J. RADIAT. RES., SUPPLEMENT, 83–98 (1991)
A Review of Forty-Five Years Study of Hiroshima and Nagasaki Atomic Bomb Survivors

I. DOSIMETRY

Physical Dosimetry at Nagasaki – Europium-152 of Stone Embankment and Electron Spin Resonance of Teeth from Atomic Bomb Survivors

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Atomic bomb/Nagasaki/^{152}Eu/ESR/Dosimetry

Gamma-rays from thermal neutron-induced radionuclide of ^{152}Eu in rocks near the ground center of the atomic bomb (A-bomb) explosion (hypocenter) in Nagasaki were measured with a pure germanium semiconductor detector. Depth profiles of ^{152}Eu activity were obtained for 22 core samples taken from stone embankments on both sides of two rivers (the River Shimono-kawa and the River Urakami-gawa) within 500 m of the hypocenter since the activity in the rocks has a history of incident neutron energy. Although the activities of the surface sections were varied from sample to sample, the slopes of depth profile in rock and the value of ^{152}Eu activity in the depth of 240–280 mm were similar among the samples taken from the same location. On the other hand, neutron penetrating experiments using both a fast and a thermal neutron reactor were performed to obtain the relationship between the incident neutron energy spectra and the depth profiles of ^{152}Eu activity in rock. The depth profiles in the bomb exposed samples were close to that obtained by using a 10 mm polyethelene moderator in the reactor experiments.

Electron spin resonance (ESR) measurements from teeth of A-bomb survivors were carried out to estimate the individual gamma-ray dose of the survivors. The absorbed dose of ten tooth samples was estimated by ESR dosimetry. The results of ESR dosimetry were consistent with the calculations of tissue dose in air of A-bombs and irradiated shielding configuration of individuals.

INTRODUCTION

An atomic bomb (A-bomb) was detonated 503 m above Nagasaki Japan on 9 August 1945, three days after the detonation at Hiroshima. The A-bomb doses of the two cities were determined by Auxier of Oak Ridge National Laboratory in 1965[1)]. The evaluated value was called T65D (Tentative 1965 dose). In 1981, Marshall reported the results of Loewe and Mendelson's studies[2)], which revealed discrepancies between the T65D and the calculated air kerma. Since that time, scientists in Japan and the United States have reassessed the dose of ionizing radiation received by survivors of those two A-bombs. 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. 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 be obtained from a variety of locations. Eu activities were first detected by Sakanoue et al.[6], and measurements have been continued by several groups[7-12]. In our previous work[11], the spatial distribution of ^{152}Eu from rock surface samples was reported for 40 locations of the embankments on both sides of two rivers within 1000 m of the hypocenter. ^{152}Eu activities of rock surfaces were decreased nearly exponentially with the slant distances.

However, the estimation of fast neutrons, which contributes tissue dose, from ^{152}Eu activities produced mainly by thermal neutron interaction at rock surface 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. 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 for the Nagasaki A-bomb neutrons, rock cores were taken from the embankments on both sides of the rivers near the hypocenter.

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

Electron Spin Resonance (ESR) dosimetry for A-bomb survivors was also carried out to estimate the individual absorbed dose by gamma-rays. The estimation of gamma-ray dose was carried out by the measurements of thermoluminescence of bricks and roof tiles exposed by the A-bombs. However, at present, few samples can be obtained and the estimation of absorbed dose is difficult since the complex exposed condition makes it difficult to estimate the dose in air measured by thermoluminescence. A new personal dosimetry, therefore, was required for the cases involving uncertainties of the exposing condition. ESR spectroscopy based on long-lived CO_3^{3-} radicals produced by irradiation in hydroxyapatite[13,14] was applied to estimate individual radiation dose directly from tooth enamel. In our previous work[15,16], we examined the characteristics of ESR signal of irradiated teeth and the methods of ESR dosimetry.

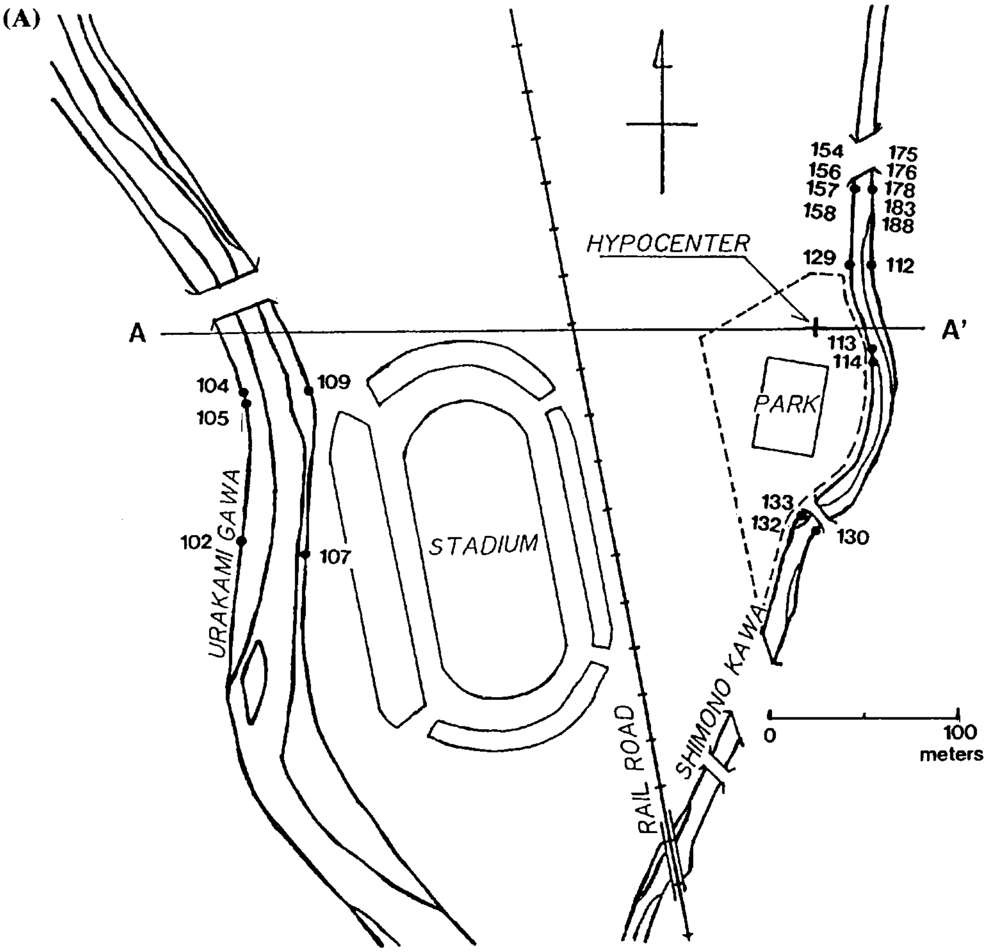
NEUTRON DOSIMETRY

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 same location which did not face the epicenter. Samples #175 to #188 were taken from a location which did face the epicenter.

The thickness of the surface sections were from 19.3 mm to 42.9 mm, since their surfaces were not flat. The cutter produced about 3 mm thick cutting losses. A typical core sample is shown in Figure 2.

Analysis of the chemical composition and water contents of three andesite samples was performed by the Shimizu-Kensetsu Co., Ltd. The averaged values of the chemical composition,



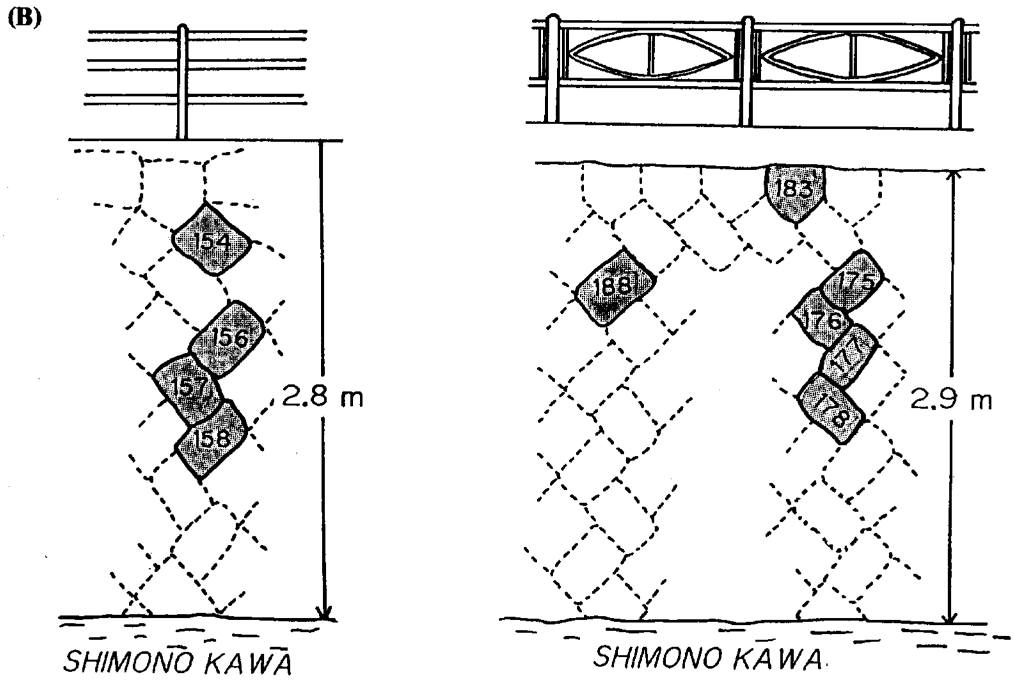


Fig. 1. (A): Locations of 22 core samples of rock in relation to the hypocenter. (B): Samples taken from the embankments on both sides of the River Shimono-kawa. Samples numbered #154 to #158 were opposite the epicenter. Samples numbered #175 to #188 were facing the epicenter. The sampling points are nearly same distance from the epicenter.

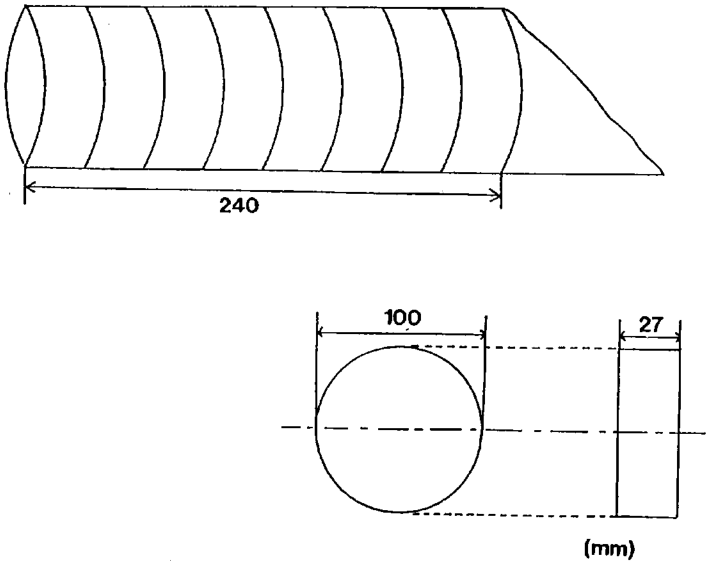


Fig. 2. A typical core sample of rock. Diameter of core samples was 99.3 ± 0.3 mm and 117.6 ± 0.4 mm for #104 to #133 and for #154 to #188, respectively. Length of core samples was 200 to 450 mm. They were cut into plates about 26.5 ± 1.5 mm thick expect for the surface sections. The cutter produced 3 mm-thick cutting losses.

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

Chemical	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

except for Eu and water, are shown in Table 1. The averaged water content of the samples was 1.2 weight-% (including crystalline bound water) except for the 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 abundance of free water at the surface.

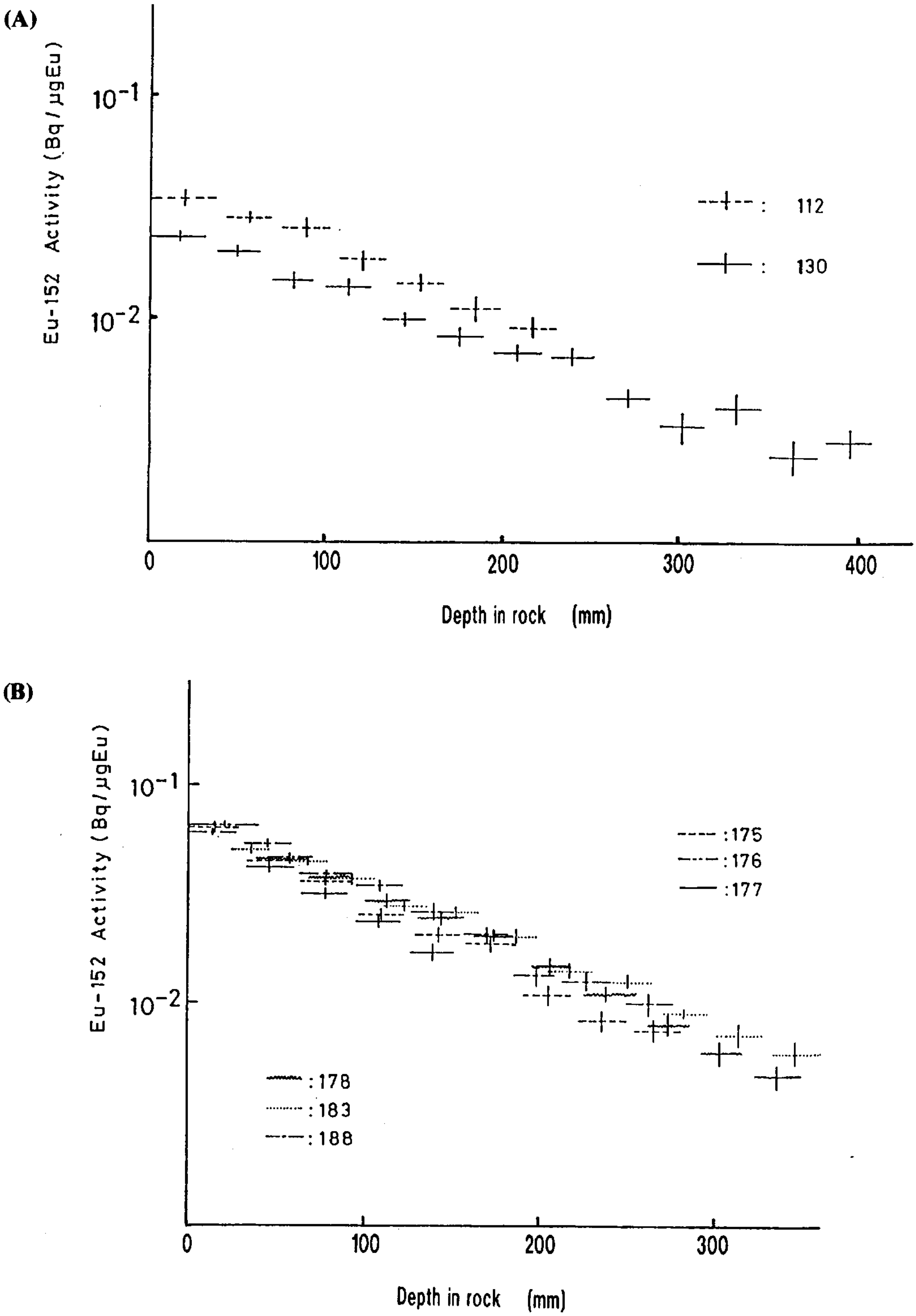
Stable Eu content of rock samples were analyzed by neutron activation methods using the research reactor of Kyoto University (KUR)[8]. The stable Eu content at various depths in a sample were nearly uniform. However, the stable Eu content of the various rocks varied from 0.3 to 2.7 ppm (μg/1g-rock) and the mean and standard deviation were 1.11 ± 0.41 ppm. Therefore, the stable Eu content of all samples were analyzed by the neutron activation methods.

A pure germanium semiconductor detector with 16% efficiency relative to a NaI(T1) detector 76 mm in diameter by 76 mm high was used in the gamma-ray measurements of the rock. 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 nondestructively measured. The ^{152}Eu peaks of 122 keV and 344 keV were identified from the gamma-ray spectrum of each core section.

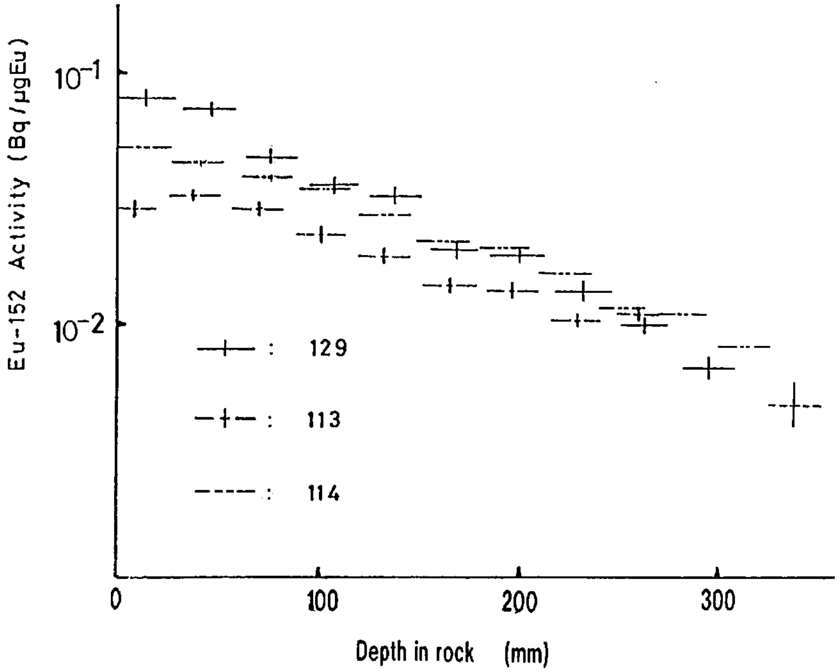
The detection efficiency of ^{152}Eu in each disk sample was calculated using the measured values of the geometrical factors in the horizontal and perpendicular axes and the absorption coefficient of 122 keV gamma-rays in the andesite rock plates.

The measured values were normalized per microgram of natural Eu and converted to activity at the time of the bombing. Figures 3(A) to 3(G) show ^{152}Eu activities at various depths in the rocks.

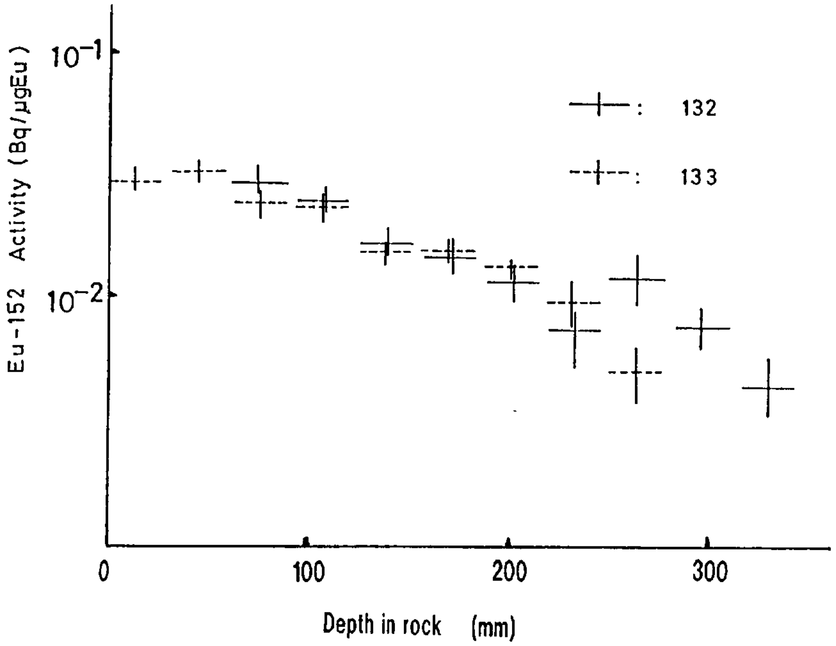
Data from samples with their surfaces facing the epicenter, taken from the River Shinono-kawa embankment, are plotted in Figures 3(A) and 3(B). Data from samples taken from the opposite embankment are plotted in Figures 3(C), 3(D) and 3(E). Data from samples with their surfaces facing the epicenter, taken from the River Urakami-gawa embankment, are plotted


(A)
Eu-152 Activity (Bq/μgEu)
10⁻¹
10⁻²
: 112
: 130
0
100
200
300
400
Depth in rock (mm)
(B)
Eu-152 Activity (Bq/μgEu)
10⁻¹
10⁻²
:175
:176
:177
:178
:183
:188
0
100
200
300
Depth in rock (mm)

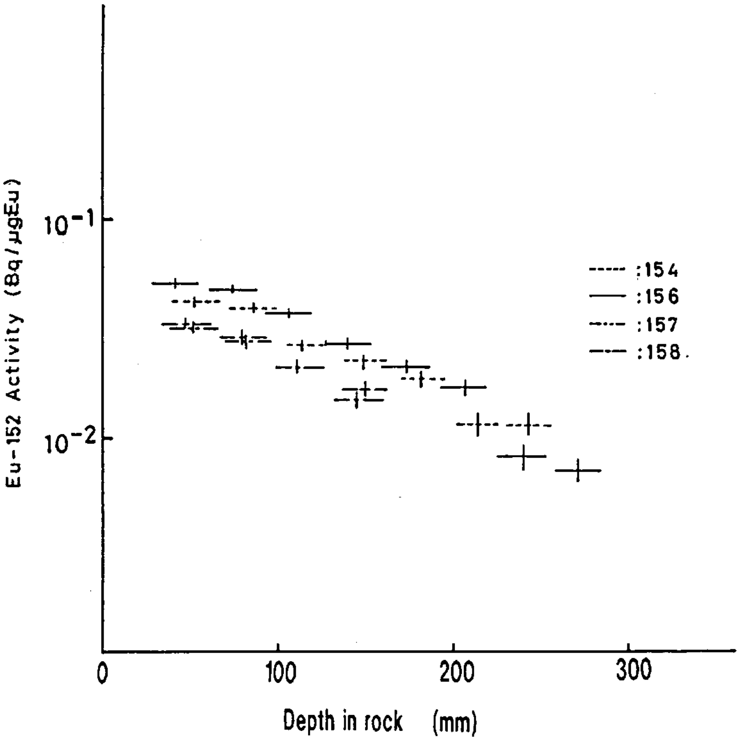
(C)


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

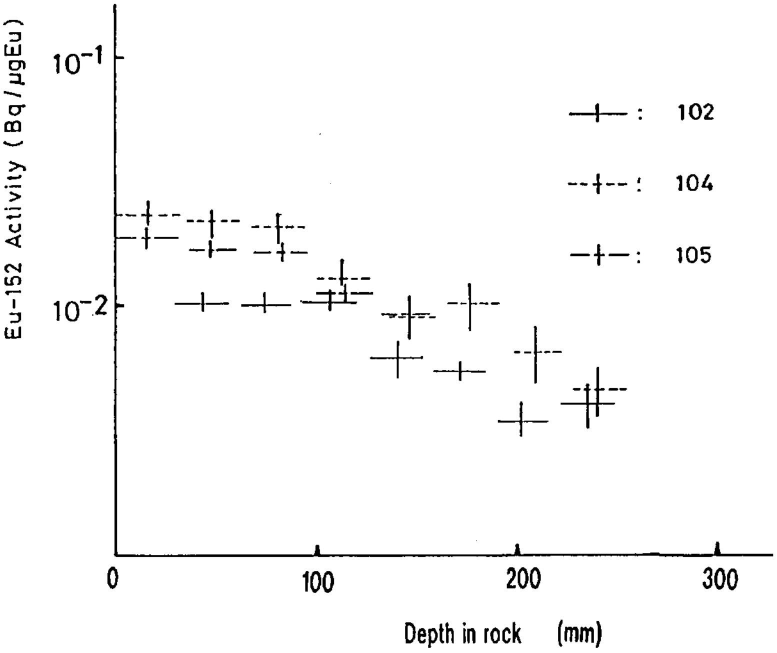
(D)


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

(E)


Eu-152 Activity (Bq/μgEu)
10⁻¹
10⁻²
:154
:156
:157
:158
0
100
200
300
Depth in rock (mm)

(F)


Eu-152 Activity (Bq/μgEu)
10⁻¹
10⁻²
: 102
: 104
: 105
0
100
200
300
Depth in rock (mm)

(G)

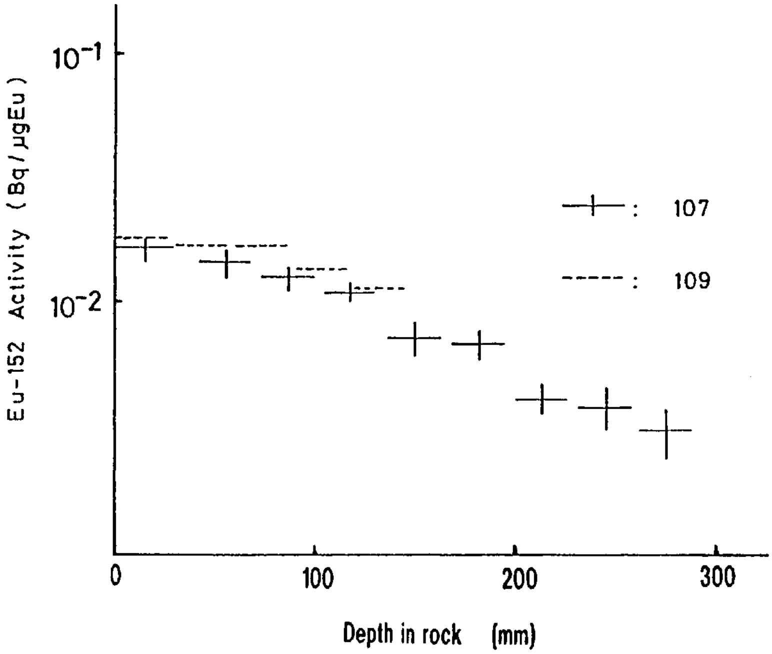


Fig. 3. Figures (A) to (G) show the ^{152}Eu activities at various depths in the rocks. The measured values were normalized per microgram of natural Eu and converted to activity at the time of the bombing. Data from samples with their surfaces facing the epicenter, taken from the Shimono-kawa embankment, are plotted in Figures (A) and (B). Data from samples taken from the opposite embankment are plotted in Figures (C), (D) and (E). Data from samples with their surfaces facing the epicenter, taken from the embankment of the Urakami-gawa are plotted in Figure (F). Data from samples taken from the opposite embankment are plotted in Figure (G).

in Figure 3(F). Samples from the opposite embankment of the same river are plotted in Figure 3(G). These samples had monotonic reductions in ^{152}Eu activities with increasing depth. There were no large discrepancies among the depth profiles of samples taken near the hypocenter (the River Shimono-kawa), as shown in Figures 3(A)–(E). That is, the neutron spectra were almost the same whether the samples faced or opposed the epicenter. There was wide variation in the relationship 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 4. The highest value of surface plate ^{152}Eu activity, 8.0×10^{-2} Bq/μgEu, was obtained from sample #129, which was taken 504 m from the apicenter. Surface ^{152}Eu activities at that location varied from sample to sample. That is, since the quantity of thermal neutrons depends on 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 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.$^{17)}$, ^{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

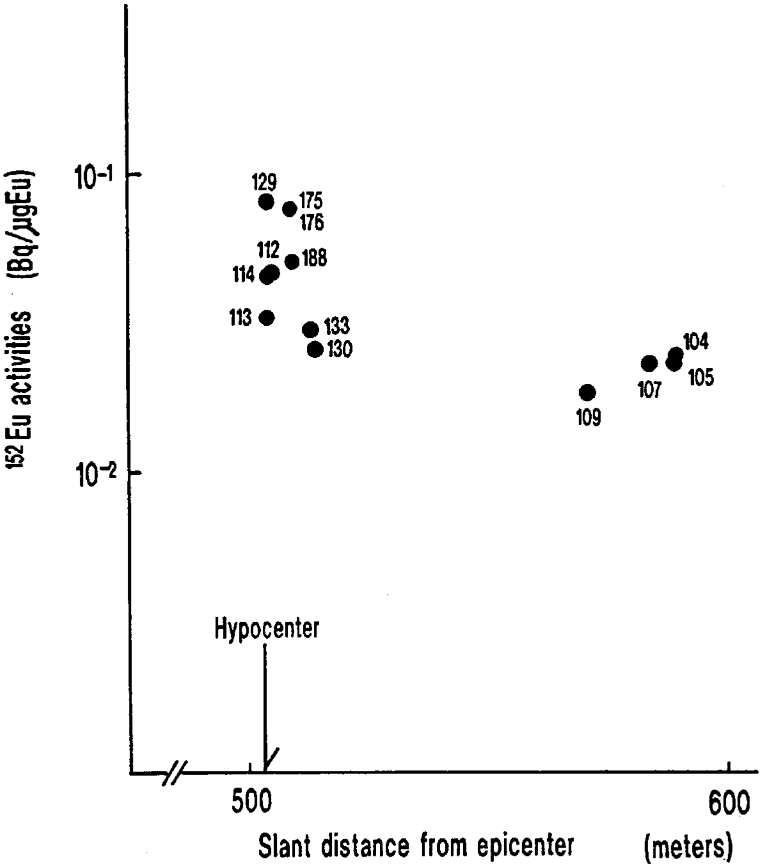


Fig. 4. ^{152}Eu activities of the surface plates plotted against the slant distance from the epicenter.

(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 activity at 280–320 mm depth in rock obtained within a radius of 100 m from hypocenter was $(7.82 \pm 0.93) \times 10^{-3}$ Bq/μgEu. With the value of the cross section of ^{151}Eu(n, γ)^{152}Eu reaction for neutron energy set at 0.025 eV, 5900 barn, thermal neutron fluence at 280–320 mm was calculated as $(4.21 \pm 0.50) \times 10^{11}$ n/cm^2. This indicates neutron fluence above intermediate energy incident at the surface.

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

2. Reactor experiments

Andesite rock similar to that exposed to the A-bomb was used in neutron transportation experiments. The rock was shaped as a cylinder (190 mm in diameter and 300 mm in length) and cut into various thickness of 10 mm to 50 mm. It was inserted into a hole (200 mm in diameter and 300 mm in length) of radiation shielding concrete between a core of nuclear fuel and a thermal column of the fast neutron reactor Yayoi. The thermal column (2 m × 2 m) was filled with air. The energy of the neutrons at the exit of the reactor was fission spectrum. The fission neutrons penetrated the rock. Eu foil sheets 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 with 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

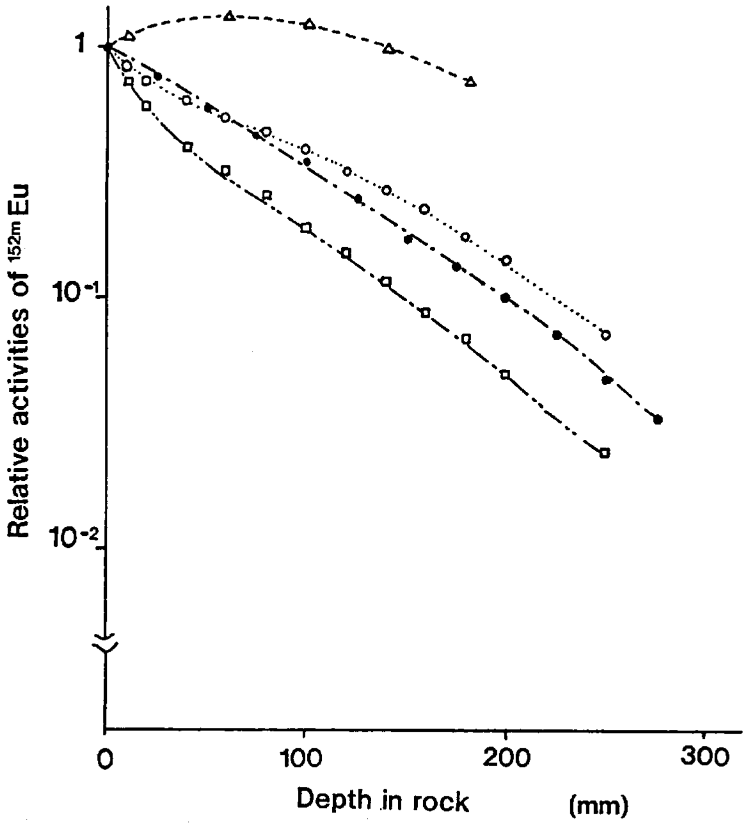


Fig. 5. Depth profiles of ^{152m}Eu activities in rock irradiated by four energy spectra in the reactors. The open symbols indicate data from the experiments at the fast neutron reactor Yayoi. The open triangles indicate the depth profile of the fission neutrons directly incident on the rock. The open 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.

by thermal neutrons moderated with water.

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 9.38 h 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.

The depth profiles of ^{152m}Eu activities in rock irradiated by four energy spectra of neutrons were obtained from the reactor experiment. The results are shown in Figure 5. 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 was 60 mm below the surface. The activity at 180 mm was reduced to 77% of the value at the surface. 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.

The average decrease was $10^{-0.34}$ per 100 mm for the A-bomb exposed samples facing the epicenter and near the hypocenter. Although the curves obtained from the exposed samples were not as steep as that obtained by using the 10 mm polyethylene plate in the reactor experiments, they were similar. The depth profiles in the rocks reflect the energy spectrum at the surface. The curve had a peak of the ^{152}Eu activity in the rock when the fast neutron component is large. And the higher energy neutrons produced the peak at a deeper position. The activities were monotonously reduced when the fast neutron component was relatively small. 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.

GAMMA DOSIMETRY

1. Materials and methods

The ESR signal intensity of the CO_3^{3-} radicals in X-ray irradiated human-tooth enamel changes with the direction of static magnetic field, because of the crystalline structure of tooth enamel[18-20]. A reproducible and simple method is required for measurements in an epidemiological study. Samples of tooth enamel were used after crushing into grains of about 1 mm in diameter to eliminate the angular dependence in the ESR measurements. Enamel grains of 100–400 mg in weight were obtained from one tooth. The samples were measured in test tubes of quartz glass of 5 mm in diameter at room temperature.

The ESR measurements were carried out by X-band spectrometer (JOEL: JES-FEXIG) under the conditions; at around 336 mT of magnetic field, at field modulation of 100 kHz and 0.32 mT. The first derivative of absorption curve was recorded.

Since the quantity of the CO_3^{3-} radicals produced by irradiation of each tooth is proportional to the content of the CO_3^{2-} ions which were trapping centers of the ionizing electrons, the increment of the ESR signal intensity per unit radiation exposure depends on each tooth. To normalize the increment of the signal intensity per unit exposure, the ESR measurements for additional irradiation of standard dose of $2.58 \times 10^{-2} - 5.16 \times 10^{-2}$ C/kg of ^{60}Co-gamma rays

Table 2. The tissue absorbed doses estimated by the ESR dosimetry for A-bomb survivors

ID No.	Place method.	Slant distance	Free-in-air tissue kerma calculation*	Irradiated configuration	Tissue absorbed dose estimated by ESR	
					Mean	± SE(%)
A- 1	N	855 (m)	33.5 (Gy)	in concrete building	2.2**(Gy) 2.0**	± 13(%) ± 14
A- 2	N	158–1074	11.5–0.84	in open?	8.5**	± 9
A- 3	N	1301	3.70	in light frame building	0.9	± 55
A- 4	N	1394	1.89	in wooden building	3.4**	± 7
A- 5	N	1459	1.70	in light frame building	1.0	± 50
A- 6	N	1488	1.27	in open, near brick wall	1.3	± 21
A- 7	N	1582	0.84	in light frame building	1.2	± 27
A- 8	N	1870	0.27	in wooden building	0.9 1.1	± 10 ± 8
A- 9	H	985–1151	0.89–0.17	in concrete building	2.7*	± 21
A-10	H	1090	4.69	in wooden building	5.4	± 7

N: Nagasaki cases. H: Hiroshima cases. *: Free-in-air tissue kerma calculated by Kerr et al.[23]. It is necessary to compare the dose to Tentative Dose evaluated in 1965 (T65D), see the reference[1]. **: The estimates were carried out in cooperation with M. Ikeya of Yamaguchi University. A-2: The man was died at 7 days after the A-bomb explosion.

were successively repeated 4 or 5 times. Assuming a linearity between the ESR signal intensity I and the exposure D, a simple regression equation of $I = a + bD$ was obtained by a least squares method. Values a and b are the intensity for zero of the additional dose and the intensity increase per unit dose, respectively. The tissue absorbed dose was obtained using the correction factor of energy dependency on the ESR signal intensity and f is the conversion factor of exposure (C/kg) to tissue absorbed dose (Gy) at the energy of the irradiated field. The conversion factors of exposure (C/kg) to the tissue absorbed dose (Gy) f were taken from the values of 36.8$^{21)}$ for A-bomb survivors.

The measurements were carried out at higher microwave power (30 mW) at the end of a week after crushing the samples.

The fading properties of the CO_3^{3-} radicals produced by the additional irradiation were examined. A test sample was measured after the irradiation of 1.29×10^{-1} C/kg of ^{60}Co-gamma rays at room temperature. The fast decay component of the signal disappeared within six hours, and the intensity approached a constant. Therefore, the ESR measurements were made more than six hours after the additional irradiation.

The ESR signal of tooth samples of the exposed persons can be measured for many years after the exposure, since the life time of the CO_3^{3-} radicals was reported as 10^6–10^7 years$^{22)}$.

2. Dose estimate for A-bomb survivors

Table 2 shows the tissue absorbed doses estimated by the ESR method for A-bomb survivors. In the table, the place of exposure, the slant distance from the center of the A-bomb explosion (hypocenter), the free-in-air tissue kerma calculated by Kerr et al.$^{23)}$, and irradiated configuration were also shown. Since individual designated A-2 died 7 days after the A-bomb explosion, the exposed distance from the hypocenter could not be exactly obtained. Comparing "the dose estimated by the ESR method" with "the free-in-air tissue kerma calculated by Kerr et al.", both of the estimates for the person exposed in open (non-shielding) condition were consistent with each other. The two estimates for the cases exposed in the concrete buildings cannot be compared directly, because of the heavy concrete shielding. In the case of A-1, a rough estimate of the concrete thickness can be obtained as 30 cm from the ratio between the free-in-air tissue kerma and the dose estimated by the ESR method. In the cases of exposure in wooden buildings (Japanese houses) and the light frame buildings (weapon factory), except the case of A-4 the two estimates are consistent with each other considering the shielding effects.

The ESR method is useful and suitable for an estimate of an accidental exposure in the absence of dosimeters. The ESR spectroscopy for tooth enamel presented good dosimeter characteristics, since a linear fit between the cumulative exposure and the signal intensity of the CO_3^{3-} radicals was obtained.

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