Residual Neutron-Induced Radionuclides in Samples Exposed to the Nuclear Explosion over Hiroshima:Comparison of the Measured Values with the Calculated Values : I. DOSIMETRY

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A Review of Forty-Five Years Study of Hiroshima and Nagasaki Atomic Bomb Survivors

I. DOSIMETRY

Residual Neutron-Induced Radionuclides in Samples Exposed to the Nuclear Explosion over Hiroshima: Comparison of the Measured Values with the Calculated Values

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^{152}Eu/^{154}Eu/^{60}Co/A-bomb neutron/Hiroshima

Residual radionuclides induced by neutrons from the Hiroshima atomic bomb have been measured at Kanazawa University for these 14 years. The results of ^{152}Eu, ^{154}Eu and ^{60}Co are reviewed in this paper. Where appropriate, an attempt is made to provide our new data with the aim of reinterpreting our published data. From the comparison of the measured values with the calculated values by DS86 methodology, we may point out here: (1) that close agreement was found between measured and calculated values for the specific radioactivity of ^{152}Eu in the samples exposed at ground ranges between 320 m and 720 m; (2) that the calculated/measured ratios for the specific radioactivity of ^{152}Eu were, however, larger than unity in the vicinity of ground zero and smaller than unity at locations more than 1000 m apart from ground zero; (3) that, in the vicinity of ground zero, epithermal neutron fluence evaluated from a set of measured specific radioactivities of ^{152}Eu, ^{154}Eu and ^{60}Co showed a close agreement with the calculated result, whereas thermal neutron fluence evaluated in the same way was different from the calculated result; and (4) that the depth distribution of the specific radioactivity of ^{152}Eu in a wall sample which was exposed at the location 320m from ground zero approximately agreed with the calculated result.

INTRODUCTION

In August 1976, when *in situ* measurements of environmental radioactivity in the Hiroshima district were carried out by Sakanoue and Komura of Kanazawa University using a high-resolution portable Ge(Li) γ-ray spectrometer, low-level γ-ray peaks of ^{152}Eu were found in a spectrum acquired in the Hiroshima Atomic Bomb Memorial Dome (what is known as Hiroshima Genbaku Dome) near ground zero[1]. The ^{152}Eu was considered to be a product of a neutron capture reaction between ^{151}Eu, a minor constituent in ordinary rock and the like, and neutrons from the nuclear explosion over Hiroshima in 1945. This assumption was based on the fact that ^{151}Eu,

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of which natural isotopic abundance is 47.9%, has an extremely large thermal neutron capture cross-section of 5.9×10^{-21} cm^2 and that the half-life of ^{152}Eu is a moderately long one (13.2 y). To confirm that the ^{152}Eu was really induced through above-mentioned process, from April 1977, we studied the correlation between the slant distance from the explosion point and the specific radioactivity of ^{152}Eu in bombed samples such as roof tiles, bricks and rocks which were collected in Hiroshima and Nagasaki. The validity of the assumption was soon verified in Hiroshima and Nagasaki[2–4]. Concomitantly, another residual neutron-induced radionuclide, ^{154}Eu (half-life: 8.5 y), was first measured in a sample from ground zero of Hiroshima together with ^{60}Co (half-life: 5.27 y) which was determined as early as 1960 in iron materials collected in Hiroshima and Nagasaki[5]. Prior to 1976, only ^{32}P (half-life: 14.3 d) and ^{60}Co had been determined in samples exposed to the nuclear explosions at Hiroshima and Nagasaki[6].

In 1981, it became clear that the T65D estimates for both γ-rays and neutrons in Hiroshima and Nagasaki were not as accurate, as previously assumed[7]. Consequently, a joint U.S.-Japan research program and review committees were established to reassess atomic bomb radiation dosimetry in Hiroshima and Nagasaki. The review was completed in 1986 for the time being, and a two-volume report was published that discussed the physical basis for a new Dosimetry System 1986 (DS86)[8].

In 1982, experimental studies for reassessment of atomic bomb radiation in Hiroshima and Nagasaki were started in Japan. From 1982, we participated in three co-operative research groups organized in Japan for the reassessment: we continued the measurement of residual neutron-induced radionuclides. Since the residual neutron-induced radionuclides such as ^{152}Eu, ^{154}Eu and ^{60}Co proved to be detectable even today by high-sensitivity γ-ray and X-ray counting techniques, it was expected that the measurement of these radionuclides would be of value to check the validity of a series of computer calculations employed for the reassessment. The use of these radionuclides for the reassessment of atomic bomb neutron dosimetry, however, has been limited by the following difficulties: (1) today, these radionuclides are found only at extremely low concentrations in materials exposed to the atomic bomb; and (2) the neutrons that induced these radionuclides were thermal and epithermal, while the neutron dose received in Hiroshima and Nagasaki is attributable mainly to fast neutrons. In order to overcome the first difficulty, we established a chemical procedure to extract Eu and Co from the samples exposed to the atomic bomb. To test the accuracy of the DS86 methodology for epithermal and fast neutrons, the depth distribution of specific radioactivity of ^{152}Eu and two sets of specific radioactivities of ^{152}Eu, ^{154}Eu and ^{60}Co were studied on samples from the vicinity of ground zero in Hiroshima[9].

While the final report of the atomic bomb dose reassessment was published in 1986, we continued to obtain additional data on residual neutron-induced radionuclides in samples exposed to the nuclear explosion in Hiroshima. In this paper, we attempt to reinterpret our previous data[4,9] of the residual neutron-induced radioactivities in Hiroshima while providing our new data.

MATERIALS AND METHODS

Samples

Table 1 provides information on the locations and types of the samples used in our study. A schematic showing the locations of these samples is given in Fig. 1. These samples were supplied by courtesy of the Research Institute for Nuclear Medicine and Biology (Hiroshima University), the Hiroshima Peace Memorial Museum, and Professor Y. Ichikawa of Nara University of Education. Except for the core sample from the outer wall of the Fukoku Life Insurance Building (sample-F)[9], specimens from the outer layers less than 3 cm thick were each subjected to analysis. The sample-F was sliced at 5–10 mm intervals parallel to the outer surface[9]. The position where each sample was exposed to the nuclear explosion was read off on a city planning map of Hiroshima which was drawn in 1979 on a scale of 1 to 2,500; the map, known as U.S. Army

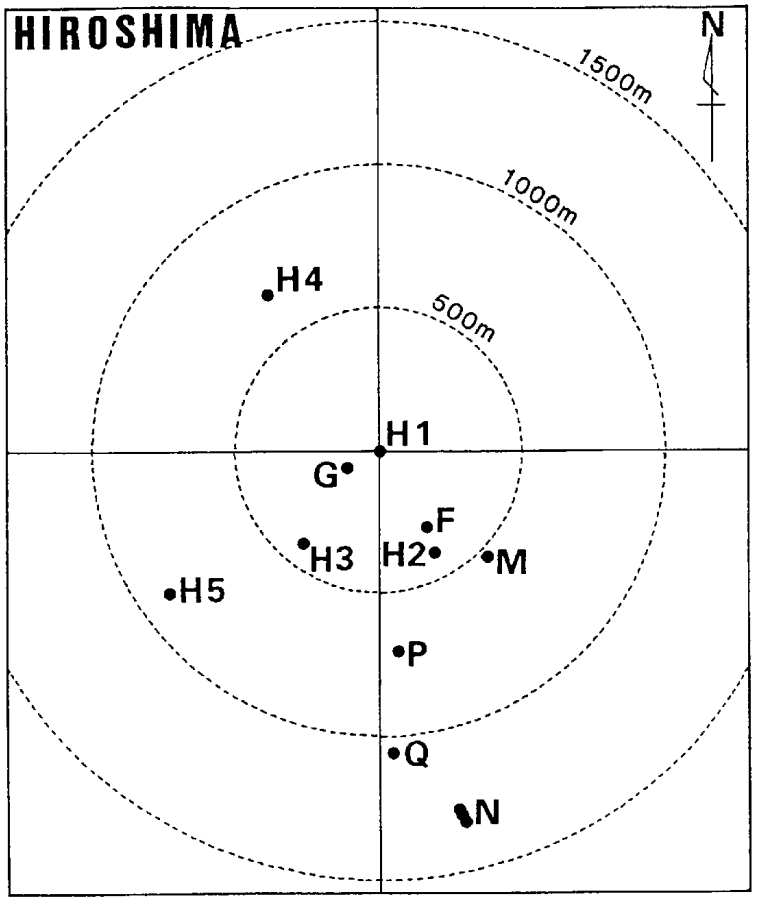

Fig. 1. Sampling locations. The concentric circles indicate ground distance from ground zero.

Map, was not used in our work, because it was said that the deviations between the newly published map and the Army Map could not be disregarded. The relatively large errors (i.e. more than 20 m) given to several values of ground distance (Table 1) are mainly attributable to the incompleteness of descriptions written for samples which were collected before the 1970's.

The samples mentioned above were each crushed and pulverized to a grain size of $<74\mu$m by the use of well checked tools.

Table 1. Samples used for measurements of residual neutron-induced radioactivities – Hiroshima –.

Sample code	Location	Sample type	Ground distance from ground zero (m)	Slant distance from the explosion point (m)
H 1	Shima Hospital	roof tile	0 + 20	580 ± 15
G	Motoyasu Bridge	rock	128 ± 5	594 ± 16
F	Fukoku Life Insurance Co. Ltd. Building	rock, concrete	320 ± 5	663 ± 16
H 2	San-yoh Memorial Hall	roof tile	410 ± 50	711 ± 41
H 3	Seigan-Ji Temple (Zaimoku-cho)	roof tile	420 ± 50	717 ± 42
M 1	Naka Telephone Exchange Office	housetop tile	533 ± 3	788 ± 13
M 2	Naka Telephone Exchange Office	wall tile	533 ± 3	788 ± 13
H 4	(Akisaya-cho)	roof tile	670 ± 90	888 ± 85
P	Chuhgoku Electric Power Co. Ltd.	wall tile	720 ± 10	925 ± 18
H 5	Mr. Tanino's House (Kawara-machi)	roof tile	880 ± 50	1054 ± 50
Q	City Hall	wall tile	1060 ± 5	1209 ± 12
N 1	Hiroshima University	housetop tile	1274 ± 5	1400 ± 11
N 2	Hiroshima University	housetop tile	1298 ± 5	1422 ± 11
N 3	Hiroshima University	housetop tile	1328 ± 5	1450 ± 11

Reagents

Unless otherwise specified below, guaranteed reagents were used in the chemistry of this work. Through a few blank runs, in which mock-up samples made of guaranteed reagents – SiO_2, Al_2O_3 and Fe_2O_3 – were processed according to the procedure mentioned below, it was confirmed that the integrated amounts of Eu and Co from the reagents and wares used in this work were less than 1.4% of the amounts of Eu and Co in the actually analyzed samples.

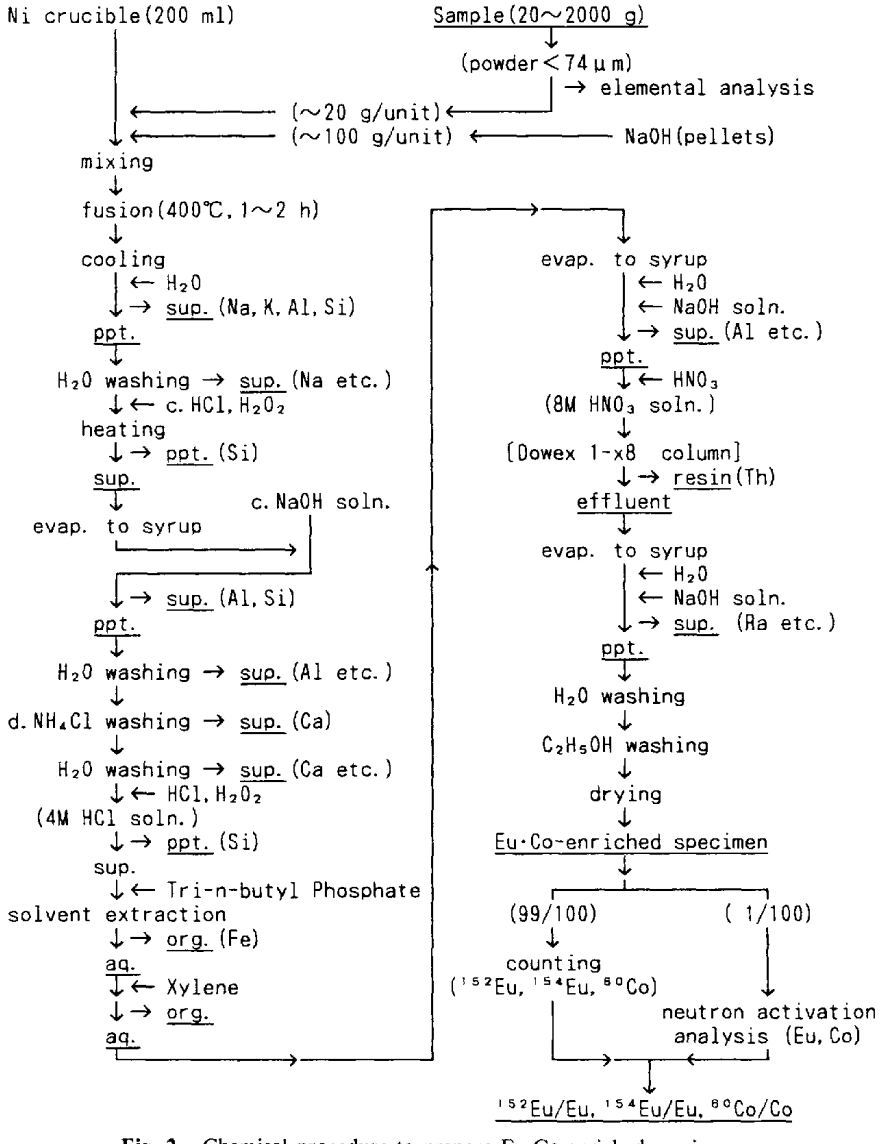

Fig. 2. Chemical procedure to prepare Eu·Co-enriched specimen.

Procedures

From each of the powdered samples of H1, F (sections), H2, H3, H4 and H5, a 20–52 g aliquot was packed in a plastic container of 54 mm inner diameter and of 19 mm depth at a density ranging from 1.4 to 1.8 g cm^{-3}. Gamma-ray and X-ray spectrometric measurements were carried out using heavily shielded Ge(Li) and/or HP-Ge detectors. Examples of the spectra were shown in previous reports[4,9]. For the determination of the counting efficiencies of the spectrometers as a function of photon energy, reference volume sources were prepared: a known amount of calibrated ^{152}Eu solution (supplied by LMRI, France) was added to 20–52 g each of powdered roof tile or rock unrelated to the atomic bombs. The residual neutron-induced radioactivities were counted for 3–10 days. Where necessary, 3–10 days counting was repeated several times.

To prepare counting specimens enriched in Eu and Co from samples of H1, G, M1, M2, P, Q, N1, N2 and N3, from 20 g to 2013 g of the powdered samples were each treated chemically according to the procedure shown in Fig. 2. The most important point of the procedure is to allow Eu and Co to coprecipitate with hydroxides at pH $\sim$11 adjusted with NaOH solution: the use of excess ammonia water must be avoided because Co forms soluble chemical species with ammonia. In the final part of the procedure, with some elements which were not removed

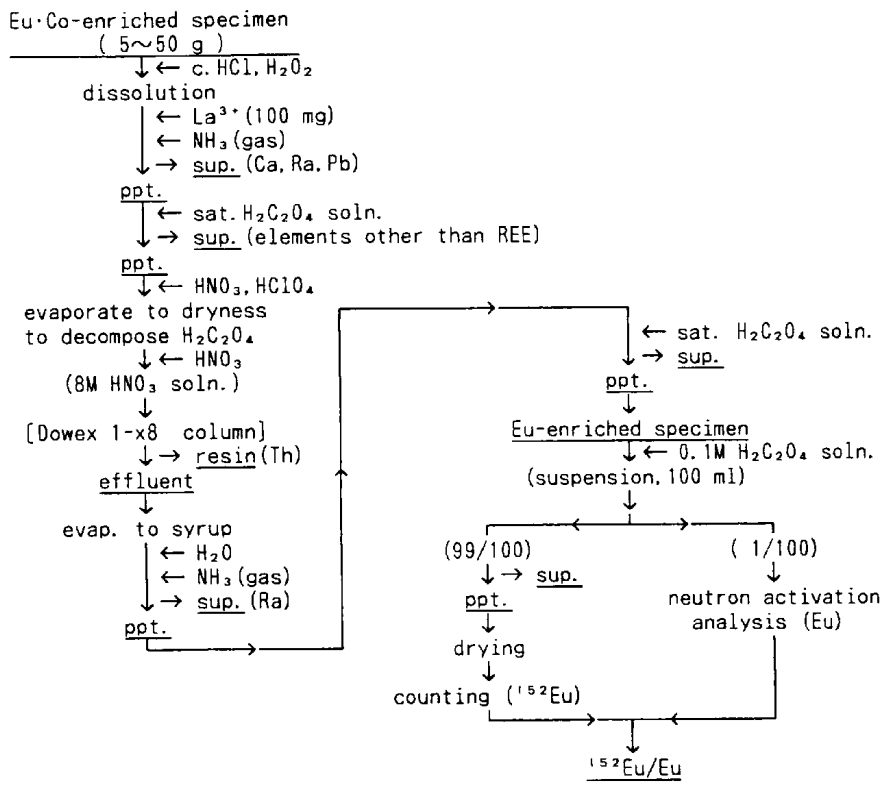

Fig. 3. Chemical procedure to prepare Eu-enriched specimen.

completely by the procedure, Eu and Co were recovered by coprecipitation with $Fe(OH)_3$, and dried. In this paper, a thusly prepared specimen (from 5 g to 50 g) is called a Eu•Co-enriched specimen. A known portion ($\sim$ 99%) of each Eu•Co-enriched specimen was formed into a disk by pressure to determine residual ^{152}Eu, ^{154}Eu and ^{60}Co simultaneously by the use of a heavily shielded Ge(Li) and/or HP-Ge γ-ray spectrometer. The counting time was given in a similar manner as above. The counting efficiencies for photons from 1173 keV to 1408 keV were measured by using mock-up samples that contained a known amount of ^{152}Eu.

Eu in the Eu•Co-enriched specimen was then further purified according to the chemical procedure shown in Fig. 3. The principal purpose of the procedure shown in Fig. 3 was to reduce further the volume of counting specimen, so as to achieve higher counting efficiency which leads to more reliable data on ^{152}Eu. In this paper, a thusly prepared specimen (from 0.05 g to 1.3 g) is called a Eu-enriched specimen. The counting of residual ^{152}Eu activity in the Eu-enriched specimen of which the main matrix was lanthanum oxalate was carried out in a similar manner to the Eu•Co-enriched specimen. Mock-up samples for the determination of counting efficiency were prepared by adding a known amount of ^{152}Eu to a known amount of lanthanum oxalate.

Elemental analysis of the pulverized samples, the Eu• Co-enriched specimens and of Eu-enriched specimens was carried out by neutron activation analysis, X-ray fluorescence analysis, and by gravimetric analysis. Oxygen content was determined by assuming oxides of the elements determined above.

Specific radioactivities of residual ^{152}Eu, ^{154}Eu and ^{60}Co at the time immediately after the nuclear explosion were calculated by using the measured results and the nuclear data* (*see Table 1 of Reference 9).

RESULTS

Throughout the chemical procedure shown in Fig. 2, the recovery of Eu was in the range 61 to 92% (average, 72%), and that of Co was in the range 56 to 88% (average, 65%). The recovery of Eu from the Eu•Co-enriched specimen through the chemical procedure shown in Fig. 3 was in the range 21 to 100% (average, 82%). By these chemical procedures. Eu and Co in the pulverized samples were concentrated in a much reduced volumes: the geometry of counting of the residual neutron-induced radioactivity thereupon was much improved. Moreover, effective removal was achieved of major natural radionuclides such as ^{40}K, ^{208}Tl and ^{214}Bi which interfered with the counting of residual neutron-induced radioactivity. As a result of the increase in the counting efficiency and of the decrease in noise level (i.e. Compton background), the factor of merit in counting the residual neutron-induced radioactivity was largely improved. Throughout the procedures which are shown in Figs. 2 and 3, ^{227}Ac (half-life: 21.77 y) – a natural radionuclide – could not be removed from Eu fraction.

Data for ^{152}Eu/Eu (i.e. specific radioactivity of ^{152}Eu) in the surface layers (< 3 cm thick) of the samples are given in Table 2 together with $\pm 1\sigma$ counting errors. The table includes both previous and new data by us. Where measurements of ^{152}Eu/Eu in one sample were carried out for more than two of the pulverized specimen, Eu• Co-enriched specimen and Eu-enriched

Table 2. Specific radioactivity of ^{152}Eu immediately after the nuclear explosion in Hiroshima.

Sample code	Specimen -Detector -Photopeak*	$^{152}Eu/Eu$ (Bq/mg) Measured: Individual value	Measured: Average value	Calculated[10]	Calc. / Meas. (Av.)
H 1	O – L – 39.5, 40.1	110.3 ±6.2			
	P – G – 1408	114.2 ±7.7			
	S – L – 122	110.5 ±7.8	111.7 ±4.2	175	1.57 ± 0.06
G	P – G – 1408	101.5 ±4.8			
	S – L – 122	93.2 ±6.7	97.4 ±4.1	145	1.49 ± 0.06
F	O – L – 39.5, 40.1	75.9 ±4.5	75.9 ±4.5	73	0.96 ± 0.06
H 2	O – L – 39.5, 40.1	22.1 ±2.2	22.1 ±2.2	45	2.0 ±0.2
H 3	O – L – 39.5, 40.1	26.7 ±2.1	26.7 ±2.1	43	1.6 ±0.1
M 1	P – L – 39.5, 40.1	14.13 ±0.79			
	S – L – 122	13.4 ±1.1	13.8 ±0.7	21	1.5 ±0.08
M 2	P – L – 39.5, 40.1	14.7 ±1.2			
	S – G – 1408	21.51 ±0.97			
	S – L – 122	21.8 ±1.2	19.3 ±0.7	21	1.1 ±0.04
H 4	O – L – 39.5, 40.1	4.5 ±2.1	4.5 ±2.1	7.7	1.7 ±0.8
P	P – L – 39.5, 40.1	5.3 ±1.4			
	S – L – 122	4.3 ±1.1	4.8 ±0.9	5.5	1.1 ±0.2
H 5	O – L – 39.5, 40.1	3.5 ±1.1	3.5 ±1.1	1.5	0.43 ± 0.13
Q	P – L – 39.5, 40.1	1.02 ±0.51			
	S – G – 1408	1.15 ±0.13			
	S – L – 122	1.15 ±0.15	1.11 ±0.18	0.37	0.33 ± 0.05
N 1	P – L – 39.5, 40.1	0.36 ±0.14			
	S – G – 1408	0.41 ±0.18			
	S – L – 122	0.48 ±0.24	0.42 ±0.11	0.057	0.14 ± 0.04
N 2	P – L – 39.5, 40.1	0.34 ±0.12	0.34 ±0.12	0.047	0.14 ± 0.05
N 3	P – L – 39.5, 40.1	0.114 ± 0.037	0.114 ± 0.037	0.035	0.31 ± 0.10

* Specimen (O: powdered specimen; P: Eu•Co-enriched specimen; S; Eu-enriched specimen)
-Detector (G: Ge(Li) or HP-Ge; L: low-energy photon detector)
-Photopeak (in keV)

specimen, the measured values were averaged. The averaged value was plotted as a function of slant range from the explosion point (Fig. 4). The averaged value was then compared with calculated value[10]. The ratio of calculated value to measured value was also plotted as a function of slant range from the explosion point (Fig. 5).

Simultanous determination of ^{152}Eu, ^{154}Eu and ^{60}Co was possible for samples H1 and G. The numerical values of the specific radioactivities were given in the previous report[9]. Since ^{152}Eu, ^{154}Eu and ^{60}Co were induced principally by thermal and epithermal neutrons, the specific radioactivity – A/W in the following equation – of ^{152}Eu, for example, at the time immediately after the nuclear explosion is given by the expression:

$$A/W = (10^{-3} \, L \, X \, \lambda / A_r) \cdot (\Phi_{th} \sigma_{th} + \Phi_{epi} I);$$

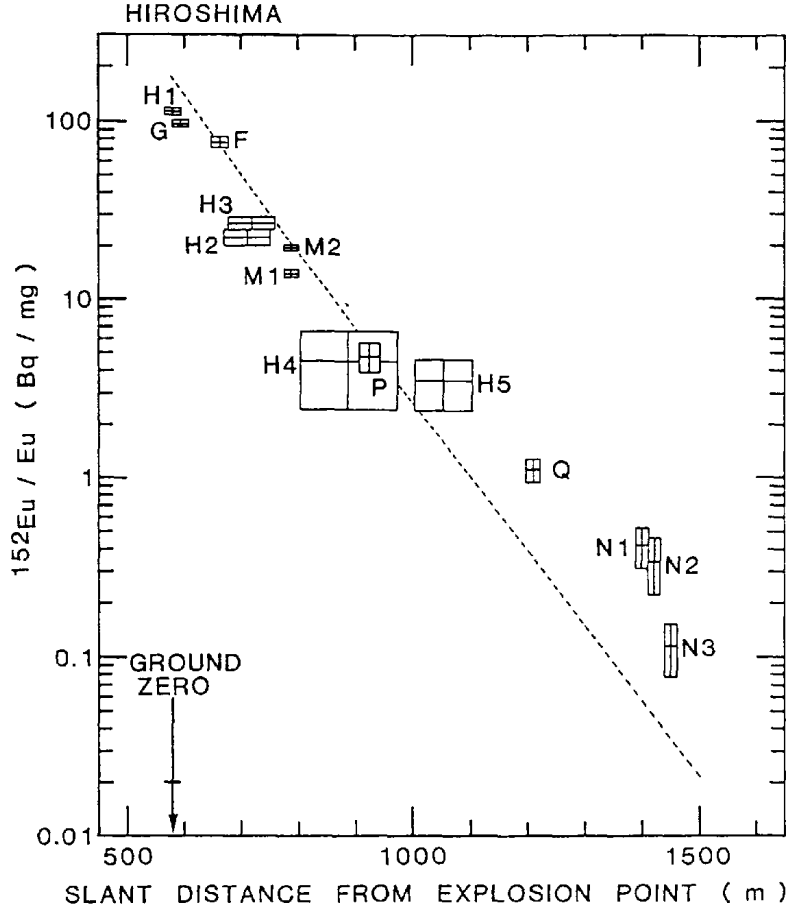

Fig. 4. Specific radioactivity of ^{152}Eu immediately after the nuclear explosion. The dashed line indicates the calculated results[10].

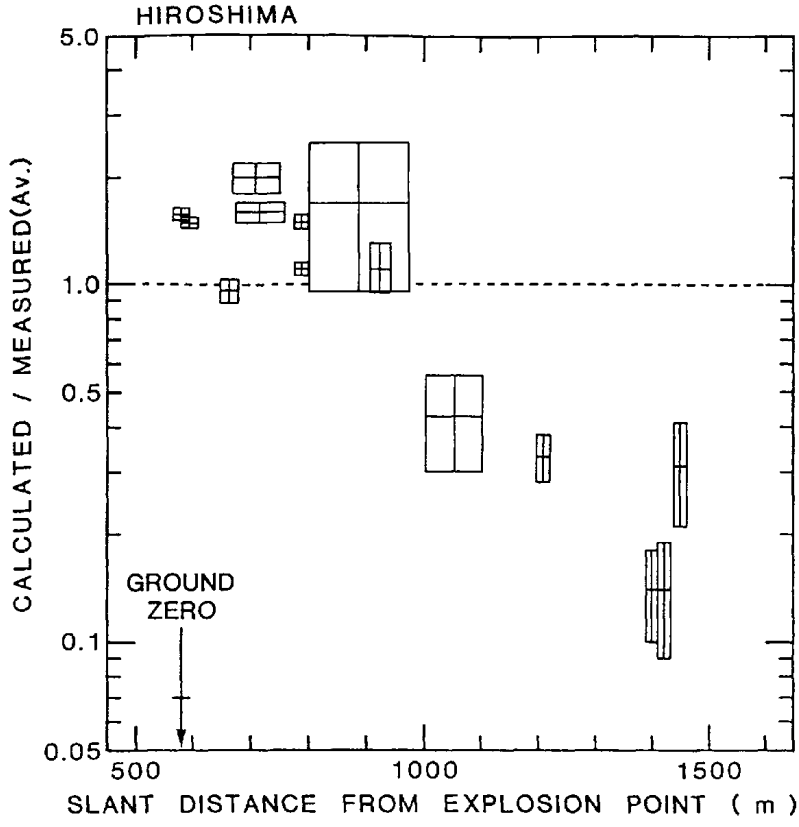

Fig. 5. Comparison of measured and calculated[10] values of specific radioactivity of ^{152}Eu immediately after the nuclear explosion.

where A in Bq (i.e. s^{-1}) is the induced radioactivity of ^{152}Eu at the time immediately after the nuclear explosion; W in mg is the mass of the target element, Eu, from which the ^{152}Eu was induced; L in mol^{-1} is the Avogadro's constant; X is the natural isotopic abundance of the target nuclide, ^{151}Eu, in atomic fraction basis: λ in s^{-1} is the decay constant of induced radionuclide, ^{152}Eu; A_r in g mol^{-1} is the atomic weight of the target element, Eu; Φ_{th} and Φ_{epi} in cm^{-2} are the thermal and epithermal neutron fluences, respectively; and σ_{th} and I in cm^2 are the thermal neutron cross section and resonance integral for the ^{151}Eu(n, γ) ^{152}Eu reaction, respectively. Since ^{151}Eu, ^{153}Eu and ^{59}Co differ from each other in the ratio of σ_{th}/I for the (n, γ)-reaction, solving the above-mentioned type of simultaneous equations for ^{152}Eu, ^{154}Eu and ^{60}Co allow the calculation of Φ_{th} and Φ_{epi}. A graphical method of solving the simultaneous equations was shown in the previous report[9]. Referring to the graphical method in which counting errors were considered, minimum and maximum values of Φ_{th} and Φ_{epi} were calculated. The results are given in Table 3 together with results calculated by DS86 methodology[11].

The data for depth distribution of ^{152}Eu/Eu in the core sample F was given in the previous

Table 3. Comparison of measured and calculated thermal – and epithermal – neutron fluences.

Sample		Measured	Calculated	Calc./Meas.
H 1	Φ_{epi}	$(1.4 \pm 0.7) \times 10^{12}$	1.51×10^{12}	1.1 ±0.5
	Φ_{th}	$(5.0 \pm 0.8) \times 10^{12}$	8.85×10^{12}	1.8 ±0.3
	Φ_{epi}/Φ_{th}	0.28 ± 0.15	0.171	0.61 ± 0.33
G	Φ_{epi}	$(1.6 \pm 0.7) \times 10^{12}$	1.14×10^{12}	0.71 ± 0.31
	Φ_{th}	$(4.5 \pm 0.8) \times 10^{12}$	7.02×10^{12}	1.6 ±0.3
	Φ_{epi}/Φ_{th}	0.36 ± 0.17	0.162	0.45 ± 0.21

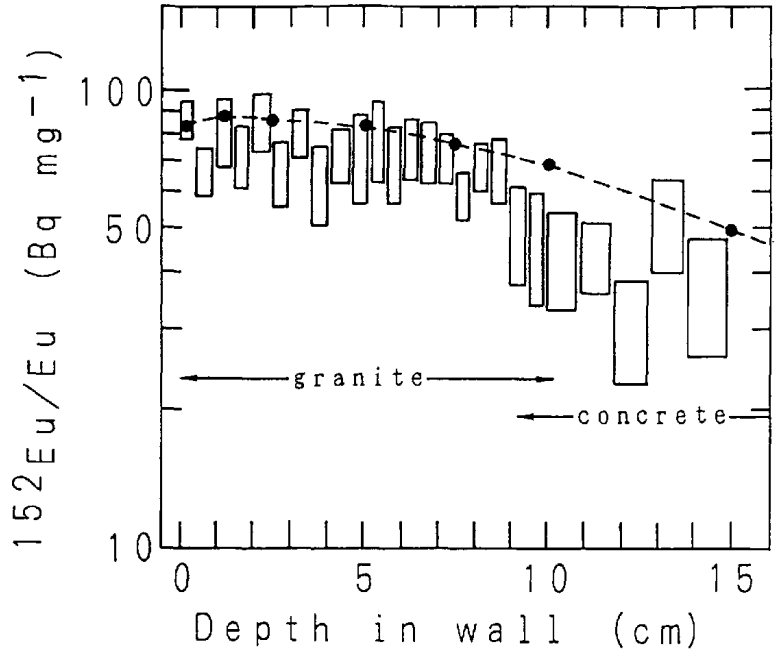

Fig. 6. Measured and calculated depth distribution of ^{152}Eu/Eu in a core sample from the outer wall of the Fukoku Life Insurance Co. Ltd. Building (ground range: 320 m) at the time immediately after the nuclear explosion.
---●--- : calculated values with dashed line for eye guide.

report together with the data for elemental composition of the core sample[9]. The measured depth profile of ^{152}Eu/Eu is reproduced in Fig. 6 to compare with the calculated result.

DISCUSSION

^{152}Eu/Eu as a function of slant range

In the previous paper[9], we reported that our data for ^{152}Eu/Eu in the surface layers of samples exposed to the nuclear explosion in Hiroshima were roughly consistent with the calculated values. Most of the improved data for ^{152}Eu/Eu presented above, however, clearly show a marked difference from calculated results (Fig. 5); close agreement is found between measured and calculated values only in the slant ranges between 650 m and 950 m (i.e. between 320 m and 720 m in the ground range). The ratio between calculated value and measured value showed a rather systematic variation with slant range: the calculated/measured ratios for the specific radioactivity of ^{152}Eu were larger than unity in the vicinity of ground zero and smaller than unity at locations more than 1000 m from ground zero. It is worth noting that the result shown in Fig. 5 generally has a tendency similar to the systematic variation as discussed for ^{60}Co data[8,12]. Although the improved data presented above are still accompanied by relative errors from 4% to 50%, and from 10% to 20% of unavoidable relative uncertainties were estimated for the calculation by DS86 methodology, we should like to point out that in DS86 methodology for Hiroshima there exists a systematic error in the calculations for neutrons responsible for the activation of ^{151}Eu. Neutrons which activated ^{151}Eu were principally at, or near, thermal energies. Neutrons at these energies therefore contribute only a small fraction of the total neutron kerma, but they reflect the intensities of these same neutrons at a point closer to ground zero where their energies were higher and they contributed significantly to the kerma. Moreover, we should like to mention that more data on ^{152}Eu for the locations farther than 1 km from ground zero of Nagasaki Atomic Bomb will be of key importance in the examination of neutrons for Hiroshima.

Φ_{th} and Φ_{epi} in the surface of H1 and G

In the previous paper[9], we presented data for simultaneously determined ^{152}Eu/Eu, ^{154}Eu/Eu and ^{60}Co/Co in the surface layers of H1 and G to conclude that not only Φ_{th} and Φ_{epi} but also Φ_{epi}/Φ_{th} showed marked differences from calculated results. Shortly afterward it was noted, however, that our calculation of DS86 Φ_{epi} had been erroneous[11]. We again compared our data on Φ_{epi} with the recommended DS86 Φ_{epi}[11]. As shown in Table 3, a close agreement was thus found between the calculated and measured values for Φ_{epi} while the calculated Φ_{th} was still larger than measured values. In the preceding discussion, we have pointed out that there exists a systematic error in the calculations for all neutrons responsible for the activation of ^{151}Eu in the vicinity of ground zero in Hiroshima; it is now pointed out that a significant error was identified in the calculation for thermal neutrons.

For the purpose of simultaneous determination of ^{152}Eu/Eu, ^{154}Eu/Eu and ^{60}Co/Co with smaller errors, we are continuing a study on a steel sample ($\sim$1.6 kg) from Aioi Bridge which

was exposed to the Hiroshima Atomic Bomb at the location 288 m from ground zero.

Depth distribution of ^{152}Eu/Eu in the core sample F

In the previous paper[9], we showed a preliminary result of analysis of the depth distribution of ^{152}Eu/Eu in core sample F. Recently, we obtained a result by DOT-MORSE coupling calculation for ^{152}Eu/Eu in the core sample F, and the measured result was compared with the calculated result (Fig. 6). In this case the calculated values fall approximately within the measured values: the calculation of neutrons by DS86 methodology can approximately reproduce the measured results at the point 320 m from ground zero (i.e. 663 m in slant range) in Hiroshima.

Thus, our results confirm the existence of a systematic error in the DS86 methodology for Hiroshima, especially in the calculations for thermal neutrons in the vicinity of ground zero (ground range < 320 m) and at locations more than 1000 m apart from ground zero.

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