Specific Activities of ⁶⁰Co and ¹⁵²Eu in Samples Collected from the Atomic-Bomb Dome in Hiroshima

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Neutron induced activity/⁶⁰Co/¹⁵²Eu/Atomic Bomb/Gamma-ray spectroscopy

Neutron-induced activities ⁶⁰Co and ¹⁵²Eu have been measured for samples collected from the Atomic-Bomb Dome locating at 161 m from the hypocenter of the Hiroshima Bomb. Specific activities ⁶⁰Co/Co and ¹⁵²Eu/Eu at the time of the detonation have been determined as 10.0±1.0 Bq mg⁻¹ (steel sample S4) and 80±9 Bq mg⁻¹ (granite sample G1), respectively. Detailed measurements of ⁶⁰Co and ¹⁵²Eu activities for samples collected from various locations of the Dome show almost no directional dependence whether the sample faced to the epicenter or not, nor vertical height dependence between 17 m height and the ground level. In addition, ¹⁵²Eu was not detected in the sample collected from the basement. It has been shown that the present ⁶⁰Co activity value, the nearest steel one to the hypocenter, as well as other short distance data are systematically lower than the calculated values based on the neutron fluence of the DS86.

INTRODUCTION

Reevaluation of Hiroshima and Nagasaki atomic-bomb dosimetry has been completed in 1987 with a final report¹) of a new dosimetry system called DS86. Concerning to the evaluation of the low energy neutron fluence, ⁶⁰Co activation data taken by Hashizume et al.²,³) both at Hiroshima and Nagasaki, systematically deviated from the calculated values by Loewe⁴). Nevertheless detailed discussion on the ⁶⁰Co activation, the discrepancy was still in open
question. Other $^{60}$Co data taken by Saito$^5$, by Hoshi and Kato$^6$ and by Nakanishi et al.$^7$ were not considered in the above discussion. No detailed consideration was made on $^{152}$Eu activation data in the final report.

Afterward, residual activity data on $^{60}$Co and $^{152}$Eu have been accumulated and a systematic deviation from the calculated activation has been revealed$^6$ in the $^{152}$Eu activation data similarly to the $^{60}$Co data. In the case of $^{60}$Co measurement, data from two locations were recently added by Kimura et al.$^9$ and by Kerr et al.$^{10}$, however, $^{60}$Co data were still scarce for detailed discussions on the neutron fluence.

In March 1990, the Atomic-Bomb Dome (the City of Hiroshima Commercial Display Building) located at 161 m from the hypocenter in Hiroshima City has been mended to keep it from crumbling. During the work of mending, we have collected steel and granite samples from various locations of the Dome. In this paper, new data of specific activities of $^{60}$Co (half-life = 5.2719 y$^{11}$) and $^{152}$Eu (half-life = 13.2 y$^{11}$) are given from samples of the Dome. A systematic discrepancy between the measured $^{60}$Co data and the calculated activities by Mendelsohn$^{12}$ is shown in short distances from the hypocenter. The $^{152}$Eu activation along vertical direction including the basement was discussed.

**MATERIALS AND METHODS**

**Steel and granite samples**

Location of the Dome is shown in Fig. 1. The coordinates of the hypocenter on the new Hiroshima City Map$^{13}$ transferred from the US Army Map$^{14}$ are also shown in the figure. Top and side view of the Dome and sampling locations are shown in Fig. 2. Four steel samples (S4, S8, S12, S16) were taken at 21 m above the ground level. The roof of the Dome was covered with the wood sheathing$^{15}$ and thin copper plates at the time of the detonation, so the steel

![Fig. 1. Location of the Atomic-Bomb Dome in Hiroshima. The coordinates of the hypocenter on the new Hiroshima city map are also shown.](image-url)
Fig. 2. Side and top views of the Atomic-Bomb Dome. Sampling locations of stone samples G1 to G6 (b and c) and steel samples S4 to S6 (d) are shown.

Fig. 3. Sketch of steel sample S4 composed of A, B, and C plates. Locations of small pieces for the γ ray measurement are shown for each plate.
samples were almost directly exposed to the neutrons. Four granite cores of 6.8 cm dia. × 30 cm leng. (G1, G2, G3, G4) were taken at 17 m above the ground level and one granite core (G5) was taken at the ground level. One concrete core (G6) of the same size was taken from the ceiling of the basement. As shown in Fig. 3, the steel sample (the base plate of the arch) was composed of three plates with 5 mm thick, i.e., two L-shaped plates and one flat plate, which were named as A, B, and C, respectively.

Sample preparation and elemental analysis

Since the Dome was near to the hypocenter, γ rays of 60Co induced in steel samples were directly measurable with a low-background Ge detector. To determine the specific activity 60Co/Co, disk sample (10.5 mm dia. × 1.6 mm thick., 1.133 g) and three stick samples (4.0 mm2 × 15 mm, 2.8 mm2 × 15 mm, 2.6 mm2 × 15 mm, total 1.101 g) were obtained from the plate SA of S4 sample shown in Fig. 3. To investigate local deviation of 60Co activity, many pieces (25 mm × 25 mm × 1 mm) were cut from the upper surface of the sample S4 as shown in Fig. 3. The depth profile of 60Co activity in steel plates were measured from the samples S4-A-3, S4-A-6, S4-C-1 and S4-C-3, where the reverse side of 1 mm thickness was taken for the samples of S4-A-6 and S4-C-3. Same size pieces were cut from samples S8, S12 and S16 at similar positions to S4-A-1. All steel sample were etched with 3M HCl, washed with water and dried prior to the measurement.

Elemental analyses in the steel samples have been done by the Kawasaki-Steel Techno-Research Co. (K-TEC) by means of the atomic absorption method. The concentration of Co was determined for each steel plate. In addition to Co content (0.26±0.02 mg/g) for steel plate S4-A, Cu and Ni contents were also determined as 0.26±0.02 mg/g and 0.27±0.02 mg/g, respectively, to estimate the yield of 60Co through the fast neutron reactions of 63Cu(n,a)60Co and 60Ni(n,p)60Co. According to the cross sections for these reactions mentioned in ref. 4, yields of 60Co were estimated negligibly small compared to the thermal neutron activation.

Granite core samples were cut into orbicular plates of 2 cm thickness. The surface plates were grained to less than 100 mesh and 30 g of the stone powder was packed in a plastic vessel. Concentration of the stable Eu in stone samples were determined by means of the neutron activation analysis. A couple of 4 g powder sample were taken and 50 μg Eu was added to one of them. Neutron activation was performed at the research reactor of Kinki-University under the thermal neutron flux of 10^7 n·cm^-2·s^-1.

Since the 152Eu activity level was very low for the sample G6, Eu was chemically enriched referring to the alkali fusion method16). Enriched sample of 3.45 g was obtained out of 45 g original sample. The Eu content in the enriched sample was determined by means of the neutron activation analysis. A couple of 4 g powder sample were taken and 50 μg Eu was added to one of them. Neutron activation was performed at the research reactor of Kinki-University under the thermal neutron flux of 10^7 n·cm^-2·s^-1.

Gamma-ray measurement

Two low-background Ge spectrometers were used for the γ-ray measurement. One was a 124 cm^3 coaxial type Ge detector17) (the CX detector, the relative efficiency: 28%) with a 20 cm
Fig. 4. Typical \( \gamma \)-ray spectrum of steel sample S4-A-1 obtained from the Dome measured with a 124 cm\(^3\) coaxial Ge detector. The \( ^{60}\)Co activity was induced by neutrons of the bomb.

Fig. 5. Typical \( \gamma \)-ray spectrum of granite sample G1 obtained from the Atomic-Bomb Dome measured with a 124 cm\(^3\) coaxial Ge detector. The \( ^{152}\)Eu activity was induced by neutrons of the bomb.
thick lead shielding and the other is a 120 cm$^3$ well type Ge detector (the WL detector, the relative efficiency: 23%) with a 25 cm thick lead shielding and an anticoincidence system to reduce the cosmic-ray background.

Steel samples were measured mainly with the CX detector at the position close to the detector's end-cap. Typical $\gamma$-ray spectrum for steel sample S4-A-1 is shown in Fig. 4. Two $\gamma$-ray peaks of $^{60}$Co are clearly seen in the figure. To determine the specific activity $^{60}$Co/Co, both the CX and the WL detector were used. The $\gamma$-ray detection efficiency for $^{60}$Co was determined using reference sources of QCR11 (produced by Amersham Int.) for the CX detector and NES-100S (produced by DuPont NEN Products) for the WL detector. The efficiency of the steel sample for the CX detector was determined from the measured dependences of the efficiency along the detector axis and the radial direction. In the case of the WL detector, it was confirmed that the efficiency in the well was constant. Self-absorption correction for the reference source and steel samples were negligible. The efficiencies of both detectors are given in Table 1.

Granite samples G1-G5 were measured with the CX detector. Typical $\gamma$-ray spectrum for sample G1 is shown in Fig. 5. Efficiency calibration for $^{152}$Eu was performed with a standard sample contained a known amount (4.47 Bq) of $^{152}$Eu. The enriched sample of G6 was pressed into a polypropylene tube and was measured with the WL detector.

RESULTS

Specific activity $^{60}$Co/Co for the steel sample S4-A is given in Table 1. Since measured values with two different detectors agreed well with each other, an average value was adopted for the specific activity of the sample. Local deviations of $^{60}$Co activation in steel plates S4-A, S4-B and S4-C were investigated comparing $^{60}$Co $\gamma$-ray counting rates corrected for the Co content in the steel plate and the results were given in Table 2. Local deviations of $^{60}$Co activation for steel samples S4 to S16 were investigated in the same manner and the results were given in Table 3. The depth profile of $^{60}$Co activity induced in steel plates is shown in Fig. 6.

Specific activity $^{152}$Eu/Eu for stone samples G1 to G6 are given in Table 4. Since $\gamma$-rays of $^{152}$Eu were not detected for the sample G6, only the upper limit was given in the table.

| Sample* | Co content (ppm) | Ge detector | $^{60}$Co/Co$^+$ (Bq mg$^{-1}$) |
|---------|-----------------|-------------|----------------------------------|
|         |                 | Type        | Efficiency** (cps/Bq) | Measured | Average  |
| S4-A, #1| 260 ± 20        | coaxial     | 0.0407 ± 0.00017         | 10.1 ± 1.6 | 10.0 ± 1.0 |
| S4-A, #2| 260 ± 20        | well        | 0.0782 ± 0.00014         | 9.8 ± 1.3  |

* The ground and the slant range were 163 ± 15 m and 602 ± 15 m and 602 ± 21 m, respectively, and the height was 21 m above the ground. Sample #1: disk of 10.5 mm dia. × 1.6 mm thick, sample #2: three sticks of 4 mm$^2$ × 15 mm, 2.8 mm$^2$ × 15 mm, 2.6 mm$^2$ × 15 mm.

** Effective efficiency for 1173 keV + 1332 keV peak counts.

$^+$ Corrected at the time of the detonation.
Table 2. Counting rates of $^{60}$Co 1173 keV + 1332 keV peaks for the steel sample S4

| Steel plate (Co content) | No. | H/V* | $^{60}$Co counting rate (cps/mgCo) |
|--------------------------|-----|------|----------------------------------|
| S4-A (260 ± 20 ppm)      | 1   | H    | 0.51 ± 0.05                      |
|                          | 2   | H    | 0.53 ± 0.06                      |
|                          | 3   | H    | 0.46 ± 0.06                      |
|                          | 4   | H    | 0.45 ± 0.05                      |
|                          | 5   | V    | 0.47 ± 0.05                      |
| S4-B (230 ± 20 ppm)      | 1   | H    | 0.44 ± 0.06                      |
|                          | 2   | H    | 0.44 ± 0.06                      |
|                          | 3   | H    | 0.52 ± 0.07                      |
|                          | 4   | V    | 0.44 ± 0.05                      |
| S4-C (80 ± 6 ppm)        | 1   | H    | 0.27 ± 0.05                      |
|                          | 2   | H    | 0.37 ± 0.06                      |

*H: horizontal plate, V: vertical plate.

Table 3. Local deviation of $^{60}$Co 1173 keV + 1332 keV peak counting rate for four steel samples.

| Steel sample | Distance (m) | Height (m) | Co content (ppm) | Counting rate (cps/mgCo) |
|--------------|--------------|------------|-------------------|--------------------------|
| S4-A-1       | 163 ± 15     | 21         | 260 ± 20          | 0.51 ± 0.05              |
| S8-A-1       | 169 ± 15     | 21         | 250 ± 20          | 0.47 ± 0.05              |
| S12-A-1      | 168 ± 15     | 21         | 250 ± 20          | 0.52 ± 0.08              |
| S16-A-1      | 163 ± 15     | 21         | 260 ± 20          | 0.50 ± 0.06              |

Fig. 6. Depth profile of the specific activity $^{60}$Co/Co for total 10 mm of steel plates S4-A and S4-C.
Table 4. \(^{152}\)Eu/Eu in stone samples obtained from the Atomic-Bomb Dome

| Sample | Height (m) | Distance (m) | Eu content (ppm) | \(^{152}\)Eu/Eu\(^+\) (Bq mg\(^{-1}\)) |
|--------|------------|-------------|------------------|------------------------------------------|
| G1     | 17         | 161±15      | 585±21           | 0.88±0.06                                 |
| G2     | 17         | 165±15      | 586±21           | 0.78±0.06                                 |
| G3     | 17         | 173±15      | 589±21           | 0.73±0.06                                 |
| G4     | 17         | 168±15      | 587±21           | 0.84±0.04                                 |
| G5     | 0.35       | 137±15      | 596±21           | 0.57±0.05                                 |
| G6*    | -1.2       | 150±15      | 600±21           | 3.3 ±0.5                                  |

\(\ast\) Eu enriched sample.  
\(**\) Upper limit.  
\(\dagger\) Corrected at the time of the detonation.

### DISCUSSION

1. **Local deviation of \(^{60}\)Co and \(^{152}\)Eu activities**

   Specific activity \(^{60}\)Co/Co were measured in detail for steel sample S4. It is shown that the \(^{60}\)Co activities are almost same for plates A and B as shown in Table 2. The \(^{60}\)Co activity for the plate C is about 3/5 of those of A or B plates since it was shielded with the 5 mm thick steel plate (A and B). The depth profile shown in Fig. 6 also represents that the \(^{60}\)Co activity for upper plate is obviously higher than the lower plate.

   The \(^{60}\)Co activities for four steel samples S4 to S16 were almost same in accord with the result for \(^{152}\)Eu activities of granite samples G1 to G4, nevertheless, steel samples S8 and S12 and stone samples G2 and G3 were not faced to the epicenter of the bomb. These results indicate that thermal neutron fluences were almost the same magnitude at sampling locations around the Dome. Similar results have been obtained in the case of depth profile measurement of \(^{152}\)Eu for the Motoyasu-bridge pillar\(^{19}\) located at 132 m from the hypocenter. It was shown that the \(^{152}\)Eu activity was almost equal at the north, west, south and east surfaces of the pillar, where the west surface did not look the epicenter. According to the neutron cross section data\(^{20}\), the (n,\(\gamma\)) cross section of \(^{151}\)Eu is dominant for thermal neutrons (5800 b). In contrast, the cross section of \(^{59}\)Co for neutron energies 0.5–1 keV is about 100 b in average, which is higher than 37 b for thermal neutrons. This indicates that the \(^{60}\)Co activity data reflects rather epithermal region than thermal region of the incident neutron energy spectrum. According to the present results, it could be said that almost no directional dependence exists for the thermal- and epithermal-energy neutrons at short distances from the hypocenter.

   As given in Table 4, the \(^{152}\)Eu/Eu specific activity of the granite sample G5 obtained at the ground level is almost equal to those of samples G1 to G4 obtained at 17 m above the ground level. The sample G5 is nearest to the hypocenter among the stone samples in the ground range, however, the slant range is almost same with others. This result indicates that the activation of Eu at the same slant range never depends whether the sample is located on the ground or 17 m
above the ground level, i.e., the reflection of neutrons from the ground is negligible. Gamma-rays of $^{152}$Eu were not detected from the sample G6 obtained in the basement. Only the upper limit is given in Table 4. The ratio of activities in basement to the ground level G5 is less than $1/50$.

2. Specific activity of $^{60}$Co against distance from the hypocenter

Measured $^{60}$Co data including the present work are summarized in Table 5. Saito’s data in the table were obtained correcting the $^{60}$Co decay at the time of the detonation of the bomb to the original data$^5$. The $^{60}$Co activity of the Fukoku Seimei Building by Hoshi and Kato$^6$ in the table is the value of the iron ring faced to the epicenter. From six $^{60}$Co activity data given for Aioi bridge by Hoshi and Kato$^6$, three data for plate A (vertical) and three data for plate B (horizontal) were respectively averaged. The $^{60}$Co data by Hashizume et al.$^3$ in the table are only for the iron rings located on rooftop for comparison with other data (their data of embedded rebar in concrete were not given here). These $^{60}$Co data are shown in Fig. 7 against ground range in comparison with the calculation by Mendelsohn$^{12}$ assuming the bomb yield to be 15 kiloton$^{21}$. This calculation has been performed using the Monte Carlo transport code on a model of Co activation in a free-field over the ground.

**Table 5. Summary of specific activity $^{60}$Co/Co data in Hiroshima**

| Location          | Distance (m) | Material      | $^{60}$Co/Co* (Bq mg$^{-1}$) | Authors              | ref. |
|-------------------|--------------|---------------|-----------------------------|----------------------|------|
|                   | Ground       | Slant         |                             |                      |      |
| Shima Hospital    | $0 \pm 20$   | $580 \pm 15$  | roof tile                   | $12.50 \pm 0.99$     | Nakanishi et al. | 7    |
| Motoyasu Bridge   | $128 \pm 20$ | $594 \pm 16$  | granite                     | $12.8 \pm 5.1$       |      |
| A-Bomb Dome       | $163 \pm 15$ | $602 \pm 21$  | steel plate                 | $10.0 \pm 1.0$       | Present work | 5    |
|                   |              | water trough  |                             | $3.96 \pm 0.28$      | Saito | 5    |
| A-Bomb Dome       |              | water trough  |                             | $4.05 \pm 0.06$      |       |      |
| Sumitomo Bank     | $250$        | $614$         | iron ring                   | $6.0 \pm 1.3$        | Hoshi, Kato | 6    |
| Aioi Bridge       | $300$        | $652$         | steel plate A               | $2.9 \pm 0.3$        |       |      |
|                   |              | steel plate B |                             | $1.8 \pm 0.2$        |       |      |
| Fukoku Seimei     | $330$        | $651$         | iron ring                   | $5.7 \pm 0.6$        | Kimura et al. | 9    |
| Chugoku Electric Co. | $687$    | $883$         | handrail                    | $0.440 \pm 0.063$    | Kerr et al. | 10   |
|                   |              |               |                             | $0.423$              |       |      |
| Yokogawa Bridge   | $1295$       | $1415$        | steel plate D               | $0.00515 \pm 0.00080$ | Kimura et al. | 9    |
|                   |              |               |                             | $0.00257$            | Kerr et al. | 10   |
|                   |              |               |                             | $0.0056$             |       |      |
| Honkawa School    | $373$        | $696$         | iron ring                   | $4.4^{**}$           | Hashizume et al. | 2    |
| Fukuromanachi School | $441$    | $719$         | iron ring                   | $3.3^{**}$           |       |      |
| Kirin Beer Hall   | $650$        | $862$         | iron ring                   | $0.98^{**}$          |       |      |
| Kodo School       | $727$        | $921$         | iron ring                   | $0.53^{**}$          |       |      |
| City Hall         | $997$        | $1147$        | iron ring                   | $0.11^{**}$          |       |      |

* Corrected at the time of the detonation.

** These absolute values were cited in ref. 10. The original values were given in ref. 2.
Fig. 7. Specific activity $^{60}\text{Co}/\text{Co}$ against ground range from the hypocenter. Symbols of the present work and other authors are indicated in the figure and each value is given in Table 5. The solid line is a calculation by Mendelsohn12).

At short distances within 300 m, present value of $^{60}\text{Co}$ is in consistent trend with those data of nonmetallic samples of Shima Hospital (rooftile) and Motoyasu Bridge (granite) by Nakanishi et al.7) and iron ring data of Sumitomo Bank and Fukoku Seimei Building by Hoshi and Kato6). It is noted that these measured $^{60}\text{Co}$ data are systematically lower than the calculation. This result agrees with those in the case of $^{152}\text{Eu}/\text{Eu}$ specific activity measurements8). As shown in Fig. 7, Saito’s data5) were lower than the neighboring data. They have estimated thermal neutron fluences of the Hiroshima and the Nagasaki bomb for the first time, however, lack of detailed information on sample makes further discussions difficult. Two data of Aioi bridge marked A and B in the figure are 1/2 to 1/3 lower than those of Fukoku Seimei Building located at almost the same ground range. A detailed calculation of Co activation including the geometry of the sample surroundings would be necessary to explain the discrepancy.
As shown in Fig. 7, recent data by Kimura et al.\(^9\) and by Kerr et al.\(^{10}\) agree well with each other, but show different trend from iron ring data by Hashizume et al. Although the calculation by Mendelsohn seems to agree with the recent \(^{60}\)Co data, a detailed calculation by Kerr et al.\(^{10}\) including the sample surroundings on the Co activation gives a slight lower value for the handrail sample of Chugoku Electric Co. and a factor of two lower value for the steel plates of Yokogawa Bridge. It is necessary to accumulate more \(^{60}\)Co data at 500–1500 m to discuss the discrepancy between the measured data and the calculation. The systematic discrepancy between the measured data and the calculation at short distances as well as those at long distances must be consistently explained to investigate whether systematic errors exist in the neutron transport calculation and the source-term calculation of the Hiroshima bomb.

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