

## Saturation of sensitization of the 110°C TL peak in quartz and its potential application in the pre-dose technique.

### *Abstract*

The increase of sensitivity of quartz by  $\beta$  or  $\gamma$  irradiation followed by high temperature activation has been investigated further. This pre-dose effect is being used for the evaluation of the natural dose given to quartz in ancient pottery. In this work we extend the method to include the cases of an exponential approach to saturation (rather than linearity) of either the reservoir or the centre or both. The results for simulated natural doses given to Norwegian  $\alpha$  quartz compare nicely with the given doses. Results with some archaeological quartz samples give good age for a "young" sample and low equivalent doses for older samples.

### *1. Introduction*

The sensitization of various thermoluminescent (TL) materials by a  $\beta$  or  $\gamma$  irradiation followed by an annealing at high temperature is a well known phenomenon (Cameron et al., 1968; Zimmerman, 1971b, Piesch et al., 1977). By sensitization one means the change of sensitivity — i.e. the response to a given (usually small) test dose — by a heavier irradiation followed by heating. In particular this effect was found (Fleming and Thompson, 1970) in quartz and therefore it is of importance when dealing with the dating of archaeological pottery. The sensitization effect, sometimes called the pre-dose effect, of the 110°C peak in quartz has further been investigated by Zimmerman (1971a) who also gave a model to account for the phenomenon. Briefly, the model deals with an electron trap T and two hole centres R and L (see Fig. 1). The cross section for trapping holes is assumed to be much bigger for R than for L, therefore, during irradiation, practically all the free holes accumulate in R whereas the created electrons, or at least some of them, concentrate in T. This trap is, however, rather shallow (yielding a peak at 110°C at a heating rate of 5°C/sec) and is emptied within hours (and certainly during antiquity) or may be emptied by heating the sample to, say 150°C.

It is assumed, however, that the recombination probability of a thermally freed electron and a hole in R is very low, so that the thermal emptying of T does not influence the concentration of holes in R. One further assumes that R is not a very "deep" state, i.e. that it is close enough to the valence band so that heating to about 500°C would release the holes from R. Although the probability of their trapping in L is rather low, as mentioned above, at this high temperature the favourable direction for the holes is to go from R to L. This is so since L is considered to be much farther from the valence band and, therefore, does not release holes at 500°C once they are captured. The increase of the number of holes in L represents an increase in the sensitivity, i.e. the response to a test dose since L is the luminescence

centre, the transition into which yields the measured 110°C peak (see appendix). The other centre, R, serves as a reservoir and its properties can be investigated only indirectly through transferring the holes into L by heating. Zimmerman (1971a) also found that applying UV light to the sensitized quartz substantially reduced the sensitivity but heating to 500°C reactivated it. The explanation was that UV light could transfer the holes back from L to R. It is to be mentioned (Thompson, 1970) that the levels corresponding to the 110°C TL peak in quartz are related to lattice defects rather than to impurities.

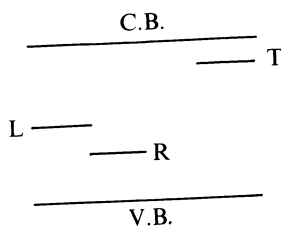


Fig. 1. Energy levels in the forbidden band needed for the explanation of sensitization (schematic). T — an electron trap. L — a hole recombination centre. R — a hole reservoir.

Fleming (1972, 1973) suggested the use of the sensitization for dating — the pre-dose technique. One can measure the change from  $S_0$ , the original sensitivity, to  $S_N$ , the increased sensitivity due to the natural  $\beta$  and  $\gamma$  dose, revealed by heating the samples to  $\sim 500^\circ\text{C}$ . An additional calibration measurement is now needed in order to evaluate the natural dose responsible for the measured sensitivity change. Fleming suggested two procedures to do this. In one procedure, two portions of the pre-dosed sample, preferably equal in weight, are used. One is activated at  $500^\circ\text{C}$  and when cooled, the sensitivity  $S_N$  is measured. The other is given an incremental dose  $\beta$  and then activated at  $500^\circ\text{C}$  and the sensitivity  $S_{N+\beta}$  is measured. The difference between the two, measures the change in sensitivity due to the additional  $\beta$ . The sensitivity enhancement rate  $p = \frac{S_{N+\beta} - S_N}{S_0} \times \frac{1000}{\beta}$  per krad is defined. In the other procedure one uses one portion of the sample only. This is activated at  $500^\circ\text{C}$  and the sensitivity  $\bar{S}_N$  is measured. An incremental dose  $\beta$  is now given and the sample is reactivated at  $500^\circ\text{C}$  and when cooled, the sensitivity  $\bar{S}_{N+\beta}$  is measured. The sensitivity enhancement rate is now defined as  $q = \frac{\bar{S}_{N+\beta} - \bar{S}_N}{S_0} \times \frac{1000}{\beta}$  per krad. The equality of  $p$  and  $q$  indicates that no saturation occurs, and therefore, a linear extrapolation between  $S_{N+\beta}$  (or  $\bar{S}_{N+\beta}$ ) and  $S_N$  down to the original sensitivity  $S_0$  should yield the natural dose  $N$  (Fleming, 1973). If, however, either the reservoir or the centre (or both) show saturation effects, this procedure should not give the correct result. As shown below,  $p = q$ , indicates the linearity of the reservoir, but not necessarily of the centre, whereas  $q > p$  occurs when the reservoir shows saturation effects, no matter whether the centre is linear or not. The saturation effects of this TL can be investigated (Fleming, 1973) by giving several incremental doses followed by activation to  $500^\circ\text{C}$  and measuring the sensitivities in between.

Aitken and Murray (1976) reported the quenching of sensitivity by an incremental dose. When, for example, after activating at 500°C the naturally pre-dosed sample which now has a sensitivity  $S_N$ , we give an additional dose of, say,  $\beta = 200$  rad, and heat to  $> 110^\circ\text{C}$  (but not too high, to prevent high temperature activation), the sensitivity measured before re-acting at 500°C is  $S'_N$  with  $S'_N < S_N$ . According to the model, this is explained to result from the reduction in the concentration of holes in L due to recombination with some of the large number of electrons released from T during the thermal cleaning of the 110°C resulting directly from the 200 rad. This seems to be a partial reason to the non-linear dependence of the sensitivity on the dose. Aitken and Murray suggested to make quenching correction, i.e. to add the value of  $S_N - S'_N$  to  $S_{N+\beta}$ , the result should represent the expected sensitivity had there been no quenching. The same correction procedure should be used for additional incremental doses to correct  $S_{N+2\beta}$ ,  $S_{N+3\beta}$  etc. It has, however, been found that even with this correction, the behaviour is very often sublinear and one should investigate the way of filling the reservoir and the centre as a function of the given doses. This has been done in the present work. A method is suggested and tested, by which the natural dose N can be evaluated in cases where the reservoir, the centre or both are showing saturation effects. Most of the work was done on Norwegian  $\alpha$  quartz (supplied by BDH) previously fired at temperatures varying between 500°C and 1000°C. The natural dose was simulated by a lab. dose, usually of 100-1000 rads followed by a slow heating to 150°C in order to bleach out the direct 110°C peak. The value of the dose evaluated by the proposed method could thus be compared to the "natural" simulated dose. Another part of the work dealt with some archaeological samples (90-150 $\mu$  grains were usually used) and the pre-dose equivalent dose was evaluated. Two recent papers on the sensitivity of quartz (Durrani et al., 1977; David et al., 1977) should be mentioned here. Durrani et al. found a strong dependence of the TL intensity on the temperature of excitation; the sensitivity decreased substantially with decreasing temperature both for x-rays and  $\gamma$  irradiations. David et. al. found changes in the sensitivity of  $\alpha$  quartz following heating to  $\sim 600^\circ\text{C}$ , a temperature beyond the transition to  $\beta$  quartz. The initial use of a test dose as high as  $4 \times 10^4$  R may mean that at least part of the effect is due to the combination of irradiation plus heating, rather than heating alone. The results are explained by David et al. on the basis of generation of vacancies in the crystal lattice.

## 2. Experimental.

The measurements were made using the apparatus described by Zimmerman (1971a, 1971b). An EMI 9635 QB photomultiplier was used for light detection, the anode pulses were counted as described by Aitken et al. (1968). Irradiations were made with beta particles from  $^{90}\text{Sr} - ^{90}\text{Y}$  plaque sources. The BDH quartz has been ground and sieved to obtain grains of 90-150 $\mu$ . An initial firing in an oven was carried out at 500°C (5,25 sample) or 700°C (7,25 sample) in air. Some of the samples, of both kinds, (5, 25 and 7,25),  $\beta$  irradiated and unirradiated were refired for several hours at  $\sim 1000^\circ\text{C}$  in air.

The heating rate used in most of the measurements was 5°C/sec. A heating rate of 1°C/sec was used while bleaching out the 110°C peak right after a high-dose irradiation (50 rad and more), prior to the application of a test dose.

### 3. Analysis of reservoir and centre saturation

Before going into the more complicated case where both the reservoir and the centre are not linear, let us start with a simpler case in which the reservoir is linear whereas the centre shows an exponential approach to saturation. (This behaviour resembles the exponential approach of TL intensity as described by Bjärngård (1967) and Huxtable and Aitken (1977)). If the number of holes made available by the reservoir during the activation is denoted by  $N_R$ , we would expect the sensitivity  $S$  to behave like  $S = S_\infty(1 - e^{-N_R/B})$  (1) where  $S_\infty$  is the maximum sensitivity attainable for very high  $N_R$ , and  $B$  a constant. In this simple case of reservoir linearity,  $N_R \propto D$  and (1) can also be written as

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

where  $D$  is the dose giving rise to  $N_R$  in the reservoir and  $B'$  a constant having dose units. In this case, there is no difference whether the dose  $D$  is given all at once followed by the activation at 500°C or in smaller amounts accumulating to  $D$  with activations in between. Once the sensitivities (corrected for quenching) are known as a function of the dose, one can use a numerical method to evaluate  $S_\infty$  and  $B'$  and to extrapolate the curve to  $S = S_0$ , thus finding the equivalent dose  $N$ . We have found that the following graphical method would be satisfactory and rather easy to use. We re-write eq. (1') as  $S_\infty - S = S_\infty e^{-D/B'}$ .

If  $S_\infty$  is known, one can plot  $\ln(S_\infty - S)$  as a function of  $D$  and get a straight line, the slope of which is  $-1/B'$ . If, however,  $S_\infty$  is not known, one can guess a value of  $S_\infty$  which should, certainly, be greater than the biggest measured  $S$ . If the plot of  $\ln(S_\infty - S)$  as a function of  $D$ , for the guessed  $S_\infty$  is linear (curve b, Fig. 2), we deduce that the chosen  $S_\infty$  was the correct one. If the curve obtained is concave (Fig. 2, a), one should choose a lower  $S_\infty$ , whereas

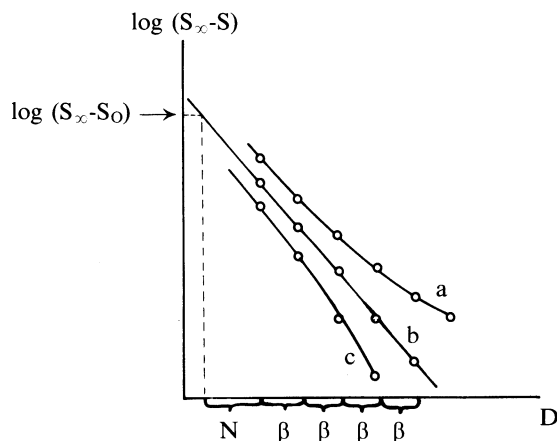


Fig. 2. The graphical method for evaluating the equivalent dose.

if it is convex (Fig. 2, c), a higher  $S_\infty$  should be chosen. By this trial and error method, one can get in a few steps the best straight line. Once this is established the value of  $S_\infty$  is known and the straight line can be extrapolated to  $S_\infty - S_0$  which would give the natural dose.

We should go now to the more common case where both the reservoir and the luminescence centre are beyond their linear range. If this is the case,  $N_R$  is an increasing function of the dose, though not a linear one. If we use a number  $n - 1$  of *equal* increments  $\beta$ , if  $N = \beta$ , and if we now denote by  $N_R$  the concentration accumulated in R during one irradiation, then after these  $n-1$  irradiations and  $n$  activations, we shall have  $S = S_\infty (1 - e^{-n \cdot N_R/\beta})$ . Of course, the previous technique should give us the correct  $N = \beta$  as a result of the extrapolation. Usually, however,  $N \neq \beta$  because we, certainly, do not know in advance the value of  $N$ . If, for example,  $N = 2\beta$ , we would have  $N_{R\beta} < N_{RN} < 2N_{R\beta}$  when  $N_{R\beta}$  is the filling of the reservoir under a dose  $\beta$  and  $N_{RN}$  is the filling under the natural dose. This is the number made available to L during the activation, and thus the previous procedure would give for example on the dose scale  $N = 1.5\beta$  (but we still would expect a straight line on the semilog graph, provided that the same increments  $\beta$  are used in one series of measurements). On the other hand, if we use  $\beta > N$ , for example  $\beta = 2N$ , we should expect to get a straight line on the semilog scale extrapolating to  $\frac{1}{2}\beta < N < \beta$ . Thus, if the result of a series of measurements is an  $N < \beta$ , we should repeat the series with a smaller increment whereas if  $N > \beta$  we should try a bigger increment until we end up with a result in which  $N = \beta$ , which should be the correct natural dose. We believe that by this procedure both the reservoir and centre saturation behaviours are taken into account.

It is to be noted that the procedure is workable just as well if either the centre or the reservoir are in the linear regions. If the reservoir is in the linear range, we should expect the correct  $N$  even if  $N \neq \beta$  (and even if non-equal increments are used in the same series). This can be detected by getting the same  $N$  in various series, each using a different  $\beta$ , including, of course,  $\beta = N$ . If, on the other hand, the filling of the centre is linear, we will see immediately that the results (corrected for quenching) are linear rather than exponential. If by using a linear extrapolation we get  $N \neq \beta$ , we should repeat the measurement with a different  $\beta$  until we get, on the linear scale,  $N = \beta$ . If both the reservoir and the centre are in the linear range, we should expect a straight line extrapolating to  $N$  even if  $N \neq \beta$ . Again, we would get the same value of  $N$  in another series using a different increment.

Finally, a word of caution, if at the end of a single set of measurements we get a "good" exponential or linear curve yielding  $N = \beta$ , we should still repeat the measurement with a different  $\beta$ . The reason is that if the reservoir is *entirely* in saturation after the natural dose  $N$  and each incremental dose  $\beta$ , then each activation transfers the same number of holes (the saturation value) to the centre, which is not a function of  $N$  or  $\beta$  any more. Experimentally, one can see this by getting, say, a result of  $N = 200$  rad (on a linear or semilog scale de-

pending on the centre properties) for  $\beta = 200$  rad,  $N = 300$  rad for  $\beta = 300$  rad etc. If this is the case one cannot, in principle, expect to extract the real natural dose  $N$  from the results.

#### 4. Results on BDH quartz

Two different kinds of BDH quartz have been used. One was previously fired to  $500^\circ\text{C}$  in air and had a high value of  $S_0$  and relatively small sensitization, about 60% for 100 rad. The other which was previously fired to  $700^\circ\text{C}$  in air had a lower value of  $S_0$  and much bigger sensitization (a factor of about 15 for 100 rad). The results were usually better in the latter. The reason seems to be that for the extrapolation one does not know which value of  $S_0$  to take. In the BDH quartz we had the advantage of being able to measure  $S_0$  before applying the natural (simulated) dose. Doing this, we found a certain amount of quenching of  $S_0$  and one should therefore ask whether to use the  $S_0$  measured before the dosing (which cannot be measured in archaeological samples!) or after. Moreover, it has been found (Bailiff, 1977) that  $S_0$  and  $S_N$  have a slightly different emission spectra which may indicate that possibly only part of  $S_0$  has to be taken into account in the present procedure. In addition, it has also been found (Thompson, 1970) that UV light could reduce  $S_0$  and it has been suggested that, perhaps, the reduced value should be used for the extrapolation. Moreover, in one high- $S_0$  BDH sample, it has been found that the sensitivity increases by irradiation without the thermal activation. These effects may, certainly, have much bigger influence on the 5.25 (high  $S_0$ , low sensitization) sample than on the 7.25 (low  $S_0$ , high sensitization) one. Another kind of BDH quartz was obtained by refiring the 5.25 samples and the 7.25 samples at  $1000^\circ\text{C}$  in air for 16 hours. In both cases, the result was a sample that could be sensitized by a factor of roughly 5 by 100 rad. Moreover, the results were practically the same when the samples were irradiated by 1 krad prior to the refiring at  $1000^\circ\text{C}$ . This fact has an important implication in archaeometry since it proves that any prior (geological) dose absorbed by the sample is "erased" once it is fired at  $1000^\circ\text{C}$ .

Table I gives some of the results of BDH quartz. As can be seen, in most cases when  $N = \beta$ , the best results are achieved using the exponential extrapolation with the quenching-corrected points. The linear extrapolation gave usually poorer results and was in most cases based on two points only,  $S_N$  and  $S_{N+\beta}$ , whereas the other points fell below this line. In the cases where  $\beta < N_{\text{given}}$ , we got, as expected  $\beta < N_{\text{eval}} < N_{\text{given}}$ . In these cases the  $N$  evaluated by the points uncorrected for quenching seems to be closer to  $N_{\text{given}}$  than  $N$  evaluated by the corrected points. This, however, seems to be a result of two errors, namely not taking  $N = \beta$  and not correcting, acting in opposite directions. In case No. 7, in which  $\beta > N_{\text{given}}$ , we got as expected  $\beta > N_{\text{eval}} > N_{\text{given}}$ . The fact that  $N$  evaluated by the corrected points and assuming exponential centre behaviour is only 25% higher than  $N$  given seems to be due to an exponential behaviour of the reservoir which deviates very little from linearity. The much higher value of the

N evaluated linearly indicates that the centre is farther from linearity. In the results on line 6, the fact that N evaluated using the exponential function is 20% off the given value (the results found using the linear extrapolation or the points uncorrected for quenching were even worse) points to the insensitivity of the method when very high doses are involved, i.e. when either the reservoir or the centre (or both) is close to total saturation right after the first (natural) irradiation.

TABLE 1. GIVEN AND EVALUATED DOSES IN NORWEGIAN  $\alpha$  QUARTZ.

Sample	$N_{\text{given}}$ (rad)	$\beta$ (rad)	$N_{\text{eval}}$ exp. corr.(rad)	$N_{\text{eval}}$ exp. uncorr.	$N_{\text{eval}}$ lin. corr.	$S_N/S_0$	
1	7,25	50	50	55		10.5	
2	7,25	100	100	105	117	12	
3	7,25	100	100	100	108	15	
4	7,25	200	200	200	300	30	
5	7,25	300	300	300	390	36.9	
6	7,25	1000	1000	1200		53.5	
7	7,25	50	150	63	70	90	11.2
8	7,25	150	50	80	135		9.5
9	7,25	250	200	250	240		12
10	7,25	300	100	150	200	190	21
11	7,25 (refired, 1000°C)	100	100	105	110		6.6
12	5,25	100	100	108			1.7
13	5,25	200	200	190			1.8

The results with the 5,25 (high  $S_0$ , low sensitization) sample are not very clear due to the relatively high  $S_0$  and to the fact that we do not know whether we should extrapolate to the measured  $S_0$  or to some reduced value of  $S_0$ . The result of No. 11 is representative of both kinds of samples, pre-irradiated and not irradiated, refired at 1000°C. Probably, due to the low value of  $S_0$ , the result here is quite good.

Preliminary measurements were done on archaeological samples. The results with the 156C1 sample — a brick sample from Fulham — are satisfactory. Using increments of 60 and 70 rads results in evaluated N values of 62 and 65 rads respectively, whereas using bigger increments results in a higher extrapolated N of 80 rads. We conclude that the equivalent dose is about 63 rad. The dose rate for this sample given by Fleming (1973) is 0.233 rad/yr. (We repeated the dose rate measurement and got practically the same result.) This yields an age of 270 years as compared to the archaeologically known age of 290-300 years and to Fleming's (1973) results of 320 ~~rad~~ <sup>year</sup>. Preliminary measurements on older samples yielded too low values.

### 5. Discussion

In the present paper, the sensitization behaviour of quartz by  $\beta$  or  $\gamma$  irradiation followed by activation at  $\sim 500^\circ\text{C}$  has been further investigated. In particular, the possibility of saturation behaviour of both the reservoir and the luminescence centre have been taken into account; the pre-dose method using the sensitization as a measure of the archaeological dose, is modified to account for the various possibilities. As opposed to the case of TLD in which one can choose the material used and calibrate it beforehand, in archaeometry, we have to use the type of quartz available and make the calibration after the natural dose  $N$  was given. It is evident that once either the reservoir is in complete saturation by the natural dose or the luminescence centre is entirely in saturation by the activation following the natural dose, no information regarding the equivalent dose can be extracted from the measurements. What one can get, at most, is a lower limit to the possible equivalent dose. It is of importance, however, to be able to extract information in the range in which either the reservoir or the centre or both are not behaving linearly provided that none of them is entirely in saturation. The procedure to evaluate the equivalent dose (E.D.) was described above. Table II summarizes the various possibilities.

TABLE 2. THE PRE-DOSE EFFECT AS MEASURED AT VARIOUS CASES.

	Reservoir			
		linear	exponential	saturated
Centre				
Linear		Straight line, correct result of $N$ irrespective of $\beta$ . (even if different $\beta$ 's are used in one series)	Straight line, correct result only for $N = \beta$	Straight line yielding $N = \beta$ irrespective of real value of $N$ .
		$q = p$	$q > p$	$q > p = 0$
Exponential		Exponential curve, correct result of $N$ irrespective of $\beta$ (even if different $\beta$ 's are used in one series).	Exponential curve, correct result only for $N = \beta$	Exponential curve yielding $N = \beta$ irrespective of real value of $N$ .
		$q = p$	$q > p$	$q > p = 0$
Saturated		No result. $q = p = 0$	No result. $q = p = 0$	No result. $q = p = 0$

The method suggested in this paper, is expected to give not only the correct natural dose  $N$  in the cases in which this is possible in principle but also gives all the necessary information regarding the reservoir and centre filling. Thus, a straight line means linear filling of the centre, exponential curve means exponential filling of the centre and no change in sensitivity with incremental dose  $\beta$  means centre saturation. As for the reservoir, if the evaluated value of  $N$  (in the linear



or exponential case) is independent of  $\beta$ , we can conclude that the reservoir is being filled linearly. If we get different values of  $N$  for different values of  $\beta$  and can find a case in which  $N = \beta$ , the reservoir is being filled non linearly, probably exponentially. If we get  $N = \beta$  for any  $\beta$  we choose it means that the reservoir is in saturation. If there is no change in the sensitivity with incremental dose, we can deduce that the centre is in saturation, but cannot say anything about the reservoir.

The question has been posed of whether the quenching correction should be applied when one is trying to fit the points to the exponential law. Although the model points to the use of quenching correction, we have tried the use of the uncorrected points as well. Certainly, another curve was found on the semilog scale, but surprisingly it sometimes extrapolated to, more or less, the same value of  $N$ . In general, however, the results were better with the corrected values, both with respect to linearity (on the semilog scale) and to the evaluated  $N$  as compared to the given dose in the BDH quartz.

The question of the complications when  $S_0$  is relatively high has already been mentioned. As a rule of thumb, one can say that the results might be doubtful when  $S_N/S_0 < 4$ . The question which is the "real"  $S_0$  to be used should be investigated further.

The fact mentioned above, that firing to  $\sim 1000^\circ\text{C}$  of the 1 krad irradiated or unirradiated BDH quartz of both kinds resulted in samples showing the same behaviour, is to be noted since it means that firing of the clay probably erased the influence of any previous irradiation or firing. Unfortunately, one has still to cope with quartz samples having different amounts of impurities and defects.

It has been found in some of the samples that heating to  $500^\circ\text{C}$  and cooling immediately did not activate completely the sample. Holding the sample for 3 minutes at  $500^\circ\text{C}$  was found to complete the activation, and it is recommended to do this routinely. The pre-dose effect of  $\beta$  irradiation was compared experimentally to  $\gamma$  and  $\alpha$ ; the  $\gamma$  radiation was found, as expected, to give the same sensitization as the  $\beta$  per rad. The  $\alpha$  irradiation was found (see also : Thompson, 1970) not to contribute much to the pre-dose effect. Another word of caution is regarding the  $\beta$  test dose used to check the various sensitivities which should be as small as possible. We used a test dose of 1 rad all along. When the doses involved were 200 rads and more, the contribution of the test dose to sensitization was negligible. In the 156C1 sample, where  $N \sim 60$  rad, the influence of the two test doses given prior to activation in each step, was taken into account. The low equivalent dose found in older archaeological samples seems to indicate a new kind of anomalous fading (Wintle, 1973), this time of the sensitivity rather than of the accumulated TL. This effect should be studied further. It is to be emphasized, however, that the results found by linear extrapolation were worse in these cases (as well as in the other cases) and, therefore, the exponential extrapolation, advocated in this paper, should be considered as a step forward, although this effect of "old" samples is not yet clarified.

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## APPENDIX : AMENDMENT OF THE THOMPSON MODEL

According to Thompson's model, the sensitivity of the sample is a measure of the concentration of holes in the luminescence centre. Thus, the more holes we have in the centre, the more TL we should expect to get for a given test dose. On the other hand, it is known that when the sensitivity is given, the TL response is proportional to the test dose. This is incompatible with the following argument. If  $n_0$  is the initial concentration of electrons in traps and  $m_0$  the initial concentration of holes in centres, the maximum TL intensity and the area under the curve would not depend on the product of the two,  $n_0 \times m_0$ , but rather on the smaller of the two,  $\min(n_0, m_0)$ . This can be briefly proved as follows :

Since  $I = -dm/dt$  (there is a proportionality factor which has been omitted) one has  $m_0 - m_\infty = \int_t^\infty I dt$ . If  $m_0 < n_0$ , the peak terminates because  $m$  is exhausted, therefore  $m_\infty = 0$  and thus

$$m_0 = \int_t^\infty I dt = S \quad (A1)$$

where  $S$  is the total area under the glow curve.

If, however,  $m_0 > n_0$ , the peak terminates because  $n$  is exhausted, thus we remain with  $m_\infty = m_0 - n_0$  and therefore

$$S = \int_t^\infty I dt = m_0 - m_\infty = n_0 \quad (A2)$$

Equations (A1) and (A2) can be summed up as :

$$S = \min(n_0, m_0) \quad (A3)$$

As shown by Kristianpoller et al. (1974), the TL maximum intensity  $I_m$  is practically proportional to the area  $S$  under the curve, even in rather complicated cases.

Rodine and Land (1971) suggested a model in which the TL intensity is proportional to both the initial concentration of electrons in traps and holes in centres due to the effect of an additional postulated trap, the retrapping into which is more efficient than both retrapping into the initial trap, and recombination. The idea was further investigated by Kristianpoller et al. (1974) who found the exact conditions leading to the dependence on both  $n_0$  and  $m_0$ . Although Rodine and Land and Kristianpoller et al., introduced the additional trap in order to explain

superlinearity in which  $S \propto D^2$ , it seems that the assumption of the additional competing trap is necessary in order to make Thompson's model compatible with the experimental evidence. Otherwise, one should say on one hand that since the sensitivity reflects the value of  $m_0$ , we must have  $m_0 < n_0$ , and on the other hand, since the TL intensity is proportional to the test dose, we must have  $n_0 < m_0$ .

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