Residual Radioactivity Measurement in Hiroshima and Nagasaki for the Evaluation of DS86 Neutron Fluence

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ABSTRACT

Residual ^{152}Eu activity produced by neutrons from the Nagasaki atomic bomb has been measured in seven mineral samples located up to 1142m from the epicenter. Europium was chemically separated from the sample and gamma-ray measurement was carried out with a well-type Ge detector. Deduced specific activities were compared with previous measurements and with activation calculation based on the DS86 neutron fluence. Present results are slightly higher than the calculation at far distances. However, systematic discrepancy as has been observed in Hiroshima is not clear. Further measurements for samples beyond 1000m from the hypocenter are necessary to ensure the discrepancy problem.

INTRODUCTION

A new dosimetry system DS86 has been assessed in 1987 for survivors of the Hiroshima and Nagasaki atomic bombings. In the low-energy neutron evaluation of activation data, a systematic discrepancy has been observed between the residual ^{60}Co data measured by Hashizume et al. (1) and activation calculation (2) for both cities. However, this problem was not clarified at that time. The ^{152}Eu data in Hiroshima and Nagasaki were not accurate enough to examine the calculation. Thereafter radioactivity data were accumulated for ^{152}Eu (3,4), ^{60}Co (5,6,7,8), and ^{36}Cl (9) up to 1500 m from the hypocenter. The results revealed a systematic discrepancy between the measured data and the calculation in Hiroshima. The discrepancy shows that calculation is over estimated near the hypocenter and vice versa beyond about 1000 m. To determine the nature of the problem, interest has been concentrated on Nagasaki data to establish whether such discrepancy exists there as well as in Hiroshima. If calculation agrees with measurements in Nagasaki, the discrepancy observed in Hiroshima is not due to uncertainties in neutron transport calculation in air but to calculation of the neutron output spectrum from the Hiroshima bomb.

Residual activity measurement of ^{152}Eu in mineral samples in Nagasaki were performed by Nakanishi et al. (10) and Okajima and Miyajima (11), but a large deviation in Nagasaki data was not enough to ensure the ^{60}Co discrepancy indicated by Hashizume's data (1). Straume et al. (12) measured ^{36}Cl at three locations up to 1250 m slant range in Nagasaki, they observed good agreement between the measurements and calculation in contrast to Hashizume's ^{60}Co data. Recently Nakanishi et al.(13) reported ^{152}Eu data at about 1100 m in Nagasaki.

We collected mineral samples to determine specific activity of ^{152}Eu:Eu up to 1140 m. The sample preparation and measurements are in progress. Preliminary results are shown in this report and compared with the calculations based on DS86 neutron.

MATERIALS AND METHODS

Seven mineral samples up to 1039 m from the hypocenter were collected for ^{152}Eu measurement. These were surface samples faced to the epicenter without any shielding. Sampling location, material, altitude from the ground level as well as sea level, distances (ground range:distance between the sample and the hypocenter, slant range:distance between the sample and the epicenter) are given in Table 1.

Mineral samples were ground to powder under 100 mesh. Because of the difficulty to detect gamma rays from ^{152}Eu for powdered samples except sample 1 and 2, all powdered samples were chemically treated to enrich the europium concentration as described previously (4). Enriched samples were pressed into a polypropylene test tube (13 mm diameter$\times$7.5 cm height) for measurement with a well-type germanium (Ge) detector. Stable europium content in the final form was determined by neutron activation analysis utilizing the Kinki University reactor and also ^{252}Cf neutron source at Research Institute for Radiation Bilology and Medicine, Hiroshima University. The europium content in enriched samples are given in Table 1.

Two low-background Ge detectors were used for the gamma-ray measurement, a 124 cm^3 coaxial Ge detector (CX) and a 120 cm^3 well-type Ge detector (WL), heavily shielded with 20 cm lead. An anticoincidence circuit was incorporated with the WL detector to suppress the cosmic-ray background. Details about these spectrometers are described elsewhere (14). The europium enriched samples were measured with the WL detector. The 344 keV gamma ray was utilized to derive the radioactivity of ^{152}Eu to avoid interference lines from natural radioactivity. Efficiency calibration and data deduction have been performed in the same way as described previously (4).

In the low-level radioactivity measurement, background contribution limits the reliance of the data and the minimum detectable limit. Origins of background for ^{152}Eu measurements are as follows: 1) radioactivity contamination of the detector and/or shielding material itself, 2) natural background gamma-rays, 3) production of ^{152}Eu in samples by neutrons not related atomic-bomb neutrons but those originated to cosmic rays.

According to the background measurement without any sample, it was confirmed that there was no ^{152}Eu contamination of the detector and shielding materials. Thus, the contamination due to possibilities of 1) and 2) were rejected. To confirm that contributions from ^{152}Eu in samples produced by cosmic-ray induced neutrons or contaminations of reagents used for chemical separation are negligible, enriched samples prepared from control samples. ^{152}Eu were not detected from control samples.

The detection limit of radioactivity depends on the background counts in the gamma-ray peak region. Peak counting rate (n_0) and detectable minimum counting rate (n') were determined for both ^{152}Eu measurements, and the ratios, n_0 / n', were larger than 2 for all samples.

RESULTS AND DISCUSSION

The specific activity of ^{152}Eu:Eu at the time of bomb explosion was obtained by correcting the elapsed time using the half-life 13.542±0.010 y (15). Results are given in Table 1. The slant range of the sample was calculated taking into account the burst height and the altitude of the sampling location. The calculations* of ^{152}Eu activation based on the DS86 neutron fluence are free field in air. The associated calculated-to-measured (C/M) ratios are given in Table 2; calculation error was not included. Specific activities of ^{152}Eu:Eu as a function of slant range are shown in Fig. 1. The C/M ratios for ^{152}Eu as a function of slant range are shown in Fig. 2.

Table 1 Nagasaki samples and specific activity ^{152}Eu/Eu

No	Place	Material	Altitude (m)		Distance(m)		Eu content (ppm)	^{152}Eu/Eu ($Bqmg^{-1}$)
			ground level	sea level	ground	slant		
1	Yana Bridge	stone wall	−2	1	311	589	3.76 ± 0.20	26.3 ± 3.0
2	Urakami Church	stone wall	1.7	15.8	465	677	4.09 ± 0.40	6.8 ± 1.7
3	Gokoku ShrineA	rock	1	16	651	812	13.1 ± 0.46	2.51 ± 0.35
4	Gokoku ShrineB	rock	2.5	16	651	812	8.04 ± 0.31	4.40 ± 0.56
5	University Hospital	tile	20	30	653	809	10.44 ± 0.92	2.47 ± 0.51
6	Nanzan School	stone wall	1.5	30	703	847	5.77 ± 0.27	1.51 ± 0.31
7	Sakamotocho	brick	1	33	1039	1142	23.7 ± 1.2	0.68 ± 0.32

* Personal communication (1996) with S.D. Egbert, Science Applications International Corp., 10260 Campus Point Dr., MS C3 San Diego, California 92121 USA.

Table 2 Calculated-to-Measured ratios of ^{152}Eu/Eu in Nagasaki

No	Place	Slant range (m)	^{152}Eu/Eu (Bqmg^{-1})	Calculation DS86	C/M
1	Yana Bridge	589	26.3 ± 3.0	17.5	0.66
2	Urakami Church	677	6.8 ± 1.7	6.2	0.91
3	Gokoku ShrineA	812	2.51 ± 0.35	1.80	0.72
4	Gokoku ShrineB	812	4.40 ± 0.56	1.80	0.40
5	University Hospital	809	2.47 ± 0.51	1.9	0.77
6	Nanzan school	847	1.51 ± 0.31	1.3	0.86
7	Sakamotocho	1142	0.68 ± 0.32	0.12	0.17

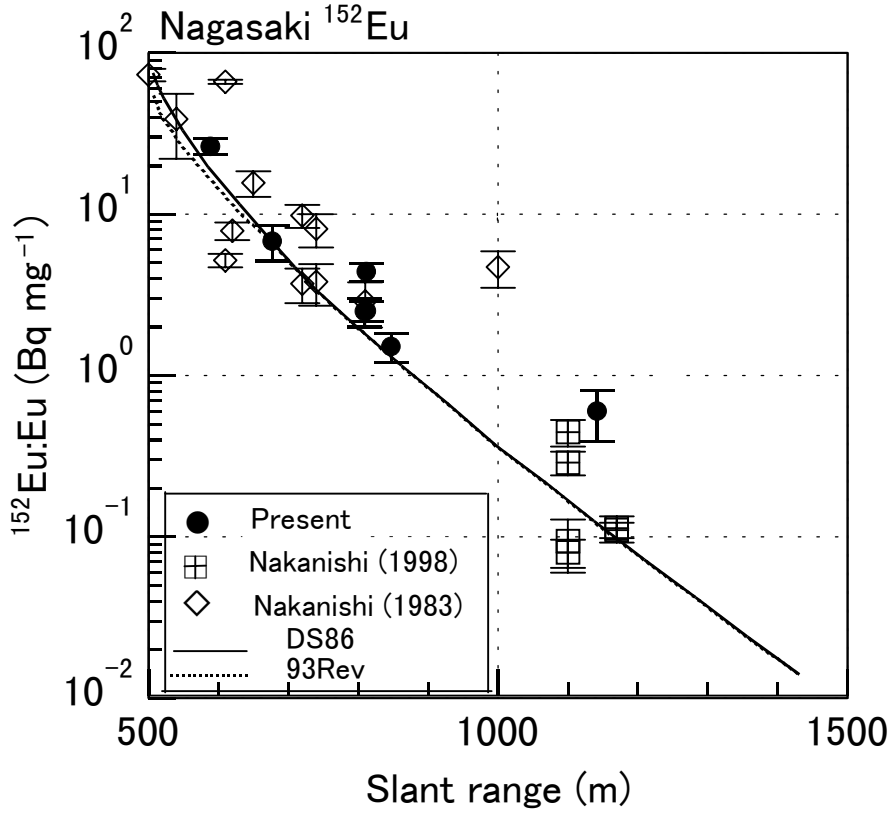

Fig.1 Specific ^{152}Eu:Eu activity at the time of the bomb explosion as a function of slant range.

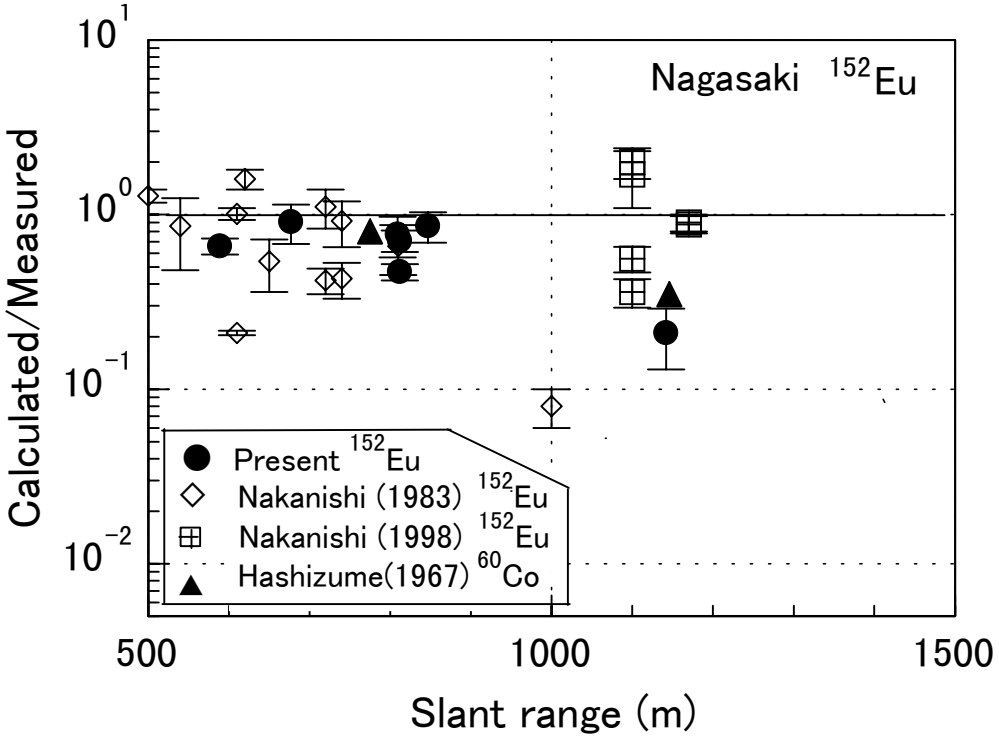

Fig.2 C/M ratios for ^{152}Eu as a function of slant range

Okajima and Miyajima (11) reported a number of ^{152}Eu activity data in rocks from embankments sampled at 76 locations within the 0-1000 m ground range. They performed gamma-ray measurement with a low-background Ge(Li) detector and deduced ^{152}Eu activity. Since their data showed a large deviations, they were not compared with the present results.

Nakanishi et al. (3) reported ^{152}Eu data for twelve rocks and roof tiles sampled at nine locations in the 500-1000 m range from the hypocenter. Gamma-ray measurement was performed with a low-background coaxial and/or a planer Ge(Li) detectors. ^{152}Eu radioactivity was deduced from the samarium X rays.

Their data were compared with the present results in Fig. 1. More recently they (13) reported six ^{152}Eu data for rooftiles sampled at 1100m and 1170m slant range. As shown in Fig.1, two of then at 1100 m are higher than the calculation, the other two are lower than the calculation and another two data at 1170m are in agreement with the calculation. Thus, their data are not enough to conclude that the measurements agree with calculation.

The ^{36}Cl data at three locations (822, 1187, 1261 m in slant range) in Nagasaki taken by Straume et al. (12) show agreement with calculation. The ^{36}Cl samples were prepared from concrete cores extracted from buildings. ^{36}Cl measurement using accelerator mass spectrometry is one sensitive technique, but the results are inconsistent with those of ^{60}Co data by Hashizume et al.(1). More data for different distances are needed to ensure the ^{36}Cl measurement.

As shown in Fig. 1, present results are slightly higher than the calculation. It can be pointed out that measurements for samples between 900m and 1100m are scarce. The C/M ratios for ^{152}Eu as a function of slant range are shown in Fig. 2. It is noted that the present ^{152}Eu are in agreement with the trend of Hashizume's ^{60}Co data. To make clear whether the discrepancy exists in Nagasaki or not , it is necessary to accumulate more activity data at distances 900-1100m.

CONCLUSION

Specific ^{152}Eu:Eu for seven samples were determined for Nagasaki samples exposed to the A-bomb. Present data support Hashizume's previous ^{60}Co data, which indicate a discrepancy between the measured data and calculation. Further measurement of activation data in Nagasaki around 1000m and evaluation of neutron transport calculation are necessary to solve the discrepancy problem.

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