Residual Radioactivity in Neutron-Exposed Objects
and Residual Alpha Radioactivity in Black Rain Areas

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Eu-152/Plutonium/Uranium isotopes/Neutron dose/Gamma spectrometry

The residual Eu-142 radioactivity due to the atomic bomb explosion in 1945 was first found in 1976 by in-situ high resolution gamma-ray spectrometry at Hiroshima. Since then, various studies on this nuclide have continued not only in Hiroshima but also in Nagasaki and they have contributed to the reevaluation of the neutron dose due to the A-bombing.

By radiochemical separation methods and alpha-ray spectrometry, rather high levels of plutonium were found in the surface soil and in the bottom sediment of water reservoir which were collected at “Black rain area” in Nagasaki.

The U-234/U-238 activity ratios for the uranium leached with 0.1 HNO3 from the surface of soil samples were found to be relatively higher for the samples which were collected at “Black rain area” in Hiroshima.

HISTORICAL INTRODUCTION:

The radiochemical laboratory, Faculty of Science, Kanazawa University began in 1966 studies on environmental plutonium due to global fallout from aboveground atomic bomb test explosions. As extension from these studies, the surface soils of Nagasaki were collected and analyzed to know the levels of plutonium at this city where the plutonium atomic bomb explosion had occurred in 1945. Prof. G.T. Seaborg mentions in his book entitled “Transuranium Elements” that the existence of plutonium was revealed by this explosion.

Since 1976, LLRL (Low Level Radioactivity Laboratory) of Kanazawa University have also been concerned with residual plutonium, Am-241 and Cs-137. The results thus obtained until 1986 were reviewed1.

For Hiroshima where the uranium (U-235) atomic bomb exploded, two uranium isotopes (U-234 and U-238) in the surface soil were studied to clarify the contribution of the fallout from the Hiroshima atomic bomb2. The essentials of the studies are presented in the second part of this paper.

On the other hand, since LLRL has been established as a unique laboratory belonging to the national university in Japan, we have carried out the intensive studies on gamma ray radiation in the environment at various interesting points in Japan with high resolution Ge(Li) gamma
In August 2nd-4th, 1976, we carried out in-situ gamma ray measurements in Hiroshima and its outskirts at the request of the late Prof. K. Takeshita of Research Institute for Nuclear Medicine and Biology, Hiroshima University, to help his study on "Dose Estimation from Residual and Fallout Radioactivity". In the in-situ gamma ray spectrum (Fig. 8 of ref. 3) taken for 150 min. in the evening of August 2nd, 1976, under the Hiroshima memorial dome "GENBAKU DOME", we found the slight gamma ray peaks (122 KeV and 338 KeV) of Eu-152 among conspicuous gamma ray peaks due to various nuclides of natural uranium and thorium series by careful computer-aided spectrum analysis made afterwards. This means that Eu-152 with a half-life of 13.2 years induced by neutron capture reaction of Eu-151, existing as a minor constituent in ordinary rocks or bricks, remained even 31 years after the atomic bomb explosion and can be utilized for the dosimetry of bomb neutron.

In order to confirm this finding, roof tile collected near the SHIMA hospital destroyed at the hypocenter was subjected to low background high resolution gamma ray spectrometry at LLRL. Gamma ray peaks due to Eu-152 were identified clearly with those of Eu-154 (half-life; 8.5 years) and Co-60 (half-life; 5.27 years). By using a thermal neutron cross section of 5900 barn for $^{151}$Eu ($n, \gamma$) $^{152}$Eu reaction, the apparent thermal neutron fluxes at the hypocenter were estimated to be $(6.1 \pm 0.5) \times 10^{12}$ n/cm$^2$, $(6 \pm 2) \times 10^{12}$ n/cm$^2$ and $(7.9 \pm 0.6) \times 10^{12}$ n/cm$^2$ from Eu-152, Eu-154 and Co-60 respectively.

While these studies were going on in Kanazawa University, the problems of reevaluation of neutron and gamma doses were advocated in U.S.A. In response to this situation, Dr. T. Nakanishi and others have developed their studies on specific radioactivities (Eu-152/Eu-151 ratios) of the samples collected from various points in Hiroshima and Nagasaki. A part of the results thus obtained was first presented at the sixth International Congress of Radiation Research held in Tokyo (Abstract B-22-3) in May, 1979, and also published.

In July, 1977, our in-situ gamma ray spectrometric studies were also carried out at Nagasaki and the existence of Eu-152 was also identified in the basic stones in the garden of Mr. Takatani residence at about 365 m distance from the hypocenter, though it was not detected at the old front gate pillar of Nagasaki Medical University and at the ruined bell tower falling from the URAGAMI church.

In December of 1979, in-situ gamma spectrometric measurements by a portable pure Ge detector (Low Energy Photon Spectrometer; LEPS) were carried out to estimate the Eu-152 concentrations in the granitic pillar of a bridge (Motoyasu) and in the walls of a concrete building (now the REST HOUSE) which are located about 150 m from the Hiroshima hypocenter. The concentrations of Eu-152 in the objects (Fig. 14 of ref. 3) were calculated from the measured counting rates of 122 keV gamma ray of Eu-152 by the method proposed by H.L. Beck et al., assuming the homogeneous distribution of Eu-152. It was interesting that the back wall of the building showed higher Eu-152 concentration than the front wall of the same building facing directly to the epicenter of the A-bomb explosion in Hiroshima.

Since 1982, the special research groups were organized with grants from the Ministry of Education to reassess, especially by experimental methods, radiation doses due to the atomic bombs explosions in Japan. In November 1982, we made our in-situ measurements again in...
Fig. 1. Locations of in-situ measurements
(a) Horizontal view  (b) Vertical view

Fig. 2. Sketches of two KOMAINUs (Guardian Dogs of the shrine) and locations of Measurements at their Pedestals.
Hiroshima at various points having different distances from the hypocenter. And four concrete core samples were sampled from the walls of the REST HOUSE to know the depth profiles of Eu-152 with the kind cooperation with Prof. K. Yokoro and others of Hiroshima University.

The essentials of the non-destructive gamma spectrometric studies on Eu-152 made by LLRL are presented in the following part (I) of this review, while the various studies accompanying radiochemical separation procedures supervised by Dr. T. Nakanishi of the Radiochemical Laboratory, Kanazawa University, are presented in his review in this supplemental issue.

RESIDUAL NEUTRON-INDUCED ACTIVITY

1. In-Situ Measurement

The locations chosen for the in-situ high resolution gamma ray spectrometric measurements in November 1982 in Hiroshima are shown in Fig. 1, horizontally (a) and vertically (b). Initially, the calibrations for three coaxial Ge detectors and one Ge-LEPS were carried out at the west-side wall of the REST HOUSE (about 150 m from the hypocenter). The in-situ measurements were made at the pedestals of two different kinds of guardian dogs (KOMAINUs) of the old Hiroshima GOKOKU SHRINE, though they have now been moved to the new campus of the shrine near the Hiroshima CASTLE. One of them is made of bronze (Bronze KOMAINU) as shown at the upper part of Fig. 2 and was located about 180 m from the hypocenter (GOKOKU SHRINE-1) at the time of atomic bomb explosion. The other is made of stone (Stone KOMAINU) as shown at the lower part of Fig. 2 and was located about 370 m from the hypocenter (GOKOKU

Fig. 3. Variation of Normalized Eu-152 Activity with Slant Distance.
The remaining stone stacked up wall of the HIROSHIMA CASTLE about 850 m from the hypocenter was also subjected to the in-situ gamma ray spectroscopic measurement. The data on 344 keV gamma ray (Eu-152) obtained by these in-situ measurements with one of the detectors (Ge-1) having the highest detection efficiency are shown in Table 1 together with the data on 352 keV gamma ray (Pb-214) belonging to U-decay series. And the counting rate ratios of these two gamma ray peaks in each spectrum are calculated to know the relative intensity of the induced specific activity of Eu-152 in each object, because it was shown by the other study that the contents of Eu-151 found by activation analyses for the core sections of the wall have fairly good correlation with the counting rates of the 352 keV gamma ray due to Pb-214 (Fig. 3 of ref. 8) and the normalized Eu-152 activity by the 352 keV gamma ray in the spectrum is considered to present the relative values of the residual Eu-152 in the objects. The variation of such normalized Eu-152 activity with the slant distance from the epicenter of A-bomb explosion is shown in Fig. 3. The normalized values decrease clearly with increasing slant distances and the slope of a straight line fitted to the data points (2, 3 and 4 in the figure) is similar to the result reported on roof tiles in Hiroshima, although the value for the REST HOUSE (1) is slightly lower than that expected owing to the shielding effect of the building or the absorption of the lower Eu-152 gamma ray by mortar on the newly resurfaced wall.

As for the data of two KOMAINUs, the points (2 and 3) in Fig. 3 are the data for the Fronts (2) and (3) in Table 1) irradiated directly from the A-bomb explosion. Seen from Table 1, for the Bronze KOMAINU, as the radiation came just over the KOMAINU (the angle of elevation to the epicenter is about 72°), the value for the Back (2) is nearly the same as that for the Front (2), though the value for the Under Part (2) under the bronze dog is lower due to

| Location of Measurement | Height of Detector (m) | Radioactivity (cpm) | Activity Ratio |  |
|-------------------------|------------------------|--------------------|----------------|-----|
|                         |                        | 344 keV            | 352 keV        |     |
|                         |                        | Eu-152             | Pb-214         |     |
| West Wall of REST HOUSE | 1.0                    | 6.19 ± 0.37        | 44.06 ± 0.72   | 0.140 ± 0.009 |
| BRONCE KOMAINU (SHRINE-1)|                        |                    |                |     |
| 2) Front of Pedestal    | 0.7                    | 14.91 ± 1.28       | 64.53 ± 1.32   | 0.231 ± 0.019 |
| 3) Back of pedestal     | 0.7                    | 14.91 ± 0.61       | 65.91 ± 1.72   | 0.226 ± 0.010 |
| 4) Under Bronze Dog     | 2.2                    | 3.11 ± 1.35        | 22.86 ± 0.41   | 0.138 ± 0.061 |
| STONE KOMAINU (SHRINE-2)|                        |                    |                |     |
| 5) Front of Pedestal    | 1.0                    | 7.77 ± 0.65        | 84.30 ± 0.90   | 0.092 ± 0.009 |
| 6) Back of Pedestal     | 1.0                    | 4.49 ± 1.64        | 89.59 ± 1.66   | 0.050 ± 0.018 |
| 7) Lower Part of Back   | 0.35                   | 6.30 ± 0.58        | 87.27 ± 1.99   | 0.072 ± 0.007 |
| 4) Stone Wall of HIROSHIMA| 1.0                    | 0.76 ± 0.17        | 73.25 ± 0.71   | 0.010 ± 0.002 |
the shielding effect by the bronze dog. On the other hand, the values of the Back (3) 6) and the Lower Part (3) 7) at the Stone KOMAINU are lower than the value of the Front (3) 5) due to the shielding effect by the stones, because the angle of elevation to the epicenter is about 57° at this point.

The data of the in-situ gamma ray spectrometries made by the other two detectors (Ge-3 and Ge-4) at the outside and the inside of the REST HOUSE shown in Fig. 4 are brought together in Table 2. As compared with the value of the 344 keV/352 keV ratios at the outside of the west wall (#1), the values of this ratio at other locations (#4 and #5 for Ge-3, #2 and #6 for Ge-4)

| Location of Measurement | Radioactivity (cpm) | Activity Ratio |
|-------------------------|--------------------|----------------|
|                         | 344 keV     | 352 keV     | 344 keV     | 352 keV     |
|                         | Eu-152      | Pb-214      |              |              |

By Ge-3 Detector

| #1 Outside of West Wall, 1st Fl. | 1.13 ± 0.20 | 8.28 ± 0.66 | 0.137 ± 0.026 |
| #4 Inside of E-N Wall, 3rd Fl.  | 0.97 ± 0.08 | 13.47 ± 0.52 | 0.072 ± 0.006 |
| #5 Center Wall, Inside of 3rd Fl. | 0.23 ± 0.03 | 15.69 ± 0.40 | 0.015 ± 0.002 |

By Ge-4 Detector (LEPS)

| #1 Outside of West Wall, 1st Fl. | 0.55 ± 0.09 | 1.77 ± 0.15 | 0.312 ± 0.056 |
| #2 Outside of South Wall, 1st Fl. | 0.29 ± 0.03 | 2.05 ± 0.06 | 0.141 ± 0.015 |
| #6 Floor Surface Center, 3rd Fl. | 0.11 ± 0.03 | 2.21 ± 0.06 | 0.049 ± 0.015 |
are definitely lower owing to the shielding effect of the building to neutron.

On the other hand, it must be remarked that, in the gamma ray spectrum for the Bronze KOMAINU, the gamma ray peaks of Co-60 was identified conspicuously. The Co-60 might be produced by the fast neutron reaction, Co-63 (n, α) Co-60, having a cross section of 23 mb for about 14-15 MeV neutron, from copper (one of the main components of bronze). However, this Co-60 is considered to be mainly produced by the thermal neutron reaction, Co-59 (n, γ) Co-60, having a larger cross section of 36 barn, from a small amount of cobalt contained as an impurity in that bronze, because the ICPF analysis for the shavings from the bronze basis of the KOMAINU, made by the courtesy of Nishin Steel Co., Kure, showed the following results and furthermore the cross section of the above-mentioned (n, α) reaction is only about 0.5 mb for fission neutron of effective energy.

Cu 79.53%; Sn 3.63%; Zn 0.64%; As 1.18%; Pb 13.25%; Fe 0.34%; Ni 0.17%; P 0.001%; Co 80 ppm.

2. Depth Profile of Eu-152 in the Core Samples

Since it is important to know information on fast neutron for reevaluation of doses due to A-bombs, four concrete wall cores were drilled out from the wall of the REST HOUSE in

Fig. 5. Depth Profiles of Normalized Eu-152 activity in Boring Cores.
the directions of arrows shown in Fig. 4(b). The cores #1, #2 and #3 were drilled out from the outer side of the wall on the first (ground) floor, while core #4 was drilled from the inner side of the wall on the third floor. Each core had a diameter of 10 cm and the borings were made to a depth of about 15 cm for cores #1, #2 and #3. For core #4, the boring was made at first to about 17 cm deep and the remaining wall of 5 cm thickness was taken off in November 1983.

The core samples were cut into sections of about 1 cm thickness and each section was subjected to nondestructive low-background high resolution gamma ray spectrometric measurements. To know the relative intensity of thermal neutron flux which induced the Eu-152 radioactivity, the variation of the normalized Eu-152 radioactivity, that is the activity ratio of 344 keV Eu-152 gamma ray peak to 352 keV Pb-214 gamma ray, is shown in Fig. 5 for each core sample with depth.

In general, these normalized Eu-152 activities decrease exponentially with depth of each core, but some differences are seen especially near the surface of each wall owing to the effect of the thermalization of fast neutron in each core taken from the different sides of the REST HOUSE. The fit of these data with the production rate of Eu-152 calculated from the spectrum of fast neutron will contribute to the reevaluation of the neutron due to the Hiroshima A-bomb explosion, considering the shielding effect of the building.

Other than these studies on neutron dosimetry, fission track studies were made for zircon crystals separated from a granite block and also three broken pieces of stained glass exposed to the radiation of A-bomb explosion in Nagasaki. As reported in detail, though the informations on annealing of fossil fission tracks in zircon crystal due to the high temperature of A-bomb radiation and the upper limit of the thermal-neutron fluence irradiated to the stained glass were obtained, the neutron dosimetry by this method was not fruitful because of the low concentration of uranium in the stained glass studied and the existence of fairly large numbers of fossil tracks in zircon of old granite.

RESIDUAL ALPHA RADIOACTIVITY IN BLACK RAIN AREAS

1. Plutonium in Nagasaki

In the surface soil samples collected from Nagasaki in 1969 at the Nishiyama area where "black rain" fell just after the nuclear explosion in 1945, a high level of residual plutonium was found. In Fig. 6, the data for surface soil samples collected from less disturbed regions such as cemeteries and grassland are shown together with the data for bottom sediment samples of four water reservoirs, kindly supplied afterwards by Nagasaki University. Depth profiles of plutonium and Cs-137 show that the level of plutonium (Bq/kg) is highest at the depth of about 24 cm, though the highest level of Cs-137 (130 Bq/kg) is found at the depth of about 12 cm (Fig. 2 of ref. 1). This peak of Cs-137 is considered to correspond to the early period 1960s when heavy global fallout due to test explosions in the strastophere occurred, while the highest plutonium level reflects the deposition of the 1945 A-bomb.

The vertical migration distributions of plutonium and Cs-137 were studied in the surface soil of Tabunokizawa cemetery at Nishiyama area in Nagasaki. The soil having lichens on the
surface and plant roots penetrating deeper were sampled by 20 cm x 20 cm squares to the depth of 20 cm (every 2 cm thickness until 10 cm deep and then every 5 cm thickness). It was found that Cs-137 remained largely with lichens near the surface and plutonium existed mainly at the depth of 5 cm - 15 cm.

The plutonium mentioned above is Pu-239 and Pu-240 which are not discriminated by alpha ray spectrometry after radiochemical separation of plutonium from the sample. The determinations of Pu-241 and other plutonium isotopes as well as Am-241 and Cs-137 were made for the soil samples collected from Nishiyama area in Nagasaki13). The observed radioactivity ratios of each pair of nuclides are obviously different from those of the corresponding pair of nuclides in the environmental samples subjected only to the global fallout due to worldwide atomic bomb test explosions. In particular, the $^{239+240}$Pu/Cs-137 ratios of Nagasaki are higher (by a factor of ten) than those (about 0.02) of the global fallout, whereas the Pu-238/$^{239+240}$Pu ratios of Nagasaki are only slightly higher than those (0.03–0.04) of global fallout. On the other hand, the Pu-241/$^{239+240}$Pu and Am-241/$^{239+240}$Pu ratios are 0.36–0.44 and 0.04–0.042 respectively for the samples taken at Nishiyama area in Nagasaki. From these data, the contribution of the plutonium of Nagasaki atomic-bomb to the measured total $^{239+240}$Pu in each sample was estimated to be as high as 95–97%14).

As for the isotopic ratio of Pu-240/Pu-239, we have developed a new method of
determination\textsuperscript{15} by measuring the LX-ray emission rate with the alpha ray emission rate, though these two isotopes emitting similar energy alpha rays can not be discriminated only by alpha ray spectrometry. By applying this methods to various samples\textsuperscript{16}, it was found that the Pu-240/Pu-239 ratio of the sample of the “black rain” area in Nagasaki was significantly lower (about 0.08) than those (the order of 0.18) of the samples subjected only to global fallout.

2. Uranium Isotope Ratios in Hiroshima

In contrast to the plutonium atomic bomb in Nagasaki, the entire Hiroshima city was subjected to the U-235 atomic bomb in 1945. Collaborative work with the Research Institute for Nuclear Medicine and Biology, Hiroshima University was carried out on the uranium isotopes in Hiroshima especially to clarify the fallout effect at the “black rain” area in Hiroshima\textsuperscript{2}. Since it is difficult to measure U-235 radiometrically, the U-234/U-238 radioactivity ratio was measured alpha-ray spectrometrically after the radiochemical separation of uranium from the sample, because U-234, a lighter natural isotope of uranium, is enriched in the process to enrich U-235.

Soil samples were collected in 1976 from the surface to 10 cm depth at 2–30 km distances from the A-bomb hypocenter in Hiroshima. Each dried soil sample of 20 grams was at first subjected to a mild leaching of soil surface uranium with 0.1 M nitric acid of 60ml for 5 hours at room temperature and then the residual soil separated by centrifugation from the suspension was treated with 8 M nitric acid of 60ml for an hour at 120°C. Uranium radiochemically separated from both solutions was electrodeposited on a stainless steel plate and measured by alpha ray spectrometry.

The data thus obtained show that the U-234/U-235 activity ratios from the solutions by mild leachings are above 1.00 for the soils taken from “black rain” area and those for the soils from the control area are below 1.00. This ratios from the solutions by 8 M HNO\textsubscript{3} are nearly 1.00 or below 1.00 for both areas. By plotting the U-234/U-238 activity ratios from the solutions by mild leachings against Cs-137 concentrations measured with a Ge(Li) gamma ray spectrometer, it was revealed that the U-234/U-238 activity ratios are tended to increase with Cs-137 concentrations for the samples from “black rain” area, though those for the samples from the control area do not increase with Cs-137 concentrations. These facts suggest that the fallout nuclides from Hiroshima A-bomb still remain in the “black rain” area and its fissile raw material was the enriched U-235.

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