Measurement of Residual $^{152}$Eu Activity Induced by Atomic Bomb Neutrons in Nagasaki and the Contribution of Environmental Neutrons to This Activity

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$^{152}$Eu activity/Atomic bomb/DS86/Neutrons.

Residual $^{152}$Eu activities induced by neutrons from the Nagasaki atomic bomb were measured for nine mineral samples located up to 1,061 m in the slant range and one control sample at 2,850 m from the hypocenter. A chemical separation to prepare europium-enriched samples was performed for all samples, and gamma ray measurements were carried out with a low background well-type germanium detector. In this paper, the measured specific activities of $^{152}$Eu are compared with activation calculations based on the DS86 neutron fluence and the 93Rev one. The calculated-to-measured ratios are also compared with those of $^{60}$Co and $^{36}$Cl. The present results indicate that the measurements agree to the calculation within a factor of three as observed in the nuclear tests at Nevada. The activation level of environmental neutrons and the detection limit for $^{152}$Eu are also discussed.

INTRODUCTION

The radiation dosimetry system (DS86) for survivors of the Hiroshima and Nagasaki atomic bombings was assessed1) in 1987. In the evaluation of low energy neutrons, a discrepancy between the residual $^{60}$Co measurement by Hashizume et al.2) and an activation calculation in both cities was pointed out in the final assessment report3). Since residual activity data were scarce at that time, $^{152}$Eu data were unable to confirm the $^{60}$Co discrepancy. After the assessment of DS86, additional residual activity data for $^{152}$Eu4–5), $^{60}$Co6–9), $^{36}$Cl10) and $^{63}$Ni11) were acquired to confirm the DS86 neutron fluence, and the evaluation of neutron transport calculation was extensively performed by U.S. and Japanese working groups. A status report 12) was published recently; however, the discrepancy problem was not clarified.

MATERIALS AND METHODS

Mineral samples

The sampling locations of nine mineral samples (NM1–NM9) are shown in Fig. 1. A control sample NM10 (at Maruo-machi) was 2,850 m southwest of the hypocenter. The sampling place, material, altitude, slant range, and sample weight are given in Table 1. Most samples were collected from the surface of old stone walls facing to the epicenter without any shielding. The surface of the rocks (andesite) was scratched to a depth of about 1 cm. The sample
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NM3 was the wall tiles obtained from the rooftop of the Nagasaki University Hospital at a site 20 m above the ground.

To calculate the ground range ($GR$) of the sample, the hypocenter coordinate on the new map of Nagasaki by Kerr et al.\(^{18}\) was used; the east-west and north-south coordinates of the hypocenter are 34.2475 and $-25.3945$. The slant range ($SR$) of the sample was calculated based on the burst height given as 503 $\pm$ 10 m\(^{18}\). Since Nagasaki lies topologically on an inclined plane, the slant range was calculated from the following equation by considering the altitude from the sea level ($S_l$) at the sampling location and the ground level ($G_l$) at the sampling site:

$$SR^2 = GR^2 + (503 + 5.2 - S_l - G_l)^2,$$

where the altitude of the hypocenter from the sea level was taken to be 5.2 m.

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**Table 1.** Mineral samples collected in Nagasaki for $^{152}$Eu measurement.

| No. | Place            | Material$^a$ | Coordinate$^b$ ($x$, $y$) | Sea level (m) | Ground level (m) | Ground range (m) | Slant range (m) | Sampling weight (g) | Enriched sample (g) | Eu content (ppm) |
|-----|------------------|--------------|---------------------------|---------------|------------------|------------------|------------------|---------------------|---------------------|-------------------|
| NM 1| Yana Bridge      | R            | 33.936, $-25.405$         | 1             | 1.0              | 311 $\pm$ 20     | 594 $\pm$ 22     | 360                 | 5.6                 | 3.8 $\pm$ 0.2     |
| NM 2| Urakami Church   | R            | 34.645, $-25.167$         | 16            | 1.7              | 458 $\pm$ 20     | 671 $\pm$ 22     | 250                 | 2.1                 | 4.1 $\pm$ 0.4     |
| NM 3| Nagasaki Univ. Hospital | T        | 34.629, $-25.925$         | 30            | 20.0             | 653 $\pm$ 21     | 809 $\pm$ 23     | 240                 | 3.5                 | 10.4 $\pm$ 0.9    |
| NM 4| Gokoku Shrine-B  | R            | 33.704, $-25.036$         | 16            | 2.5              | 651 $\pm$ 20     | 815 $\pm$ 22     | 1000                | 4.3                 | 13.1 $\pm$ 0.5    |
| NM 5| Gokoku Shrine-A  | R            | 33.704, $-25.036$         | 16            | 1.0              | 651 $\pm$ 20     | 816 $\pm$ 22     | 800                 | 3.5                 | 8.1 $\pm$ 0.3     |
| NM 6| Nanzan School-A  | R            | 34.509, $-24.741$         | 30            | 1.5              | 704 $\pm$ 20     | 850 $\pm$ 22     | 650                 | 5.4                 | 5.7 $\pm$ 0.3     |
| NM 7| Shimoda House    | R            | 34.600, $-26.093$         | 9             | 4.0              | 782 $\pm$ 20     | 926 $\pm$ 22     | 963                 | 12.8                | 4.4 $\pm$ 0.2     |
| NM 8| Prefectural Gymnasium-B | R       | 33.504, $-24.896$         | 10            | 1.5              | 895 $\pm$ 20     | 1024 $\pm$ 22    | 1000                | 5.9                 | 5.0 $\pm$ 0.2     |
| NM 9| St. Maria School | R            | 33.323, $-25.210$         | 21            | 1.0              | 943 $\pm$ 20     | 1061 $\pm$ 22    | 1000                | 3.8                 | 5.0 $\pm$ 0.2     |
| NM10| Maruo-machi (control) | R        | 33.964, $-28.230$         | 30            | 0.5              | 2850 $\pm$ 20    | 2889 $\pm$ 22    | 1400                | 4.8                 | 2.1 $\pm$ 0.2     |

$^a$R = rock, T = tile. $^b$New city map of Nagasaki.

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**Fig. 1.** The locations of mineral samples (NM1–NM9) and the hypocenter of the atomic bomb in Nagasaki.

**Fig. 2.** The chemical procedure to prepare the europium-enriched sample.

**Sample preparation**

All mineral samples were ground to powder under 100 mesh. A preliminary gamma ray measurement indicated that gamma rays from $^{152}$Eu were not observed for any sample except NM1, which was the sample nearest the hypocenter. Thus the chemical separation procedure shown in Fig. 2 was performed for all samples to enrich the europium concentration. The process was as follows. A powdered sample of 50 g was fused with about 120 g of NaOH pellets in a nickel crucible by heating at 450°C for 2 h.
The fused mass was dissolved in 1 liter of water, and the precipitate (hydroxide) was filtered with a filter paper (Toyo Advantec Co., No. 5A). The precipitate was dissolved in hydrochloric acid, and NaOH solution was added to adjust the pH to 3. After centrifugation, the supernatant was collected, and the NaOH solution added to adjust the pH to 13. Then the precipitate was filtered, washed with water, and dried, and about 1 g of enriched sample was obtained. During this process, the natural activities of uranium and thorium chains included in the sample were also enriched. To remove these activities, small amounts of Na$_2$SO$_4$ and BaCl$_2$ were added to the supernatant to form BaSO$_4$ for coprecipitating the background activities, where a CeCl$_3$ solution was added as a carrier for europium. All these processes were repeated four times and 2–13 g of enriched sample was obtained.

A portion of the enriched sample was used for the activation analysis of the stable europium and the rest was pressed into a polypropylene test tube of 13-mm diameter and 75-mm height for the gamma-ray measurement with well-type Ge detector.

Stable europium content in the enriched sample was determined by neutron activation to produce $^{152}$mEu (half-life = 9.3 h) by using $^{252}$Cf neutron source at the Research Institute for Radiation Biology and Medicine, Hiroshima University: A couple of 0.3 g samples were taken from the enriched sample and 50 $\mu$g of europium was added to one of them. These samples were irradiated about two days, and the gamma ray measurement was performed immediately after the irradiation. The weights of enriched samples and the europium contents are given in Table 1.

Gamma-ray measurement

The europium samples were measured with a low-background spectrometer composed of a well-type Ge detector, which has a 120 cm$^3$ crystal volume, then shielded with 20-cm-thick lead and incorporated with an anticoincidence circuit to suppress the cosmic ray background. The 344 keV gamma ray of the radioactivity of $^{152}$Eu was used to avoid interference lines from natural radioactivities. One run was continued for more than 10$^6$ s, and each sample was measured three times. An example of the gamma-ray spectrum of the sample NM7 is shown in Fig. 3. Partial gamma-ray spectra around the 344 keV gamma ray from $^{152}$Eu for mineral samples. NM1: Yana Bridge (GR = 311 m); NM5: Gokoku Shrine-B (651 m); NM6: Nanzan school-A (GR = 704 m); NM9: St. Maria school (943 m); and NM10: control sample (GR = 2,850 m).

Data deduction and results

The specific activity of $^{152}$Eu immediately after the bomb explosion was obtained by correcting the elapsed time and using a half-life of 13.542 ± 0.010 yr for $^{152}$Eu. The results are given in Table 2. The errors concerned with the specific activities are due to the peak counts (8–49%), the stable europium concentra-
tion (4%), and the detection efficiency (10%). The total error range was from 13% to 50%. The specific activities of $^{152}$Eu as a function of the slant range are shown in Fig. 5. The results by Nakanishi et al. are also shown in the figure. The calculation of activation to produce $^{152}$Eu in a free field in air based on the DS86 neutron fluence and revised neutron fluence $93Rev^*$ is shown in the figure by solid and dotted lines, respectively. The associated calculated-to-measured (C/M) ratios are also given in Table 2.

### DISCUSSION

#### Background contribution and detection limit

In the previous work\(^1\), the environmental neutron activation to produce $^{60}$Co was experimentally examined by measuring 4 g of cobalt oxide. It was found that actual thermal neutron flux might be a factor of about three lower than the value in the UNSCEAR report\(^2\). Since the amount of enriched europium from the atomic bomb exposed samples is several tens of micrograms, the activity level induced by environmental neutrons is about three order lower than that of the atomic-bomb induced activity in both cities.

The detection limit of interested radioactivity depends on the background counts in the gamma ray peak region. It is important to know the detection limit of the spectrometer; whether the peak counts are really reliable. The critical level $L_C = 2.33 \sigma_b$\(^2\), where $\sigma_b$ is a standard deviation of background counts, was adopted as detectable minimum counting rate (d.m.c). The peak counting rate ($n_0$) and d.m.c ($n^*$) for the Nagasaki samples and some of the Hiroshima samples are given in Table 4. According to the ratios of $n_0/n^*$ as given in this table, the measurements are almost at detection limit of about 1,100 m in the slant range in Nagasaki and 1,300 m in Hiroshima.

#### $^{152}$Eu activity as a function of distance

The calculation of neutron fluence of DS86 was revised in 1993. In the neutron fluence $93Rev$, the neutron energy group of the source term was increased to 46 groups from 27 groups, and the data base of the neutron cross sections was changed to ENDF/B-6, from ENDF/B-5. This change reduced the thermal activation rate near the hypocenter at Nagasaki by nearly a factor of two.

The measured specific activities of $^{152}$Eu as a function of the slant range are shown in Fig. 5. Nakanishi et al.\(^{13,16}\) reported the $^{152}$Eu data for twelve rocks and roof tiles sampled at nine locations in the 500–1,000 m slant range from the hypocenter. They performed gamma ray measurements with a low-background coaxial and/or a planer Ge(Li) detectors. The $^{152}$Eu radioactivity was deduced from the samarium X rays. More recently, Nakanishi et al.\(^{16}\) reported six $^{152}$Eu data for the roof tiles sampled at 1,100 m and 1,170 m in slant range. As shown in Fig. 5, the present data are consistent with the Nakanishi’s data within about 900 m, except for two anomaly high data at 600 m and

### Table 2. Results of $^{152}$Eu measurement and calculated-to-measured ratios (C/M) for Nagasaki samples.

| No. | Place            | Slant range (m) | $^{152}$Eu/$^{60}$Co (Bq mg$^{-1}$) | Calculated C/M ratio |
|-----|------------------|-----------------|-------------------------------------|----------------------|
|     |                  |                 | $^{152}$Eu                           |                      |
|     |                  |                 | $^{152}$Eu/$^{60}$Co (Bq mg$^{-1}$) | $^{DS86}$ 93Rev      |
|     |                  |                 | $^{152}$Eu/$^{60}$Co (Bq mg$^{-1}$) | $^{DS86}$ 93Rev      |
| NM1 | Yana Bridge      | 594 ± 22        | 24.8 ± 2.9                          | 16.0 ± 1.7           |
| NM2 | Urakami Church   | 671 ± 22        | 6.50 ± 1.10                         | 6.20 ± 1.7           |
| NM3 | Nagasaki Univ.   | 809 ± 23        | 2.07 ± 0.28                         | 1.90 ± 0.10          |
| NM4 | Gokoku Shrine B  | 815 ± 22        | 3.57 ± 0.63                         | 1.70 ± 0.10          |
| NM5 | Gokoku Shrine A  | 816 ± 22        | 2.73 ± 0.36                         | 1.70 ± 0.10          |
| NM6 | Nanzan School    | 850 ± 22        | 1.58 ± 0.33                         | 1.30 ± 0.10          |
| NM7 | Shimoda House    | 926 ± 22        | 1.25 ± 0.42                         | 0.67 ± 0.10          |
| NM8 | Prefectual Gymnasium A | 1024 ± 22 | 0.96 ± 0.30                         | 0.26 ± 0.10          |
| NM9 | St. Maria School | 1061 ± 22       | 0.86 ± 0.40                         | 0.22 ± 0.10          |

$^{60}$Co specific activity immediately after bomb explosion.

Hiroshima samples are summarized in Table 3 assuming that $^{152}$Eu reaches to saturation activity of 88 mBq g$^{-1}$ with the neutron flux of $8.0 \times 10^{-3}$ n cm$^{-2}$ s$^{-1}$ reported in the UNSCEAR report\(^21\). Since the amount of enriched europium from the atomic bomb exposed samples is several tens of micrograms, the activity level induced by environmental neutrons is about three order lower than that of the atomic-bomb induced activity in both cities.

The detection limit of interested radioactivity depends on the background counts in the gamma ray peak region. It is important to know the detection limit of the spectrometer; whether the peak counts are really reliable. The critical level $L_C = 2.33 \sigma_b$\(^2\), where $\sigma_b$ is a standard deviation of background counts, was adopted as detectable minimum counting rate (d.m.c). The peak counting rate ($n_0$) and d.m.c ($n^*$) for the Nagasaki samples and some of the Hiroshima samples are given in Table 4. According to the ratios of $n_0/n^*$ as given in this table, the measurements are almost at detection limit of about 1,100 m in the slant range in Nagasaki and 1,300 m in Hiroshima.

### Fig. 5. Specific activity of $^{152}$Eu at the time of the explosion as a function of the slant range. Data from Nakanishi et al.\(^{13,16}\) are shown for comparison. The solid and dotted lines indicate the calculation in a free field in the air.
AMS method is a very sensitive technique, however, the range) are in agreement with the overall calculations. Since the data has some problems. One problem is that its values in higher surface: the surface cement in a depth of a few cm typically gives profiles of $^{152}$Eu for 22 cores taken from stone embankments. 1,000 m in the ground range. They also measured depth in rocks from embankments sampled at 76 locations within scattered; two are higher than the calculation and others lower. A possible reason of their data scattering at 1,100 m might be that two of the roof tiles were not directly exposed to the bomb. Beyond 900 m, present data are comparable with Nakanishi’s two higher values at 1,100 m.

Okajima and Miyajima\textsuperscript{14} reported several $^{152}$Eu activity data in rocks from embankments sampled at 76 locations within 1,000 m in the ground range. They also measured depth profiles\textsuperscript{23} of $^{152}$Eu for 22 cores taken from stone embankments. Their data, however, cannot be compared with the present results because of their insufficient accuracy as a result of extensive scattering.

\textbf{Comparison between calculated and measured activities}

The C/M ratios of $^{152}$Eu as a function of the slant range and of $^{60}$Co and $^{36}$Cl are shown in Fig. 6. The $^{36}$Cl data taken by Straume \textit{et al.}\textsuperscript{55} at three locations (822, 1,187, and 1,261 m in the slant range) are in agreement with the overall calculations. Since the AMS method is a very sensitive technique, however, the $^{36}$Cl data has some problems. One problem is that its $^{36}$Cl values in concrete strongly depend on the depth of the sample from the surface: the surface cement in a depth of a few cm typically gives higher $^{36}$Cl values than the inside concrete. Another problem results is coming from its very long half-life ($3 \times 10^7$ y) of $^{36}$Cl. Since $^{36}$Cl is also induced by environmental neutrons, the background activity must be subtracted correctly. Recently, Ruehm \textit{et al.}\textsuperscript{26} reported preliminary $^{36}$Cl data by AMS for granite samples collected in Hiroshima. Their data were collected from 590 to 1,484 m in the slant range. The results at short distances were lower than the DS86 calculation, and those at intermediate distances (1,045–1,350 m) were close to the calculation. In a comparison of their data with $^{152}$Eu and $^{60}$Co at long distances, the background $^{36}$Cl in each sample must be subtracted correctly because the background level determination might be different for each sample as a result of its very long half-life.

As described in our previous work\textsuperscript{17}, the $^{60}$Co data determined for five steel samples were in agreement with Hashizume’s $^{60}$Co data. The results were roughly in agreement with the calculation.

Nagasaki-type bombs, Ranger-Fox Shot and Buster-Dog Shot, were tested at the Nevada Test Site and gold activation was measured\textsuperscript{25,26}. The measurement was compared with the values reported by Okajima and Miyajima\textsuperscript{14}. The agreement was improved for the calculation after addition of contribution from delayed neutrons. As a result, the agreement between the gold activation values and calculations was better than a factor of three over all ranges\textsuperscript{27}. The present $^{152}$Eu data and our previous $^{60}$Co data\textsuperscript{17} are somewhat similar to

\begin{table}[h]
\centering
\caption{Contribution of environmental neutron activation for residual $^{152}$Eu measurement in Nagasaki and Hiroshima.}
\begin{tabular}{|c|c|c|c|c|c|c|}
\hline
Sample & Place & Date of measurement & Slant range (m) & Sample weight (g) & Europium content (ppm) & Europium weight (µg) & BG activity\textsuperscript{a} B (mBq) & $^{152}$Eu activity\textsuperscript{b} A₀ (mBq) & Ratio B/A₀ \\
\hline
\textbf{Nagasaki} & & & & & & & & & \\
NM1 & Yana Bridge & 990908 & 594 & 4.0 & 3.8 & 15 & $3.1 \times 10^{-3}$ & 6.3 ± 0.1 & 2.1 ± 0.4 \\
NM2 & Urakami Church & 940906 & 671 & 2.1 & 4.1 & 8.6 & $8.0 \times 10^{-4}$ & 4.7 ± 1.2 & 1.6 ± 1.4 \\
NM3 & Nagasaki Univ. Hospital & 940917 & 809 & 3.0 & 10 & 31 & $2.7 \times 10^{-4}$ & 5.2 ± 1.7 & 5.2 ± 1.4 \\
NM4 & Gokoku Shrine-B & 991026 & 815 & 3.3 & 13 & 43 & $3.8 \times 10^{-3}$ & 2.1 ± 1.0 & 5.3 ± 1.3 \\
NM5 & Gokoku Shrine-A & 991012 & 816 & 2.5 & 8.1 & 33 & $2.9 \times 10^{-4}$ & 5.5 ± 0.7 & 1.8 ± 0.4 \\
NM6 & Nanzan School-A & 990922 & 850 & 4.0 & 5.8 & 23 & $2.0 \times 10^{-3}$ & 0.6 ± 0.1 & 3.6 ± 1.0 \\
NM7 & Shimoda House & 000128 & 926 & 4.0 & 4.4 & 18 & $1.6 \times 10^{-4}$ & 1.4 ± 0.5 & 1.1 ± 0.4 \\
NM8 & Prefectural Gymnasium & 000331 & 1,024 & 4.0 & 5.0 & 20 & $1.7 \times 10^{-3}$ & 0.3 ± 0.1 & 4.9 ± 1.3 \\
NM9 & St. Maria School & 000114 & 1,061 & 4.0 & 3.8 & 20 & $1.8 \times 10^{-4}$ & 1.1 ± 0.5 & 1.6 ± 0.4 \\
\hline
\textbf{Hiroshima}\textsuperscript{c} & & & & & & & & & \\
1 & Shima Hospital (7-4) & 910117 & 579 & 2.1 & 3.5 & 7.2 & $6.3 \times 10^{-4}$ & 73 ± 10 & 8.7 ± 1.0 \\
45 & Naka Telephone Office & 901114 & 774 & 3.4 & 8.0 & 27 & $2.4 \times 10^{-3}$ & 52 ± 5 & 4.6 ± 0.5 \\
56 & Kodo Primary School & 910525 & 916 & 3.5 & 2.8 & 10 & $8.8 \times 10^{-4}$ & 4.1 ± 1.2 & 1.9 ± 0.4 \\
65 & City Hall & 910221 & 1,171 & 1.7 & 12.4 & 21 & $1.8 \times 10^{-3}$ & 2.1 ± 1.1 & 8.6 ± 1.4 \\
68 & Primary School & 901119 & 1,450 & 3.4 & 10.6 & 36 & $3.2 \times 10^{-3}$ & 1.7 ± 1.2 & 1.9 ± 0.3 \\
\hline
\end{tabular}
\footnotesize{\textsuperscript{a}Saturation activity of $^{152}$Eu assuming 88mBq/g. \textsuperscript{b}Radioactivity in the Eu-enriched sample at the time of the measurement. \textsuperscript{c}Taken from Ref. 4.}
\end{table}
Specific $^{152}$Eu activities were determined for nine Nagasaki mineral samples exposed to the atomic bomb. The present data indicate a slight deviation from the calculation, but the measured data agree with the calculation within a factor of three overall ranges.

### CONCLUSION

Specific $^{152}$Eu activities were determined for nine Nagasaki mineral samples exposed to the atomic bomb. The present data indicate a slight deviation from the calculation, but the measured data agree within a factor of three overall ranges. The measured data agree with the calculations within a factor of three up to the slant range of 1,061 m. Further measurements of activation data should be necessary to clarify the discrepancy problem.

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