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Specific Radioactivity of Europium-152 in Roof Tiles Exposed to Atomic Bomb Radiation in Nagasaki

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Specific radioactivities of residual europium (Eu)-152 were measured in six roof tile samples exposed to the Nagasaki atomic bomb at two locations. The ground distances of the two locations from the hypocenter are 1020 m and 1060 m. In order to obtain reliable data, Eu-enriched samples (from 207 to 855 mg) were prepared by separating Eu from each roof tile sample (from 1 to 2 kg). For the major aliquot of the Eu-enriched sample, residual radioactivity of ^{152}Eu was measured using a low-energy photon spectrometer. For the minor aliquot of the Eu-enriched sample, Eu content was determined by neutron activation analysis. Results of the specific radioactivity (^{152}Eu/Eu, Bq mg^{-1}) corrected to the time of bombing were in a range from 0.080 to 0.446. Although the measured values showed some scattering, they are moderately consistent with the calculated values by the DS86 methodology, *i.e.* the average ratio of the calculated to measured values is 1.3 ± 0.8.

INTRODUCTION

After the publication of the "Dosimetry System 1986 (DS86)" report[1], it was independently pointed out by our group and by the Hiroshima dosimetry group that the measured specific radioactivities of ^{152}Eu (^{152}Eu/Eu, Bq mg^{-1}) induced by the Hiroshima atomic bomb (A-bomb) neutrons were inconsistent with the values calculated by the DS86 methodology. The ratio of calculated-value to measured-value near the hypocenter is 1–2, whereas that beyond 1 km from the hypocenter is 0.1–0.5[2,3]. This inconsistency became more defined as more measured values were subsequently added[4–6]. The specific radioactivities of ^{60}Co[3–7] and ^{36}Cl[8] also showed inconsistencies similar to ^{152}Eu[2,6]. Inconsistencies in ^{60}Co values were recognized before the publication of DS86, but the reason for this has remained unclear. Accumulated data has conclusively shown that the calculations according to the DS86 methodology for radionuclides induced by thermal (and resonance) neutrons from the Hiroshima A-bomb are inconsistent with the

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measured values. What has caused such inconsistencies between the measured values and the calculated values for the Hiroshima A-bomb? To answer this, measurements of residual neutron-induced radionuclides, the source terms (*i.e.*, the bomb yield and the neutron spectrum) used for developing DS86, and air-transport of neutrons have been reinvestigated.

This work was undertaken to substantiate whether measured data for ^{152}Eu in samples exposed to the Nagasaki A-bomb more than 1 km from the hypocenter deviate from calculated values in a similar manner as Hiroshima bomb values. Therefore we analyzed samples exposed to the Nagasaki A-bomb at little-unambiguous positions more than 1 km from the hypocenter. If these discrepancies exist in both Hiroshima and Nagasaki data, the problem arises from the neutron transport calculations or from an unaccounted for variable common to both cities. If the discrepancies are not seen in Nagasaki, the problem arises from the source terms of the Hiroshima A-bomb, such as neutron spectrum or neutron yield. Our present results should aid in narrowing down the cause of the problem.

MATERIALS AND METHODS

Samples

We collected exposed roof tiles for this study from five places located on high ground in a donut belt at a horizontal ground distance of 700–1500 m from the hypocenter of the Nagasaki A-bomb in 1991. The sample size for the determination of ^{152}Eu needs to be more than 1 kg, hence we tried to collect only slightly damaged blocks of roof tiles. Although some non-exposed materials or materials of uncertain locations of exposure may have been collected because of inaccurate recollections or hearsay, we initially collected large samples of roof tiles (1–2 kg). The exposure conditions of the collected roof tiles of the Anakoboji Temple and of the house of Mr. Murataro Ide were very reliable. The sampling locations of these roof tiles are shown in Figure 1 as NA for Anakoboji Temple (368, Ebira-cho, horizontal ground distance of 1020 m from the hypocenter, 100 m above sea level) and NI for the house of Mr. Murataro Ide (9–34, Hanazono-cho, horizontal ground distance of 1060 m from the hypocenter, 25 m above sea level). There were no measurements of ^{152}Eu at such distant places from the hypocenter in Nagasaki.

Chemical separation of Eu

Since Eu is a trace component in roof tiles and the half-life of ^{152}Eu is not so long (13.33 y), the levels of residual radioactivity of ^{152}Eu in these samples were expected to be extremely weak. Therefore, Eu was chemically separated from the sample matrix to improve the geometrical efficiency of counting the residual radioactivity of ^{152}Eu as well as to obtain a counting source with little self absorption and to eliminate the natural radionuclides which may disturb the measurement of extreme low-levels of ^{152}Eu radioactivity. The chemical procedure used in this work is a somewhat simplified version of a method published elsewhere[2]. Each roof tile block was washed, dried and pulverized to <125 μm (hereinafter referred to as the original sample). Based on the data of stable Eu concentration (Table 1) determined by neutron activation analysis, the necessary amount of the original sample for chemical separation was estimated to obtain >1 mg of Eu.

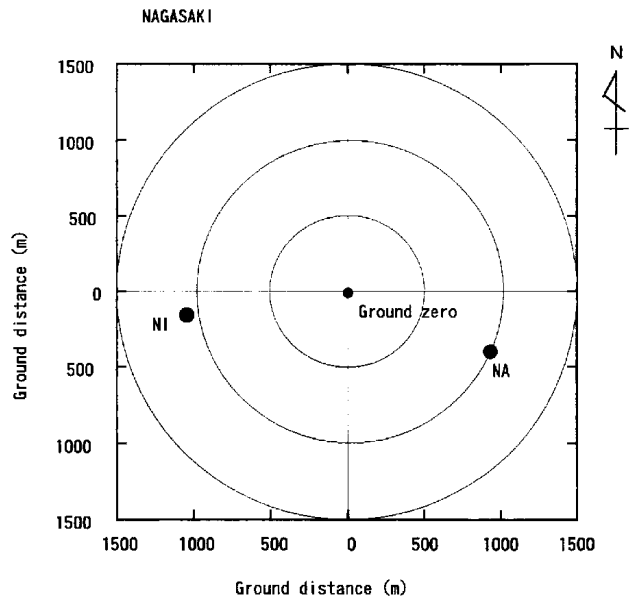

Figure 1. The locations of roof tile sampling in Nagasaki in 1991.

Table 1. Results of the measurements

Place of exposure (Sample code)	Distance from the burst point (m)	Amount of original sample analyzed (g) [Eu content (mg)]	Eu-enriched sample (mg) [Eu content (mg)]	Eu recovery (%)	Counting rate of 39–40 keV X-ray of Eu-152 in Eu-enriched sample (10^{-4} cps)	Interval between exposure and start of counting (y)	Eu-152/Eu ATB[a] by measurement (Bq mg^{-1})	Eu-152/Eu ATB[a] by DS86 calculation (Bq mg^{-1})	Calculation /measurement (: C/M ratio)
Anakoboji Temple	1,100								
(NA-1)		1,680 [2.51 ± 0.11]	855.3 [1.72 ± 0.05]	68.5 ± 3.6	1.19 ± 0.43	46.75	0.094 ± 0.034	0.195	2.07 ± 0.75
(NA-2)		1,710 [2.39 ± 0.11]	207.5 [1.17 ± 0.03]	49.0 ± 2.6	1.28 ± 0.24	46.95	0.080 ± 0.016	0.195	2.44 ± 0.49
(NA-3)		976 [1.34 ± 0.07]	509.6 [1.16 ± 0.04]	86.6 ± 5.4	3.42 ± 0.56	47.54	0.288 ± 0.048	0.195	0.68 ± 0.11
(NA-4)		1,722 [2.43 ± 0.11]	850.5 [1.91 ± 0.06]	78.6 ± 4.3	5.99 ± 1.11	47.58	0.446 ± 0.084	0.195	0.44 ± 0.08
Ide residence	1,170								
(NI-1)		1,900 [2.45 ± 0.10]	395.3 [1.51 ± 0.04]	61.6 ± 3.0	1.95 ± 0.20	47.26	0.110 ± 0.012	0.116	1.05 ± 0.11
(NI-3)		1,290 [1.76 ± 0.09]	419.2 [1.56 ± 0.04]	88.6 ± 5.1	2.35 ± 0.35	47.47	0.113 ± 0.021	0.116	1.03 ± 0.19

[a]ATB: at the time of bombing

From ~50 to ~90 % of Eu in 1–1.9 kg of each original sample was recovered in 207–855 mg of Eu-enriched sample (major matrix is lanthanum oxalate) (Table 1). The Eu-enriched sample was then uniformly suspended in about 15 cm^3 of methanol and 0.5–0.8% aliquots (two aliquots for each sample) of the suspension were placed in polyethylene tubes (8 mm ϕ × 15 mm) and dried for neutron activation analysis of stable Eu content in the Eu-enriched sample. The remain-

ing suspension was placed in a polyethylene tube (21 mm ϕ) and allowed to stand in order to settle the oxalate in a uniform thickness. After drying, a thin layer of oxalate formed at the bottom of the polyethylene tube (21 mm ϕ) was subjected to the measurement of residual ^{152}Eu radioactivity.

Neutron activation analysis of Eu

Neutron irradiation for activation analysis of Eu was carried out using a TRIGA-II nuclear reactor (100 kW) at the Atomic Energy Research Institute of Rikkyo University. The samples for neutron activation analysis were irradiated on the rotary specimen rack in the reactor together with standard reference samples prepared using a Eu standard solution for atomic absorption analysis. For the correction of the relative neutron fluence among the irradiation samples, a weighed stainless-steel wire ring (about 40 mg) was attached around each sample as a flux monitor. After neutron irradiation for 24 h (intermittent irradiation) and appropriate cooling, γ-ray spectrometry was carried out.

Measurement of radioactivity of residual ^{152}Eu

The residual radioactivity of ^{152}Eu produced by A-bomb neutrons was determined by measuring (one cycle: 7–54 days) 39.52 keV and 40.12 keV X-rays and 121.78 keV γ-rays using a heavily shielded HP-Ge low-energy photon spectrometer (HP-Ge LEPS).

Mock-up samples of various thickness were prepared as follows to measure the counting efficiency for the measurement of residual ^{152}Eu in each Eu-enriched counting source: oxalate precipitate was prepared from a solution containing a known amount of La, 1 mg of Eu and a known amount of ^{152}Eu standard solution (calibrated at LMRI, France). The ^{152}Eu-doped precipitate thus prepared was then allowed to settle at the bottom of a polyethylene tube (21 mm ϕ), dried and weighed for counting.

RESULTS AND DISCUSSION

Figure 2 shows the results of comparison of "factor of merit" for counting of major X-rays and γ-rays of ^{152}Eu using our HP-Ge LEPS (active volume: > 200 mm$^2 \times > 10$ mm t; thickness of Be window: 0.127 mm) and our normal HP-Ge γ-ray spectrometer (relative efficiency of the detector: 30%). It is more advantageous to use the HP-Ge LEPS to determine the residual radioactivity of ^{152}Eu in the Eu-enriched samples obtained in this work. In Figure 3, the factor for converting the counting rate (cps) of our HP-Ge LEPS to the disintegration rate (Bq) is shown for 39.52 keV and 40.12 keV X-rays and 121.78 keV γ-rays from ^{152}Eu in relation to the thickness of the counting sample. It is obvious from Figure 3 that the counting of 39.52 keV and 40.12 keV X-rays is more efficient than counting of 121.78 keV γ-rays using our HP-Ge LEPS for the determination of low-level radioactivity (around 0.01 Bq) of ^{152}Eu in Eu-enriched samples prepared in this work.

Data for ^{152}Eu and Eu together with selected description on the original and Eu-enriched samples are given in Table 1. The results of ^{152}Eu/Eu (corrected to the time of bombing) obtained

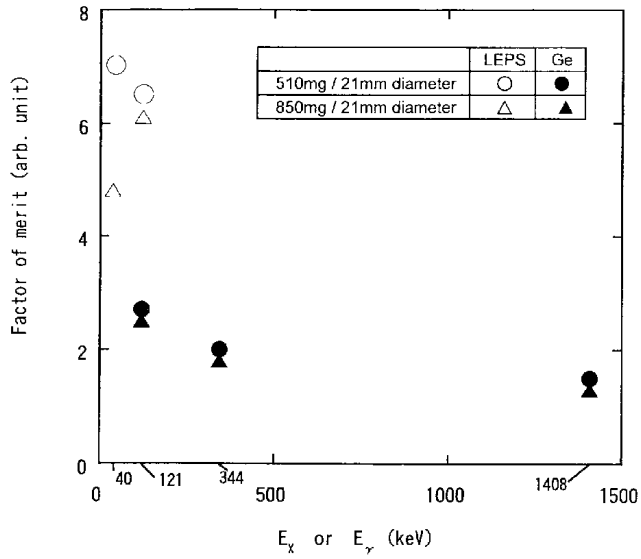

Figure 2. Comparison of "factor of merit" in the measurement of major X-rays and γ-rays for the determination of low-level ^{152}Eu radioactivity in lanthanum oxalate samples with thicknesses of 510 or 850 mg/21 mm ϕ. LEPS: HP-Ge low-energy photon spectrometer. Ge: HP-Ge γ-ray spectrometer.

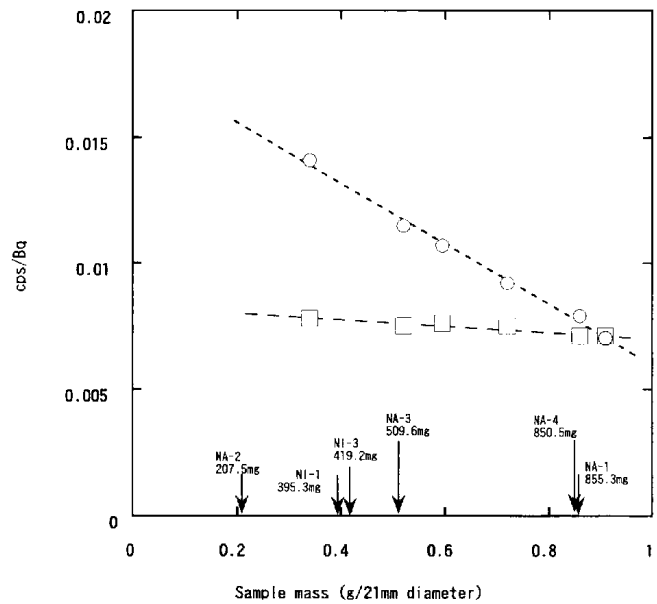

Figure 3. Relationship between "counting rate (cps)" and "disintegration rate (Bq)" as a function of the thickness of 21 mm ϕ samples in counting the (39.52 keV + 40.12 keV) X-rays (○) and 121.78 keV γ-rays (□) of ^{152}Eu by HP-Ge LEPS.

in this work (marked with ○) are shown in Figure 4 together with reported results of measurements[9–12] and the calculated values (shown by solid curve) according to the DS86 methodology.

The slant distance between the sample and the burst point of the Nagasaki A-bomb was calculated assuming the burst point to be 500 m above the hypocenter which is 5 m above sea level. However, an error of about ±15 m was assumed, because the sample roof tiles were scattered around at the time of the bombing.

Since the graphic representation in the DS86 report[9] are the sole official values calculated by the DS86 methodology for the specific radioactivity of ^{152}Eu in Nagasaki, we converted the graphic representation into numeric data and reproduced the curve shown in Figure 4.

The half-life value of ^{152}Eu used to calculate the radioactivity of the nuclide at the time of bombing (ATB) was the conventional value of 13.33 y[13] instead of the newly reported value of

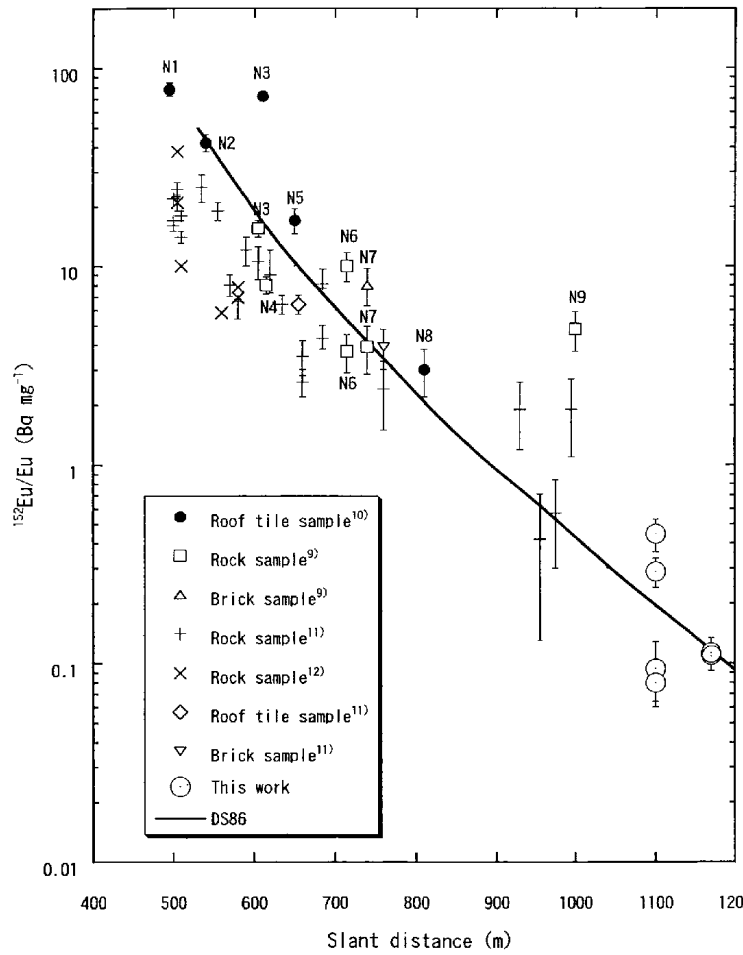

Figure 4. Specific radioactivity of ^{152}Eu at the time of bombing (ATB) in Nagasaki.

13.542 y[14]. The effect of the difference in the half-life value on the present decay correction of ^{152}Eu is less than half of the overall error ($\pm$ 1 σ) of the present measurement.

Measurements of residual radioactivity of ^{152}Eu in the Eu-enriched samples have been intermittently repeated for the last several years, and the accuracy of the data has been confirmed. In spite of the moderately good reproducibility in measurement of residual ^{152}Eu in the individual Eu-enriched samples, rather scattered ^{152}Eu/Eu data were obtained for the samples from the NA site. Although the reasons for the scatter are unclear, the mean of the scattered ^{152}Eu/Eu data for NA falls within the range of the data for NI and that predicted by DS86 methodology. Such scatter may be unavoidable in measurement of extreme low-levels of radioactivity.

For each ^{152}Eu/Eu data point obtained, the ratio of the calculated value to the measured value (hereinafter referred to as C/M ratio) was calculated together with associated error due to counting statistics ($\pm$ 1 σ) (Table 1). The mean of C/M ratios and standard deviation for the scatter of C/M ratios were calculated to be 1.3 ± 0.8 without considering the error associated with each C/M ratio. DS86 reproduced each ^{152}Eu measurement within a factor of two for locations at a slant distance of 1100 m to 1200 m from the burst point of the Nagasaki A-bomb. On the other hand, the C/M ratios for ^{152}Eu specific radioactivity at slant distances of 1100 m to 1200 m from the burst point of the Hiroshima A-bomb are around 0.2–0.3. Therefore, it may be concluded that the relation of the C/M ratio for ^{152}Eu specific radioactivity to distance from the burst point tends to be different between Hiroshima and Nagasaki.

In order to further clarify the conclusions mentioned above, samples exposed to the Nagasaki A-bomb at a slant distance of about 1500 m from the burst point (ground distance of 1400 m from the hypocenter) will have to be measured. For this, procurement of more than 5 kg of exposed sample and chemical processing as done in this study are essential to obtain > 5 mg of Eu.

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