Title	Measurements of Europium-152 Depth Profile of Stone Embankments Exposed the Nagasaki Atomic Bomb for Neutron Spectrum Analysis (Commemoration Issue Dedicated to Professor Hidekuni Takekoshi On the Occasion of His Retirement)
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Measurements of Europium-152 Depth Profile of Stone Embankments Exposed the Nagasaki Atomic Bomb for Neutron Spectrum Analysis

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Quantitative measurement of neutron-induced radionuclide of ^{152}Eu in rocks near the hypocenter (ground center of the atomic bomb explosion) in Nagasaki was performed to obtain the depth profiles and calculate the neutron energy spectrum. Core samples were drilled and taken from the stone embankments on both sides of river within a radius of 500 m from the hypocenter. After cutting each core into about 27 mm-thick sections, each section was measured its gamma-ray spectrum with a pure germanium semiconductor detector and analyzed a content of natural europium by the activation method. The highest value 8.0×10^{-2} Bq/μg of ^{152}Eu at the time of the blast was obtained from the surface plates of rock cores collected near the hypocenter. The surface activity of cores was reduced with increasing the slant distances from the hypocenter. The slopes of the depth profiles were similar among samples taken from the same location.

In order to analyze the depth profile of ^{152}Eu activity in rock andesite, experiments using a fast neutron reactor and thermal neutron reactor were carried out. Comparing the measurements on the A-bomb exposure rock with the simulated results at the reactors, among the experiments, the depth profile using the neutron moderator of 10 mm polyethylene was closed to that obtained from the A-bomb exposed samples. The experiment of thermal neutron incidence only could not reproduce the profiles from the A-bomb exposed samples. This fact indicates that the depth profiles of ^{152}Eu in rock exposed to the A-bomb include valuable information concerning the neutron spectrum and intensity.

KEY WORDS: Atomic Bomb/ Residual Activity/ ^{152}Eu/ Dosimetry/ Nagasaki/ Neutron/

INTRODUCTION

An atomic bomb (A-bomb) was detonated 503 m above Nagasaki, Japan on 9 August 1945, three days after the detonation over Hiroshima.

The A-bomb doses of two cities were determined by Auxier of Oak Ridge National Laboratory in 1965[1)]. The evaluated value was called T65D (Tentative 1965 dose). In

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1981, Marshall reported the results of Loewe and Mendelson's studies[2)], which revealed discrepancies between the T65D and the calculated air kerma. According to their work, the neutron dose in Hiroshima 1 km from the hypocenter was about one-tenth that of T65D. Since that time, the efforts of reassessing the dose of ionizing radiation received by survivors of atomic bombs dropped on two cities have been performed among the scientists in Japan and United States under the Radiation Effects Research Foundation (RERF) auspices. In 1986, the final report on A-bomb dosimetry was published[3)] and Dosimetry System 1986 (DS86) was recommended. DS86 is mainly based on computer calculation. In addition, there were very few measured values of neutron dose and these few data were radioactivity measurements of neutron induced ^{32}P, ^{60}Co and ^{152}Eu. The measurement of ^{32}P in sulfur of electric wire insulators is the only method for evaluating high energy neutrons (above 3MeV)[4)]. However, the activity of ^{32}P can not be measured today because of its short half-life (14.2 days). The radioactivities which can still be measured with statistical significance are those of ^{152}Eu, ^{154}Eu and ^{60}Co, whose half-lives are 13.2, 8.5 and 5.3 y, respectively. Europium and cobalt have large cross sections for thermal neutrons. Therefore, tissue kerma contributed by fast neutrons cannot be obtained directly from the radioactivity of ^{60}Co, ^{152}Eu or ^{154}Eu. Data for ^{60}Co were obtained using Fe rings on rooftops and Fe rods embedded in concrete in Hiroshima and Nagasaki[5)]. However, suitable samples can not obtained from various locations. Europium activities were first detected by Sakanoue et al.[6)], and measurements have been made by several groups[7−12)]. Since Eu is contained naturally in rock, many samples are obtainable from stone embankments along the rivers in Nagasaki. The nuclide ^{151}Eu is regarded as a highly sensitive neutron detector because of its large thermal neutron capture cross section, 5900 barn at 0.025 eV. That is, with a moderator for fast neutrons, ^{151}Eu can be used as a detector with fast neutron response. Moreover, the sensitivity is controllable by changing the thickness of the moderator so as to detect preferentially neutrons with various energies. Therefore the incident neutron energy on the rock surface can be estimated by measuring the change of the activity of ^{152}Eu with increasing depth in the rock. To obtain the energy spectra and tissue kerma of Nagasaki A-bomb neutrons, rock cores were taken from the embankments on both sides of the rivers near the hypocenter.

Since ^{152}Eu activities are produced mainly by thermal neutron capture, estimation of fast neutron fluences is not simple. Fast neutrons incident on a massive material, such as stone embankments, lose their energy by scattering with the rock elements until they become thermal neutrons. Then these neutrons induce ^{152}Eu activity in the rock. Therefore, the activity caused by these neutrons in the rock has a history of incident neutron energy. The depth profile of ^{152}Eu activities in the stone embankments reflects the energy spectrum of the A-bomb.

Thus, we have performed the experiments to compare the depth profile of ^{152}Eu activity in Nagasaki rock samples with that in the rock irradiated by known energies of neutrons using a fast neutron reactor at Tokyo University (Yayoi) and a thermal neutron reactor at the Japanese Atomic Energy Research Institute (JRR-4).

MATERIALS AND METHODS

1. Rock samples exposed to the Nagasaki A-bomb.

Core samples of rock (andesite) were drilled out from the stone embankments near the hypocenter in Nagasaki. The depth profiles of ^{152}Eu activities were measured at 22 locations which are shown Figures 1(A) and 1(B). Samples # 154 to # 158 were taken from a location which did not face the epicenter. Samples # 175 to # 188 were taken from a location which did face the epicenter. A cross-sectional view of the line A-A' in Figure 1(A) is shown in Figure 2.

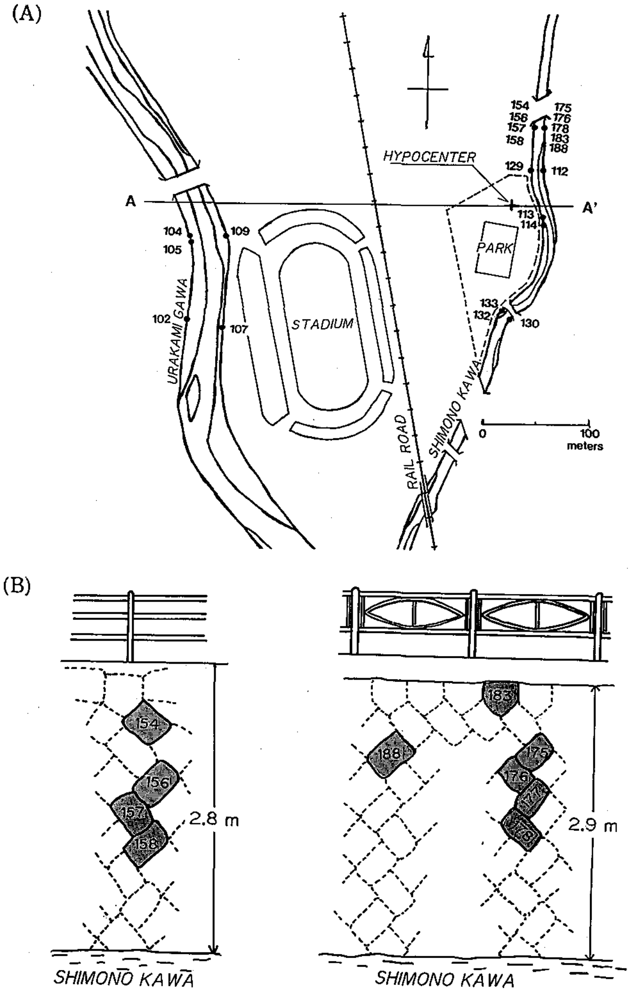


Fig. 1. (A): Locations of 21 core samples of rock in relation to the hypocenter.
(B): Samples taken from the embankments on both sides of the river Shimono-kawa. Samples numbered # 154 to # 158 were opposite the epicenter. Samples numbered # 175 to # 188 were facing the epicenter. The sampling points are nearly same distance from the epicenter.

The thickness of the sections were varied from 19.3 mm, to 42.9 mm, since the their surfaces were not flat. The cutter produced about 3 mm thick cutting losses. A typical core sample is shown in Figure 3.

Analysis of the chemical composition and water contents of three samples was performed by the Shimuzu-Kensetsu Co., Ltd. The averaged values of the chemical composition, except Eu and water are shown in Table 1. The averaged water content of the samples was 1.2 weight % (include crystalline bound water) except for the

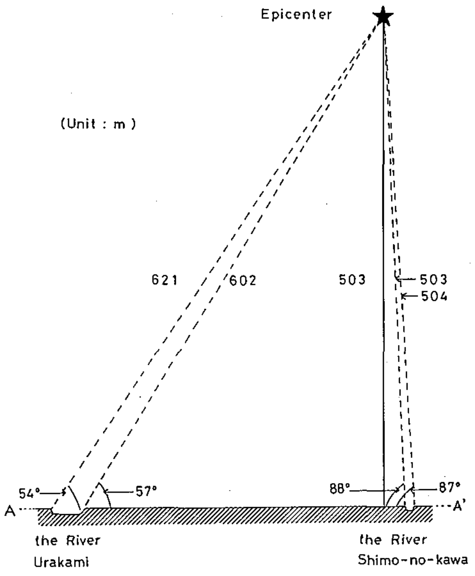


Fig. 2. A Cross-sectional view of the line A-A' in Fig. 1(A).

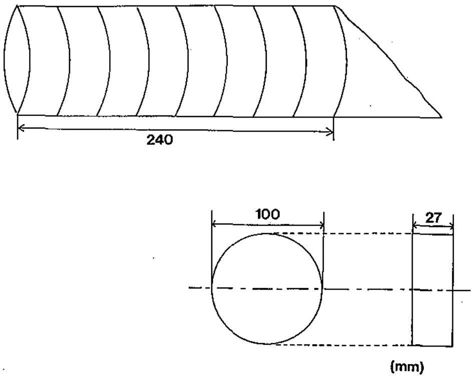


Fig. 3. A typical core sample of rock. Diameter of core samples was 99.3± 0.3 mm and 117.5±0.4 mm for #104 to #133 and for #154 to #188, respectively. This is because using two boring cylinders, 100 mm and 120 *mm in diameter*, were used. Length of core samples was from 200 mm to 450 mm. They were cut into plates about 26.5±1.5 mm thick except for the surface sections. The cutter produced 0.3 cm-thick cutting losses.

Table 1. The averaged values of the chemical composition of andesite taken from the hypocenter except Eu and water.

Chemical conposition	Content (weight-%)
SiO_2	62.0
TiO_2	0.72
Al_2O_3	16.7
Fe_2O_3	1.97
FeO	3.50
MnO	0.09
MgO	3.25
CaO	5.66
Na_2O	3.67
K_2O	2.19
P_2O	0.25

surface section. The water content of the surface section was 3.4 weight-%. This value is higher than that obtained from inside the rock because of the aboundance of free water on the surface.

Natural Eu contents of each core sample and the distribution in a sample were analyzed by neutron activation methods using the research reactor of Kyoto University (KUR). The distribution of stable Eu concentrations in sample # 114 are shown in Figure 4. The mean and standard deviation were 1.11 ±0.49 ppm (μg/1g-rock). Thus, the Eu concentrations in each sample were nearly uniform. However, the mean Eu concentrations of the various samples were deviated from 0.3 to 2.7 ppm as shown in Figure 5. Therefore, we measured the Eu concentration of all samples by neutron activation analysis using the reactor KUR.

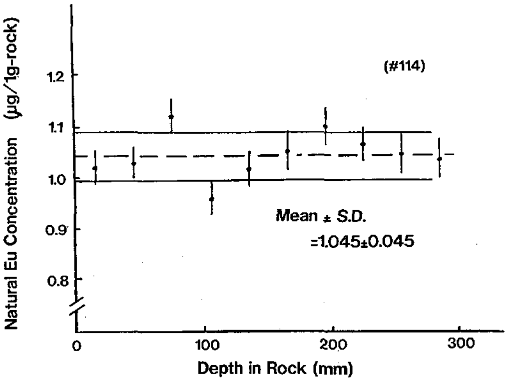


Fig. 4. Stable Eu concentrations at various locations in a core sample (# 114). Natural Eu content of the core sample was analyzed by neutron activation methods using the research reactor of Kyoto University (KUR). The mean and standard deviation were 1.045±0.045 ppm (μg/1g-rock).

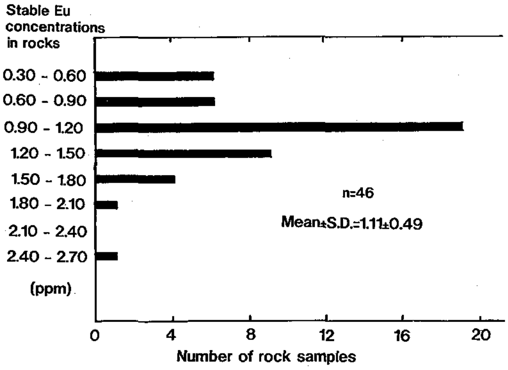


Fig. 5. Eu concentrations of the various rock samples. The concentrations ranged from 0.3 to 2.7 ppm. Eu concentration of all samples by the neutron activation analysis using the reactor KUR.

2. Reactor Experiments

Andesite rock similar to that exposed to the A-bomb was used in neutron transportation experiments. The rock was shaped as cylinder (190 mm in diameter and 300 mm in length) and cut as shown in Figure 6. It was inserted between a core of nuclear fuel and a thermal column of the fast neutron reactor Yayoi as shown in Figure 7. The rock was surrounded by concrete. The fission neutrons penetrated from left to right of Figure 7. The output energy spectrum of fission neutrons was calculated by the following formula,

$$N(E)=0.484 sinh \sqrt{2E\ exp(-E)}, \quad (E \text{ in MeV})$$

where, $N(E)$ is number of neutrons of energy E Mev.
The thermal column was filled with air.

Three sheets of Eu sheet (the weight of each sheet was about 20mg) and one sheet

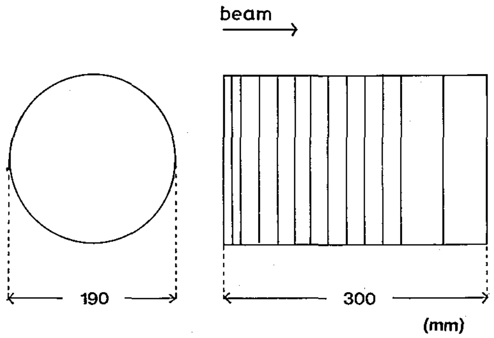


Fig. 6. Andesite rock similar to the A-bomb exposured rock used in neutron transportation experiments in the reactor. The rock was shaped as cylinder (190 mm in diameter and 300 mm in length) and cut.

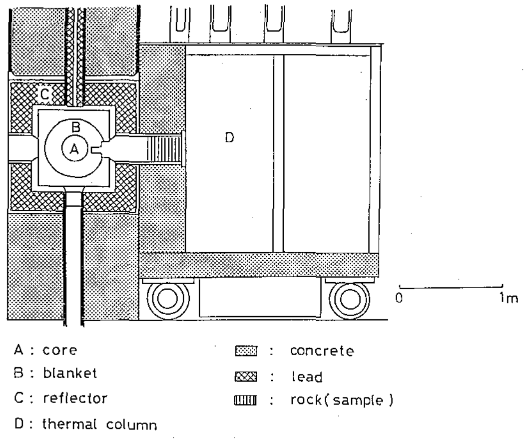


Fig. 7. The rock was inserted in the hole between a core of nuclear fuel and a thermal column of the fast neutron reactor Yayoi.

of Au foil monitoring the neutron fluence were attached to the surface of each section of rock. The depth profile in the rock was obtained by measuring the activated Eu foil.

Polyethylene plates of 10 mm and 30 mm were inserted in front of the rock to moderate the energy of the neutrons. The neutron energy spectrum from Yayoi was changed by scattering due to hydrogen atoms in the polyethylene. Thus, the neutron penetrating experiments were carried out using three kinds of neutron energy spectra.

Similar experiments were also carried out at the thermal reactor JRR-4 to allow comparison with the penetration of fast neutrons. In the experiments at JRR-4, the rock was irradiated only by thermal neutrons moderated with water.

MEASUREMENTS

1. Exposed rock from Nagasaki

A pure germanium semiconductor detector with 16 % efficiency relative to a NaI(Tl) detector 76 mm in diameter by 76 mm high was used in the gamma-ray measurements. The detector was shielded by 100 mm thick lead bricks, 20 mm thick iron plates, 5 mm thick copper plates and 5mm thick plastic plates on the inner sides. All core sections were nondestructively measured as shown in Figure 8. The side indicated by the arrow is the front of a core section. The ^{152}Eu peaks of 122 keV and 344 keV were identified from the gamma-ray spectrum of each core section.

The detection efficiency of ^{152}Eu in each disk sample was determined as follows.

1) A geometrical efficiency factor was determined by measuring the changes of gamma-ray counts when the position of a point source of ^{152}Eu was moved in the horizontal (r) and perpendicular (h) axes of the detector as shown in Figure 9. The relation between the detection efficiency (E) and the perpendicular distance (h) from the source at r=0 was computed as follows:

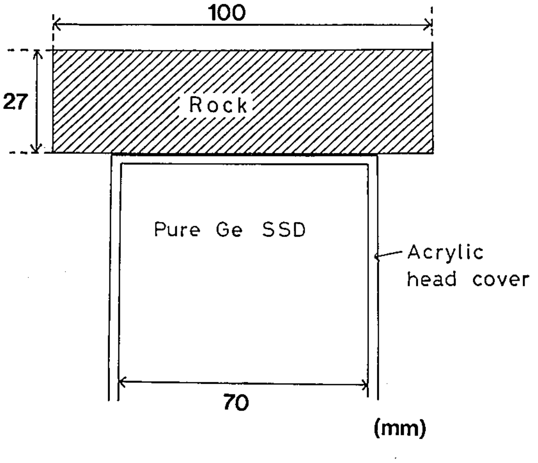


Fig. 8. The detector was shielded by 100 mm thick lead bricks, 20 mm thick iron plates, 5 mm thick copper plates and 5 mm thick plastic plates on the inner sides. All core sections were measured nondestructively.

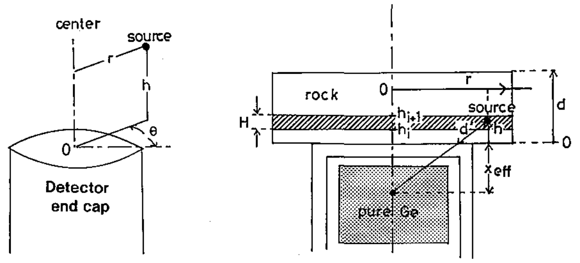


Fig. 9. Parameters for determining of detection efficiency of ^{152}Eu activity of disk samples.
(A) r: horizontal distance from the center of the detector surface to a point source, h: perpendicular distance from the center of detector surface to a point source.
(B) R: radius of the disk, H: a thickness of the disk, d': absorption distance, $d' = (h \cdot \sqrt{(X_{eff})+r})/(h+X_{eff})$ were, X_{eff} is the distance between the detector surface and the effective center of the detector.

$$E_{r=0}(h) = a_0 + a_1 \cdot h + a_2 \cdot h^2 + a_3 \cdot h^3 \tag{1}$$

where, a_0 to a_3 are constants in mm. The relation between the detection efficiency (E) and horizontal distance (r) at h=0 was computed as follows :

$$E_{h=1}(r) = \exp[\{-b_1 \cdot \exp(-b_2 \cdot r)\} \cdot r] \tag{2}$$

where, b_1 and b_2 are constant parameter.

2) The absorption coefficient of 122 keV gamma-rays in the andesite was measured by using andesite plates of 4.5 mm in thickness. The effective linear energy absorption coefficient (μ) of 122 keV for the rock (andesite) was determined to be 0.04279 mm^{-1}.

3) From above, the relative detection efficiency of a disk sample of rock is given as follows:

$$E(r, h, \mu) = \frac{2}{RH} \int_{h_i}^{h_{i+1}} E_{r=0}(h) \cdot \int_{0}^{R} r \cdot E_{h=1}(r) \cdot \exp(-\mu d')\,dr\,dh \tag{3}$$

where,
R=radius of the disk,
H=thickness of the disk,
d'=absorption distance d' was computed as follows:

$$d' = (h \cdot \sqrt{(h+X_{eff})+r}) \,/\, (h+X_{eff})$$

where, X_{eff} is the distance between the detector surface and the effective center of the detector.

4) The following measurements were made to determine the absolute ^{152}Eu content from the 122 keV gamma-ray counts of each sample.

The core rock sample taken near the hypocenter was cut into 4.5 mm thick plates and the gamma-rays of 122 keV were measured. Then the sample was milled. The rock powder was kept in a cylindrical polyethylene container and compared with a control sample of the same volume of rock powder with standard ^{152}Eu activity kept in the similar container. Thus, the absolute detection efficiency of ^{152}Eu activity for the 4.5 mm rock plate was determined. The relative detection efficiency of eq.(3) when $H=4.5$ mm was normalized the above value. The correction of the detection efficiency for each disk sample was made by using the calculation eq.(3) considered geometry and absorption effects since the thickness and the diameter of sample were varied.

To determine the amount of natural Eu contained in each rock sample, activation analysis was performed using the Research Reactor of Kyoto University (KUR)$^{8)}$.

2. Reactor Experiments

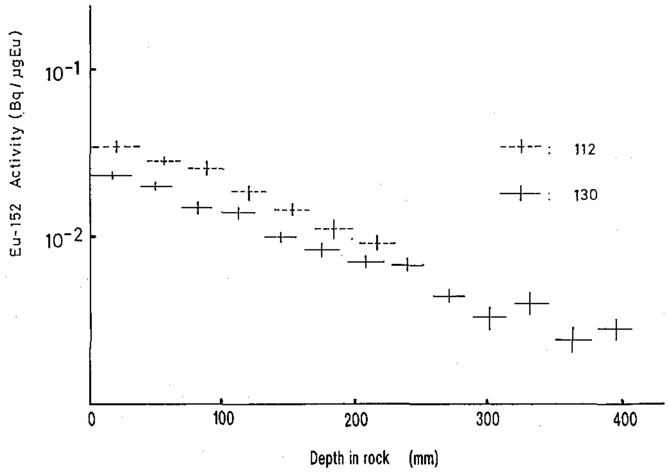
The gamma-rays from ^{152m}Eu, were measured instead of those from ^{152}Eu, to reduce statistical errors due to the small decay constant (1.67×10^{-9} sec^{-1}) of ^{152}Eu. The cross section spectra of the capture reaction of ^{151}Eu(n, γ)^{152m}Eu for neutron energy is almost the same as that of ^{151}Eu(n, γ)^{152}Eu. The half-life of ^{152m}Eu is 9.38h and the decay constant is 2.98×10^{-5} sec^{-1}. 841.6 keV of gamma-rays from ^{152m}Eu were measured by a pure germanium semiconductor detector.

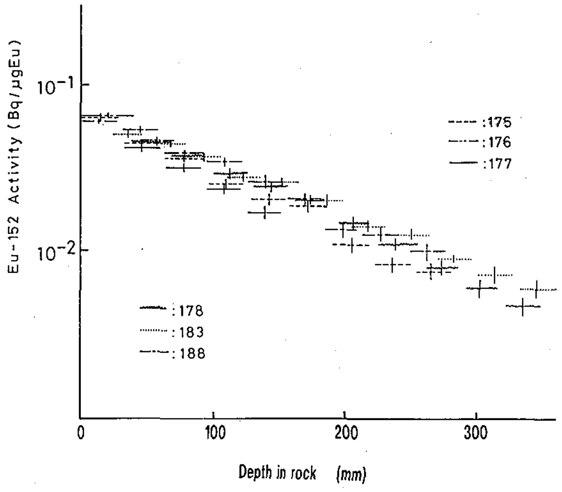
RESULTS AND DISCUSSION

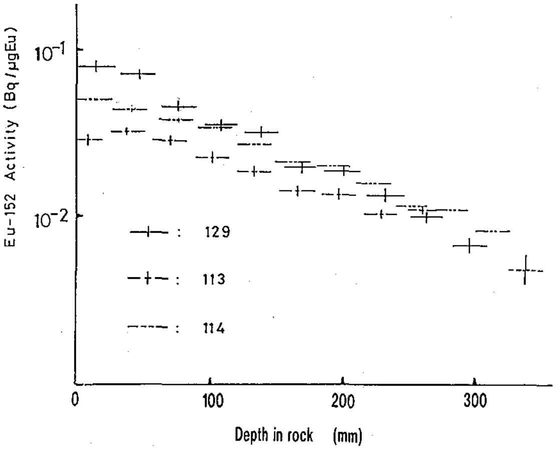
1. Depth profiles obtained from rock exposed to the A-bomb

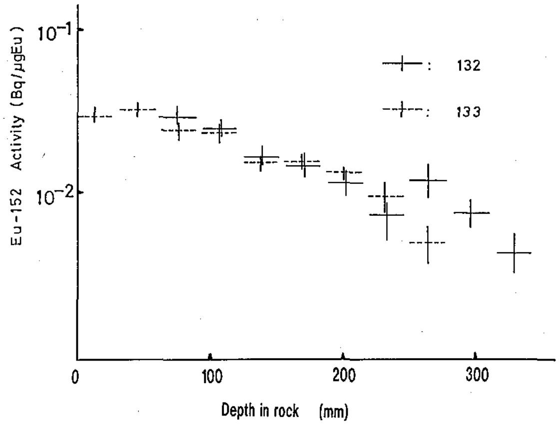
The measured values were normalized per microgram of natural Eu and converted to activity at the time of the bombing. Figures 10(A) to 10(G) show the depth distribution of ^{152}Eu activity in the rocks.

Data from samples with their surfaces facing the epicenter, taken from the embankment of the Shimono-kawa are plotted in Figures 10(A) and 10(B). Data from samples with their surface opposing the epicenter taken from the embankment of the Shimono-kawa are plotted in Figures 10(C), 10(D) and 10(E). Data from samples with their surface facing the epicenter taken from the embankment of the Urakami-gawa are plotted in Figure 10(F). Data from amples with their surface opposing the




Eu-152 Activity (Bq/µgEu)
10⁻¹
10⁻²
:175
:176
:177
:178
:183
:188
0
100
200
300
Depth in rock (mm)


Eu-152 Activity (Bq/µgEu)
10⁻¹
10⁻²
: 129
: 113
: 114
0
100
200
300
Depth in rock (mm)


Eu-152 Activity (Bq/µgEu)
10⁻¹
10⁻²
: 132
: 133
0
100
200
300
Depth in rock (mm)

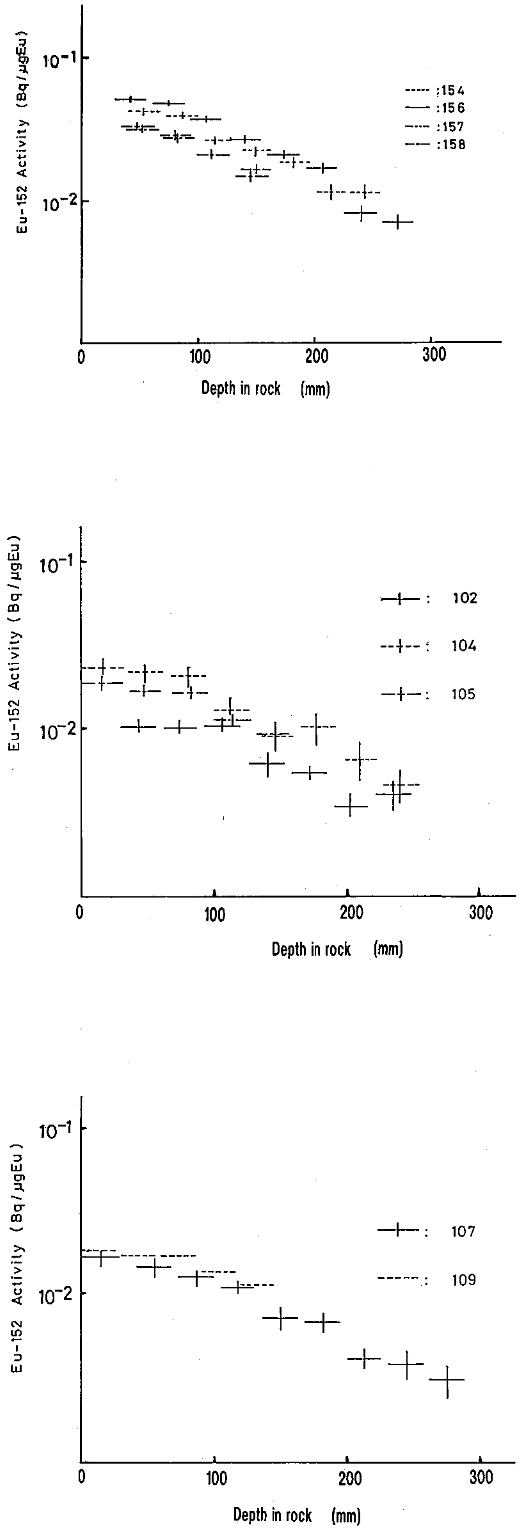


Fig. 10. Figures (A) to (G) show the depth distribution of ^{152}Eu activities in the rocks. The measured values were normalized per microgram of natural europium and converted to activity at the time of the bombing. Data from samples with their surfaces facing the epicenter taken from the embankment of the Shimono-kawa are plotted in Figures (A) and (B). Data from samples with their surface opposing the epicenter taken from the embankment of the Shimono-kawa are plotted in Figures (C), (D) and (E). Data from samples with their surface facing the epicenter taken from the embankment of the Urakamigawa are plotted in Figure (F). Data from samples taken from the opposite embankmant are plotted in Figure (G).

epicenter taken from the Urakami-gawa are plotted in Figures 10(G). These samples had monotonic reductions in ^{152}Eu activity with increasing depth. There were no large discrepancies among the depth profiles of samples taken near the hypocentor (Shimono-kawa) as shown in Figures 10(A)-(E). That is, the neutron spectra were almost same whether the samples faced or opposed the epicenter. There was wide variation in the relation between measured values of the surface plates and the slant distance from the epicenter; nevertheless, the ^{152}Eu activity tended to decrease with increasing slant distance, as shown in Figure 11. The highest value of ^{152}Eu activity of the surface plates of cores, 8.0×10^{-2} Bq/μgEu, was obtained from sample #129, which had been taken at 504 meter from the epicenter. ^{152}Eu activities of surface at that location were varied sample to sample. That is, since the quantity of thermal neutrons depends on by the surroundings, ^{152}Eu activities induced by thermal neutrons are also variable. However, at the depths greater than 80 mm, the activities were relatively constant, since the fast neutrons were not be affected by the surroundings. In the study of neutron response function on reaction rates of ^{151}Eu(n, γ) ^{152}Eu by Kosako et al[13)], ^{152}Eu activities at depths from 280 mm to 320 mm reflect the quantity of incident neutrons whose energies are from intermediate (100 eV) to fast (above 1 MeV) at the surface. That is, the thermalized neutrons with energies above intermediate at the surface are captured by ^{151}Eu with higher probability deeper than 280 mm. The average ^{152}Eu activities at 280 mm-320 mm depth in rock obtained within a radius of 100 m from hypocenter was $(7.82 \pm 0.93) \mathrm{x} 10^{-3}$ Bq/μgEu. With the value of the cross section of ^{151}Eu(n, γ) ^{152}Eu reaction for neutron energy of 0.025 eV, 5900 barn, thermal neutron fluence at the depth 280 mm-320 mm was calculated as 4.21 ± 0.50 x10^{11} n/cm^2. The thermal neutron fluence at the depth of 280 mm-320 mm corresponds to the neutron fluence above intermediate energy incident at the surface.

Among samples taken along the Urakami-gawa, the ^{152}Eu activities of those with their surfaces facing the epicenter was higher than that of samples from the opposite embankment, i. e. #107 and #109.

2. Depth profiles obtained from reactor experiments

The depth profiles of ^{152m}Eu activities in rock irradiated by four energy spectra of

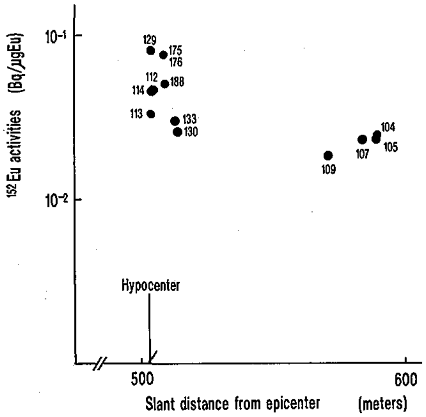


Fig. 11. ^{152}Eu activities of the surface plates plotted against the slant distance from the epicenter.

neutrons were obtained from the experiment using the reactors. The results are shown in Figure 12. The open symbols indicate the dataa from the experiments at the fast neutron reactor Yayoi. The depth profile of the fission neutrons incident on the rock is indicated by open triangles. Fission neutrons at the surface lose their energy by collision with the elements of the rock and become thermal neutrons. Therefore, the number of thermal neutrons at the surface was relatively small. The peak of ^{152m}Eu activity were at 60 mm below the surface. The activity at 180 mm was reduced to 77 % of the value at the surface. The open circles and the open squares indicate the experiments using polyethylene moderators plates, respectively. The spectrum of neutrons incident on the rock became soft since the fission neutrons are thermalized by hydrogen atoms in the polyethylene moderators. The ^{152m}Eu activity decreases because of the reduction of thermal neutrons. The decrease was $10^{-0.44}$ per 100 mm, and $10^{-0.76}$ per 100 mm for the experiments using 10 mm and 30 mm moderators, respectively.

The decrease from the results of JRR-4 was $10^{-0.52}$ per 100 mm, which was between the values obtained using 10 mm and 30 mm polyethylene moderators.

Comparing the measurements on the A-bomb exposure rock with the simulated results at the reactors, the curves obtained from the A-bomb exposed samples were closed to that obtained by using the neutron moderator of 10 mm polyethylene among the experirmntal results. The decrease was $10^{-0.34}$ per 100 mm for the samples facing the epicenter and near the hypocenter. Since the humidity in Nagasaki at the time of the bombing was 71 %, the number of hydrogen atoms in the air from the epicenter to the rock surface (503 m) was calculated to have been 7.6×10^{24}. The number of hydrogen atoms in 10 mm polyethylene plates was calculated to be about 2.1×10^{25}. Therefore, the slope obtained from the depth profiles of samples exposed to the A-bomb is not as steep as that obtained using the 10 mm polyethylene plate.

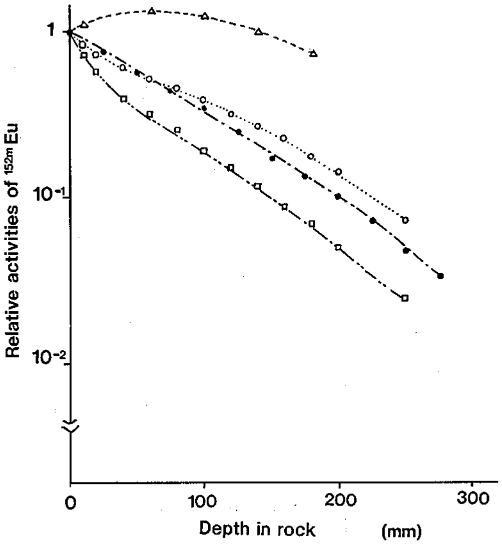


Fig. 12. Depth profiles of ^{152m}Eu activities in rock irradiated four energy spectra a in the reactors. The open symbols indicate the data from the experiments at the fast neutron reactor Yayoi. The open triangles indicate the depth profile of the fission neutrons directly incident on the rock. The open circles and the open squares indicate the experiments using 10 mm and 30 mm polyethylene moderators, respectively. The closed circles indicate the results from the thermal reactor JRR-4.

CONCLUSION

The depth profiles of ^{152}Eu in the andesite stone embankments of the Shimonokawa and Urakami-gawa exposed to the Nagasaki A-bomb were measured in detail. Moreover, experiments using reactors were carried out to compare the depth profiles and to determine if the profiles can be used to derive spectral information. Since the neutron absorption cross section for the ^{151}Eu(n, γ) ^{152}Eu reaction is large for thermal neutrons, the depth profile of ^{152}Eu indicates the fluences of thermal neutrons. However ^{152}Eu activity at large depletion length reflects the fast neutrons at the surface indirectly because they lose energy while slowing down in the rock. Comparing the measurements of the A-bomb-exposed rock with the results from the reactors, the depth profile using a 10 mm polyethy lene neutron moderator 10 mm polyethylene was close to that obtained from the A-bomb exposed samples. The experiments using the 30 mm polyethylene moderator, which make the spectrum softer, or using thermal neutron incidence only, did not reproduce the profiles from the A-bomb exposed samples. This fact indicates that the depth profiles of ^{152}Eu in rock exposed to the A-bomb can be used to determine the neutron spectrum and intensity of the blast.

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