Chapter 4 Appendix 8

CONVENTIONAL THERMOLUMINESCENCE CHARACTERISTICS OF A HIROSHIMA TILE AND A NAGASAKI BRICK

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Previous work on four tiles from Hiroshima, which was reported at the 1984 thermoluminescence (TL) workshop,[1] showed that the fine grain method was not applicable to this material because of problems of spurious TL and nonreproducibility. The work on quartz inclusions did not give meaningful results because sufficient quartz could not be extracted from the small samples initially supplied. For this study larger samples were supplied, and it has been possible to extract sufficient quartz to apply the standard pottery techniques of inclusion dating using etched quartz grains.[2]

Sample Preparation and Method

The outer 0.2 cm of the sample was removed using a diamond impregnated wheel saw and sections approximately 2 × 2 × 3 cm were crushed in an enclosed pestle and mortar (piston and cylinder type, cylinder internal diameter 2.65 cm) using a hand operated hydraulic press (Type Apex P3010/4, with a 4.45 cm diameter ram set to a maximum pressure of one tonne). Grains in the range 90 to 150 μm were separated by seiving, washed in analar acetone, dried at 50°C and passed through a Franz magnetic separator. The resulting nonmagnetic fraction was then etched for 40 minutes in 40% hydrofluoric acid. It was then repeatedly washed in distilled water, methanol, and analar acetone, and finally dried at 50°C. Quartz grains in the range 90 to 125 μm were then selected for analysis.

The standard Oxford Laboratory TL set was used for the analysis,[3] the photomultiplier (Type EMI 9635QA) being fitted with a blue-pass 5-60 Corning filter and a Chance Pilkington HA3 filter to reject the infrared. The heating rate used was 5 to 500°C/s. The glow oven was evacuated to 0.01 Torr before high purity argon (<2 ppm oxygen) was introduced. All the irradiations used a 40 mCi ^{90}Sr plaque source (Type SIP from the Radiochemical Centre, Amersham) in a housing with a hand operated shutter and they were done "on plate". The samples were normalized by weighing, the average weight being 2.5 mg, spread in a monolayer on 1-cm diameter stainless steel disks, and stuck down with silicone oil (Willy Rusch-Silkospray). The plateau region of the glow curve usually extended for 50°C, spanning the region of the glow curve in which the 375°C peak is to be expected and the accrued dose deposited from natural plus bomb radiation was evaluated in this region using the normal additive dose method. There were usually 16 samples of accrued (i.e., natural + bomb) dose, and these were then used for the second glow growth curve, and there were 16 samples of (accrued + additonal beta-particle doses). The growth curve was pursued to an added beta-particle dose of twice the accrued dose and the light level then was three times the accrued dose light level. Tests on stored (accrued dose + beta-particle dose) material were used to check for the presence of anomalous fading.[4] The material was stored for six weeks and no fading was found (<3%).

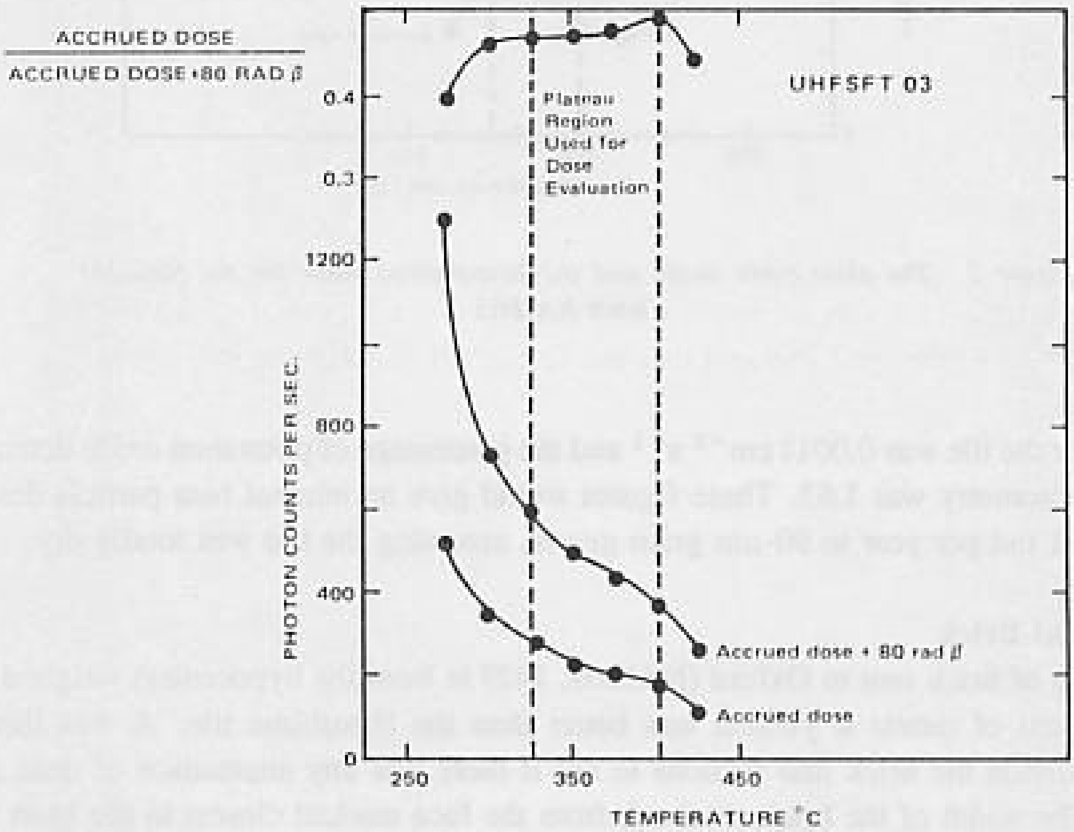

Figure 1. The glow curve shape and the beta-plateau shape for the Hiroshima tile UHFSFT03

The Hiroshima Tile

The portion of tile sent to Oxford (UHFSFT03, 1433 m from the hypocenter) weighed 950 g and from this amount of sample 105 mg of etched quartz was produced for analysis. The glow curve shape and the beta-particle plateau shape are illustrated in Figure 1.

For this material the accrued dose, made up of equivalent dose (ED = 75 rad) and second glow intercept (I = 9 rad) was found to be 84 ± 10 rad. The ratio of the first glow growth curve slope to the second glow growth curve slope was 0.9. The thick-source alpha particle

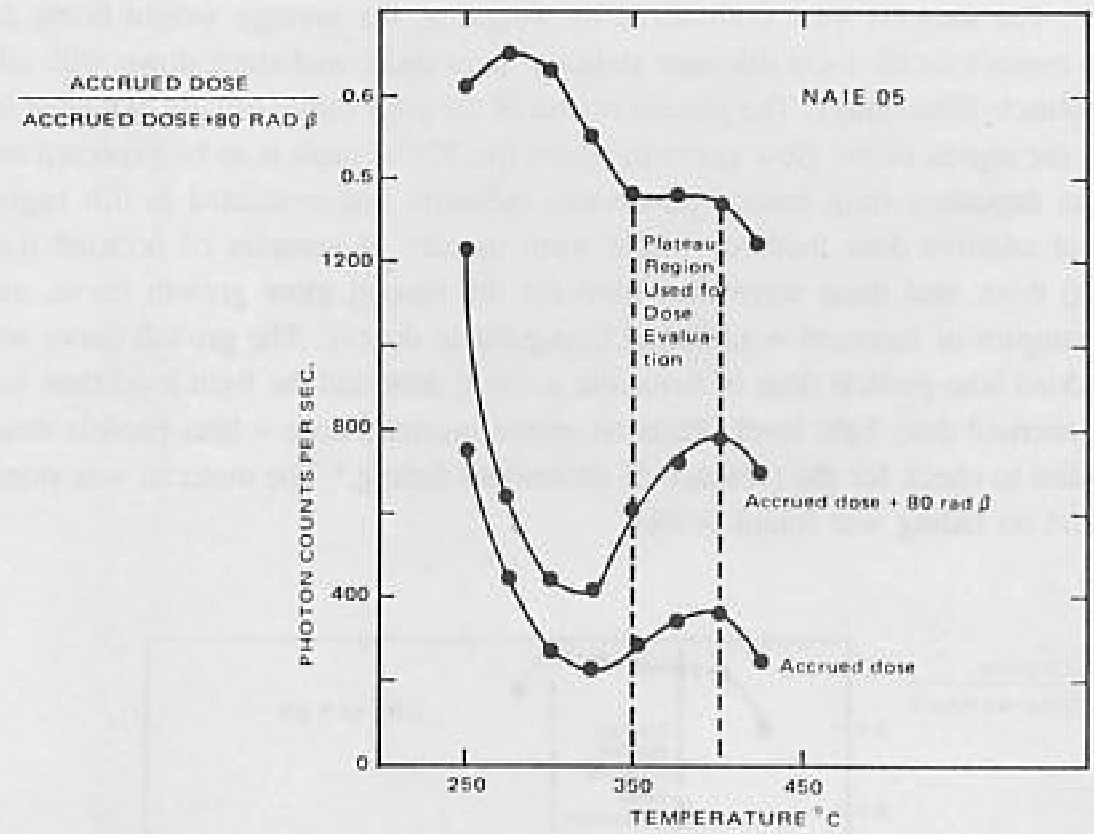

Figure 2. The glow curve shape and the beta-plateau shape for the Nagasaki brick NAIE05

count rate for the tile was 0.0011 cm^{-2} s^{-1} and the percentage of potassium oxide determined by flame photometry was 1.63. These figures would give an internal beta-particle dose rate of about 0.21 rad per year to 90-μm grain quartz, assuming the tile was totally dry.

The Nagasaki Brick

The piece of brick sent to Oxford (NAIE05, 1427 m from the hypocenter) weighed 925 g but the amount of quartz it yielded was better than the Hiroshima tile. It was therefore possible to divide the brick into sections to see if there was any attenuation of dose across the brick. The width of the brick received, from the face marked closest to the blast to the perpendicularly opposite face was 11 cm. After removal of the outer layer it was divided into three slices A, B, and C, 3.5 cm wide, A being closest to the blast and C farthest away.

The glow curve shape is shown in Figure 2. The beta-particle plateau shape, glow curve shape, and plateau were similar for all pieces of the brick. The results are given in Table 1. The thick-source alpha particle count rate for the brick was 0.0008 cm^{-2} s^{-1}, and the percentage of potassium oxide determined by flame photometry was 1.2. These figures yield an internal beta-particle dose rate of about 0.15 rad per year to 90-μm grains in the brick and an infinite-matrix gamma-ray dose of 0.13 rad per year, assuming the brick was totally dry.

Calibration Measurements

Some material extracted from Japanese samples and drained at the University of Utah was

Table 1. Results for Nagasaki Brick NAIE05

Sample	Weight[a] (mg)	ED + I (rad)	Accrued Dose (rad)	S_1/S_2[b]
A	80	74 ± 21	95 ± 10	0.84
B	90	60 ± 20	80 ± 9	0.81
C	95	47 ± 26	73 ± 8	1.00

[a]Weight of quartz produced for analysis.
[b]The ratio of first to second glow growth curve slopes.

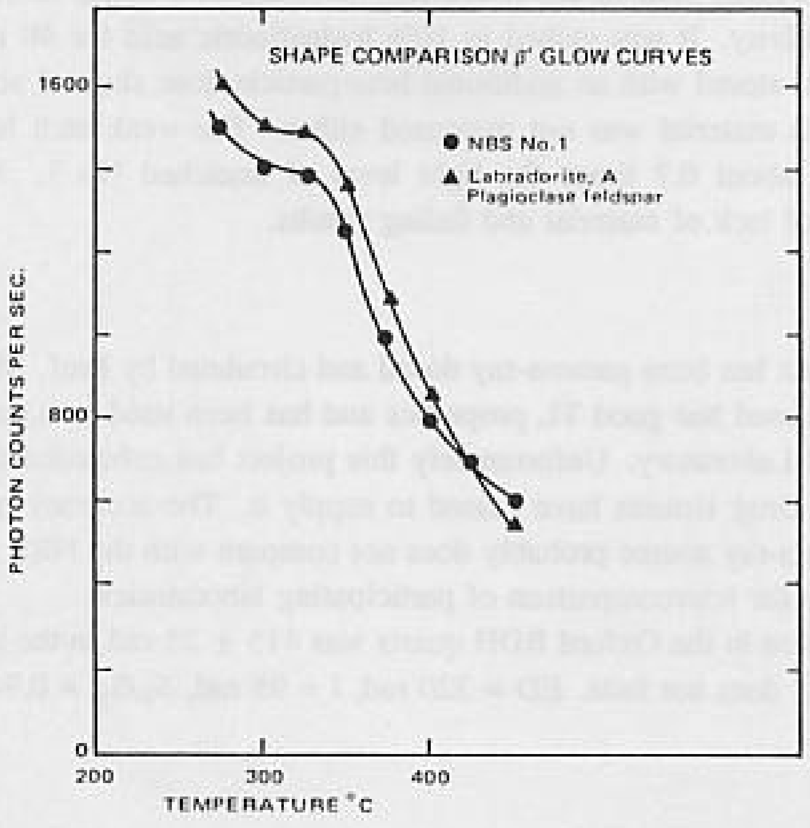

Figure 3. Comparison the glow curves for an NBS sample and for labradorite, a plagioclase feldspar

given three known but undisclosed gamma-ray doses by the National Bureau of Standards (NBS No. 2 to 4) and sent as a blind test for calibration purposes, along with an undosed portion (NBS No. 1). Dose measurements both in the unetched material and after normal etching treatment were requested.

Initial measurement on this unetched material indicated doses in the region of approximately 10 rad for No. 2, 15 rad for No. 3, and 35 rad for No. 4. However, examination of the glow curves suggested possible feldspar as well as quartz components (Figure 3). The glow curves were not clean in the high temperature region either, sometimes the 325°C quartz peak was evident and sometimes it was not. The light levels were about five times higher for this untreated material than for the etched quartz from Hiroshima and Nagasaki. Fading tests on the unetched, undosed material (No. 1) of stored beta-particle doses given in Oxford indicated that the material faded by 15% over a six-week period, so any dose evaluated on unetched material is meaningless.

NBS No. 2 and 4 were then given a 40-minute etch in 40% hydrofluoric acid, which removed most of the sensitivity, bringing it into line with the previously measured etched quartz. The very low dose in No. 2 meant that it was not accurately measureable by the previously described method because the light level was too low. NBS No. 4 was measured and the result obtained was ED = 32.5 rad, I = 1.5 rad, total 34 ± 8.5 rad. $S_1/S_2 = 0.7$. Fading tests on stored (etched No. 4 + additional beta-particle dose) showed no fading (<3%) over a six-week period.

After discussion with Dr. Haskell about the impossibility of measuring etched No. 2, NBS No. 3 was given a weaker etch to see if the fading component could be removed without destroying the sensitivity. It was etched in 10% hydrofluoric acid for 40 minutes. Fading tests on this material stored with an additional beta-particle dose showed about 15% fading over a week, so this material was not measured either. The weak etch had not removed much light; it was about 0.7 times the light level of unetched No. 3. No other etches were tried because of lack of material and fading results.

Oxford Quartz

Some more quartz has been gamma-ray dosed and circulated by Prof. M. J. Aitken from Oxford. The quartz used has good TL properties and has been used as a standard for many years in the Oxford Laboratory. Unfortunately this project has exhausted the stock held in Oxford and British Drug Houses have ceased to supply it. The accuracy of the calibration of the Oxford gamma-ray source probably does not compare with the NBS sources, but this material is adequate for intercomparison of participating laboratories.

The dose measured in the Oxford BDH quartz was 415 ± 25 rad in the high temperature region. The material does not fade. ED = 320 rad, I = 95 rad, $S_1/S_2 = 0.94$.

Conclusions

Nagasaki and Hiroshima. If sufficiently large initial samples are available there is no difficulty in measuring the extracted quartz sample by conventional TL methods in the high temperature region. The problem is the initial extraction of the quartz as the yield is low and the material is often high fired. The process is both laborious and very time consuming. The yield of quartz from the two different types of material supplied for this analysis is worth noting, the brick yielding two and a half times more quartz.

The Calibration Quartz. The material circulated for calibration by the University of Utah in an unetched or lightly etched state was found to fade so it was not suitable for the calibration. If it was fully etched, then because of the low dose given to it there was too little light for measurement, and doses of 100 rad or more would have been a more suitable choice.

The calibration experiment should be repeated using quartz which has good TL properties, of which there is a large stock. Values at least commensurate with those of the accrued doses in the Hiroshima and Nagasaki material and preferable larger should be selected for the gamma-ray doses, and they should be given by sources such as are available at NBS.

The fine detail of interlaboratory calibration should not be sought at the expense of the high dose-rate experiment, which should have preceded all these measurements.

References

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