# Chapter 5 Appendix 14

# RESIDUAL NEUTRON-INDUCED RADIO-ACTIVITIES IN SAMPLES EXPOSED IN HIROSHIMA

Takashi Nakanishi, Kenji Kobayashi, Takuo Yamamoto, and Kunio Miyaji Kanazawa University

Among the radionuclides that have been detected in materials exposed to the nuclear explosions in Hiroshima and Nagasaki, <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co can be measured even today by using high-sensitivity gamma-ray counting techniques. <sup>1,2</sup> Since <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co are produced principally from stable isotopes of europium and cobalt by neutron capture reactions, they are useful radionuclides for checking the validity of a series of computer calculations employed for the reassessment of atomic bomb neutron dosimetry in Hiroshima and Nagasaki (Chapter 5). The use of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co for the reassessment of Abomb 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 nuclear explosions; 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 the present work, for the purpose of obtaining information on fast neutron fluence in Hiroshima, the depth distribution of the activity of <sup>152</sup>Eu to the abundance of stable Eu, <sup>152</sup>Eu/Eu, was studied, using a thick sample exposed to the Hiroshima A-bomb. Furthermore, a proposal to analyze thermal and epithermal neutron fluences from a set of data of <sup>152</sup>Eu/Eu, <sup>154</sup>Eu/Eu, and <sup>60</sup>Co/Co was examined, using samples exposed to the Hiroshima A-bomb in the vicinity of the hypocenter. For the reliable and simultaneous determination of <sup>152</sup>Eu/Eu, <sup>154</sup>Eu/Eu, and <sup>60</sup>Co/Co, chemical extraction of Eu and Co from the exposed sample was studied. The procedure to prepare samples enriched in Eu and Co was also applied to samples exposed to the Hiroshima A-bomb at locations 1300 m from the hypocenter, because determination of <sup>152</sup>Eu is extremely difficult without chemical enrichment of Eu from samples exposed to the A-bomb at locations greater than 1000 m from the hypocenter.

Samples. The following samples exposed to the Hiroshima A-bomb were supplied to this

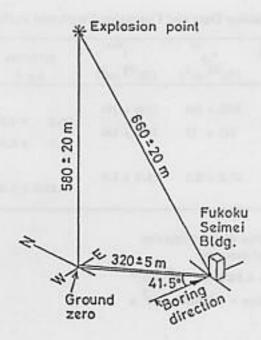


Figure 1. Relative position of the Fukoku Seimei Building to the explosion point and ground zero of Hiroshima A-bomb

work by courtesy of the Research Institute for Nuclear Medicine and Biology, Hiroshima University:

- Core sample from the outer wall of the Fukoku Seimei Building (320 m from the hypocenter, see Figure 1; 10 cm in diameter and 22.6 cm in length);
- 2. Roof tile of Shima Hospital (hypocenter; 60 g supplied);
- Surface layer of granite newel of Motoyasu Bridge (128 m from the hypocenter; 250 g supplied);
- Housetop tiles of Naka Denwa-Kyoku (533 m from the hypocenter; two separate samples, weighing 180 and 470 g, were supplied); and
- Housetop tiles of Hiroshima University (from about 1255 to 1312 m from the hypocenter; three samples, weighing 590, 570, and 570 g, were supplied).

The core sample (1) was bored on the wall facing the hypocenter at a position 6 m above the ground. The surface layer of the wall was made of granite 10 cm in thickness, and the inner part was made of concrete. For the present work, the granite layer of the core was sliced at 5 mm intervals and the concrete layer at 10 mm intervals.

The samples mentioned above were each crushed and pulverized to a grain size of  $< 74 \mu m$ .

Procedures. For the determination of <sup>152</sup>Eu in the core sample (1), 27 to 52 g each of the pulverized samples was packed in a plastic container having an inner diameter of 54 mm and a depth of 19 mm at a density ranging from 1.4 to 1.8 g cm<sup>-3</sup>. X rays attributable to <sup>152</sup>Eu (39.52 and 40.12 keV) were counted by the use of a heavily-shielded high-purity germanium low-energy photon spectrometer. To determine the counting efficiency of the spectrometer for 39.52 and 40.12 keV photons from the volume sample, reference volume

Table 1. Nuclear Data and Constants Employed in this We	Table 1.	Nuclear Data ar	d Constants E	Employed	in this Work
---------------------------------------------------------	----------	-----------------	---------------	----------	--------------

Element. Isotope	Hatural isotopic abundance (atom %)	<sup>σ</sup> th (10 <sup>-24</sup> cn <sup>2</sup> )	o <sub>th</sub> I		y-ray (photons/100-decay -events)
Eu(151.96*) Eu-151 Eu-152 Eu-153 Eu-154	47.9 52.1	5900 ± 200 320 ± 12	3700 ± 340 1635 ± 200	13.2 ± 0.3 8.5 ± 0.5	39.52, 40.12 keV(45.00) 1408.03 keV(21.21) 1274.80 keV(35.50)
Co(58.9332*) Co-59 Co-60	100	37.2 ± 0.2	75.5 ± 1.5	5.2719 ± 0.00	11 1173.23 keY(99.86) 1332.51 keY(99.98)

<sup>\*</sup> atomic weight (g mol-1)

Avogadro constant = (6.022045 ± 0.000031) x 10<sup>23</sup> mol-1

sources were prepared by adding a known amount of <sup>152</sup>Eu standard solution (available from LMRI, France) to about 27 to 52 g each of powdered granite or concrete unrelated to the A-bomb.

For samples (2) to (5), a chemical procedure was emplyed to prepare counting specimens enriched in Eu and Co. The pulverized samples were each fused with NaOH at 400°C for one hour, and the resulting cake was taken up in distilled water. Then, chemical separation of Eu and Co from major elements (Si, Al, Na, K, CA, and Fe) and Th was carried out in a usual way. Eu and Co, with other elements that were not removed in the chemical procedure, were then recovered by coprecipitation with Fe(OH)<sub>3</sub>, dried, and formed into a disk by pressure.

The simultaneous gamma-ray counting of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co in the specimen enriched in Eu and Co was carried out by the use of a heavily shielded Ge(Li) gamma-ray spectrometer. The counting efficiencies for 1173 to 1408 keV photons were measured using mock-up samples that contained known amounts of <sup>152</sup>Eu.

Elemental analysis of the samples was carried out by nondestructive neutron activation analysis (Na, Al, K, Sc, Mn, Fe, Co, Cs, La, Ce, Eu, and Tb), by x-ray fluorescence analysis (Si, K, Ca, Ti, and Fe), and by thermogravimetric analysis (H<sub>2</sub>O). Oxygen was determined by assuming it was present as oxides of the elements determined above.

Specific activities of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co at the time immediately after the nuclear explosion were calculated by using the measured results and the nuclear data listed in Table 1.

## Results and Discussion

Examples of the gamma-ray spectra are shown in Figures 2 and 3. Data for <sup>152</sup>Eu and Eu in the core sample (1) are given in Table 2 together with the standard deviations due to counting statistics. The depth distribution of <sup>152</sup>Eu/Eu in the core sample (Figure 4) showed a significant decrease of A-bomb neutrons, which induced <sup>152</sup>Eu, at depths more than about 7 cm in the wall. The decreasing part of the <sup>152</sup>Eu/Eu curve in the concrete layer was then compared with the calculated attenuation curve of thermal neutron fluence (solid and

<sup>\*\*</sup> thermal neutron cross section of (n,y)-reaction

<sup>\*\*\*</sup> resonance integral of (n,y)-reaction

<sup>1</sup> year (sidereal) = 365.25636 days = 3.1558150 x 107 s

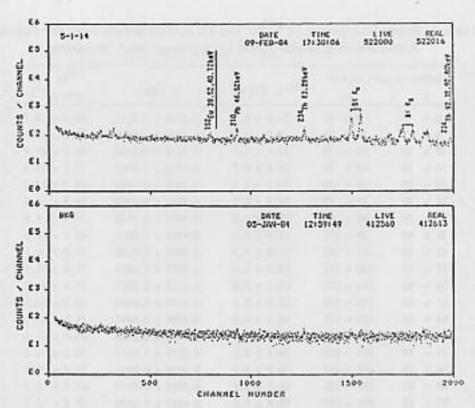


Figure 2. Photon spectra (with low-energy photon spectrometer) of wall sample from the Fukoku Seimei Building (upper) and background (lower)

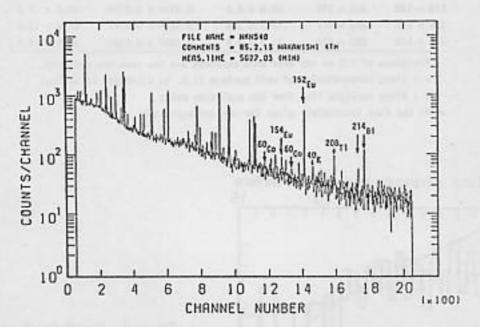


Figure 3. Gamma-ray spectrum with Ge(Li) coaxial detecter of Eu-and-Coenriched specimen prepared from granite of Motoyasu Bridge

broken semilogarithmic-linear curves in the lower part of Figure 4). The attenuation curves of thermal neutron fluence were drawn on the basis of the calculated diffusion lengths of thermal neutrons in the granite and concrete layers, respectively, where the diffusion lengths were evaluated from attenuation coefficients of thermal neutrons in the core sample (summed values in Table 3). The decreasing part of the <sup>152</sup>Eu/Eu curve was found to be consistent with the calculated attenuation of thermal neutrons in the concrete layer; this implies that the

Table 2. Depth Distribution of <sup>152</sup>Eu and Eu in Wall Sample from the Fukoku Seimei Building (about 320m from ground zero), Hiroshima

Dept	h in s	rall (mm)* b	<sup>152</sup> Eu (Bq kg <sup>-1</sup> )**	Eu (ppm)	<sup>152</sup> Eu / Eu (Bq mg <sup>-1</sup> )**
0 +	3	0 ~ 10	30.8 ± 2.9	0.3598 ± 0.0062	85.6 ± 8.2
4 .	9	12 ~ 25	24.9 ± 3.0	0.3764 ± 0.0064	66.2 ± 8.1
10 %	14	28 ~ 39	27.7 ± 4.6	0.3414 ± 0.0138	81.1 ± 13.9
15 %	19	42 ~ 52	29.4 ± 4.7	0.4099 ± 0.0061	71.7 ± 11.5
20 ~	25	54 ∼ 68	30.7 ± 4.4	0.3610 ± 0.0089	85.0 ± 12.3
26 %	30	71 ~ 82	26.4 ± 4.2	0.4004 ± 0.0160	65.9 ± 10.8
31 ~	35	85 ~ 96	31.6 ± 3.8	0.3954 ± 0.0125	79.9 ± 9.8
36 ∿	40	99 ~ 111	25.5 ± 5.0	0.4101 ± 0.0070	62.2 ± 12.2
41 ~	46	114 ~ 127	27.9 ± 3.7	0.3894 ± 0.0128	71.6 ± 5.7
47 ~	51	130 ~ 141	23.3 ± 4.9	0.3252 ± 0.0224	71.6 ± 15.9
52 ~	56	143 ~ 153	24.7 ± 5.0	0.3173 ± 0.0081	77.8 ± 15.9
57 ~	60	155 ~ 165	22.6 ± 4.4	0.3260 ± 0.0045	69.3 ± 13.4
61 ~	65	168 ~ 178	26.3 ± 4.0	0.3554 ± 0.0071	74.0 ± 11.3
66 ~	70	181 ~ 192	27.4 ± 4.1	0.3752 ± 0.0180	73.0 ± 11.4
71 ~	74	195 ~ 204	25.1 ± 3.2	0.3713 ± 0.0147	70.3 ± 9.2
75 %	79	207 ~ 217	23.2 ± 2.9	0.3975 ± 0.0055	58.4 ± 7.2
≥ 08	84	220 ~ 231	26.7 ± 3.1	0.3956 ± 0.0179	67.5 ± 8.3
85 %	89	233 ~ 245	28.8 ± 4.6	0.4343 ± 0.0108	66.3 ± 10.7
90 -	94	247 ~ 259	20.2 ± 4.9	0.4128 ± 0.0160	48.9 ± 11.9
95 *	99	261 ~ 273	15.1 ± 4.2	0.3260 ± 0.0074	46.3 ± 12.9
100 ~	108	275 ~ 298	13.9 ± 3.3	0.3216 # 0.0128	43.2 ± 10.3
109 ~	117	300 ~ 323	14.7 ± 2.6	0.3407 ± 0.0076	43.1 ± 7.7
118 ~	128	325 ~ 351	10.0 ± 2.6	0.3314 ± 0.0069	30.2 ± 7.8
29 ~	137	353 ~ 377	16.9 ± 3.8	0.3277 ± 0.0143	51.6 ± 11.9
38 ~	149	380 ~ 410	11.9 ± 3.4	0.3262 ± 0.0100	36.5 ± 10.5

<sup>\*</sup> Fractions of 0.5 mm and over were reckoned and the rest was cut away.

<sup>..</sup> At the time immediately after the nuclear explosion.

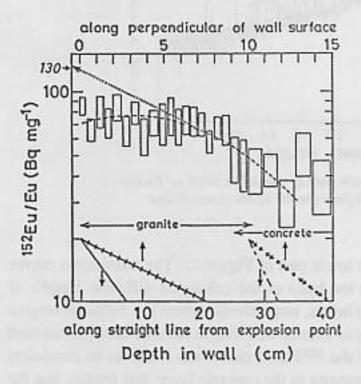


Figure 4. Depth distribution of 152Eu/Eu in wall sample from the Fukoku Seimei Building immediately after the nuclear explosion in Hiroshima

a : along perpendicular of wall surface (i.e. in direction of boring)

b : along straight line from the explosion point

Table 3. Element Composition of Wall Sample from the Fukoku Seimei Building and Calculated Attenuation Coefficient (Σ) for Thermal Neutron in the Wall

		Atomic cross for thermal r	eutrons	In granite (	o = 2.693 g c	m <sup>-3</sup> )	In concrete (* - 2.19) g cm <sup>-3</sup> )		
	4, (g no1 <sup>-1</sup> )	(10-24	m <sup>2</sup> )	Mean concentration* (in weight fraction)	Ia (cn-1)	Is (cn*1)	Mean concentration*	fa (cm <sup>-1</sup> )	I <sub>3</sub> (cm <sup>-1</sup> )
H	1,008	0.332	81.5	2.16(13) x 10 <sup>-4</sup>	1.15 x 10 <sup>-4</sup>	2.83 x 10 <sup>-2</sup>	(in weight fraction) 4.57 (23) x 10 <sup>-3</sup>	1.99 x 10 <sup>-3</sup>	4.88 x 10 <sup>-1</sup>
							9454 C 44 4 5 Miles 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1		
0	15.00	0.00027	4.24	4.71(13) x 10 <sup>-1</sup>	1.29 x 10 <sup>-5</sup>	2.20 x 10 <sup>-1</sup>	4.47 (14) x 10 <sup>-1</sup>	9.95 x 10 <sup>-6</sup>	1.56 x 10 <sup>-1</sup>
Na	22.99	0.53	3.4	2.53(17) x 10 <sup>-2</sup>	9.45 x 10-4	6.07 x 10 <sup>-3</sup>	8.74 (57) x 10 <sup>-3</sup>	2.66 x 10-4	1.71 x 10 <sup>-3</sup>
Al	26.98	0.232	1.51	6.56/60) x 10-2	9.15 x 10-4	5.95 x 10 <sup>-3</sup>	3.52 (35) × 10 <sup>-2</sup>	3.99 x 10 <sup>-4</sup>	4.32 x 10 <sup>-3</sup>
51	28.09	0.160	2.4	3.39(30) x 10 <sup>-1</sup>	3.13 x 10 <sup>-3</sup>	4.70 x 10 <sup>-2</sup>	2.020/58) x 10-1	1.52 x 10 <sup>-3</sup>	2.28 x 10 <sup>-2</sup>
E	39.10	2.1	2.2	4.01(37) x 10 <sup>-2</sup>	3.49 x 10 <sup>-3</sup>	3.66 x 10 <sup>-3</sup>	1.91 (52) x 10-2	1.35 x 10 <sup>-3</sup>	1.42 x 10 <sup>-3</sup>
Ca	40.08	0.43	3.2	6.92(49) x 10 <sup>-3</sup>	1.20 x 10-4	8.96 x 10-4	2.56 (28) x 10-1	3.62 x 10 <sup>-3</sup>	2.70 x 10-2
Se	44.96	26.5	24	3.09(40) x 10 <sup>-5</sup>	2.95 x 10 <sup>-6</sup>	2.67 x 10 <sup>-6</sup>	3.25 (20) x 10 <sup>-5</sup>	2.53 x 10 <sup>-6</sup>	2.29 x 10 <sup>-6</sup>
71	47.88	5.1	4.4	5.86(85) x 10-4	1.21 x 10 <sup>-4</sup>	8.73 x 10 <sup>-5</sup>	7.2 (39) x 10 <sup>-4</sup>	1.23 x 10 <sup>-4</sup>	8.85 x 10 <sup>-5</sup>
Hn	14,94	13.3	2.0	2.67((0) x 10-4	1.05 x 10-4	1.58 x 10 <sup>-5</sup>	4.43 (22) x 10 <sup>-4</sup>	1.41 x 10 <sup>-4</sup>	2.13 x 10-5
Fe	55.85	2.55	11.8	8.8 (32) x 10 <sup>-3</sup>	6.51 x 10 <sup>-4</sup>	3.01 x 10 <sup>-3</sup>	1.310/#37 x 10 <sup>-2</sup>	7.89 x 10 <sup>-4</sup>	3.65 x 10 <sup>-3</sup>
Co	58.93	37.2	6	5.65(10) x 10-7	5.78 x 10 <sup>-7</sup>	9.33 x 10 <sup>-8</sup>	5.49 (44) x 10 <sup>-6</sup>	4.74 x 10 <sup>-6</sup>	7.64 x 10-7
Cs	132.9	29	7	4.97(40) x 10-6	1.76 x 10 <sup>-6</sup>	4.25 x 10"7	2.11 (25) x 10 <sup>-6</sup>	6.07 x 10 <sup>-7</sup>	1.47 x 10-7
Li	138.9	9.2	9.3	1.72(43) x 10 <sup>-5</sup>	1.85 x 10 <sup>-6</sup>	1.87 x 10 <sup>-6</sup>	1.55 (72) x 10 <sup>-5</sup>	1.35 x 10 <sup>-6</sup>	1.37 x 10 <sup>-6</sup>
Ce	140.1	0.43	2.8	3.93(43) x 10 <sup>-5</sup>	2.87 x 10 <sup>-7</sup>	1.27 x 10 <sup>-6</sup>	1.937(##) x 10 <sup>-5</sup>	1.15 x 10 <sup>-7</sup>	5.11 x 10-7
Eu	152.0	4600	8	3.75(33) x 10 <sup>-7</sup>	1.84 x 10 <sup>-5</sup>	3.20 x 10 <sup>-8</sup>	3.315(65) x 10 <sup>-7</sup>	1.32 x 10-5	2.30 x 10 <sup>-8</sup>
Th	158.9	25.5	7	1.82(27) x 10 <sup>-6</sup>	4.74 x 10 <sup>-7</sup>	1.30 x 10 <sup>-7</sup>	4.15 (72) x 10 <sup>-7</sup>	8.79 x 10 <sup>-8</sup>	2.41 x 10 <sup>-8</sup>
sum				0.958 ± 0.018	9.63 x 10 <sup>-3</sup>	0.297	0.987 ± 0.032	1.02 x 10 <sup>-2</sup>	0.705

For example, 2.16/33) x 10<sup>-6</sup> stands for (2.16 ± 0.13)x10<sup>-4</sup>, 8.8/33) x 10<sup>-3</sup> for (8.8 ± 1.1)x10<sup>-3</sup>, anabsorption. Excuttering

neutrons entering the outer wall surface of the Fukoku Seimei Building were well thermalized at a depth of about 7 cm from the surface. Preliminary analysis of the depth distribution of  $^{152}$ Eu/Eu has led us to estimate that 60 to  $90\,\mathrm{Bq\,mg^{-1}}$  of  $^{152}$ Eu/Eu, measured at the wall surface, would result from thermal and epithermal neutrons at the wall surface. Another 40 to  $70\,\mathrm{Bq\,mg^{-1}}$  of  $^{152}$ Eu/Eu, roughly estimated by extrapolating the decreasing part of the  $^{152}$ Eu/Eu curve at a depth of about 7 cm back to the surface along the attenuation curve of thermal neutrons in the granite layer, would be added to the 60 to  $90\,\mathrm{Bq\,mg^{-1}}$ , if every neutron entering the wall surface were a thermal neutron. To produce 60 to  $90\,\mathrm{Bq\,mg^{-1}}$  of  $^{152}$ Eu/Eu requires a thermal and epithermal (neutron energy  $\leq 1\,\mathrm{keV}$ ) neutron fluence of 3.2 to  $4.8 \times 10^{12}\,\mathrm{cm^{-2}}$ ; to produce the 40 to  $70\,\mathrm{Bq\,mg^{-1}}$  requires 2.2 to  $3.8 \times 10^{12}\,\mathrm{cm^{-2}}$  of fast or intermediate neutrons (neutron energy  $\geq 1\,\mathrm{keV}$ ). Although the analysis given here is preliminary, the results are consistent with the neutron spectrum calculated at a position  $250\,\mathrm{m}$  from the hypocenter in Hiroshima.

The simultaneous determination of <sup>152</sup>Eu/Eu, <sup>154</sup>Eu/Eu, and <sup>60</sup>Co/Co was performed using the following counting specimens enriched in Eu and Co: a counting specimen of about 6.0 g, which contained Eu of 4.2 ppm and Co of 64 ppm and was obtained from about 20 g of the roof tile of Shima Hospital (sample (2), original concentration of Eu and Co were 1.4 ppm and 23 ppm, respectively). Another counting specimen of 5.5 g, which contained Eu of 32 ppm and Co of 37 ppm and was obtained from 200 g of granite of Motoyasu Bridge (sample (3), original concentration of Eu and Co were 0.89 ppm and 1.6 ppm, respectively). Effective removal of <sup>40</sup>K, <sup>208</sup>Tl, and <sup>214</sup>Bi, which interfered with the counting of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co radioactivities at extremely low levels, from the counting specimens was

Table 4. Neutron-induced Radioactivity and Target Elements of (n,γ)-reactions in Samples Exposed Near Ground Zero, Hiroshima

20	Induced radioactivity**						get el			Specific activity**					
	(Bq/kg-sample)					(ng/kg-sample)					(Bq/mg)				
	Eu-152	:	463	. 2	23	Eu	:	4.20	1	0.11	Eu-152/Eu	:	110.3	:	6.2
(a)*	Eu-154	:	87	*	18	Eu		4.20	*	0.11	Eu-154/Eu	:	20.8	±	4.2
	Co- 60	:	801	*	61	Co	1	64.1	t	1.3	Co- 60/Co	:	12,50	*	0.95
	Eu-152		3250	1	120	Eu		32.01	•	0.96	Eu-152/Eu	:	101.5	1	4.8
(b)*	Eu-154	:	690	*	150	Eu	:	32.01	1	0.96	Eu-154/Eu	:	21.6	*	4.7
	Co- 60	:	470	#	190	Co	1	36.6	±	1.1	Co- 60/Co	:	12.8		5.1

- . (a) : Eu, Co enriched sample from roof tile of Shima Hospital
  - (b) : Eu, Co enriched sample from granite of Motoyasu Bridge
- \*\* at the time immediately after the nuclear explosion

achieved with the enrichment of Eu and Co. Data for <sup>152</sup>Eu/Eu, <sup>154</sup>Eu/Eu, and <sup>60</sup>Co/Co in samples (2) and (3) are given in Table 4. Since radioactivities of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>60</sup>Co were induced principally by thermal and epithermal neutrons, radioactivity of <sup>152</sup>Eu, for example, at the time immediately after the nuclear explosion is given by the expression

$$A(^{152}Eu) = \frac{10^{-3}W(Eu)}{Ar(Eu)}L \times (^{151}Eu/Eu)\lambda(^{152}Eu) \times \{\phi_{th} \sigma_{th}(^{151}Eu) + \phi_{epi} I(^{151}Eu)\}$$
 (1)

where A is the activity in Bq immediately after the nuclear explosion, W the mass of the target element in mg in which the activity was induced, Ar is the atomic weight in g mol<sup>-1</sup> of the target element, L is the Avogadro constant in mol<sup>-1</sup>, X is the natural isotopic abundance (atomic fraction basis) of the nuclide of interest in the target element,  $\lambda$  is the decay constant of the neutron-induced radionuclide,  $\phi_{th}$  is the thermal neutron fluence in cm<sup>-2</sup>,  $\sigma_{th}$  is the thermal neutron cross section for the  $(n, \gamma)$  reaction in cm<sup>2</sup>,  $\phi_{epi}$  is the epithermal neutron fluence in cm<sup>-2</sup>, and I is the resonance integral for the  $(n, \gamma)$  reaction in cm2. By substituting the measured values (Table 4), nuclear data and constants (Table 1) into Equation (1) for 152 Eu/Eu, 154 Eu/Eu, and 60 Co/Co, respectively, three similar equations were obtained. Then by solving the simultaneous equations for  $\phi_{th}$  and  $\phi_{epi}$ , the thermal and epithermal fluences that can explain the 152Eu/Eu, 154Eu/Eu, and 60Co/Co simultaneously, were evaluated (Figure 5). The solutions for  $\phi_{th}$  and  $\phi_{epi}$ , with errors, are shown by the parallelograms in Figure 5. The  $\phi_{epi}/\phi_{th}$  ratios evaluated in the present work, 0.12 to 0.50 at Shima Hospital and 0.17 to 0.67 at Motoyasu Bridge, are significantly lower than the calculated results,  $\phi_{epi}/\phi_{th}$  2.5, for the position 250 m from the hypocenter in Hiroshima (closed circle in Figure 5). The present method of analyzing thermal neutron fluence and epithermal neutron fluence was examined for validity by a reactor experiment in which a mixture sample of Eu and Co reagents was irradiated in a TRIGA-II nuclear reactor and the results by the present method were compared with those by neutron activation with and without a cadmium cover (Tables 5 and 6).4 The results presented in Table 6 show that the present method of analyzing thermal and epithermal neutron fluences is reasonably reliable.

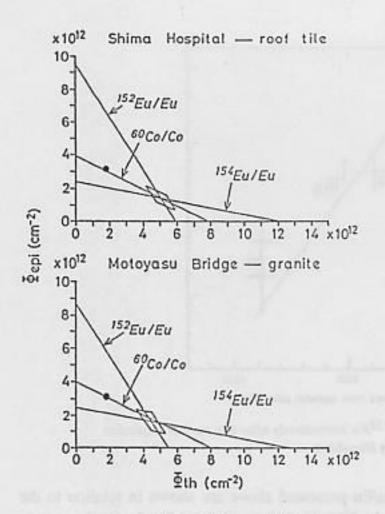


Figure 5. Analyses of Φth and Φepi by 152Eu/Eu, 154Eu/Eu, and 60Co/Co

Table 5. 152Eu, 154Eu, and 60Co Activity in Mixture Sample of Eu and Co Reagents Irradiated in RSR of TRIGA-II Nuclear Reactor for Four Hours 100kW

Induced r	adioa sampl				1000	et element g/sample)	Specific activity* (Bq/ng)			
Eu-152 :	1746	0 ±	420	Eu	t	0.1001266	Eu-152/Eu : (174.4 ± 4.2)×10			
Eu-154 :	173	3 ±	47	Eu	:	0.1001266	Eu-154/Eu : (173.1 ± 4.7)×10			
Co- 60 :	153	8 ±	42	Co	:	0.1002246	Co- 60/Co : (153.5 ± 4.1)×10			

<sup>.</sup> at the time immediately after neutron irradiation

Table 6. Thermal and Epithermal Neutron Fluence Rates Measured for RSR of TRIGA-II Nuclear Reactor at 100 kW

PER I	thi	ls work	Hone	ricos.	
o <sub>th</sub>	(6.48 :	0.06)×10 <sup>11</sup>		= 0.88)×10 <sup>11</sup>	
70X			(7.16	± 0.71)x10 <sup>11</sup>	[Au]
opi	(1.3 :	0.3 )x10 <sup>10</sup>		± 0.11)×10 <sup>10</sup>	
300			10.70	= 0.13)x10 <sup>10</sup>	[Au]

Measurements of  $^{152}$ Eu/Eu in housetop tiles, which were exposed to the Hiroshima Abomb at Naka Denwa-Kyoku (sample (4) and at Hiroshima University (sample (5)), were carried out using specimens enriched in Eu and Co, and the following specific activities of  $^{152}$ Eu at the time immediately after the nuclear explosion were obtained:  $14.1 \pm 0.8$  Bq mg $^{-1}$  in 180 g of original sample from Naka Denwa-Kyoku; and  $0.18 \pm 0.13$  and  $0.53 \pm 0.24$  Bq mg $^{-1}$  in 590 g of original sample from Hiroshima University (1255 m from the hypocen-

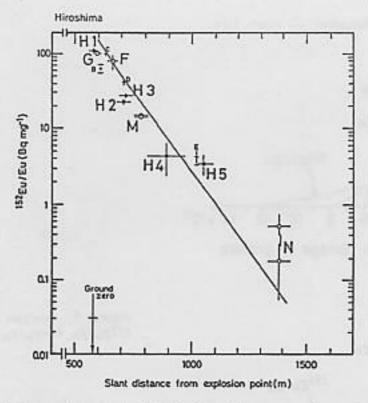


Figure 6. Specific radioactivity of <sup>152</sup>Eu immediately after the nuclear explosion in Hiroshima

ter). In Figure 6, the results of the <sup>152</sup>Eu/Eu presented above are shown in relation to the slant distance from the explosion point of the Hiroshima A-bomb (sign F refers to the sample (1), G the sample (3), M the sample (4), and N the sample (5)), together with our previous results (H1 to H5; H1 was the same as the present sample (2)).<sup>2</sup> The solid curve in Figure 6 was drawn on the basis of values calculated by Loewe.<sup>5</sup> So far as <sup>152</sup>Eu/Eu is concerned, the calculated values are roughly consistent with our measured values.

### Acknowledgments

We thank Prof. K. Yokoro, Drs. M. Hoshi and K. Katoh of the Research Institute for Nuclear Medicine and Biology, Hiroshima University, and Prof. Y. Ichikawa of Nara University of Education for the samples. We also wish to acknowledge with thanks the assistance of the members of the Atomic Energy Research Laboratory, Musashi Institute of Technology in neutron irradiation, that of Prof. M. Sakanoue, Dr. K. Komura, and Mr. K. L. Tan of the Low-Level Radioactivity Laboratory, Kanazawa University, in the gamma-ray spectrometry, that of members of the Research Center for Radioisotopes, Kanazawa University, in low-energy photon spectrometry, that of Prof. K. Terada and Dr. K. Matsumoto of the Faculty of Science, Kanazawa University, in fluorescence x-ray analysis, and that of Mr. H. Kasajima of the Faculty of Science, Kanazawa University, in slicing of the core sample. The encouragement extended throughout this study by Prof. K. Sakamoto of the Faculty of Science, Kanazawa University, is acknowledged. This work was supported financially by Grants-in-Aid for Scientific Research from the Ministry of Education, Science and Culture, Japan, under grants No. 57030060, 58030050, 59030057, 58380022, and 60304096. Further details of this work have been published elsewhere in Japanese. 6,7

#### References

- Sakanoue, M., Maruo, Y., and Komura, K., 1981. In Int. Symp. Methods Low-Level Counting and Spectrometry, p. 105. Vienna: International Atomic Energy Agency, publication IAEA STI/PUB/592.
- Nakanishi, T., Imura, T., Komura, K., and Sakanoue, M., 1983. <sup>152</sup>Eu in samples exposed to nuclear explosions at Hiroshima and Nagasaki. Nature 302:132-134.
- 3. Loewe, W. E., 1981. Private communication.
- Honda, T., Karisawa, Y., and Nozaki, T., 1985. Bull. Atomic Energy Res. Lab., Musashi Inst. of Technology, No. 9, 64.
- 5. Loewe, W. E., 1986. Private communication.
- Nakanishi, T., 1985. Summary Reports of Grants-in-Aid Special Research (1982-1984; Chairman: Prof. K. Yokoro; Grant No. 57030060, 58030050, and 59030057 to Hiroshima University), The Ministry of Education, Science and Culture, Japan, 25.
- Nakanishi, T., 1986. Summary Reports of Grant-in-Aid Comprehensive Project Research (A) (1983-1985; Chairman: Prof. A. Sekiguchi; Grant No. 58380022 to the University of Tokyo), The Ministry of Education, Science and Culture, Japan, 62.