A Regression Method for Evaluation of Paleodose in the Pre-dose Technique

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***Abstract**: A regression method for expanding the application range of the pre-dose technique is introduced. Comparing methods on the basis of a common acceptable prediction uncertainty, the limiting value of the allowable paleodose N in the regression method is found ($N \leq 1.2B$) to be at least twice that applicable to other methods. Uncertainties arising from the non-linear filling of R traps are also discussed.*

I Introduction

The pre-dose technique in thermoluminescence (TL) dating was established more than twenty years ago following the reports of Fleming (1969, 1973) and Zimmerman (1971) and has attracted continuing study. Essentially Fleming reported that the 110°C TL peak in quartz retains a "memory " of previous irradiation and this "memory" can be used to evaluate the paleodose of archaeological samples. A model for the interpretation of the pre-dose effect was originally proposed by Zimmerman (1971). Studies and further developments of the potential for application of the method have since been reported (Chen, 1979, Aitken, 1985, Bailiff and Haskell, 1984). A detailed description of the procedure used in this technique and the proposed model are provided by Aitken (1985) and Bailiff (1991).

In Zimmerman's model, two electron traps are involved and two hole traps— the luminescence centre L and the reservoir trap R. Although a saturating exponential sensitivity change was pointed out long ago by Chen (1979), in most earlier reports about the practical evaluation of the paleodose it is assumed that both hole traps are linearly filled through the irradiation dose. As this assumption is correct only when the dose is very small, i.e. within the range of a few Gy, this significantly limits the age for archaeological samples suited to the pre-dose method.

In recent years, growing attention is being paid to the effects of non-linear filling of the hole traps. In the review paper of Bailiff (1991), techniques for reducing the experimental errors introduced by the non-linearity are presented.

A regression method that expands the range of application of the pre-dose technique is suggested and discussed in this work.

II Experimental Work

The natural quartz and the ancient brick samples used in this work are from China.

Two Sr^{90} β sources each 50mCi were used for the additive dose and test dose, set up at different irradiation distances. All TL measurements were performed on a 7188 thermoluminescence dating system manufactured by Littlemore Scientific Engineering Company, Oxford. Thermal activation temperatures are 500°C for the natural quartz and 470°C for the quartz extracted from the ancient brick (Fig. 1 shows the thermal activation characteristics). The test dose was 0.01Gy, the heating rate 10°C/s, and sensitivity was recorded by integrating the TL intensity from 80°C to 140°C.

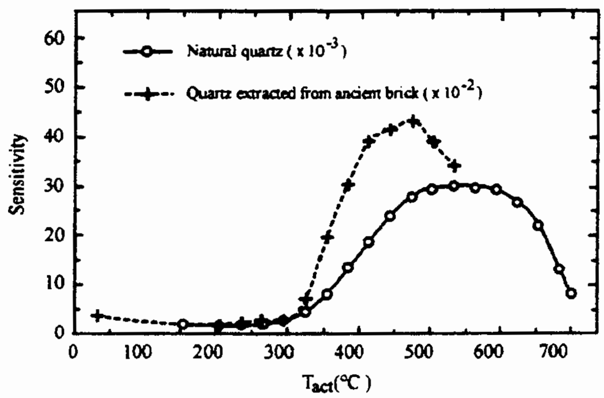

***Figure 1.** Thermal activation characteristic curves of natural quartz (solid line) and quartz extracted from the 1022 years old brick (dashed line)*

III The Regression Method

1. *Principle*

If it is assumed that the reservoir traps are filled linearly, the sensitivity S can be represented as:

$$S = S_\infty (1 - e^{-D/B}) \qquad (1)$$

where S_∞ is the saturation value of S in the absence of quenching, B is a constant and D is the irradiation dose (Chen, 1979).

Quenching occurs if the sample has been successively irradiated and activated. The sensitization of one step in the dose/activation cycles, ΔS, is given by:

$$\Delta S = S_\infty (1 - S{\downarrow}/S_\infty)(1 - e^{-\beta/B}) \qquad (2)$$

where $S{\downarrow}$ is the quenched value of S and β is the dose increment for this step (Bailiff, 1991).

Equation (2) can be written in an alternative form:

$$\Delta S = S_\infty (1 - e^{-\beta/B}) - (1 - e^{-\beta/B}) S{\downarrow} \qquad (2a)$$

If β remains unchanged in the successive dose/activation cycles, $(1 - e^{-\beta/B})$ will be constant. Consequently, a linear relation between $S{\downarrow}$ and ΔS should be found with gradient $b = -(1 - e^{-\beta/B})$ and intercept $a = S_\infty (1 - e^{-\beta/B})$.

This suggests a method enabling the paleodose N to be evaluated. Serial measurements of $S{\downarrow}$ and ΔS separated by equal dose increments β will give ($S{\downarrow}$, ΔS) data pairs. Regression of the data will give gradient and intercept values from which S_∞ and B can be obtained. The paleodose N can then be found from the equation:

$$\Delta S_N = S_N - S_0 = S_\infty (1 - S_0/S_\infty)(1 - e^{-N/B}) \qquad (2b)$$

2. Examples

(i) *Natural quartz* - the natural dose of an archaeological sample cannot be precisely known so, to test the method, fine grain natural quartz annealed at 800°C for 15 hours is used. The "natural" dose is replaced by a laboratory dose, giving an equivalent paleodose N followed by warming to 150°C. It has been found that the characteristics and range of linearity varies widely in quartz with various origins. More than ten groups of quartz samples found in China, either natural or extracted from ancient potterys, were tested. The linear range of these samples was found to be 2-3Gy only.

First, an equivalent paleodose N=3.38Gy was used to test the proposed method.

Measurements of the sensitivities of the 110 °C peak in the successive dose/activation cycles were performed with a 2.70Gy dose increment. Results are given in Table 1.

Dose(Gy)	S ($\times10^4$)	$S{\downarrow}$ ($\times10^4$)	ΔS ($\times10^4$)
N	3.56	2.37	1.84
N+2.70	4.21	3.08	1.51
N+5.40	4.58	3.51	1.31
N+8.10	4.82	3.89	1.08
N+10.80	4.97		

Table1. *Sensitivities and sensitizations of natural quartz samples with a "paleodose" administered in successive dose/activation cycles.* ($S_0 = 0.14\times10^4$)

The ΔS ~ $S{\downarrow}$ curve is shown by the solid line in Fig. 2. As predicted, there is a good linear relation between ΔS and $S{\downarrow}$. The regression parameters from eqs. (2a) and (2b) give $S_\infty = 6.11\times10^4$; B = 3.94Gy and N = 3.35Gy.

It can be seen that the calculated value of N lies very close to the actual laboratory "paleodose" — 3.38Gy. Repeated examinations have been performed with similar quartz samples but varying the "paleodose" and the dose increment β. The largest "paleodose" used was 4.73Gy. It is shown by the experimental results that the uncertainty in the calculated N can be kept within 10% by using an appropriate value of β (this will be discussed in the following section).

(ii) *Quartz extracted from ancient brick* - measurements were also taken with fine grain quartz extracted from an ancient brick sample produced in the Five Dynasties. The age of the sample is 1022 years. The results are given in Table 2.

Dose(Gy)	S($\times10^3$)	$S{\downarrow}$($\times10^3$)	ΔS($\times10^3$)
N	4.55	3.77	1.97
N+4.75	5.74	5.03	1.17
N+9.50	6.20	5.66	0.73
N+14.25	6.39	6.15	0.33
N+19.00	6.48		

Table2. *Sensitivities and sensitizations of quartz samples extracted from ancient brick in successive dose/activation cycles* ($S_0 = 0.22\times10^3$)

The ΔS ~ $S{\downarrow}$ curve of this sample is shown by the dashed line in Fig. 2. The values obtained by

The $\Delta S \sim S\downarrow$ curve of this sample is shown by the dashed line in Fig. 2. The values obtained by regression are: S_∞ = 6.69x10^3; B = 4.15Gy; N = 4.54Gy.

This result appears reliable as the average annual dose of Chinese ceramics for pre-dose method, in which the α-dose should be neglected, is about 4.23mGy (Stoneham, 1987), then the age of that ancient brick obtained from the result N = 4.54Gy is about 1073 years.

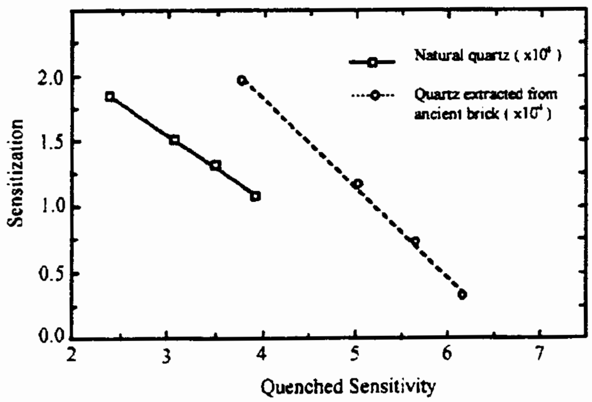

***Figure** 2.Regression curves of the sensitization and quenched sensitivities of natural quartz (solid line) and quartz extracted from 1022 years old brick (dashed line) in the dose/activation cycles*

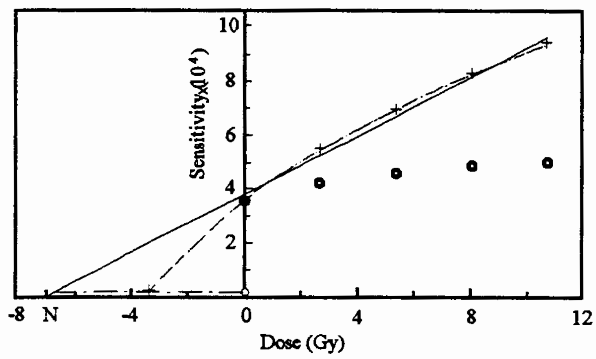

***Figure** 3.The relation between sensitivity and irradiation dose, details in text.*

IV Discussions

1. *Comparison between the regression method and the standard methods*

In previous work (Aitken, 1985, Bailiff, 1991), several methods for evaluating the paleodose N have been reported. The equivalent paleodose in example 1 presented in the above section is also evaluted with those methods for comparison.

(i) *by the multiple activation technique* Data processing of example 1 is shown in Fig.3, where the empty cycles represent the data of S given in Tab.1, the plus symbols represent the points corrected from quenching. The principle of the correction for quenching is described by Atiken(1985).

It is found from the solid line and the dots in Fig.3 that the calculated paleodose N is about 7.0Gy. Comparing with the true paleodose N=3.38Gy, the error is larger than 100%.

(ii) *by putting the first two rows of Table 1 in the equation*:

$$N = \frac{S_N - S_0}{S_{N+\beta} - S_N\downarrow} \bullet (\beta - \beta_T) \qquad (3),$$

where β_T is the test dose. The calculated value of N is 4.97Gy — this is 50% above the actual value.

For the discussion of those errors, equation (1) $S = S_\infty (1 - e^{-D/B})$ is rewritten:

$$S = S_\infty [D/B - 1/2(D/B)^2 + 1/6(D/B)^3 - \ldots] \quad (1a)$$

i.e. the linear relation only holds when D << B, then $S \approx S_\infty (D/B)$.

The multiple activation technique and eq.(3) are both based on the assumption of a linear relation between S and D, though a pseudo non-linearity is introduced by quenching when multiple activations are employed.

The real relation between sensitivity and dose in example 1 is shown by the dashed line in Figure 3, which indicates that the dose used in this example is far beyond the linear range. Large errors were introduced by the non-linearity. The error in method (ii) is smaller than that in method (i), as less non-linear part of the S-D curve is used in method (ii) case.

It can be shown from equation (1a) that the error produced when using equation (3) will be larger than 20% when $D/B \geq 0.7$. That means that equation (3) is properly valid only for the case of D/B<0.7, i.e. $(N+\beta) < 0.7B$ or $N < 0.5B$. And in the case of multiple activation technique, the value of N is even more limited.

In the regression method, the paleodose N is evaluated from the values of the intercept *a* and the gradient *b*. The uncertainties in *a* and *b* in the present work are about 5%. It can be shown that the main contribution to the uncertainty of the final result is caused due to the uncertainty in B. As $b = -(1 - e^{-\beta/B})$, i.e. $-\beta/B = \ln(1+b)$, it follows that:

$$|\Delta B/B| = |\Delta b/b|\ |[(1+1/b)\ln(1+b)]^{-1}| \quad (4)$$

where $-1 < b < 0$. It is seen from eq.(4) that the relative error in B increases with the increase of the absolute value of b. For example, suppose $|\Delta b/b| \approx 5\%$. If $b = -0.7$ then $|\Delta B/B| \approx 10\%$; if $b = -0.8$ $|\Delta B/B| \approx 12.5\%$; and $|\Delta B/B|$ will be about 45% when $b = -0.97$.

It is suggested that $|b|$ should not be larger than 0.7 (i.e. $\beta/B \leq 1.2$) if the error of the final result is expected to be within 15%. As discussed in the following section, the value of β should in fact be as close as possible to the paleodose N. Thus the regression method would give acceptable results within the range $N \leq 1.2B$.

This expands the application range by a factor of at least two compared to the standard method.

(ii) *The effects of non-linear filling of the reservoir traps*

Equation (1), which is the foundation of the regression method, is based on the assumption that the traps R are filled linearly. In fact the population of R traps themselves would be better described by an expression similar to equation (1) (Chen, 1979). The assumption of linearity is in fact only correct when the irradiation dose is small. In the work reported here both N and β are outside this condition.

The non-linear filling of R traps will give a further contribution E_R to the estimated uncertainty. It can be shown that E_R is determined largely by the ratio $|(N-\beta)/\beta|$ and also the ratio between the natural dose N and the saturation (i.e., 90% of R traps are filled) dose of the R traps, D_s^R.

Table 3 gives an indication of the relationships involved.

N/D_s^R	$\|(N-\beta)/\beta\|$	E_R
17%	0	0
	10%	2%
	20%	5%
33%	0	0
	10%	4%
	20%	10%
67%	0	0
	10%	7%
	20%	15%

Table3. *The relation between E_R, N/D_s^R and $|(N-\beta)/\beta|$*

It can be seen from Table 3 that E_R increases with the ratio $|(N-\beta)/\beta|$. Obviously its value can be reduced if β is close to N, and can even be neglected when $\beta \approx N$. This is in agreement with Bailiff's report (1991). This indicates that, before measurements are taken for an archaeological sample, the approximate natural dose should be estimated in order to select the appropriate dose increment β. When the pre-dose technique is used for a paleodose outside the linear range, the result should be considered unreliable if the calculated value of N in the final result is much larger or much smaller than β. In this case it is advisable to use an adjusted dose increment closer to the value of the obtained N and reiterate the whole experimental procedure until the difference obtained between N and β is within 20%.

When the dose is near the saturation dose of R traps, a small difference between N and β will induce a serious error in the final result. This is the main reason to limit the values of the paleodose and the dose increment in the regression method. It is not easy to estimate the saturation dose of R traps. However in all the measurements reported in this work no serious error caused by the non-linear filling effects of R traps has been detected within the conditions of $N \leq 1.2B$ and $\beta = (1 \pm 20\%)N$.

V Summary

Using the pre-dose technique, the paleodose can be evaluated from the parameters obtained in the regression of the the sensitizations and quenched sensitivities recorded during the dose/activation cycles. Compared with the standard method, the range of application is expanded by a factor of at least two using the regression method. The conditions $N \leq 1.2B$ and $\beta = (1\pm20\%)$ N are suggested for obtaining reliable predictions.

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