I. DOSIMETRY

Reassessment of Gamma-ray Doses in Hiroshima and Nagasaki

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Reassessment of gamma-ray doses from the atomic bombs in Hiroshima and Nagasaki has been carried out with the thermoluminescent measurements of bricks and decorative tiles which were collected from the buildings that remain as they were at the time of the explosions. The thermoluminescent measurements were performed using a dating technique generally used in archaeology. Annual background dose rates from natural radionuclides in the brick and tile samples and from environmental radiations around the samples were determined with commercially available thermoluminescent detectors. The resultant gamma-ray doses in Hiroshima and Nagasaki are given as a function of distance from the hypocenters and are compared with the DS86 (Dosimetry System 1986) and the T65D (Tentative 1965 Dose).

INTRODUCTION

In 1980, Kerr and Pace¹⁾ and Loewe and Mendelsohn²⁾ reported preliminary results of gamma-ray and neutron doses in Hiroshima and Nagasaki which they independently recalculated using the radiation spectra calculated by Preeg. The dose assessment was discussed at the Late Effects Workshop on Dosimetry of the Atomic Bomb Survivors on May, 1981 and a Symposium on Atomic Bomb Dosimetry on September, 1981. No conclusion was reached, in spite of all our efforts at the two meetings. In Japan, dose reevaluation studies were started with financial help from the Ministry of Health and Welfare in August 1981.

The individual radiation dose received by the atomic bomb survivors is one of the most fundamental parameters in the Life Span Study (LSS) conducted at the Radiation Effects Research Foundation (RERF). Consequently, for the investigation of dose reassessment, atomic bomb dosimetry groups were established in 1981 in the United State and Japan. A total of four US-Japan joint workshops were held for an exchange of data and views. As a result of investigating many data, the Dosimetry System 1986 (DS86) was developed.

Formerly most scientists have studied atomic bomb dosimetry, using physical and chemical changes induced in various materials exposed to atomic bomb radiations in Hiroshima and Nagasaki. The resultant data played a very important role for the determination of the absolute value of atomic bomb doses in developing the Tentative 1965 Dose³⁾. The previous gamma-ray

doses had been carried out mostly for tile and brick samples exposed within about 1 km from the hypocenters, because an apparatus used for the thermoluminescence (TL) measurements was inferior in measuring small amounts of TL dosimeters are available for low dose measurements. The preliminary result of Loewe and Mendelsohn showed that their data were in good agreement with the gamma-ray doses previously evaluated by the TL measurements of ceramic materials within about 1 km from the hypocenters in Hiroshima and Nagasaki²⁾. In the present dose reassessment, the TL measurements were performed for the brick and decorative tile samples from existing buildings located at various distances more than 1 km from the hypocenters in Hiroshima and Nagasaki⁴⁾.

This paper will describe the methodology of TL measurement and some dosimetric parameters required for the atomic bomb dosimetry. The resultant gamma-ray doses will be compared with the data from the DS86.

MATERIALS AND METHODS

According to the preliminary data²⁾, the gamma-ray doses at a distance of 1.5 km from the hypocenters in Hiroshima and Nagasaki were estimated to be about 0.4 and 0.8 Gy, respectively²⁾. Annual background dose-rate of ceramic materials was about 4 mGy per year, roughly 80 per cent of which comes from natural radionuclides contained within the ceramic materials, and 20 per cent from the environment. Since it was estimated that most of brick and tile samples were manufactured more than 40 years before, the value of background dose was expected to be about 0.2 Gy for a typical sample. In the present dose estimation, a very precise determination of background radiation dose was required for the brick and tile samples. The background doses come from radiation emitted by the natural radionuclides of uranium, thorium and potassium contained within the brick and tile samples, and from the environment.

The TL output of bricks and tiles is dominated by light emitted mainly by quartz. The radionuclides and quartz inclusions distribute inhomogeneously in bricks and tiles. The average range of the alpha particles from uranium and thorium is only about 25 μm and as a consequence the average dose received by quartz inclusions decreases with grain size. In TL dating, various techniques have been developed in order to simplify the measurements of background dose from natural radionuclides. The TL measurement techniques most often used for atomic bomb dosimetry are the quartz inclusion technique and the per-dose technique, which are widely applied for the TL dating⁵⁾. In the present paper, an additive pre-dose technique was used for the determination of gamma-ray dose less than about 2 Gy. In the standard pre-dose technique, a high level radioactive beta-ray source is required to provide a laboratory calibration dose. The present TL measurements had to be carried out in a cold area of laboratory. No radioactive materials were permitted in the cold area (according to the national regulation concerning the radiation protection). In the additive pre-dose technique, the TL samples were irradiated with given doses of ^{60}Co gamma rays as the calibration doses instead of beta-ray doses. In the pre-dose technique, linear relationships between TL response and gamma-ray dose are not obtained for the high dose region above about 2 Gy, although the linear dose-response range was slightly different

from sample to sample. For atomic bomb dosimetry, the stability of TL signal storage is very important. The TL storage can be checked with a plateau test⁵⁾. Gamma-ray dosimetry for the high dose region was carried with a high temperature technique, in which the gamma-ray doses were determined using the area under the glow curve from 275 to 310°C. In all TL measurements with the high temperature technique, the stability of TL storage in the samples was checked with the plateau test.

The brick and decorative tile samples were prepared in the laboratory by removing a 5-mm thickness of the outer and inner surfaces with a diamond blade. The removal of surfaces was required to decontaminate mud and dirt and to ensure charged particle equilibrium in the measured volume. The small pieces of ceramic samples were crushed gently into grains in a mortar. By passing the grains through a mesh of 210 μm and a 105 μm sieve, unwanted materials than 210 μm and smaller than 105 μm were sifted out. The remaining samples with a grain size from 105 to 210 μm were washed with water and acetone in an ultrasonic cleaner. Unwanted clay matrix material was removed from the dried samples by a magnetic separator. Finally, the existence of quartz in the sample was confirmed with an optical microscope. For the TL measurements the sample was divided into at least three aliquots to be irradiated with different given doses of ^{60}Co gamma rays for the calibration.

In the additive pre-dose technique, a beta-ray dose of 12 mGy (1.2 rad) from a small ^{90}Sr beta-ray source was used as a test dose. The ^{60}Co gamma-ray exposure was measured in roentgen with an ionization chamber (0.6 cm^3 ; Ohyo Giken Co., Ltd.); and calibrated with the National Standard Dosimeter at the Electrotechnical Laboratory in Tsukuba, Japan. Powdered TL samples were irradiated with gamma-rays under secondary electron equilibrium by using a 1-cm thick brick plate, monitoring gamma-ray doses with MSO-TL detectors (Mg_2SiO_4 ; Kasei Optonics Co., Ltd.). Tissue dose in air was determined using a conversion factor of 9.5 mGy/R for ^{60}Co gamma-rays.

In the high temperature technique, at low doses the TL output per unit dose for quartz is small but increases gradually with increasing dose until a linear region is obtained. This is defined as a supralinearity. In order to determine a correction factor for supralinearity, the samples were irradiated with various gamma-ray doses after the samples were heated at 500°C for 8 hours. Their TL outputs were measured and plotted as a function of the doses. The correction factor was evaluated from the curve. The TL output obtained by original heating was corrected for supralinearity using the factor.

The background dose of beta-rays coming from natural radionuclides in brick and tile samples was measured with the experimental arrangement shown in Fig. 1, using MSO-TL powder with a grain size of about 100 to 150 μm . The background doses of gamma rays from environmental radiations and from building materials including brick and tile samples were determined with the MSO-TL detectors put at various depths in an aluminium plate 2-cm thick. The dosimetric plates were set for 6 months at the places from which the samples were collected. The background doses measured at the various depths in the plate were almost same.

TL powder for background measurement
in a lead shield container

Lucite(beta-ray shield)

Brick sample

Plastic film(alpha-ray shield)
(0.1 mm thick)

TL powder for beta-ray background
measurement from brick sample

Fig. 1. Experimental arrangement for background measurement of beta-rays. Beta-ray doses from natural radionuclides in the ceramic samples were determined with TL powder (Mg_2SiO_4 , manufactured by Kasei Optonics, Co., Ltd.). These dosimetric cells of all samples were kept in a lead shield container for 6 months.

RESULTS AND DISCUSSION

Triboluminescence resulting from the processes of crushing and mechanical handling when making powdered samples was investigated with an experiment using two types of brick sample. One set of samples was irradiated with a given dose after it was crushed into powder. The other set was crushed after irradiation then, the TL measurements were made with the additive pre-dose technique and the high temperature technique. From the experiment, it was concluded that there was no significant effects of triboluminescence during the sample preparation.

Table 1 gives the results of present TL measurements and background dose estimations. The gamma-ray doses of atomic bombs in Hiroshima and Nagasaki were evaluated by the following equation:

$$D = T_{\text{TL}} - AT_{\text{B}} \quad \dots\dots\dots (1)$$

where

D : gamma-ray dose from atomic bombs;

T_{TL} : total dose estimated from the TL measurements;

A : the age of ceramic samples; and

T_{B} : total background dose rates from natural radioactivities and environmental radiations including cosmic rays.

The standard error of the dose estimates was derived using the following equation:

$$\sigma_D = [\sigma_{\text{TL}}^2 + (\sigma_A)^2(T_{\text{B}})^2 + A^2 (\sigma_{\text{B}})^2 + (\sigma_{\text{r}})^2]^{1/2} \quad \dots\dots\dots (2)$$

where

σ_D : standard error in the atomic bomb dose estimate;

σ_{TL} : error in total dose estimated from the TL measurements;

σ_A : error in age estimate of ceramic samples;

Table 1. The results of thermoluminescence measurements.

Place and sample ^b	Distance from hypocenter (m)	Total dose (cGy)	Annual background dose		Time after original firing ^c (years)	Total back-ground dose (cGy)	Atomic bomb γ dose (cGy)
			β dose (μ Gy/year)	γ dose (μ Gy/year)			
Hiroshima							
Naka Tel. A	507	3030 $\pm$ 270 ^d	Neglected	Neglected	—	Neglected	3030 $\pm$ 270
Naka Tel. B	523	3330 $\pm$ 170 ^d	Neglected	Neglected	—	Neglected	3330 $\pm$ 170
San-in Bank	621	1960 $\pm$ 170 ^d	Neglected	Neglected	—	Neglected	1960 $\pm$ 170
Chugoku Electric A	665	800 $\pm$ 110 ^{d,e}	Neglected	Neglected	—	Neglected	800 $\pm$ 110
Chugoku Electric B	691	1050 $\pm$ 170 ^d	Neglected	Neglected	—	Neglected	1050 $\pm$ 170
Hiroshima Univ. A	1377	115 $\pm$ 7	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	95 $\pm$ 7
Hiroshima Univ. B	1387	127 $\pm$ 20	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	107 $\pm$ 20
Hiroshima Univ. C	1401	11 $\pm$ 2 ^a	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	-8 $\pm$ 2
Hiroshima Univ. D.	1425	120 $\pm$ 13	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	100 $\pm$ 13
Hiroshima Univ. E	1426	97 $\pm$ 5	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	77 $\pm$ 5
Hiroshima Univ. F.	1428	14 $\pm$ 5 ^a	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	-5 $\pm$ 5
Hiroshima Univ. G	1450	123 $\pm$ 17	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	103 $\pm$ 17
Hiroshima Univ. H	1457	20 $\pm$ 4 ^a	2650 $\pm$ 320	1250 $\pm$ 40	52 $\pm$ 2	20 $\pm$ 2	0 $\pm$ 4
Red Cross Hospital	1451	75 $\pm$ 10	2750 $\pm$ 360	1210 $\pm$ 70	44 $\pm$ 2	17 $\pm$ 2	58 $\pm$ 10
Postal Savings	1613	52 $\pm$ 6	2960 $\pm$ 450	1250 $\pm$ 90	47 $\pm$ 2	20 $\pm$ 2	32 $\pm$ 6
Provision Depot	3133	23 $\pm$ 5	1910 $\pm$ 380	1200 $\pm$ 80	73 $\pm$ 2	23 $\pm$ 3	0 $\pm$ 5
Nagasaki							
Urakami	459	4430 $\pm$ 480 ^d	Neglected	Neglected	—	Neglected	4430 $\pm$ 480
Sakamoto	1079	775 $\pm$ 96 ^d	Neglected	Neglected	—	Neglected	775 $\pm$ 96
Ieno A	1427	132 $\pm$ 21	2180 $\pm$ 280	1090 $\pm$ 30	50 $\pm$ 5	16 $\pm$ 2	116 $\pm$ 21
Ieno B	1427	130 $\pm$ 22	2180 $\pm$ 280	1090 $\pm$ 30	50 $\pm$ 5	16 $\pm$ 2	114 $\pm$ 22
Ieno C	1437	71 $\pm$ 7 ^a	2180 $\pm$ 280	1090 $\pm$ 30	50 $\pm$ 5	16 $\pm$ 2	55 $\pm$ 7
Zenza	1437	116 $\pm$ 24	1820 $\pm$ 220	1050 $\pm$ 60	60 $\pm$ 3	17 $\pm$ 2	99 $\pm$ 24
Inasa A	2026	45 $\pm$ 7	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	13 $\pm$ 9
Inasa B	2026	33 $\pm$ 4 ^a	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	1 $\pm$ 7
Inasa C	2026	27 $\pm$ 5 ^a	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	-4 $\pm$ 8
Inasa D	2036	32 $\pm$ 13 ^a	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	0 $\pm$ 14
Inasa E	2036	24 $\pm$ 8 ^a	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	-7 $\pm$ 10
Inasa F	2036	21 $\pm$ 7 ^a	2950 $\pm$ 380	1100 $\pm$ 40	80 $\pm$ 15	32 $\pm$ 7	-10 $\pm$ 9
Chikugo	2369	21 $\pm$ 15	2620 $\pm$ 470	1110 $\pm$ 50	60 $\pm$ 12	22 $\pm$ 5	0 $\pm$ 15

Note. Tel, telephone office; Urakami, chapel; Sakamoto, wall around the cemetery; Ieno, wall around a private house; Zenza, wall around the cemetery; Inasa, warehouse; Chikugo, wall around the cemetery.

^a The dose was estimated for the samples 5–15 mm deep.

^b For Hiroshima sample from the provision depot was brick, and others were ornamental tiles. For Nagasaki, the samples were brick.

^c The number of years from the original firing to the time of TL measurement estimated from the age of the building.

^d High temperature technique.

^e Shielded.

σ_{β} : error in annual beta-ray dose; and

σ_{γ} : error in annual gamma-ray dose.

It was difficult to set precisely a time-zero point of the original firing for each ceramic for the determination of background doses. However, information on the original firing for each building was obtained from the register book of building. Before World War II, the brick and

Fig. 2. Gamma-ray doses from the atomic bombs in Hiroshima and Nagasaki. a: Hiroshima and b: Nagasaki. The solid lines show the DS86 gamma-ray dose curve and the dotted lines show the T65D gamma-ray dose curve. The open circles show the present data and closed the previous data.

the decorative tile were most likely manufactured about one year before the construction of the building, because they were rare building materials at that time.

The total background dose was estimated taking into account only beta and gamma rays due to natural radioactivities and environmental radiations. According to the TL dating studies, alpha rays are less effective in producing TL output per unit dose than beta and gamma rays. In the pre-dose technique, the TL response from alpha rays can be negligibly small⁵⁾.

The distances from ground zero at which the ceramic samples were collected were determined with the new maps in Hiroshima and Nagasaki⁶⁾. Figs. 2a and 2b show the gamma-ray doses as a function of distance from ground zero in Hiroshima and Nagasaki, respectively. In these figures, solid lines show the DS86 gamma ray doses⁷⁾ and the dotted lines show the T65D gamma-ray doses. The open circles show present data given in Table 1 and the closed circles show the previous data⁸⁾. In Hiroshima, the six points around 1.4 km show the measured values of decorative tiles on the Hiroshima University building. Three of them exceed calculations by about 30%. Except for these points, present data in Hiroshima are in agreement with the DS86 dose curve. In Nagasaki, however, the DS86 and T65D gamma-ray doses are essentially the same and the TL data are in close agreement with the two sets of data.

In this reassessment of gamma-ray doses in Hiroshima and Nagasaki, five laboratories from three countries participated, namely Utah University from the United States, Nara University of Education and National Institute of Radiological Sciences from Japan, and Durham University and Oxford University from the United Kingdom. The measured values by the five laboratories were significantly lower than calculated values by DS86 in Nagasaki, where the principal source of data was from the leno-cho bricks, and the measured values were significantly higher than calculated values for the tiles collected from the Hiroshima University at virtually all locations. It is not clear yet whether the significant discrepancies between the calculated values and the measured values are due to a bias in the DS86 dose calculations or a potential characteristics in the TL measurements. Since the area beyond 1 km from ground zero in both cities is the region of greater importance for the LSS in RERF, further measurements will be required to confirm the validity of the DS86 and the TL measurements.

REFERENCES

1. Kerr, G.D. (1981) Review of dosimetry for the atomic bomb survivors. In proceedings of the Fourth Conference on Neutron Dosimetry, Neuherburg. Commission of the European Communities, Luxembourg.
2. Loewe, W.E. and Mendelsohn (1980) Revised estimates of dose at Hiroshima and Nagasaki and possible consequences of radiation induced leukemia (Preliminary). Technical Report D-80-14, Lawrence Livermore National Laboratory, Livermore, CA.
3. Miton, R.C. and Shohoji, R. (1968) Tentative 1965 radiation dose estimation for atomic bomb survivors. Report TR 1-68. Atomic Bomb Casualty Commission (ABCC).
4. Maruyama, T., Kumamoto, Y. and Noda, Y. (1988) Reassessment of gamma doses from the atomic bombs in Hiroshima and Nagasaki. *Radiat. Res.* **113**: 1-14.
5. Fleming, S. (1979) Thermoluminescence techniques in archaeology. Clarendon Press, Oxford.
6. Radiation Effects Research Foundation (Ed. Roesch, W.C.) (1987) US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki. Vols. I and II. Radiation Effects Research

Foundation, Hiroshima and Nagasaki.

7. Auxier, J.A., Cheka, J.S., Hatwood, F.F., Jones, T.D. and Thorngate, J.D. (1966) Free-field radiation dose distributions from Hiroshima and Nagasaki. *Health Phys.* **12**: 425-429.
8. Hashizume, T., Maruyama, T., Shiragai, A., Tanaka, E., Izawa, M., Kawamura, S. and Nagaoka, S. (1967) Estimation of the air dose from atomic bombs in Hiroshima and Nagasaki. *Health Phys.* **13**: 149-161.