i.e. $N = n_{\infty_L}$, formula (11) may be rewritten in the following way:

$$\beta = \frac{\gamma P}{n_{\infty_L}} \qquad (20)$$

As γ is the palaeodosimetry dose sensitivity, $\frac{dn_o}{dD}$, and, in this case $\beta = \alpha = 1/\tau$, then,

$$\frac{1}{\tau} = \frac{dn_o\, P_{crit}}{dD\, n_{\infty_L}} \qquad (21)$$

$$P_{crit} = \frac{dD\, n_{\infty_L}}{dn_o \tau} \qquad (22)$$

The critical dose rate value for traps with different life times may be calculated from formula (22) when τ is known.

The model presented furnishes a basis for estimating effective lifetimes, τ_r, of charge carriers in traps considering the difference between natural and laboratory saturation values. Leaving aside mathematical calculations, we confine ourselves with presenting the final formula only:

$$\tau_r = \frac{dD\, n_{\infty_L}}{dn_o\, P_N}\left(\frac{n_{\infty_L}}{n_{\infty_N}} - 1\right) \qquad (23)$$

where,

τ_r - is the effective lifetime of charge carriers in traps valid for individual samples in natural conditions of burial.

Below is a table of age correction coefficients obtained from laboratory reconstruction of palaeodose.

Table 1. Age correction coefficients for laboratory reconstruction of palaeodose.

$\frac{I_N}{I_{\infty N}}$	$I_{\infty L}/I_{\infty N}$						
	1.1	1.2	1.5	2	3	5	10
10	1.002	1.005	1.03	1.04	1.04	1.04	1.05
25	1.015	1.03	1.06	1.09	1.10	1.12	1.18
50	1.033	1.06	1.15	1.23	1.26	1.31	1.38
75	1.10	1.17	1.33	1.46	1.61	1.69	1.78
90	1.23	1.38	1.57	1.72	1.90	2.32	2.45
99	1.82	2.20	2.78	3.33	3.38	4.18	4.42

Discussion

The main problem is the determination of the natural saturation value (I_{∞_N}) to get the ratio $I_{\infty_L}/I_{\infty_N}$. Laboratory and natural saturation values have been found to be equal equal ($I_{\infty_L}/I_{\infty_N} = 1$) for numerous quartz samples from different climatic regions. For each sample, added laboratory doses did not result in an increase in TL, from which we conclude that the laboratory saturation value had been obtained.

As discussed above, laboratory and natural saturation values are functions of the number of traps, but the latter additionally depends on τ of the corresponding traps. τ for the relevant traps in quartz is $\sim 10^{13}$ a, assuming a monomolecular model (Hütt et al., 1979), and this accounts for the similarity of the laboratory and natural saturation values. Consequently, the laboratory reconstructed palaeodose needs no correction. In the case of other materials (feldspars, carbonates), the problem is more complicated. Investigations (Mejdahl, 1988; and our results using OSL) of old samples have shown that for alkali feldspars the laboratory saturation value is greater than the natural saturation value. Consequently for dating samples, the theoretical value, $\tau = 3.10^7$ a, is not realised for a quasi-continuous energy trap spectrum (Strickertsson, 1985) of alkali feldspars, and generally a much lower value applies.

The difference between natural and laboratory saturation values was measured by ESR for aragonite shells (Hütt and Jaek, in press). The important problem of constancy of the natural saturation level, independent of the geological region of sampling, needs additional investigation for old samples from different climatic regions. If processes of age information "degradation" are thermally dependent, this value will be constant only for definite climatic regions. In cases where these processes are not strongly thermally dependent (like non-radiative transitions) this ratio may be effectively constant.

As an illustration of usage of the proposed model we analysed an experimental dose response curve for alkali feldspars from a marine sediment sample to estimate P_{crit} and τ_n (see equations 22, 23). Using the following values; $\Delta D = 300$ Gy; $\Delta I = 171$, $I_{\infty_L} = 896$, $I_{\infty_L}/I_{\infty_N} = 1.2$ (pers. comm. Vagn Mejdahl, 1988) yields

$$P_{crit} = 5.25.10^{-5}\ \text{Gy a}^{-1} \qquad \tau = 3 \times 10^6\ \text{a}$$

The fact that P_{crit} is two orders of magnitudes smaller than the annual dose rate ($2.5.10^{-3}$ Gy.a^{-1}) demonstrates that dependency of saturation value dependency on dose rate is absent within the range from natural to laboratory rates. However, the effective thermal stability of carriers in the particular traps confirms Mejdahl's conclusion that the upper limits of dating are lower than values deduced from laboratory reconstructed dose response curves.

Conclusion

1. The standard plateau test is a necessary but insufficient condition for obtaining reliable results.
2. The saturation test is not valid for alkali feldspars, carbonates and, perhaps, other minerals, because the growth of signal may be observed for naturally saturated samples too.
3. The fact that $I_{\infty_L}/I_{\infty_N}>1$ decreases the upper dating limit requires corrections to be applied to TL dates.
4. Experimental studies of samples having ages beyond the dating limits from wider climatic regions are necessary.

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References

Aitken, M. J. (1985) *Thermoluminescence dating*. Academic Press, London.

Debenham, N.C. (1985) Use of U.V. emissions in TL dating of sediments, *Nucl. Tracks and Radn. Measts.*, **10**, 717-725.

Hütt, G., Smirnov, A. and Tale, I., (1979), On the application of TL of natural quartz to the study of geochronology of sedimentary deposits, *PACT J.*, **3**, 362-374.

Hütt, G., and Jaek, I., Laboratory reconstruction of palaeodose: problems of dating accuracy and upper limitation, *Appl. Rad. and Isotopes*, in press.

Kortov, V.C., (1986), radiation defects on the surface of silica and aluminium oxides crystals, Abstracts, The VI All Union Conference on Radiation Physics and Chemistry of Ion Crystals, Riga, Oct 9-11, 1986, (in Russian), 27.

Mejdahl, V., (1988), Long term stability of the TL signal in alkali feldspars, *Quarternary Science Reviews*, **7**, 357-360.

Shljukov, A., and Shakhovets, S. (1986), The validity of radiation calibration of quartz in TL dating, International TL Workshop on Methodology of TL Dating, Tallinn, April 20-24, 2986. (Unpublished).

Strickertsson, K., (1985), The thermoluminescence of potassium feldspars, *Nucl. Tracks and Radn. Measts.*, **10**, 613-617.

PR Reviewer's comments (Vagn Mejdahl)

The use and definition of the term critical dose-rate might benefit from some reconsideration. As the natural dose-rate is increased, the saturation level will gradually approach the laboratory saturation level, and it becomes a matter of definition when the natural dose-rate is sufficiently high for the application of a fading correction to be no longer required. Thus the concept and evaluation of a "critical" step in this situation appears inappropriate.

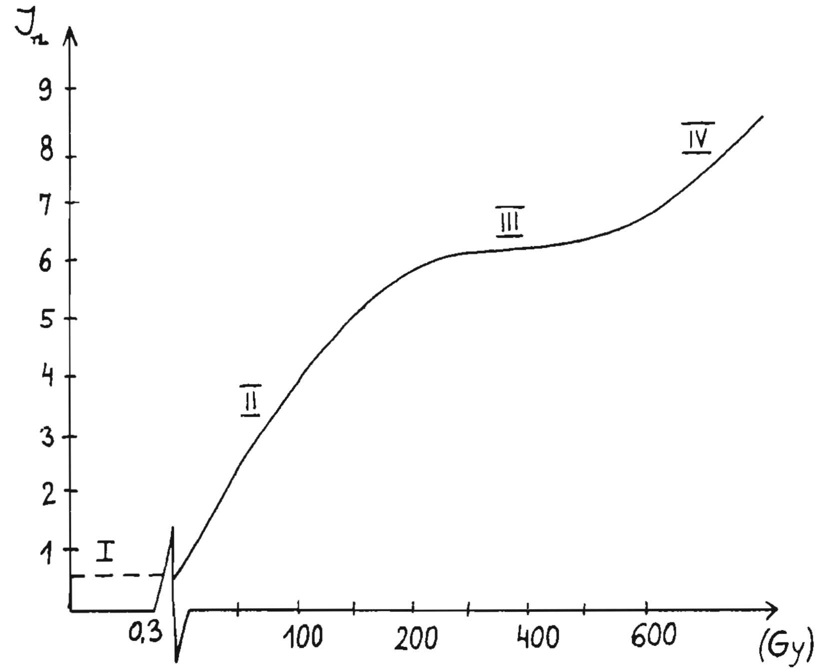

Figure 1. Dose response curve. Axes; the ordinate represents TL signal (arb. units), and the abscissa represents laboratory dose (Gy).