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Measurement of Residual ^{60}Co Activity Induced by Atomic-bomb Neutrons in Nagasaki and Background Contribution by Environmental Neutrons

KIYOSHI SHIZUMA[1*], SATORU ENDO[1], MASAHARU HOSHI[2], JUN TAKADA[2], KAZUO IWATANI[3], HIROMI HASAI[4], TAKAMITSU OKA[5], TATSUYA SHIMAZAKI[6], YUTAKA OKUMURA[7], SHOICHIRO FUJITA[8], TADAAKI WATANABE[8], and TETSUJI IMANAKA[9]

^{60}Co / Specific activity / Atomic bomb / DS86 / Neutrons

Residual ^{60}Co activity in five steel samples induced by neutrons from the Nagasaki atomic bomb has been measured within about 1000 m from the hypocenter. The chemical separation of cobalt and nickel from steel samples was performed, and cobalt-enriched samples were prepared for all samples. Gamma-ray measurements were carried out with a low-background well-type germanium detector.

The gamma-ray spectra for five samples were compared with the spectrum of a control sample to ensure that the observed ^{60}Co was actually induced by A-bomb neutrons. The activation of cobalt by environmental neutrons was also investigated. It has been shown that the present ^{60}Co data are consistent with earlier Hashizume's data.

INTRODUCTION

A new dosimetry system (DS86) for survivors of the Hiroshima and Nagasaki atomic bombs was assessed in 1987. It was mentioned in the final report of the US-Japan joint assessment that a systematic discrepancy had been observed between the residual ^{60}Co data measured by Hashizume *et al.*[1)] and an activation calculation[2)] based on DS86 neutrons in the low-energy neutron region. Residual activity data for ^{152}Eu[3,4)], ^{60}Co[5–8)], and ^{36}Cl[9)] were acquired in Hiroshima, and the discrepancy was confirmed. To clarify the nature of the Hiroshima problem, interest has been concentrated on the Nagasaki data to establish whether such a discrepancy exists there as well as in Hiroshima. If the calculation agrees with the measurement in Nagasaki, the discrepancy observed in Hiroshima is not due to any uncertainties in the neutron transport calculation in air, but to the calculated neutron output

*Corresponding author: Phone ; +81–824–7614,
Fax ; +81–824-2–7192,
E-mail: shizuma@hiroshima-u.ac.jp
[1] Graduate School of Engineering, Hiroshima University, 1–4–1 Kagamiyama, Higashi -Hiroshima 739–8527, Japan
[2] Research Institute for Radiation Biology and Medicine, Hiroshima University, 1–2–3 Kasumi, Minami-ku, Hiroshima 734–8557, Japan
[3] Hiroshima Prefectural College of Health and Welfare, 1–1 Gakuencho, Mihara 723–0053, Japan
[4] Department of Electrical Engineering, Hiroshima Kokusai Gakuin University, Nakano, Aki-ku, Hiroshima 739–0321, Japan
[5] Kure University, 1–1–1 Gohara, Kure 724–0729, Japan
[6] Kumamoto University Radioisotope Center, 2–2–1 Honjo, Kumamoto 860–0811, Japan
[7] Atomic Disease Institute, Nagasaki University School of Medicine, Sakamoto-machi, Nagasaki 852–8102, Japan
[8] Radiation Effects Research Foundation, 5–2 Hijiyama Park, Minami-ku, Hiroshima 732–0815, Japan
[9] Research Reactor Institute, Kyoto University, Kumatori-cho, Osaka 590–0494, Japan

spectrum from the Hiroshima bomb.

In this work, we present specific activity measurements of ^{60}Co for five samples up to 1063 m in the slant range. The results were compared with calculations based on the neutron fluence (93Rev)* as well as DS86 neutrons. We have also investigated the activation of ^{60}Co by environmental neutrons.

MATERIALS AND METHODS

Steel samples

Five steel samples up to 935 m from the hypocenter were collected for ^{60}Co measurements. The locations of these samples (NS1-NS5) are shown in Fig. 1. The sampling place, material, altitude, slant range, and sample weight are given in Table 1.

The samples are indicated as follows. NS1: 2.2 cm-diameter reinforcing bars from the Takatani house located 290 m east of the hypocenter. The samples were embedded in a concrete pillar at 3.3 cm depth from the surface. NS2: reinforcing bars from Shiroyama School located 540 m west of the hypocenter. The samples were embedded in a concrete pillar at 1.5 cm depth from the surface. NS3: hand rail from the rooftop of Nagasaki University Hospital located 653 m south-east of the hypocenter. NS4: reinforcing bars of one of the pillars supporting the Motoki Bridge located 780 m north of the hypocenter. The samples were embedded at 4.5 cm depth in concrete from the surface. NS5: steel rails from Mitsubishi Steel Company located 935 m south of the hypocenter. The samples lay on a concrete bed for carts at a pier.

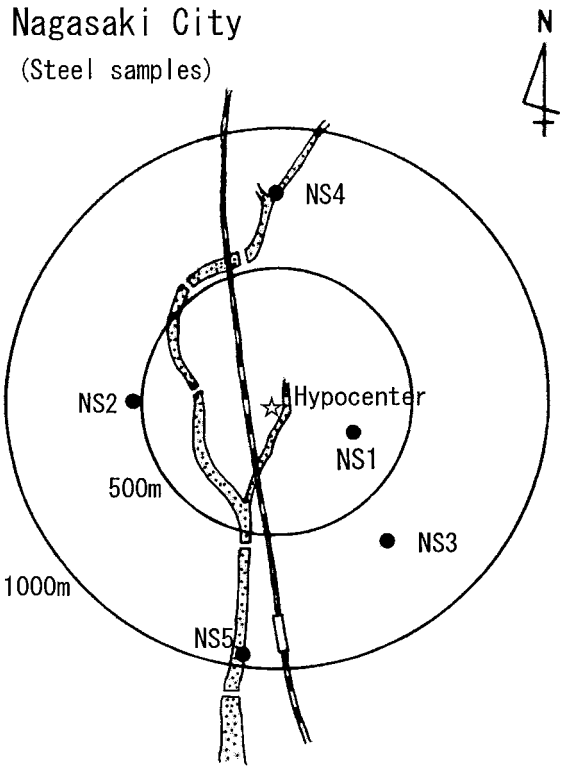

Fig. 1. Locations of steel samples (NS1-NS5) and the hypocenter of the atomic bomb in Nagasaki.

Sample preparation

All steel samples were chemically processed to extract cobalt in the same way as in previous work[8]. Steel samples were scratched into small chips, and chips were washed with dilutred HCl and dissolved in hot HCl. No carrier was added in the chemical pro-

Table 1. Steel samples collected in Nagasaki City for ^{60}Co measurement.

No	Place	Material	Depth[a] (cm)	Altitude (m)		Distance (m)		Steel Weight (g)	Enriched sample	
				Ground	Sea	Ground	Slant		Weight(g)	Co (mg g^{-1})
NS1	Takatani house	rebars	3.3	1	27	290 ± 21	561 ± 23	260	0.46	13.4 ± 0.7
NS2	Shiroyama School	rebars	1.5	10	22	540 ± 21	721 ± 23	600	0.80	27.2 ± 1.4
NS3	Nagasaki Univ. Hospital	hand rail	0	20	10	653 ± 21	809 ± 23	1000	0.66	14.8 ± 0.7
NS4	Motoki Bridge	rebars	4.5	–1.5	7	780 ± 21	927 ± 23	950	0.82	18.7 ± 0.9
NS5	Mitsubishi Steel	steel rail	0	0	2	935 ± 21	1063 ± 23	760	1.06	28.8 ± 1.4

[a] Depth from the concrete surface

cess. Small amounts of HNO_3 and H_2O_2 were added to the solution to oxidize Fe^{2+} to Fe^{3+}. The solvent-extraction method described by Kawamura *et al.*[10] with isopropyl ether was applied to extract major matrix iron ions, leaving nickel and cobalt ions in the solution. The solution was then heated to reduce the volume. This procedure was repeated again until 200 ml of the solution was obtained. For further purification, the solution was passed through an anion exchange containing 300 g of Dowex 1-X8 (100–200 mesh, type Cl) resin. The resin was then washed with 8 M HCl to collect nickel ions and 4 M HCl to collect cobalt ions. The cobalt fraction was precipitated by adding NaOH. The precipitate was collected by filtration, washed well with water, and dried. Finally, a few grams of enriched cobalt sample were obtained. The content of stable cobalt in the final form was analyzed by the Kawasaki Steel Techno-Research Co. Ltd. by atomic-absorption analysis. Cobalt recovery from the steel sample was typically 20%. The weight and cobalt content of enriched samples are given in Table 1.

Gamma-ray measurement

Samples were measured with a well-type Ge detector having a 120 cm^3 crystal volume and shielded with 20-cm-thick lead incorporated with an anticoincidence circuit to suppress the cosmic-ray background[11]. About one gram of cobalt-enriched sample was pressed into a polypropylene test tube of 13-mm-diameter and 75-mm-height. Each sample was measured twice, and one run continued for 10^5–10^6 s. Examples of gamma-ray spectra around the1173 and 1332 keV regions are shown in Fig. 2. The gamma-ray spectrum of a control sample is also shown in Fig. 2. The control sample described in previous work was obtained from Army Food Storehouse in Hiroshima, which was located 4571 m from the Hiroshima hypocenter. Five steel plates (25 cm wide and 25 cm long and 1.5 mm thick (total 1500 g)) were utilized. As shown in Fig. 2, no ^{60}Co peaks were observed in the control sample, whereas both 1173 and 1332 keV peaks were identified for samples NS1 to NS5.

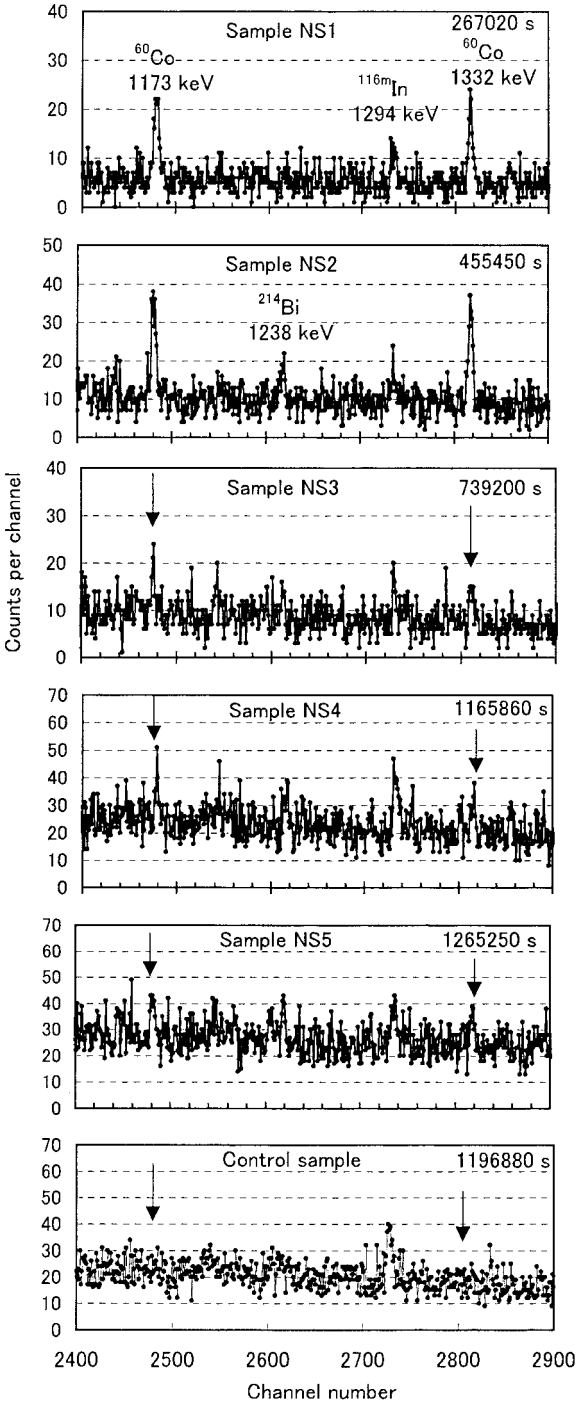

Fig. 2. Partial spectra around the 1173 and the 1332 keV gamma rays from ^{60}Co for steel samples. Takatani house (GR = 290 m), Shiroyama School (GR = 540 m), Nagasaki University Hospital (GR = 653 m), Motoki Bridge (GR = 780 m), Mitsubishi Steel (GR = 935 m), and a control sample[8] of Army Food storehouse in Hiroshima.

Data deduction

An efficiency calibration was performed in the same way as in previous work[8]. The sum of the peak counts of the 1173 and 1332 keV lines was used for an efficiency calibration and for determining the ^{60}Co activity. A known amount of ^{60}Co 102 Bq (Amersham CKZ24 3.587 kBq gsol^{-1} with total error ± 0.5%) was deposited to each 0.5, 1.0, 2.0, and 3.0 g of the sample, which was prepared by the same chemical procedure as a steel sample not exposed to the A-bomb. The efficiency for each standard sample is given in Table 2.

The peak counts were obtained from the total number of counts in the peak region (7 keV) by subtracting the background counts under the peak. The background was determined by drawing a line passing through the middle point of the low-, and high-energy regions (7 keV) of the peak. The error of the peak counts can be obtained from the error propagation formula. The specific activity of ^{60}Co at the time of the bomb explosion was obtained by correcting the elapsed time using a half life 5.2719 ± 0.0014 years[12]. The results are given in Table 3. The associated errors were as follows: the counting error of the peak counts (8–20%), the cobalt concentration (5%), and the detection efficiency (1.3%). The total error range was from 7 to 30%. The calculation of ^{60}Co activation in free field in air based on the DS86 neutron fluence and the revised neutron fluence 93Rev* and associated calculated-to-measured (C/M) ratios are also given in the

Table 2. Efficiency of the well-type Ge detector for the 1173+1332 keV lines from ^{60}Co.

Sample weight (g)	Efficiency (cps Bq^{-1})
0.5	0.0802 ± 0.001
1	0.0797 ± 0.001
2	0.0774 ± 0.001
3	0.0772 ± 0.001

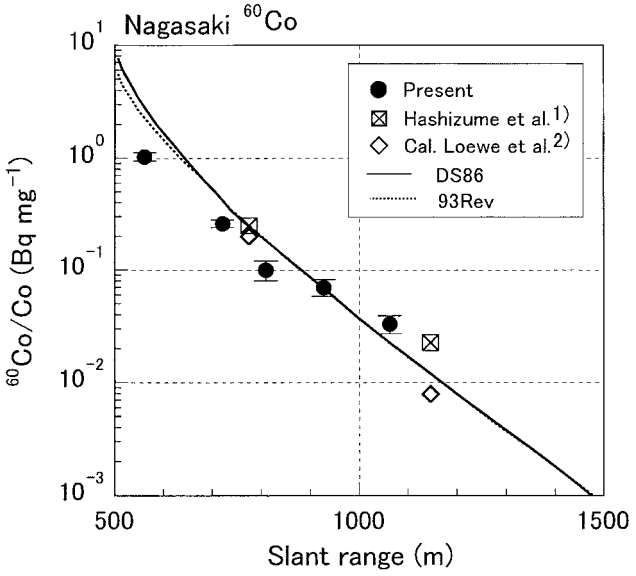

Fig. 3. Specific ^{60}Co activity (at the time of the explosion) as a function of the slant range. Data were taken by Hashizume *et al.*[1] for steel bars 8 cm depth in concrete. A corresponding calculation by Loewe *et al.*[2] is also shown. The solid and dotted lines indicate the activation calculation in a free field in air.

Table 3. Results of ^{60}Co specific activity measurement and Calculated-to-Measured (C/M) ratios in Nagasaki.

No	^{60}Co/Co[a] (Bq mg^{-1})	Calculated (Bq mg^{-1})		C/M ratio	
		DS86	93Rev	DS86	93Rev
NS1	1.03 ± 0.09	2.6	2.1	2.5 ± 0.2	2.1 ± 0.2
NS2	0.26 ± 0.02	0.47	0.47	1.80 ± 0.14	1.80 ± 0.14
NS3	0.10 ± 0.02	0.19	0.19	1.90 ± 0.40	1.90 ± 0.40
NS4	0.070 ± 0.012	0.066	0.066	0.94 ± 0.16	0.94 ± 0.16
NS5	0.033 ± 0.006	0.023	0.023	0.70 ± 0.13	0.70 ± 0.13

[a] Specific activity immediately after the bomb explosion

table. Specific activities of ^{60}Co as a function of the slant range are shown in Fig. 3.

ENVIRONMENTAL NEUTRON ACTIVATION

Environmental neutrons originating from cosmic-rays can activate materials at the ground level. It is important to estimate the background activity levels of materials by environmental neutrons to determine whether they are comparable to the residual activity level. Up to now, the environmental neutron flux and spectrum over the ground have been measured by many authors: Yamashita *et al.*[13], Nakamura *et al.*[14], Schraube *et al.*[15], and an UNSCEAR report[16].

The environmental neutron flux was directly estimated by measuring activated ^{60}Co in a cobalt regent. Four grams of a commercially purchased regent of cobalt oxide CoO were measured with a Ge detector. The measurement was continued for 1200640 s. Two peaks with energies of 1173 and 1332 keV from ^{60}Co were observed in the spectrum, as shown in Fig. 4. The other peaks were the 1238 keV of the natural background of ^{214}Bi and the 1294 keV of ^{116m}In produced by the ^{115}In (n, γ)^{116m}In reaction of indium included in the detector material.

The detection efficiency of ^{60}Co for 4 g of CoO was determined by measuring reference samples prepared by adding a known amount (24.9 Bq) of ^{60}Co into 4 g of CoO and mixed well. The ^{60}Co activity produced by the environmental neutrons in 4 g of CoO was determined to be 2.40 ± 0.31 Bq, which corresponds to a specific activity of 0.78 ± 0.10 mBq (gCo)$^{-1}$.

If one assumes the thermal neutron flux at ground level as being $\phi = 8.0 \times 10^{-3}$ ncm^{-2}s^{-1} from the UNSCEAR report[16], the saturation activity per 1g of cobalt is calculated from $A = N_{59}\ \sigma\phi = 3.0$ mBq (gCo)$^{-1}$, where $N_{59} = 7.87 \times 10^{21}$ is the number of

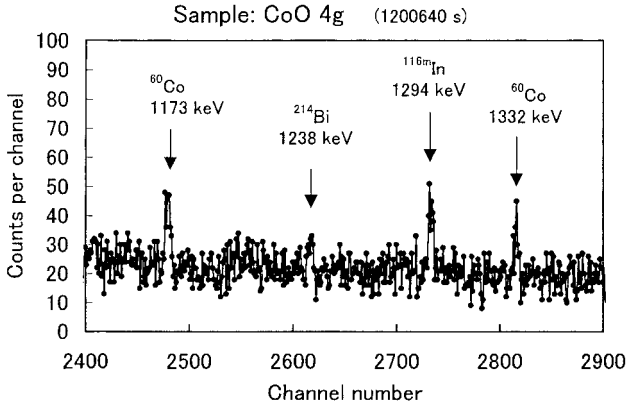

Fig. 4. Gamma-ray spectra of cobalt activation by environmental neutrons. The sample was four grams of CoO.

Table 4. Cobalt activation by environmental neutrons.

Sample (CoO)	4 g
Calculation	
^{59}Co atoms in 1 g of CoO	7.87×10^{21}
Cross sections	
^{59}Co(n,γ)^{60m}Co	20 b
^{59}Co(n,γ)^{60g}Co	17 b
total	37 b
Half lives	
^{60m}Co	10.47 m
^{60g}Co	5.27 y
Thermal neutron flux (assumed)	8.0×10^{-3} ncm^{-2}s^{-1}
Saturation activity for 1 g Co	3.0 mBq (gCo)$^{-1}$
Measurement	
Date	Oct. 19, 98
Measureing time	1200640 s
Peak counts	
1173 keV gamma-ray peak	140 ± 19
1332 keV gamma-ray peak	80 ± 17
1173 + 1332 keV	224 ± 25
Efficiency for 4 g of sample	
1173 keV	0.0419 ± 0.0026
1332 keV	0.0359 ± 0.0049
1173+1332 keV	0.0777 ± 0.0049 cps(Bq)$^{-1}$
Activity	
4 g of CoO	2.40 ± 0.31 mBq (4 gCoO)$^{-1}$
1 g of Co	0.78 ± 0.10 mBq (gCo)$^{-1}$

cobalt 59 atoms in the sample and the cross section of ^{59}Co(n, γ) ^{60}Co is taken to be σ= 37 b. The cross section is described as follows: the activation cross section, ^{59}Co(n, γ)^{60m}Co ($T_{1/2}$ = 10.47 m), is σ= 20 b, and ^{59}Co(n, γ)^{60g}Co ($T_{1/2}$ = 5.27 y) is σ= 17 b. Since all ^{60m}Co decays to ^{60g}Co, the actual activation cross section of ^{60}Co was assumed to be 37 b. Measurements of the ^{60}Co activity in CoO and the calculation are summarized in Table 4. The calculation is about a factor of 3 higher than the measured value, which indicates that the thermal neutron flux is about 2.7×10^{-3} ncm^{-2}s^{-1}. This result is rather near to Yamashita's results[13] of $(1.07 \pm 0.06) \times 10^{-3}$.

DETECTION LIMIT

Since the residual radioactivity levels are very low at present, it is important to know whether the peak identification in the gamma-ray measurement is really reliable. General discussions of the detection limit have been made by many authors: Currier[17], Tsoulfanidis[18], Brodsky[19]. In the case of gamma-ray spectrometry, peak counting of the residual radioactivity and background counting were simultaneously carried out, i.e., the background can be graphically subtracted from the peak region. If we let the gross counts in the interested peak region be G and background counts be B during the measuring time (T (s)), the net counts can be given as

$$S = G - B \quad \text{(counts)}, \tag{1}$$

or in the form of the counting rate,

$$ST^{-1} = GT^{-1} - BT^{-1}. \tag{2}$$

The errors of G and B are expressed as and $\sigma_G = \sqrt{G}$, $\sigma_B = \sqrt{B}$, respectively. Then, the error of S becomes $\sigma_S = \sqrt{\sigma_G^2 + \sigma_B^2} = \sqrt{G+B}$. When the residual radio-

Table 5. Detection limit for the ^{60}Co measurement.

No.	Place	Measuring time T (s)	Peak counts (1173 + 1332) N_S (counts)	Peak counting rate n_0 (counts s^{-1})	Background counts N_B	Background counts σ_B	d.m.c[a] n* (counts s^{-1})	Ratio n_0 / n*
[Nagasaki]								
NS1	Takatani	267020	199 ± 17	7.5×10^{-4}	50	7.0	6.1×10^{-5}	12
NS2	Shiroyama	455450	300 ± 23	6.6×10^{-4}	160	12.6	6.4×10^{-5}	10
NS3	Nagasaki Univ.	739200	75 ± 15	1.0×10^{-4}	100	10.0	3.2×10^{-5}	3.2
NS4	Motoki Bridge	1165860	140 ± 22	1.2×10^{-4}	280	16.7	3.3×10^{-5}	3.6
NS5	Mitsubishi Steel	1265260	143 ± 24	1.1×10^{-4}	300	17.3	3.2×10^{-5}	3.6
[Hiroshima]								
S1	A-Bomb Dome	269150	2901 ± 54	10.8×10^{-3}	44	6.6	5.7×10^{-5}	188
S2	Kirin Beer Hall	278530	580 ± 27	2.1×10^{-3}	58	7.6	6.4×10^{-5}	33
S3	Kodokan Building	607140	353 ± 21	5.8×10^{-4}	101	10	3.9×10^{-5}	15
S4	City Hall	611420	275 ± 24	4.5×10^{-4}	102	10.1	3.8×10^{-5}	12
S5	Red Cross Hospital (pipe)	823710	79 ± 21	0.96×10^{-4}	148	12.2	3.4×10^{-5}	2.8
S6	Red Cross Hospital (ladder)	938660	122 ± 18	1.3×10^{-4}	168	13.0	3.2×10^{-5}	4.0
S7	Hiroshima Back of Credit	1313690	151 ± 21	1.1×10^{-4}	221	14.9	2.6×10^{-5}	4.1
S8	Army Foods Storehouse	1288030	–	–	230	15.2	2.7×10^{-5}	–

[a] Detectable minimum counting rate n* = 2.326 σ_B/T

activity level is very low, the gross counts G becomes near to B, and S distributes as a mean value, $\mu_S \approx 0$, with a standard deviation of $\sigma_S = \sqrt{G+B} \approx \sqrt{2B}$. To judge whether the obtained net count (S) is significant, statistical hypothesis testing can be applicable. The critical level (L_C) can be given as follows:[17)]

$$L_C = k_a \sigma_S, \tag{3}$$

where k_a is a significance level which corresponds to a one-tailed probability of $1-\alpha$. Generally, k_a is taken as $k_a = 1.645$ with a significance level $\alpha = 0.05$. Thus, the critical level can be expressed as

$$L_C = 1.645\sigma_S = 2.236\ \sigma_B. \tag{4}$$

In the present work, the critical level was taken as the detectable minimum counting rate (d.m.c) and compared with the counting rate of residual activity measurements. The results are given in Table 5 for Nagasaki and Hiroshima samples. The observed ^{60}Co counting rates were more than a factor of 3 higher than the d.m.c.

DISCUSSIONS

Contribution of environmental neutron activation

Possible origins of the background for ^{60}Co measurements are as follows: 1) natural background gamma-rays, 2) radioactive ^{60}Co contamination of the detector and/or shielding material, 3) radioactive ^{60}Co contamination of the reagents used for chemical separation, 4) production of ^{60}Co in samples by environmental neutrons. Based on a background measurement without any sample, it was confirmed that there was no ^{60}Co contamination of the detector and/or shielding materials. In previous work[8)], an enriched sample pre-

Table 6. Contribution of environmental neutron activation for the residual ^{60}Co activity at the time of the measurements in Nagasaki and Hiroshima.

Sample	Place	Date of measurement	Slant range (m)	Sample (g)	Co content (%)	Co (mg)	BG activation[a] B (mBq)	^{60}Co activity[b] A_0 (mBq)	Ratio B / A_0
[Nagasaki]									
NS1	Takatani House	960216	561	0.48	1.34	6.36	0.019	8.49 ± 0.71	2.2×10^{-3}
NS2	Shiroyama School	960303	721	0.80	2.77	22.2	0.067	7.53 ± 0.69	8.8×10^{-3}
NS3	Nagasaki Univ. Hospital	961024	809	0.66	1.48	9.57	0.029	1.16 ± 0.25	2.5×10^{-2}
NS4	Motoki Bridge	960402	927	0.82	1.87	15.3	0.046	1.37 ± 0.22	3.3×10^{-2}
NS5	Mitsubishi Steel	960417	1063	1.06	2.88	30.5	0.091	1.29 ± 0.22	7.1×10^{-2}
[Hiroshima]									
S1	Atomic-Bomb Dome	950405	582	0.94	0.99	9.3	0.028	135.2 ± 8.4	2.1×10^{-5}
S2	Kirin Beer Hall	950401	874	2.6	1.89	49.1	0.147	26.8 ± 1.3	5.5×10^{-3}
S3	Kodokan Building	950704	916	0.93	3.72	16.2	0.112	6.57 ± 0.47	1.7×10^{-2}
S4	City Hall	950408	1159	0.52	7.00	36.4	0.109	5.60 ± 0.49	1.9×10^{-2}
S5	Red Cross Hospital (pipe)	950516	1580	1.37	4.10	56.0	0.168	1.21 ± 0.32	1.4×10^{-1}
S6	Red Cross Hospital (ladder)	950506	1582	0.71	3.70	26.0	0.078	1.33 ± 0.26	5.8×10^{-2}
S7	Bank of Credit, Yokogawa	950526	1703	1.13	6.46	73.0	0.219	2.50 ± 0.29	8.7×10^{-2}

[a] Environmental thermal neutron flux was assumed to be 8.0×10^{-3} ncm^{-2}s^{-1}.
[b] ^{60}Co activity in sample at the time of measurement.

pared from control samples (S8, GR = 4571 m) was measured, but no ^{60}Co was detected. Thus, the possibilities of 1–3) were rejected. Concerning possibility 4), environmental neutrons are able to induce ^{60}Co, as described above. The contribution of environmental neutron activation to the residual activity is summarized in Table 6. Since the actual thermal-neutron flux is thought to be less than 1/3 of 8.0×10^{-3}ncm^{-2}s^{-1}, the activity levels induced in 6–73 mg cobalt in enriched samples are two orders of magnitude lower than the residual activity level at the time of the measurement.

Kerr *et al.*[6] also measured two control samples for a ^{60}Co measurement: one from a light steel-frame building erected in the early 1940's at the Aluminum Company of America, Aloca-Mayville, Tennessee, and another from an old rail track at the Homestack Mine, a deep underground mine in South Dakota. Since they did not detect any significant difference in counts from the two samples, they concluded that environmental neutrons did not contribute significantly to the cobalt activation of the Hiroshima bridge sample.

Discrepancy between the measurement and calculation

Earlier measurements by Hashizume *et al.*[1] and corresponding calculations by Loewe[2] are compared with the present results in Fig. 4. The solid and dotted lines indicate an activation calculation based on the DS86 and 93Rev neutrons, respectively. Present steel samples were two surface samples (NS3, NS5) and three samples embedded 1.5–4.5 cm deep in concrete. A detailed calculation considering to neutron attenuation in concrete is necessary; however, such corrections were not included in the present work. According to depth profiles measured in concrete cores[20], the decrease in the activation from the surface to 4 cm depth in concrete is supposed to be at most 10%. The present data show fairly good agreement with the calculation, only the trend is slightly sallower than that of the calculations.

The C/M ratios for ^{60}Co as a function of the slant range are shown in Fig. 5. They were compared with Hashizume data[1] in Nagasaki and also with those of

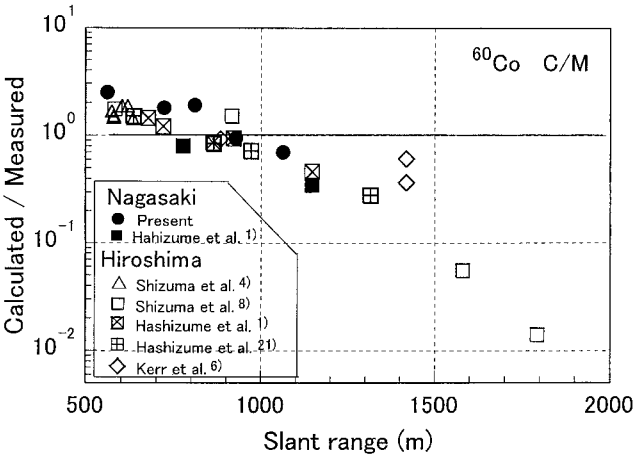

Fig. 5. Comparison of the C/M ratios of ^{60}Co for Nagasaki and Hiroshima.

Hiroshima ^{60}Co data[1,21]. The C/M ratios show a similar trend of Hashizume's two datum points. However, the present data are roughly in agreement with the calculations at a slant range of about 1000 m. Since the present ^{60}Co data were obtained for samples at a slant ranges within about 1063 m, the discrepancy is still not clarified due to a lack of available data beyond 1100 m.

CONCLUSION

Specific ^{60}Co activities were determined for five Nagasaki samples exposed to the A-bomb. Although the present data support Hashizume's ^{60}Co data, the possibility of discrepancy from the calculation is not clarified. It has been shown that the environmental neutron activation is negligibly small compared to the residual activity. Further measurements of the residual activities in Nagasaki are necessary to clarify the discrepancy problem.

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*Personal communication (1996) with S. D. Egbert, Science Applications International Corp., 10260 Campus Point Dr., MS C3 San Diego, California 92121 USA. 93Rev is based on the same assumption as DS86, with improved source energy/angle resolution, more recent air cross sections, and finer energy resolution in transport calculations.

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