Chapter 4 Appendix 3

THERMOLUMINESCENCE MEASUREMENT OF GAMMA RAYS

BY THE QUARTZ INCLUSION METHOD

Yoneta Ichikawa, Tsuneto Nagatomo
Nara University of Education

Masaharu Hoshi
Hiroshima University

Since reassessment of atomic bomb radiation doses was proposed, efforts have been made to obtain accurate gamma-ray doses with thermoluminescence (TL) methods using ceramic samples. TL methods were also used in the 1960s to estimate gamma-ray doses at ground distances within 1000 m from the Hiroshima and Nagasaki hypocenters. The TL methods used are essentially the same as the techniques applied to the dating of ancient pottery excavated from archaeological sites. In the two decades after the 1960 estimates, the accuracy of TL dating has been greatly improved by the use of the quartz inclusion technique. TL dosimetry of gamma rays from the A-bombs in Hiroshima and Nagasaki by the quartz inclusion method is reported.

Materials and Methods

Samples. Discrepancies between new calculated estimates of dose and the T65D estimates are significant in the range of ground distances beyond 1000 m. Therefore, gamma-ray dosimetry using samples collected beyond 1000 m is essential to certifying that the new dose values are reliable.

TL samples should satisfy the following conditions: (1) there should have been no shielding objects between the sample and the A-bomb epicenter; if the sample was not directly exposed to the A-bomb radiation, the shielding conditions should be known, (2) the

The authors appreciate the help of Messrs. S. Nagata and I. Ueda of the Osaka Branch of the National Electro-Technical Institute in irradiating with calibrated ^{60}Co gamma rays.

sample should not have been heated since the kiln firing, and (3) the date of the kiln firing of the sample or the age of the building should be known. The samples used were as follows:

Hiroshima
Tile from the I-shaped building of Hiroshima University Faculty of Science (H1 to H5
Tile from the E-shaped building of Hiroshima University Faculty of Science (H6 to H11 and No. 4)
Tile from Naka Telephone Office
Tile from Chugoku Electric Co. Building
Tile from Chokin-kyoku (Postal Saving Bureau)
Japanese tile from a Japanese house in Nobori-machi

Nagasaki
Brick from Ieno-cho wall
Tile from Japanese house in Ieno-cho
Ceramic sherd from Nishi-machi
Brazier from Shiroyama-cho

In order to check for discrepancies between the experimental values obtained by the different measuring groups, samples which had been systematically treated and irradiated were distributed to several laboratories. The samples were a Nagasaki Ieno-cho brick sample, US National Bureau of Standards (NBS)-irradiated samples (No. 1 to 4), Hiroshima University tile sample (No.4), and an Oxford quartz sample.

Sample Preparation. The 2-mm thick layer of the front surface and 1-mm thick layer of the underside of the tile were removed using a diamond saw. Then the tile was crushed into grains by a rolling jaw crusher. The crushed grains were sieved into three groups with sizes of 100 to 200 mesh (74 to 149 μm), 200 to 300 mesh (49 to 74 μm), and 300 mesh (49 μm) or smaller. Quartz grains were extracted from the 49 to 149 μm grains in the following way. The grains in the size range 74 to 149 μm were subjected to ultrasonic cleaning in water and in acetone. After the grain samples were treated with aqua regia at room temperature for 24 hours, they were separated into magnetic and nonmagnetic fractions. The nonmagnetic fraction was treated with 10% hydrofluoric (HF) acid for one hour to remove residual nonquartz materials and to mildly etch the surface of each quartz grain. The quartz grains obtained were repeatedly washed in water and acetone.

Powdered samples at sizes smaller than 49 μm were pressed into two circular disks of 45 mm diameter and 3 mm thickness and used as sources of natural beta particle emitting radionuclides for the estimation of background (BG) doses.

A piece of Nagasaki Ieno-cho brick was divided into three portions A, B, and C after removal of 2-mm thick front layer and 1-mm thick back layer. Small quantities of feldspars were observed in nonmagnetic minerals which were etched with HF acid for one hour at room temperature.

Measurements. A Harshaw Model 2000 TL reader was used for the measurements. Quartz

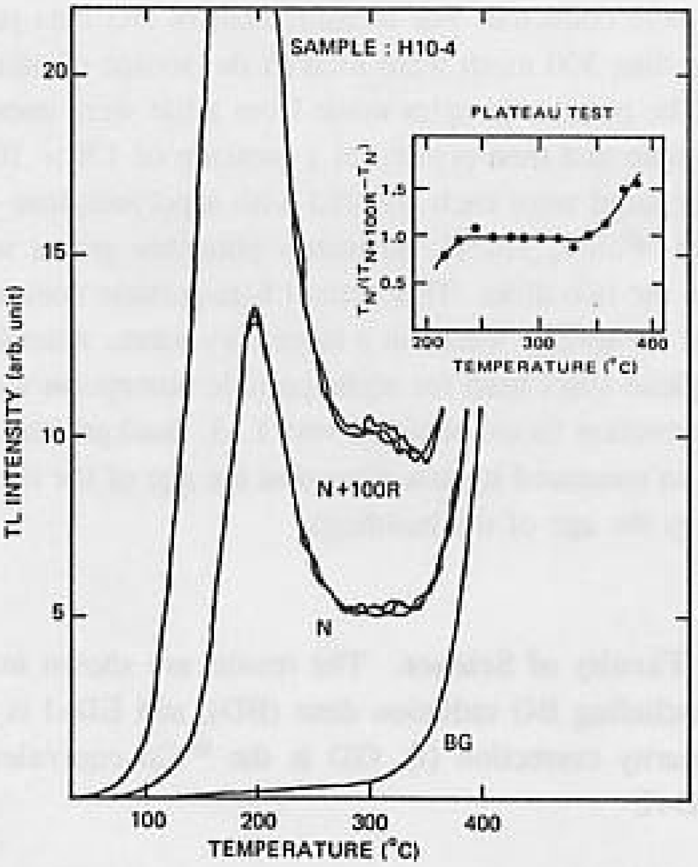

Figure 1. Thermoluminescence glow curves and a plateau test. N: TL induced by A-bomb and BG natural radiations. N+100R: TR induced by A-bomb and BG natural radiation plus ^{60}Co gamma rays for calibration. BG: glow due to incandescence

samples of 20 mg were heated from room temperature to 500°C at a heating rate of 20°C/s in an atmosphere of oxygen-free nitrogen. Glow curves of tile sample H10 from the Hiroshima University Faculty of Science (HUFS) are shown in Figure 1. Curve N was measured without calibration doses, while the curves N+100R were measured with a 100 R ^{60}Co gamma-ray exposure in addition to the A-bomb radiation and natural BG radiations. The gamma-ray exposures for calibrations were made by irradiating thin-layered quartz samples placed between 4-mm thick Lucite plates with ^{60}Co gamma rays at the Osaka Branch of the National Electro-Technical Institute, calibrated within an accuracy of ±0.5%.

A plateau test was made for each sample (see insert in Figure 1). The area under the glow curve in the plateau region gave the measure of TL intensity. The linearity between TL intensities and absorbed dose was also checked for each sample after it was annealed at 500°C for 30 minutes and exposed to ^{60}Co gamma rays. No supralinearity was observed for the tile samples collected from the buildings of HUFS. Several samples (e.g., the Chokin-kyoku tile in Hiroshima and the brazier sherd in Nagasaki) showed supralinearity.

BG Measurements. BG radiations consist of gamma rays from environmental materials, beta particles from radioactive elements in the tile material itself, and cosmic rays. For measurement of BG dose of cosmic rays and gamma rays from the environment, 74 to 149 μm grains of $CaSO_4$:Tm dosimeter were used. A polyethylene tube (5 mm diameter × 30 mm) containing the $CaSO_4$:Tm grains was encased in a 1-mm thick copper tube (7 mm diameter × 10 cm). The copper tubes were placed for several months at the sites where the tested tile samples were collected. For measurement of BG beta-particle dose, crushed powder samples smaller than 300 mesh were used as the source of natural beta particles as mentioned previously. The powder samples made from a tile were encased in an aluminum ring placed on a metal plate and then pressed at a pressure of 1.8×10^4 psi into a circular disk. Two disks thus prepared were each covered with a polyethylene sheet of 3.5 mg/cm^2 to absorb alpha particles. $CaSO_4$:Tm TL dosimetry phosphor grains were placed in a thin layer uniformly between the two disks. This natural beta-particle dosimetry set was encased in a lead box and placed for several weeks in a laboratory room. Attenuation of natural beta particles in the polyethylene sheet used for alpha-particle absorption was estimated from an empirical curve. The correction factor obtained was 1.13. Background radiation doses were obtained from the rates so measured by assuming that the age of the tile samples taken from the buildings are equal to the age of the buildings.

Results

Hiroshima University Faculty of Science. The results are shown in Table 1. ED is the ^{60}Co equivalent dose including BG radiation dose (BD), and ED+I is the ^{60}Co equivalent dose with the supralinearity correction (I). GD is the ^{60}Co equivalent dose obtained by subtracting BD from ED+I.

Table 1. Gamma-ray Doses for the Sample Collected from Hiroshima University, Faculty of Science

Sample	Ground Distance (m)	ED (rad)	ED+I[a] (rad)	BD (rad)	GD (rad)
H1	1271	158± 8	158	14	144
H2	1282	163±14	163	14	149
H3	1298	159±21	159	14	145
H4	1316	127±24	127	14	113
H5	1338	116±15	116	14	102
H6	1388	105± 5	105	18	87±5[b]
H7	1393	97±11	97	18	79
H8	1422	89± 6	89	18	71
H9	1428	81± 6	81	18	63
H10	1451	91± 5	91	18	73±5[c]
H11	1461	67± 6	67	18	49

a. No supralinearities were observed for any of the samples.
b. Mean value of 3 tile samples.
c. Mean value of 4 tile samples.

Other Samples in Hiroshima. Two tiles from the same sampling point on the roof of the Naka Telephone Office building were analyzed. Gamma-ray doses were obtained from the TL intensities in the 375°C peak. The BG radiation dose was not estimated, however, it is negligible because the tile was only 520 m from the hypocenter.

Wall tiles from the Chugoku Electric Co. building were measured. Plateau was observed

Table 2. Gamma-ray Doses of Hiroshima Samples Other than HUFS

Sample	Ground distance (m)	ED (rad)	ED+I (rad)	BG (rad)	ED (rad)
Naka Telephone Office[a,b]					
204-2	523	2945±352	-	-	2945
204-3	523	2898±418	-	-	2898
203-3	523	3088±276	-	-	3088
		±		average	2977± 99[c]
Chugoku Electric Co.[a,b]					
3-1-3	692	1281±150	-	21±3	1260±153
3-2-2	692	1089± 85	-	21±3	1068± 88
	692	1104± 72	-	21±3	1082± 75
3-2-3	692	1137± 77	-	22±3	1115± 80
	692	1068±100	-	22±3	1046±103
		±		average	1114± 85[c]
Japanese house at Nobori-machi[a,d]					
	1131	192± 11	-	-	-
Chokin-kyoku (Postal Saving Bureau)[a,e]					
	1597	50± 6	55± 6	14±2	40± 8

a. Supralinearity was not checked.
b. BG radiation dose is negligible compared with ED.
c. The error is the standard deviation.
d. BG radiation dose was not measured.
e. BG beta dose rate was 0.323 rad/yr. BG gamma-ray dose rate was assumed to be 0.082 rad/yr.

between 260 and 390°C. Good reproducibility in TL glow curves was observed.

A roof tile that is called "Oni-gawara", on the top corner of a roof of a Japanese house at Nobori-machi, was used. Plateau was between 270 and 350°C.

A tile at 1590 m from the hypocenter was collected from the roof of the Chokin-kyoku (Postal Saving Bureau) building. Supralinearity correction was 5 rad. The annual beta particle dose rate measured was 0.323 rad/yr. The BG dose was calculated assuming that annual gamma-ray dose is the same as that measured at the E-shaped building of HUFS. The results obtained are shown in Table 2.

Nagasaki. Quartz grains were extracted from a brazier in a Japanese house at 731 m from the hypocenter. The result obtained for this sample can give information on the effect of shielding by a Japanese house, if it is compared with new calculated gamma-ray doses. Grains treated in undiluted HF acid were used.

A ceramic sample collected at 1075 m from the hypocenter was used. The sample was shielded from A-bomb radiation at the time of the explosion, but details of the shielding conditions were not known.

Table 3. Gamma-ray Doses of Nagasaki Samples[a]

Sample	Ground distance (m)	ED (rad)	ED+I (rad)
Brazier[b]	731	583±41	624±41
Ceramic sherd	1075	409±67	-
Ieno-cho roof tile	1565	56±12	-

a. BG radiation doses were not measured.
b. Shielded at the time of the explosion.

Table 4. Gamma-ray Doses of Nagasaki Ieno-cho Brick

Sample	Depth from surface (mm)	ED (rad)	ED+I (rad)
A	3- 36	119	119
B	36- 70	106	106
C	70-104	102	102

A roof tile of a Japanese house was obtained at the 1565 m from the hypocenter in Nabori-machi.

The gamma-ray doses obtained from these sample are shown in Table 3.

Interlaboratory Comparison. Samples for interlaboratory comparison had been systematically treated and irradiated and were as follows.

Nagasaki Ieno-cho Brick. The sample was cut into three sections (A, B, and C) as shown in Table 4. Reproducibility of TL intensity was better for grains treated with 47% HF acid for 30 minutes than for grains treated with 10% HF acid for one hour. However, the grains with 10% HF acid treatment have about 10 times larger TL output than that with 47% HF acid treatment. Since the linear relationship between TL intensity and dose was certified with the sample treated with 10% HF acid in the plateau region between 290 and 360°C, radiation doses were estimated using the grains with 10 % HF treatment. The fading was 20% at the 180°C peak, and no fading was observed at the 220°C peak over a two-week period. Supralinearity was not observed.

NBS No. 1 - No. 4 Samples. Samples were selected by the University of Utah and were irradiated at NBS. The quantity of the distributed sample was too small to treat using the standard sample preparation procedure. Sample grains were washed in 10% HF acid for three minutes and only one or two TL glow curves were recorded for each sample. Assuming that sample No. 1 was not exposed at NBS, it was exposed to 20 R ^{60}Co gamma rays. The TL intensities of No. 2, 3, and 4 were compared at 220°C with that of No. 1. Gamma-ray doses

Table 5. Gamma-ray Doses of NBS Sample

Sample	ED (rad)
No. 2	6.7
No. 3	16.3
No. 4	38.1

obtained for these samples are shown in Table 5. Supralinearity and plateau tests could not be made.

Hiroshima University Tile No. 4. Plateau was observed in the range between 250 and 350°C. No supralinearity was found. The sample was divided into four portions and exposed at 0, 50, 100, and 200 R with ^{60}Co gamma rays. The result obtained by these four samples were 75 ± 15 rad.

Oxford Quarts Sample. Plateau was observed between 300 and 350°C. Supralinearity correction of about 85 rad was obtained. Additional ^{60}Co gamma-ray exposures of 400, 800, and 1200 R were given to three portions of them. The result obtained was 387 ± 18 rad.

Discussion

Since the samples used were exposed to A-bomb radiations about 40 years before the measurements, the plateau test was made for each sample. About 20% of the TL fading was observed at 220°C peak in TL glow curves. The triboluminescence in the high temperature region in TL glow curves could not be neglected compared with TL due to rather small quantities of A-bomb radiation doses. Therefore, only the TL output in the plateau region was used in the present dosimetry.

The contribution of alpha particles from radioactive nuclides in the tile itself to the TL observed could be neglected, because the quartz grains were measured after their surface layers were removed by HF acid treatment.

The variations in the measured values were, for example, 5% of the mean for the three H6 tile samples and 6% of the mean for the four H10 tile samples. The errors in the present estimates of BG doses are considered to be less than 10%.[1] In order to get the gamma-ray dose in air from the present ^{60}Co equivalent radiation dose, the effects such as thickness of the tile sample, radiations backscattered by concrete on which the tile was placed, slant angle of incident radiations, and energy dependence of TL response of gamma rays should be taken into consideration.

Using tiles of HUFS irradiated with ^{60}Co gamma rays, reduction in the average TL intensity due to self-shielding was estimated. The correction factor obtained was 1.05 for the 2.6-cm thick tile and 1.02 for the 2.0-cm thick tile. However, the tile samples had been exposed to A-bomb radiations while placed on the concrete floor of the buildings. Therefore, the actual build-up effect of the A-bomb gamma rays in the tile could have been larger than those in the tile exposed to ^{60}Co gamma rays without backscattered gamma rays by concrete blocks. Furthermore, the effective thicknesses of the tile samples should be considered, since the major fraction of the A-bomb gamma rays must have impinged at large slant angles on the tile samples.

The TL response of quartz to gamma rays at 40 and 100 keV are 3 to 1.2 times as large as that to ^{60}Co gamma rays, respectivley. Fortunately, the contribution of such low-energy gamma rays in the A-bomb radiation dose were much smaller than those of photons of 1 MeV or higher energy.

These problems are being studied in more detail.

References

1. Ichikawa, Y. and Nagatomo, T., 1983. Thermoluminescent dating and its application to gamma ray dosimetry. In *U.S.-Japan Joint Workshop for Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki*, pp. 104-114. Hiroshima: Radiation Effects Research Foundation.
2. Ichikawa, Y., Nagatomo, T., Hoshi, M. and Kondo, S., 1987. Thermoluminescence Dosimetry of Gamma Rays from the Hiroshima Atomic Bomb at Distances of 1270 to 1460 meters from the Hypocenter. Health Physics (in press).