Intercomparison of Luminescence Measurements of Bricks from Dolon’ Village: Experimental Methodology and Results from Japanese Laboratory

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Thermoluminescence dosimetry/External dose/Semipalatinsk Nuclear Test Site/Brick.

We have applied the thermoluminescence dosimetry technique to measure the total external dose from all the nuclear explosions in a few locations near the Semipalatinsk nuclear test site in Kazakhstan. The technique was optimized at our laboratory by fundamental study of the method of thermoluminescence dosimetry. The measured values of each sample at 10 mm depth were 248 ± 102 mGy (KSD-1), 30 ± 76 mGy (2(1-3)), 222 ± 63 mGy (2(3-2)), 217 ± 55 mGy (2(4-1)). The results are part of an international intercomparison exercise using bricks collected from the areas surrounding the Semipalatinsk nuclear test site.

INTRODUCTION

The former Union of Soviet Socialist Republics (USSR) conducted more than 450 nuclear explosions, including 86 atmospheric, 30 on the ground and 346 underground nuclear tests at the test site near Semipalatinsk City in Eastern Kazakhstan, during the period 1949–1989.1−3 The effects on human health from ionizing radiation, particularly the effect of exposure to prolonged low-dose radiation, have been a serious issue for people living in the surrounding areas.

The dose of radiation in the quartz grains included in ceramic bricks, accumulated since the time of manufacture can be measured by thermoluminescence dosimetry. The radiation dose in the bricks provides a measure of the external gamma radiation field and can thus be used to obtain information on the cumulative external radiation exposure at specific locations, as was also successfully applied by us to evaluate dosimetry long after the Hiroshima and Nagasaki atomic bombs.4,5

We have applied this technique to measure the total external dose from all the nuclear explosions in a few locations near the Semipalatinsk nuclear test site in Kazakhstan.6 This method should help reveal the total external dose originating from Semipalatinsk nuclear tests.7 The estimation of external radiation doses to the residents near Semipalatinsk nuclear test site has been performed by model calculations, a biological measuring method, etc, and it is important to verify the consistency and dosimetric accuracy of these different data. A group of scientists from a number of countries with experience in the use of thermoluminescence dosimetry has set up an intercomparison study based on samples of bricks collected near the Semipalatinsk nuclear test site.

This paper presents some fundamental investigations of the method of thermoluminescence dosimetry that aims to improve and optimize the technique at our laboratory. The results of the measured accumulated external dose are our contribution to the international comparison of thermoluminescence dosimetry using bricks collected from the areas surrounding the Semipalatinsk nuclear test site.

MATERIALS AND METHODS

Preparation of quartz samples for thermoluminescence measurement

The experimental procedure for estimation of external gamma-ray dose by the thermoluminescence dosimetry technique has been previously described.5,8,9 To apply the quartz inclusion technique, sand size grains were extracted from bricks by essentially the method described by Ichikawa10 and Fleming11 as follows.

The steps of sample preparation procedures for quartz samples for thermoluminescence measurement are shown in Fig. 1. First a 2-mm-thick layer from the front surface of the brick and 1-mm-thick layers from the four sides and the
back of the brick were removed using a water-cooled diamond saw. The front surface layer had to be removed because the secondary electrons due to gamma rays could not have reached their equilibrium state, and direct sunlight might have caused a reduction of the thermoluminescence signal in quartz.

The required brick sections were cut using the water-cooled diamond saw to produce a series of slices of increasing depth range from the front surface. The remaining brick was cut into 6 pieces along planes parallel to the surface and the depth profile of the radiation dose was estimated. The central depths from the surface of the brick were located at 3.5, 10, 20, 60, 100 and 120 mm. Then, the remaining brick pieces were gently crushed into grains by a rolling jaw crusher, and particles 53–250 \( \mu \text{m} \) in size were selected using standard sieves.

The particles were treated with hydrochloric acid to dissolve iron and other metals. The powdered grains were stirred in 100 mL of 20% hydrochloric acid with a magnetic stirrer for 40 min, washed with water 3 times, stirred in 100 mL acetone with a magnetic stirrer for 15 min, and washed once with water. After the treatment, any remaining magnetic bodies were removed by a magnetic separator (Nihon Chikagaku-Sha Company, Japan) several times after inspection under a microscope.

The collected nonmagnetic grains were further etched for 60 min by stirring in 100 mL of hydrofluoric acid with a magnetic stirrer (preventing heating above 30°C), and washed in acetone with an ultrasonic washer. By etching the outer layer of the grain, the absorbed dose contribution from alpha particles emitted by radionuclides in the brick is reduced to negligible levels. After drying, particles 90–125 \( \mu \text{m} \) in size were selected using standard sieves. This treatment was performed under an LED dark room lamp of wave length 670 nm.

**Measurement of thermoluminescence**

The quartz grains from each cut of the sample bricks were weighed and divided into about 100-mg portions, wrapped in wax paper 0.01 mm thick, and sandwiched between two polymethylmethacrylate plates 4 mm thick. The thickness of the quartz grain sample was controlled to 2 mm. The portions were irradiated with 0, 0.2, 0.4, 0.6, 0.8, and 1.0 Gy with \( ^{60}\text{Co} \) gamma rays. The \( ^{60}\text{Co} \) gamma rays were produced with a medical irradiation system (Shimadzu Corporation, Japan: RTGS-2DM 2.4 \( \times 10^{13} \) Bq). The center of the sample was located 80 cm downward from the \( ^{60}\text{Co} \) radiation source at a height of 120 cm from the floor. The sample was supported by a 0.25-mm-thick Mylar sheet. The exposure values were measured with a cavity-type ionization chamber (Toyo Medic CO., Ltd., Japan: RAMTEC1000D, PTW, Germany: N30001) calibrated within +/-2%.

Thermoluminescence intensity measurement was performed after irradiation keeping samples at 50°C for 24 h using a TLD reader (Kyokko Co., Ltd., Japan: Model 2500). Each sample measured weighed 25 mg and was heated from room temperature to 410°C at a heating rate of 10°C s\(^{-1}\) in a nitrogen gas flow.

The natural background doses \( D_N \) in the brick samples collected were estimated by the method previously described. For estimation of natural gamma-ray exposure for bricks in thermoluminescence dosimetry analysis, we used the data from in situ gamma-ray dose rate measure-
The beta-ray internal dose rate for quartz grain in the brick was measured for each brick sample in the laboratory. We applied the measurement technique of sandwiching $\alpha$-Al$_2$O$_3$:C optical stimulated luminescence (OSL) sheet type detector (Nagase Co., Ltd, Japan) between two brick samples, which were stored in a 10 cm thick Pb shielding box. The thickness of the OSL sheet detector was 100 $\mu$m. Laboratory irradiations for calibration were performed using a 370 kBq $^{90}$Sr/$^{90}$Y beta source. The protective layer of 0.1 mm thickness polystyrene sheet of the OSL detector was removed for beta-ray measurement and covered with 2 mm aluminum plate in order to evaluate the gamma-ray component from the interior brick samples. The dose component due to alpha particles originating within the clay matrix was reduced to negligible by etching the surface of the quartz. Therefore, the gamma-ray dose to quartz grains, $D_X$ is determined using the following equation:

$$D_X = D_T - T \cdot D_N$$

where,

$D_T =$ accumulated dose to quartz grains

$T =$ age of brick since firing

$D_N =$ dose rate from natural radioactivity of brick and ground and cosmic radiation

RESULTS AND DISCUSSION

The reason for the etching process is removal of the dose contribution from alpha particles emitted by radionuclides in the brick. The range for the alpha ray energy in the quartz was obtained by Monte Carlo calculation. Monte Carlo cal-

![Fig. 3. Micrography of quartz grains after the etching process.](image)

![Fig. 4. Histograms of the quartz grain diameter after the etching process.](image)
Calculations for this study were performed with the TRIM\textsuperscript{(13)} code for energies from 0.5 to 15 MeV to cover a wide range of natural radioactive elements. The molecular composition of 500-µm-thick quartz was considered to be Si (33.3\%) + O (66.7\%), and its density 2.0 g/m\textsuperscript{3}. Fig. 2 shows a calculated alpha-ray range in quartz from a well-collimated beam. The range in quartz of an alpha ray of energy 8.8 MeV that corresponded to the maximum energy of natural radiation is 60 µm.

Figure 3 shows the quartz grain appearance after etching by stirring in 100-mL of 20% hydrofluoric acid for 30 and 60 minutes. The histogram of the size of the quartz grains after the etching process is shown in Fig. 4. The average size of the quartz grains was 193 µm after a processing time of 30 minutes and 112 µm after a processing time of 60 minutes, and if the processing time was over 70 minutes the quartz grains disintegrated. Therefore the standard etching time in the present case was from 60 to 70 minutes.

The brick samples for intercomparison, which were delivered to our laboratory and the extracted quartz grain from bricks, are listed in Table 1. Examples of glow curves for samples 2(4-1) with or without additional doses of \textsuperscript{60}Co gamma rays are shown in Fig. 5. TL intensities in the glow curves obtained were integrated from 245°C to a temperature of, for example, 330°C in the samples.

Measured values of accumulated doses in the quartz grains from each brick sample are summarized in Table 2. The data obtained for natural background dose are summarized in Table 3 as the background doses for each brick. The total background doses were obtained by multiplying the measured background dose rates by the brick ages. The depth dose distributions are shown Fig. 6. The results of the mea-

| Sample   | Weight of the sample (g) | Extracted quartz grain (g) | Extraction efficiency (%) |
|----------|--------------------------|-----------------------------|---------------------------|
| KSD-1    | 720                      | 0.819                       | 0.11                      |
| 2(1-3)   | 490                      | 0.792                       | 0.16                      |
| 2(3-2)   | 420                      | 0.844                       | 0.20                      |
| 2(4-1)   | 530                      | 0.900                       | 0.17                      |

Fig. 5. Thermoluminescence glow curves and curve constructed for the plateau test. The TL intensity on the vertical axis is readings from a TL reader. The temperature is indicated in °C. Notation N in the figure indicates natural glow curves. 0.2, 0.4, 0.6, 0.8 and 1.0 indicate the glow curves for samples irradiated with \textsuperscript{60}Co gamma-ray doses of 0.2, 0.4, 0.6, 0.8 and 1.0 Gy, respectively. The values on the vertical axis in the inserted figure are \((T_N - BG)/(T_{N+\gamma} - T_N)\).
The values of the accumulated dose in the bricks collected from the areas surrounding the Semipalatinsk nuclear test site, measured by the thermoluminescence dosimetry method, have been submitted for the international intercomparison, which will clarify their validity.

CONCLUSION

We have applied the thermoluminescence dosimetry technique to measure the total external dose from all the nuclear explosions in a few locations near the Semipalatinsk nuclear test site in Kazakhstan. The technique was optimized at our laboratory by fundamental study of the method. The measured values of each sample at the 10 mm depth were 248 ± 102 mGy (KSD-1), 309 ± 76 mGy (2(1-3)), 222 ± 63 mGy (2(3-2)), 217 ± 55 mGy (2(4-1)). The results of the accumulated external dose are our contribution to the international comparison of thermoluminescence dosimetry using bricks collected from the areas surrounding the Semipalatinsk nuclear test site.

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REFERENCES

1. Gusev, B. I, Rosenson, R. I., Abylkassimova Z. N. (1997) The Semipalatinsk nuclear test site: a first assessment of the radiological situation and the test-related radiation doses in the surrounding territories. Radiat. Environ. Biophys. 36: 201–204.

2. Lindholm, C., Shimon, S.L., Makar, B., Baerstock, K.(eds.) (2002) Workshop on dosimetry of the population living in the proximity of the Semipalatinsk atomic weapons test site. STUK-A187, February 2002

3. Imanaka, T., Fukutani, S., Yamamoto, M., Sakaguchi, A., Hoshi, M., (2005) Width and Center-axis Location of the Radioactive Plume That Passed over Dolon and Nearby Villages on the occasion of the First USSR A-bomb Test in 1949. J. Radiat. Res. 46: 395–399.

4. Ichikawa, Y., Higashimura T., Sidei, T. (1966) Thermoluminescence dosimetry of gamma rays from atomic bombs in Hiroshima and Nagasaki. Health Phys. 12: 395–405

5. Ichikawa, Y., Nagatomo, T., Hoshi, M., Kondo, S. (1987) Thermoluminescence dosimetry of gamma rays from the Hiroshima atomic bomb at distance of 1.27 to 1.46 km from the hypocenter. Health Phys. 52: 443–451

6. Takada, J., Hoshi, M., Nagatomo, T., Yamamoto, M., Endo, S., Takatsuji, T., Yoshikawa, I., Gusev, B. I., Sekerbaev, A. K., Tchaijunusova, N. J. (1999) External doses of residents near Semipalatinsk nuclear test site. J. Radiat. Res. 40: 337–344.

7. Bailiff, I. K., Stepanenko, V. F., Goksu, H. Y., Jungner, H., Balmukhanov, S. B., Balmukhanov, T. S., Khamidova, L. G., Kisilev, V. I., Kolyado, I. B., Kolizhenkov, T. V., Shoikhet, Y. N., Tsyb, A. F. (2004) The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk nuclear test site: an evaluation of potential. Health Phys. 87: 625–41.

8. Nagatomo, T., Hoshi, M., Ichikawa, Y. (1992) Comparison of the measured gamma ray dose and the DS86 estimate at 2.05 km ground distance in Hiroshima. J. Radiat. Res. 33: 211–217

9. Nagatomo, T., Ichikawa, Y., Ishii, H., Hoshi, M. (1988) Thermoluminescence dosimetry of gamma-rays from the atomic bomb at Hiroshima using the predose technique. J. Radiat. Res. 113: 217–234

10. Ichikawa, Y. (1965) Dating of ancient ceramics by thermoluminescence. Bull. Inst. Chem. Res. Kyoto Univ. 43: 1–8

11. Fleming, S. J. (1970) Thermoluminescent dating: Refinement of the quartz inclusion method. Archaeometry 12(2): 133–145

12. Takada, J., Hoshi, M., Endo, S., Yamamoto, M., Nagatomo, T., Gusev, B. I., Rozenson, R. I., Apsalikov, K. N., Tchaijunusova, N. J. (1996) Thermoluminescence dosimetry of gamma rays from the fallout of the Semipalatinsk nuclear tests. Effects of low level radiation for residents near Semipalatinsk nuclear test site. Proceed. Second Hiroshima International Symposium: 195–199

13. Biersack, J. P., Haggmark, L. G. (1980) A Monte Carlo computer program for the transport of energetic ions in amorphous targets. Nucl. Instr. Meth. 174: 257.

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