Chapter 8

ACTIVATION MEASUREMENTS FOR THERMAL NEUTRONS

Part I. Ultra-Low-Background Measurements of ^{152}Eu in Hiroshima Samples

Kazuhisa Komura, Masaharu Hoshi, Satoru Endo, Tetsuji Imanaka, Hiroto Fukushima

Introduction

^{152}Eu activities induced by atomic-bomb neutrons were discussed in the DS86 Final Report (Roesch 1987). However, the ^{152}Eu data were not adopted as essential data, because they were rather scattered, and only few data were available for samples collected at more than 1 km from the hypocenter. Since the publication of DS86, a number of ^{152}Eu measurements have been performed by Shizuma et al. (1992, 1993), who developed an extremely low-level ^{152}Eu measurement method by chemical enrichment of the europium element. The ^{152}Eu activities thus obtained were found to be lower than the theoretical value near the hypocenter, but became higher with increase in the distance. The discrepancy between measured and calculated ^{152}Eu activities amounted to more than an order of magnitude in remote samples. The cause of the discrepancy had not yet been solved.

In 1998, existence of naturally induced ^{152}Eu (and ^{60}Co) was first recognized in chemical reagents by Komura and Yousef (2000) by ultra-low-background gamma spectrometry at Ogoya Underground Laboratory (OUL) (Komura 1998). Natural production of ^{152}Eu was explained by $^{151}Eu(n,\gamma)^{152}Eu$ reactions induced by environmental neutrons. Contribution of natural ^{152}Eu, however, was found to be extremely low and could not adequately explain the excess ^{152}Eu activity observed in samples at more than 1 km distance from the hypocenter. In order to verify previous ^{152}Eu measurements, 17 samples from Hiroshima and 7 from Nagasaki, which were already prepared and measured by Shizuma et al. (1992, 1993), were re-measured at OUL. The 344 keV gamma ray from ^{152}Eu was not detected except in the samples collected within 1 km and 0.6 km distances from the hypocenter of Hiroshima and Nagasaki, respectively. This strongly suggested that ^{152}Eu activities in the previous measurements were lower than the detection limits

even of the ultra-low-background Ge detectors, and/or the strong interference of the 342.7 keV gamma ray from ^{227}Ac made it difficult to detect the extremely low-level ^{152}Eu activities.

At the U.S.-Japan Workshop held in November 2001, it was decided to carry out new ^{152}Eu measurements using large amounts of granite samples and to compare the results with those of ^{36}Cl data by AMS measurement. This section describes the results of ^{152}Eu measurements obtained from large granite samples of up to 2 kg.

Samples and Chemical Enrichment of Europium

Sixteen granite rocks collected in Hiroshima were included in the intercomparison study of ^{152}Eu and ^{36}Cl. Thirteen samples from within 1.4 km of the hypocenter were intended for detection of bomb-induced ^{152}Eu activity, and the remaining 4 samples collected from more than 3 km distance were used to evaluate the background level of natural ^{152}Eu. The surface portion of 5 cm of each granite sample was removed by sawing, and the major fraction (340-2,000 g) was subjected to ^{152}Eu measurement with the residual amount used for AMS measurement of ^{36}Cl at three laboratories at the University of Utah in the United States, Ludwig Maximilians University in Germany and the University of Tsukuba in Japan.

Chemical enrichment of europium was performed at the Japan Chemical Analysis Center (JCAC) in Chiba, Japan, based on the method developed by Shizuma et al. (1993). About 40-times enrichment with a chemical yield of 65-88% was attained for europium by this chemical separation method.

Gamma Ray Measurement of ^{152}Eu Activity

The europium fraction thus separated was oven-dried at 450°C and compressed into disc shape of 19 mm in diameter. Gamma rays of ^{152}Eu were measured by two large-volume ultra-low-background well-type Ge detectors in OUL (Komura 2002) with detection efficiencies of 73.5% and 70.5% at 1,332 keV relative to 7.6 cm $\phi \times$ 7.6 cm NaI(Tl) scintillation detector. By the chemical separation, interferences of the ^{238}U- and ^{232}Th-series nuclides and ^{40}K were reduced to about 1/2,000, 1/100 and 1/10,000 on average, respectively. However, the contribution of ^{227}Ac was reduced to only 1/5-1/50, because chemical properties of actinium are quite similar to europium.

Examples of gamma-ray spectra are shown in Figure 1. Gamma-ray spectra recorded were analyzed by a least squares fitting program. In the cases of extremely low ^{152}Eu activity, the peak area was calculated also by a manual method after the subtraction of contributions of ^{238}U-series, ^{232}Th, ^{40}K and ^{227}Ac, referring to standard spectra obtained by the same counting conditions.

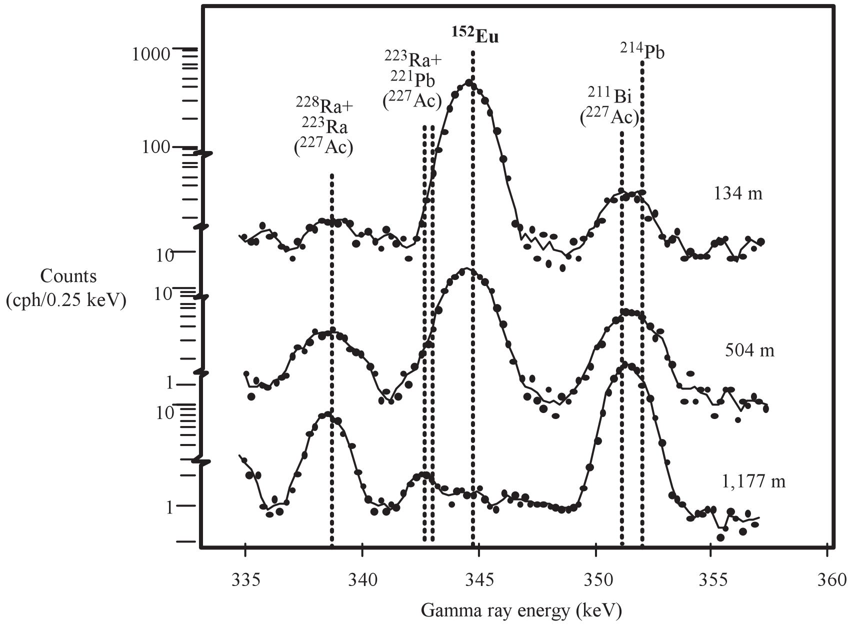

Figure 1. *Examples of gamma-ray spectra of samples from Motoyasu Bridge (134 m), Shirakami Shrine (504 m) and Kozenji Temple (1,177 m) measured by ultra-low-background well-type Ge detectors.*

Results and Discussion

Because of larger sample weights (factor of 5-10), lower background (factor >50) and higher detection efficiencies (factor >3), the present measurements could be performed with an improvement factor of higher than 100 compared with previous measurements. The lowest positive count was 0.0028 ± 0.0007 cpm (counts per minute) in the sample collected at 1,424 m from the hypocenter. The ^{152}Eu activity (Bq mg^{-1}) at the time of measurements was calculated first and then the ^{152}Eu activity on August 6, 1945 was obtained by correcting both the contribution of natural ^{152}Eu and 57 years of decay after the bombing. The value of 2×10^{-5} Bq mg^{-1} Eu was adopted as the natural ^{152}Eu activity induced by environmental neutrons by assuming that 1/3 of neutron flux (8×10^{-3} cm^{-2}s^{-1}) (Hewitt and Hughes 1980) contributes to the production of ^{152}Eu.

The ^{152}Eu activities thus obtained are given in Table 1 and plotted in Figure 2 together with previous measurements and calculated values based on DS86 and DS02. Errors given in Table 1 and Figure 2 show 1 σ of standard deviation including uncertainties of chemical yields, counting statistics of gamma-ray measurements and systematic error of detection efficiency. As is seen in Figure 2, ^{152}Eu activities obtained from granite samples in Hiroshima are well reproduced by theoretical calculation based on the DS02. Discrepancy between measured ^{152}Eu and calculated values was successfully reconciled by the present work.

Table 1. ^{152}Eu activity in granite samples in Hiroshima

No.	Sample	Ground range (m)	^{152}Eu on Aug.6, 1945 (Bq/mg-Eu)
1	Motoyasu Bridge Railing	134	99.4 ± 5.5
2	Shirakami Shrine Fence	504	15.2 ± 0.89
4	Myochoji Temple	639	6.2 ± 0.38
5	Old Prefectural Office	877	1.57 ± 0.1
3	Honkeiji Temple	896	0.99 ± 0.07
6	Enryuji Temple	925	1.06 ± 0.09
7	Shingyoji Temple	915	0.78 ± 0.06
8	City Office pavement	1022	0.27 ± 0.09
9	Kozenji Temple	1177	0.15 ± 0.03
18A	Hiroshima Univ. “E” Bldg.-1	1385	ND
18B	Hiroshima Univ. “E” Bldg.-2	1385	ND
17	Kikkawa Ryokan	1424	0.038 ± 0.019
10	Senngyoji Temple	4295	ND
11	Kannnonji Temple	5302	ND
14	Senzoubo local stone	8791	ND
15	Myokenji Temple	7611	ND

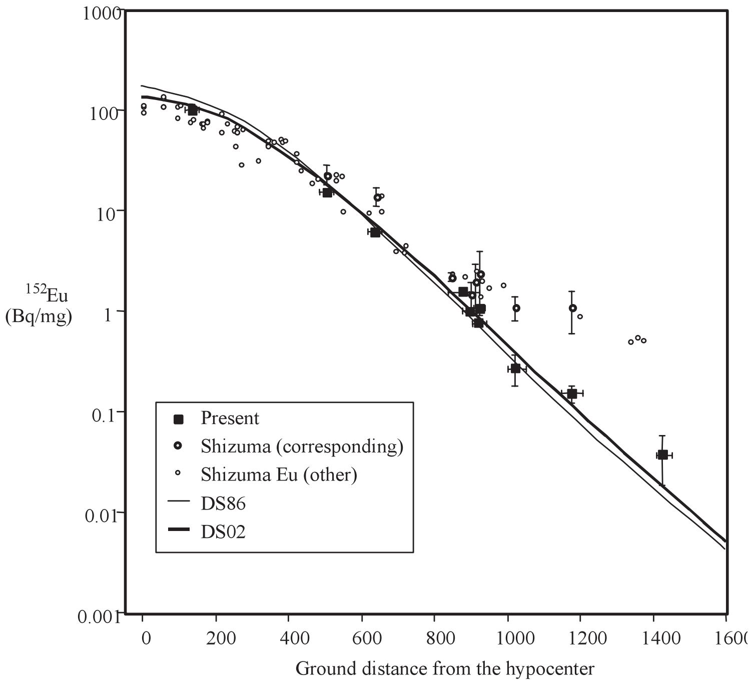

Figure 2. *^{152}Eu activity as a function of distance from the hypocenter. Results for similar granite samples from the same locations are referred to as “corresponding”; results from other locations are referred to as “other.”*

Acknowledgement

The authors express their sincere thanks to the Radiation Effects Research Foundation for financial support in chemical separation.

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