Chapter 4 Appendix 9

THERMOLUMINESCENCE RESULTS ON SLICES FROM A HIROSHIMA TILE UHFSFT03

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As was reported at the May 1984 Utah thermoluminescence (TL) workshop, high fired tiles and porcelain fragments can be sliced into 200 μm sections with constant surface area. When conventional pre-dose measurements were carried out on these slices the doses evaluated were in good agreement with results obtained by other workers using conventional quartz separation techniques.[1]

There are several advantages in using slices. First, less sample is needed as about 50 consecutive slices can be cut from a block measuring typically 1 cm^2 cross section and 2 cm in length. There are no problems with securing grains to the plate or loss of grains during measurement. Hypothetically there is less damage to the grains when they are cut slowly under cold water than when they are crushed. The disadvantage is that other minerals besides quartz are present in the slice and the signal is weaker than that obtained using quartz inclusions.

Preparation

A cuboid was cut from front to back of the tile. The front 2 mm and outer surfaces were removed leaving a portion that was 1 cm^2 cross section and 2 cm long. This was cut into about 50 slices of 200 μm thickness using a microslice.[1] The slices were washed in acetone and dried before use.

Measurements

Measurements were made on a standard TL set (Alldred design) using an EMI 9635 QA photomultiplier tube fitted with a Corning 7-59 filter. On-plate irradiations were made using a 40 mCi ^{90}Sr beta-particle source. The heating rate was 10 °C/s. Natural and artificially induced TL signals are shown in Figure 1.

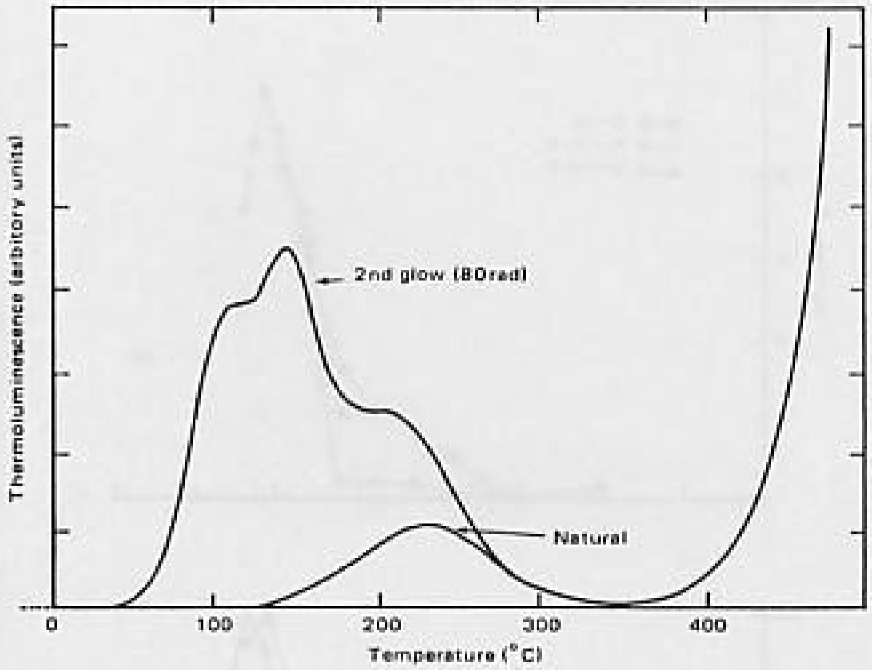

Figure 1. Glow curves obtained from a 200 μm slice of tile UHFSFT03

The natural TL has its maximum between 210 and 225 °C. It is mainly due to the 210 °C peak, which will have thermally faded over the intervening 40 years. The maximum peak height is around 4.5 kHz. In the artificial TL there is a double peak compounded of one peak at 110 °C and the other at 145 °C. In all pre-dose measurements, only the peak height at 110 °C was recorded.

Pre-dose Measurements

Thermal activation characteristics (TAC) are shown in Figure 2. Test doses of 2.6 rad were used throughout the work and the sample heated to each annealing termperature without holding. Three main problems presented themselves:

High S_o. S_o/S_N was between 0.3 and 0.6 (generally the ratio was around 0.4). To check whether substraction of S_o was valid, the procedure laid down by Bailiff[2] was followed. After completion of measurements on each sample, the slice was placed under an ultraviolet light source for periods between 10 minutes and one hour. In all cases the signal was reversed to the original S_o value, so it was assumed that this value could be safely used in calculations.

Low Sensitivity. S_N was on average 1.5 kHz using a test dose of 2.6 rad and heating rate of 10 °C/s.

Temperature for Maximum Activation. There was difficulty in determining the best annealing temperature for maximum activation. To obtain the greatest accuracy it is essential that maximum activation be achieved for each measurement (in particular when there are

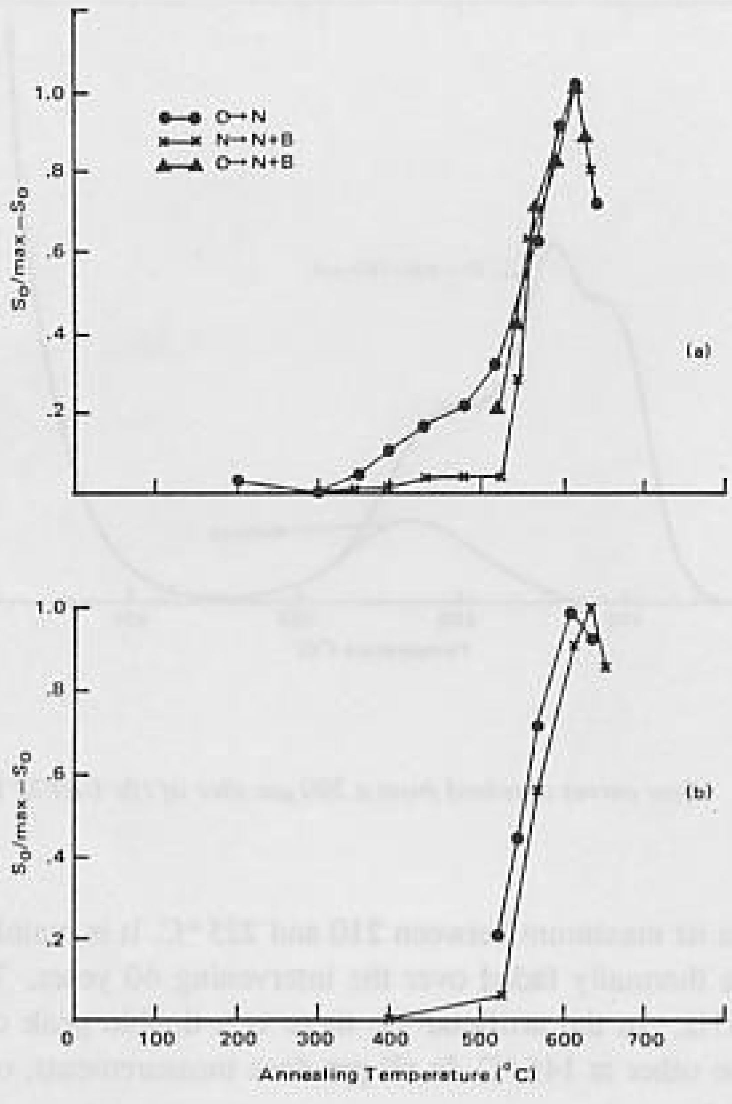

Figure 2. Tile UHFSFT03. (a) TAC. (b) TAC showing 25°C displacement

the two problems detailed above). As described previously,[1] the thermocouple that records the plate temperature as well as driving the ramp is below the nichrome plate. The slice is 200 μm thick and a good heat insulator. In addition, it is reasonable to suppose that most of the signal comes from the surface of the slice, which in this case is dark gray in colour. It is estimated that there is a 30 to 40 °C temperature difference between the surface of the slice and the plate bottom at 630 °C. The thermocouple reading for maximum activation was determined from a single TAC (this proved correct for one series of measurements); but, when these were repeated two days later, it was found to be too high and deactivation occurred. This can be seen from the TAC shown in Figure 2b where there is an apparent constant 25 °C difference between them and, in one deactivation, occurs 25 °C earlier in one than the other.

Multiple Activation

Four evaluations of the dose were made for different values of the beta particle calibrating dose (Figure 3). The accrued dose is calculated from:

$$Dose = \frac{S_N - S_o}{S_{N+\beta} - S_N}\beta \qquad (1)$$

Linearity was observed for an additional dose of up to 100 rad on top of the natural: Mean accrued dose = 87 ± 7 rad.

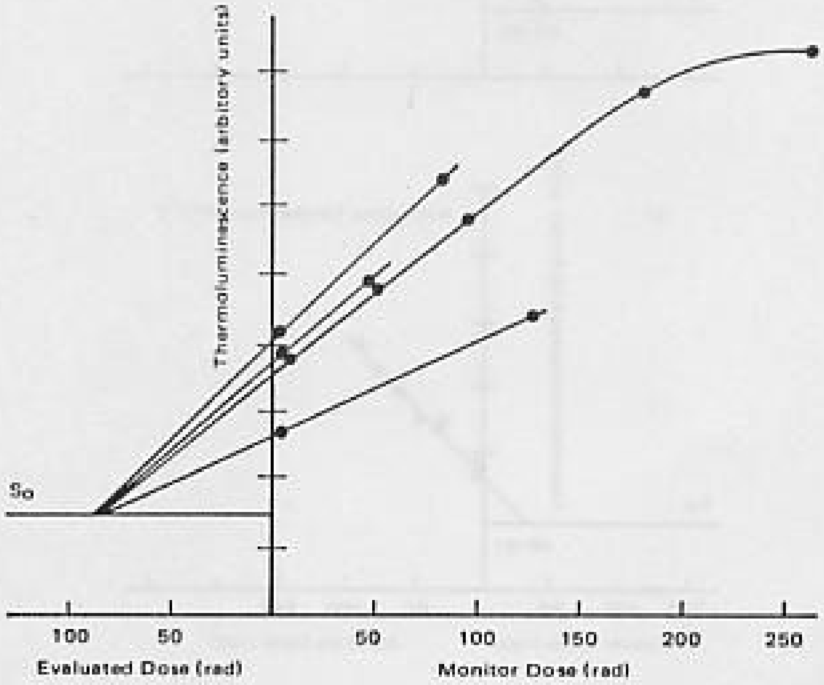

Figure 3. Pre-dose multiple activation for the UHFSFT03

Additive Dose

Two problems presented themselves. The problem of maximum annealing temperature required to obtain maximum activation has already been mentioned. Two evaluations were made on different days using different annealing temperatures (Figures 4a and b).

The second problem was that of normalization between different slices. This was achieved in the following manner. Each slice was glowed to 315 °C so that there was a record of the 200 to 300 °C TL. Slices with the same shape glow curve and peak height within 10% of the mean were selected. S_o was measured using a 2.6 rad test-dose. All S_o values were within 18% of the mean and these were used for normalizing $S_{N+\beta}$. The annealing temperatures were 615 and 595 °C. Although reasonable straight line fits were obtained for additional doses up to 100 rad in both cases, it will be noted that two different equivalent doses (ED) were obtained.

The ED corresponding to an annealing temperature of 595 °C (Figure 4a) was higher than that corresponding to a temperature of 615 °C (Figure 4b). There are reasons to believe that the ED in the second case is lower than it should be because some deactivation occurred. In case (a) care was taken to ensure that deactivation did not occur by repeating each measurement 5 °C higher: Accrued dose (a) = 83 ± 20 rad; Accrued dose (b) = 46 ± 7 rad.

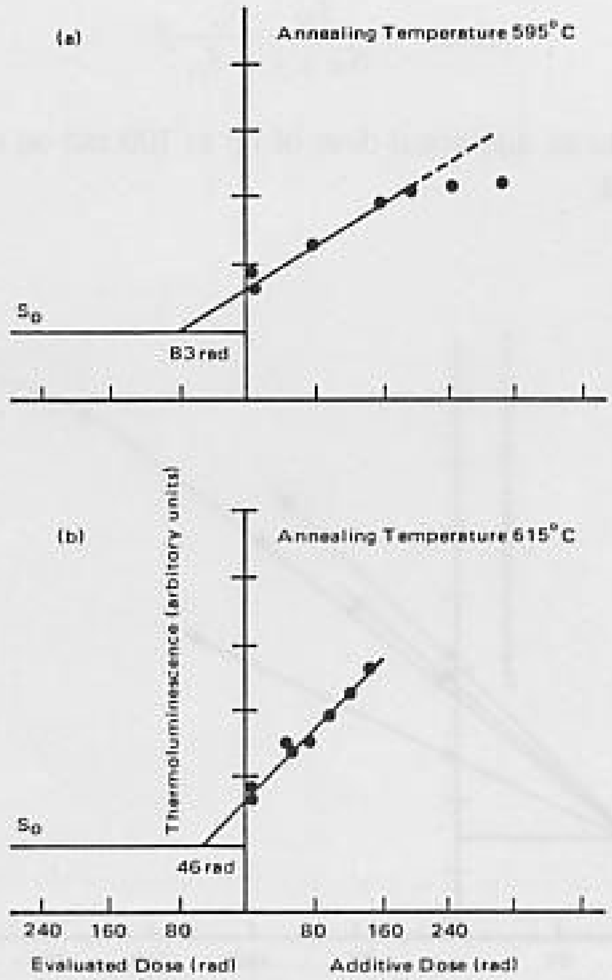

Figure 4. Pre-dose: additive dose for tile UHFST03. (a) annealing temperature 595°C. (b) annealing temperature 615°C

High Temperature Analysis

The advantages of analyzing between 200 and 275 °C is that the TL signal is brighter than the pre-dose signal. In addition one is free from the problems of high S_o values, maximum activations, and normalization. The major problem is that of thermal fading.

The mean life of the 210 °C peak has been put at times from a few hundred to a few thousand years[3,4] depending on the ambient temperature. It is highly sensitive to temperatures above 20 °C (the range encompassing the maximum summer temperatures in Hiroshima is around 26 to 38 °C). Ichikawa and Nagatomo[5] estimate that at the peak about 20% fading occurred in 40 years.

Three ordinates were chosen (225, 250, and 275 °C) and growth curves plotted (Figure 5). Linear growth was observed and three different ED obtained.

Fading. Current work at Oxford by Templer[6] indicates that TL in the dating plateau of zircon consists of a stable and an unstable component. Decay of the unstable component gives rise to what is known as anomalous fading. The natural TL contains only the stable component as all the unstable portion has decayed over the archaeological lifetime. Artificial irradiations (either second glows or first glows consisting of natural plus artificial TL) contain

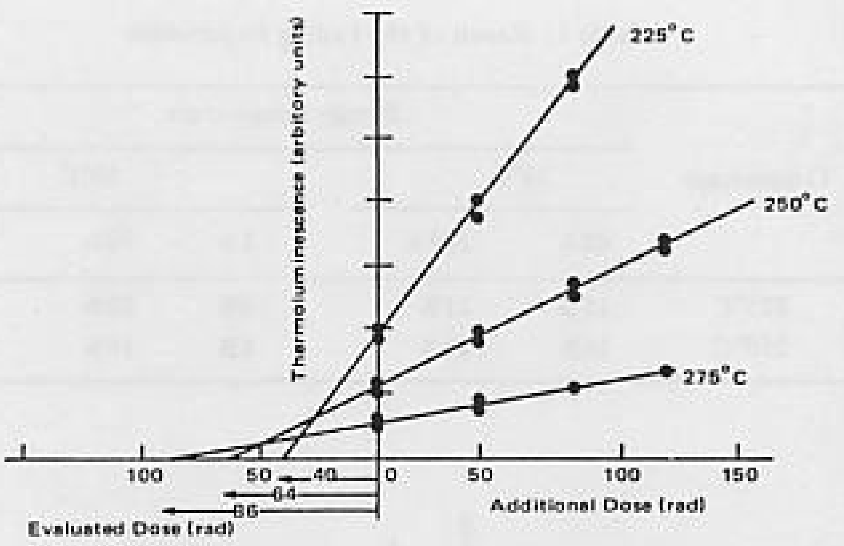

Figure 5. Accrued dose estimates at three different temperatures uncorrected for fading. Tile UHFSFT03

both the stable and unstable components - hence the evaluated dose will be too low. Work is in progress to show that some feldspars behave similarly to zircon. As yet anomalous fading has not been reported in quartz.

Templer[6] showed that the unstable component can be removed by heating and the higher the holding temperature, the more rapid the decay. Furthermore a plateau level is reached where all the unstable component has decayed and only the stable component is left. The plateau level is the same whatever the holding temperature. The holding temperature should be carefully selected so as not to be so high that it erodes some of the stable component.

The experimental procedure involves choosing various holding temperatures and measuring the amount of TL remaining in artificially irradiated samples after varying lengths of time until no further decay is observed. The signal then consists only of the stable component. If the stable TL is a fraction f of the total signal,

$$ED = \frac{N}{[(N + \beta) - N]\,f}\beta \qquad (2)$$

N consists only of the stable component. $[(N+\beta) - N]f$ is the stable part of the artificial irradiation.

An attempt was made to obtain some quantitative estimate of the fading. This was achieved in the following manner.

There are strong indications from previous work (unpublished) that the 210°C peak shares the same luminescent center as the 110°C peak. Hence this peak pre-doses under the same conditions as the 110°C peak. However, it was ascertained that if each sample was only heated to 310°C, pre-dosing did not occur and it was possible to use successive beta particle doses of 80 rad for monitoring.

Slices were drained of their natural TL to 310°C, cooled to room temperature and given

Table 1. Result of the Fading Experiment

Temperature	Storage temperature				
	38°C		50°C		
	45 h	167 h	1 h	70 h	168 h
225°C	15%	21%	6%	20%	28%
250°C	15%	20%	6%	16%	20%

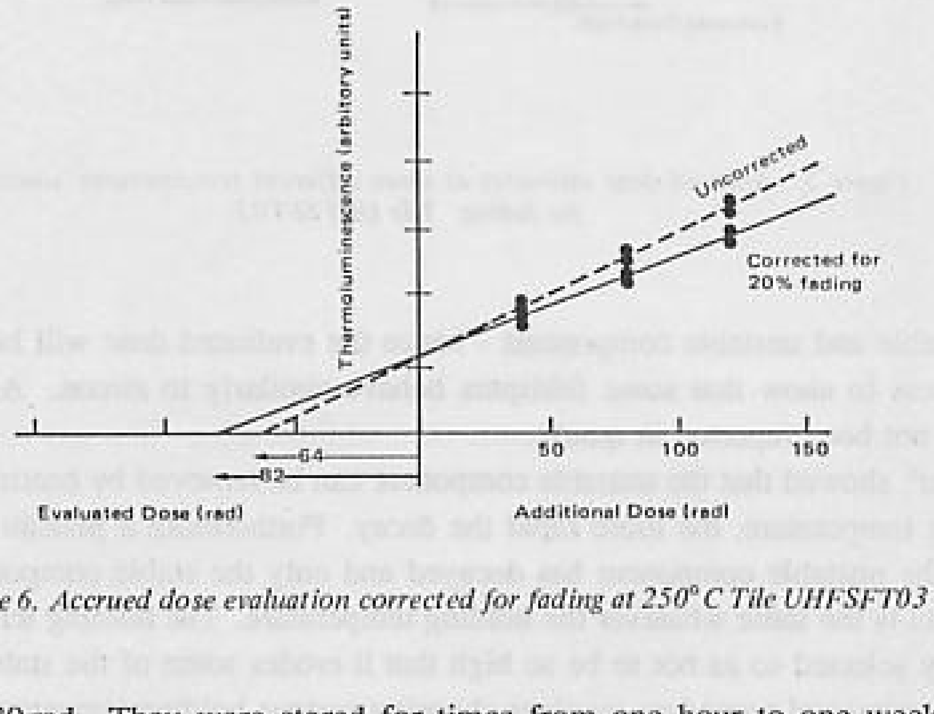

Figure 6. Accrued dose evaluation corrected for fading at 250° C Tile UHFSFT03

irradiations of 80 rad. They were stored for times from one hour to one week at 38 and 50 °C. After storage, the slices were glowed to 310 °C to measure the TL intensity (I_s). Next the slices were cooled, given a monitored dose of 80 rad, and immediately glowed to 310 °C to measure the TL intensity (I_i).

The percentage by which I_s/I_i falls below unity is shown as a function of storage temperature and time in Table 1. The light level at 275 °C was low and although there was probably some fading at this temperature, it was within the limits of the accuracy of TL measurement (around 5%). With no fading correction, the accrued dose is 87 ± 9 rad.

At 250 °C, stability appears to be reached at ambient temperatures of 38 and 50 °C after the same holding time, both reaching a level of 20% fading. However, data is not precise enough to reach an unambiguous conclusion. At 225 °C, signal loss is both greater and more rapid, especially when held at 50 °C.

Assuming little or no further loss occurs beyond 20% at the 250 °C TL ordinate, the fading can be corrected. An accrued dose of 82 ± 4 rad was evaluated (Figure 6). The error quoted is the random error of measurement only.

Results and Discussion

The results are shown in Table 2. All doses are doses to quartz and are in rad. Only

Table 2. Results of Thermoluminescence Measurements

Method	Accrued dose (rad)	Number of data points
Pre-dose MA	87± 7	4
Pre-dose		
AD (595°C)	83±20	5
AD (615°C)	46± 7	8
250°C (with 20% fading correction)	82± 4	8
275°C (no account of fading)	87± 9	6

random errors are quoted; these are based on least squares fits.

There is good agreement for four of the five evaluations - only the additive dose evaluation (615 °C) is much lower than the mean. Although in principle, the last two results should be regarded as minimum accrued doses, they are in line with each other and the preferred pre-dose measurements. Of the five results, it is considered that the most reliable are the multiple activation and the 250 °C analysis, although both have their limitations. In the case of multiple activation there is the risk of underestimating the dose because of sensitization of R-traps after the first heating.[7] However, previous work with porcelain slices indicated that multiple activation was more reliable than additive dose[1] and gave results that agreed well with those obtained using more established TL methods.

With the 250 °C analysis one cannot be sure that there will not be further fading, but more fading experiments are in hand and slices are being stored at ambient temperatures of 38 and 50 °C for one month and one year to see if the fading has really stabilized. It should be noted that there was no indication of a decrease in dose through the 1 cm thickness of the tile.

References

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