Journal of Hiroshima Prefectural College of Health Sciences Humanity and Science 4 (1) 9－20 2004

Neutrons due to Natural Radioactivities in Granite Rock Samples in Hiroshima City and the Surrounding Areas

Kazuo KATO

Department of Radiological Sciences, Hiroshima Prefectural College of Health Sciences

Abstract

In order to prepare data for estimating the precise amounts of the naturally produced long life nuclide of ^{36}Cl (3 x 10^5 y of half life) in granites distantly exposed to the Hiroshima atomic bomb, contents of potassium (K), uranium (U) and thorium (Th) were determined by measurements of gamma rays from natural radioactivities. The contents of K, Th and U in granites not exposed to the atomic bomb were also determined for examining how the contents varied amongst different lithospheres around Hiroshima. This yielded interesting facts, such as that the K contents in Iyo stones from Iyo-Ohshima Island, Ehime Prefecture were much lower than those in Giin stones from Kurahashi Island near Hiroshima City. Seventeen samples were taken out at various depths in a large granite body near the top of the mountain for the Giin quarry. The results showed that the U and Th contents were very high in a small part of the body. The maximum values were 6.5 ppm U and 55 ppm Th. This suggests that the U and Th contents vary widely according to location in the Giin quarry.

Key words : uranium, thorium, potassium, Hiroshima atomic bomb, neutrons

Introduction

The materials exposed to the Hiroshima atomic bomb retain several kinds of radioactivity induced by the atomic bomb neutrons. In order to experimentally evaluate the neutron fluences, ^{36}Cl and ^{152}Eu residual radio-activities have been measured[1,2]. Although the ^{36}Cl activities with long half life (3 x 10^5 y) are weak in the samples exposed to the atomic bomb, it was possible to detect the ^{36}Cl using accelerator mass spectrometry (AMS) at the Munich tandem laboratory[1]. The results showed that the $^{36}Cl/Cl$ ratios in the granite samples near the hypocenter were clearly lower than those estimated by the $^{152}Eu/Eu$ ratios in the same sample[1,2]. The detection limit of the ^{36}Cl AMS at the Munich tandem laboratory was sufficiently low for measuring $^{36}Cl/Cl$ in the granite samples with ground distance beyond 1 km[3]. As reported by Rühm *et al.*[4], the measured $^{36}Cl/Cl$ ratios in granite samples decreased smoothly as the distance increased, where the background $^{36}Cl/Cl$ ratios were estimated from the measurements of $^{36}Cl/Cl$ ratios in the quarry samples. The $^{152}Eu/Eu$ and $^{60}Co/Co$ measured in distant samples showed that the thermal neutron fluence was much larger than that estimated by the $^{36}Cl/Cl$ in granite in the slant range greater than 1.2 km[4,5,6]. The $^{36}Cl/Cl$ in the concrete sample distantly exposed to the atomic bomb was also much larger than the $^{36}Cl/Cl$ in granite[4,7]. If the atomic bomb thermal neutron fluences were much higher than those estimated by the $^{36}Cl/Cl$ ratio by Rühm *et al.*[4], the neutron doses to the atomic bomb survivors were probably high, as shown by Katayama *et al.*[8] Shizuma *et al.*[9] revealed that the high $^{60}Co/Co$ ratios measured in the distant samples were not due to background neutrons. The half-lives of ^{60}Co and ^{152}Eu are so short that naturally produced ^{60}Co and ^{152}Eu are usually negligible. Since the half-life of ^{36}Cl is very long (3x10^5 y), the amount of ^{36}Cl naturally produced in distant samples may be comparable to those produced by the atomic bomb neutrons. Many investigations have been performed on cosmogenic radionuclides including ^{36}Cl[10,11,12,13]. Natural neutrons also produced ^{36}Cl, as well as the atomic bomb neutrons. The natural neutrons are produced by (α, n) reactions and by spontaneous fission of ^{238}U[14]. The α particles are produced from α-decay of natural radioisotopes mainly in the thorium (Th), uranium (U) and actinium series[14]. For estimating the production rates of cosmogenic ^{36}Cl in granite rock, it is necessary to determine the elemental composition, depth of the quarried location in the lithosphere, and erosion rate at the quarry. However, the ^{36}Cl production in a sufficiently deep position is mainly caused by natural radioactivities. Identification of the quarry for the granite samples exposed to the atomic bomb is necessary for estimating the depths where the granite samples were quarried. Granite rocks are formed at various depths under the ground. It is known that potassium (K) content relates to the depth[15]. Yonehara *et al.*[16] showed that the ^{222}Rn air-concentrations of several houses in Hiroshima were so high that frequent exchange of air is desirable. This suggests that a set of K and U contents may be a good index to indicate the location of a quarry near Hiroshima City. For the quarries around Hiroshima, the depths of stones taken before the 2nd World War can be estimated through discussions with veteran specialists on quarries.

In this study, a relatively large amount of granite rock (25 g) is used for determining the U and Th contents. Since the distribution of minerals is inhomogeneous in ordinary granites, such a large volume of a powdered sample is necessary for estimating the mean contents of these elements over a large volume of rock material. Owing to the simplicity of the present method, the numerous samples could be examined to reveal how the U, Th and K contents varied amongst lithospheres. The main purpose of this paper is to report the results.

Materials and Methods

Calibration of Ge-detector

The gamma rays are measured with a high purity co-axial type Ge-detector (Oxford CPVDS30-0190, volume of 160 cc) shielded with lead blocks thicker than 15 cm. Gamma rays were measured by mounting 25 g of powdered rock sample (4.5 cm in diameter and 7 mm in thickness) in a cylindrical plastic capsule on the window of the Ge-detector. The detection efficiency of the Ge-detector is determined by using two rock reference samples (GJ1a and JR1) from the Geological Survey of Japan (GSJ). The powdered rock reference samples were pressed with 2700 [atm] by a cold isostatic pressing machine (Mitsubishi, MCT 100). The GSJ rock reference samples were previously used for developing the methods to quantitatively isolate europium and chloride elements from silicate rock samples[17,18]. The details of the reference samples are described in Table 1. Numerous investigations have been conducted for determining the U and Th contents in the GSJ reference rock samples[19]. This is the reason why the samples are available as calibration sources for the Ge detector. In order to ensure determined detection efficiency, a ^{152}Eu weak standard source (Japan Isotope

Cooperation, EU-402) with 3% of uncertainty of activity was also used. The ^{152}Eu standard source is point-like and available for examining how the detection efficiency varies with the distance from the detector window. The absorptions of gamma rays in the powdered rock sample were also examined, locating the ^{152}Eu standard source on the GSJ rock reference sample, JR-1. In the estimation, the detection efficiency, ε, at height, h, from the bottom of the sample case was assumed to be expressed by the equation:

$$\varepsilon = \varepsilon_0 \frac{e^{-\mu h}}{(h+d_0)^2}$$

, where ε_0, μ and d_0 are experimentally determined. Thus, the gamma-ray detection efficiency based on the ^{152}Eu standard source measurements was compared with the detection efficiency determined by the GSJ rock reference samples.

Table 1 Potassium, thorium and uranium in the reference rock samples of the Geological Survey of Japan[19]. For Th and U, the value in parenthesis is the number of compiled values, and the standard deviation is indicated as the uncertainty[19]. The uncertainty of K content is ignored because it could be precisely determined[20].

Sample	Species	K_2O (%)	Th (ppm)	U (ppm)
JG1a	Granodiorite	3.96	12.8 ±1.5 (27)	4.69 ±1.09 (28)
JR1	Rhyolite	4.41	26.7 ±2.6 (36)	8.88 ±1.32 (40)

Granite samples

The remaining granite samples exposed to the Hiroshima atomic bomb are used as gravestones, handrails of bridges, walls of buildings, pavements, etc. In this study, granite samples from the quarries shown in Figure 1 in Hiroshima City and the surrounding areas were used in order to find out where the granite stones were produced. It is well known that many Aji and Iyo stones were transported into Hiroshima City for use as gravestones. Iyo-stone is hornblende biotite granodiorite, and Aji-stone is a fine grain biotite granite. Aji-stone is the granite from Aji-cho in the eastern Sanuki district, Kagawa Prefecture, Ryoke belt, Southwest Japan, 155 km from Hiroshima. Iyo-stone used as gravestones in Hiroshima usually indicates granite from Iyo-Ohshima Island in Ehime Prefecture. Kitagi quarry is in Kasaoka, Okayama Prefecture. The distance from Hiroshima is approximately 100 km, and the north latitude is nearly equal to that of Hiroshima. It is well known that Kitagi stones have been used for the materials of buildings in Osaka and Tokyo.

Granites in Kurahashi Island are typical Hiroshima granites. Giin and Odachi quarries are at Kurahashi-cho, Hiroshima Prefecture, which is 33 km from Hiroshima city. The Giin quarry dates from before the 2nd World War, and it produced a lot of stones for buildings, bridges and pavements in Hiroshima City. Stones at Kurahashi Island are easily damaged by weathering effects and so are inappropriate for gravestones. Hiroshima granites were produced at three stages; older Hiroshima (100-110 [10^6

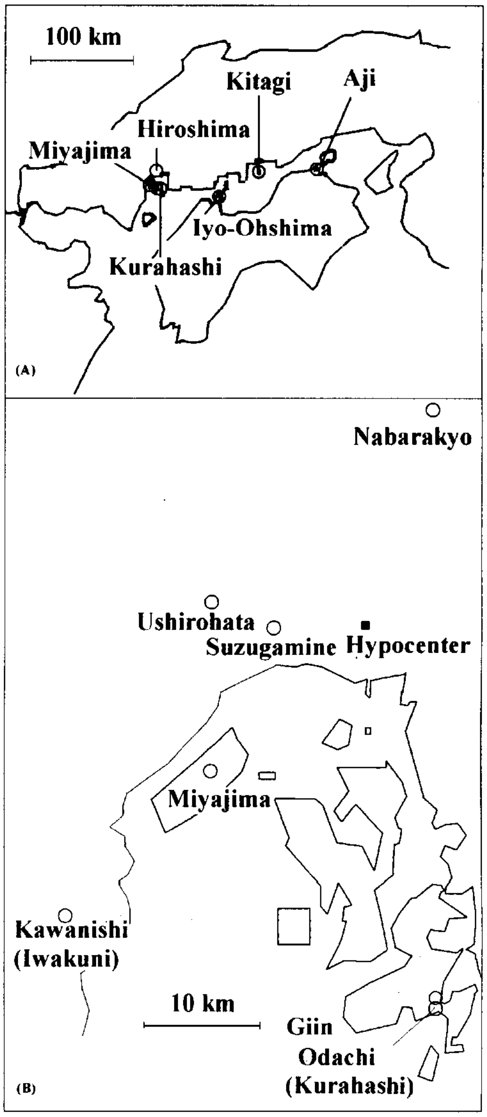

Figure 1 Locations of quarries.
(A) Quarries at Kurahashi Island, Kitagi Island, Iyo-Ohshima Island, and Aji-cho.
(B) Granite samples in the vicinity of Hiroshima City.

y]), middle Hiroshima (85-100 [10^6 y]) and younger Hiroshima (70-85 [10^6 y])[21]. The older Hiroshima granite is middle-sized grain hornblende biotite granite which frequently includes large grain potassium feldspar, like the Giin stone[21]. Giin stones are rich in large pink potassium-feldspar, while Odachi stones often include white potassium-feldspars. Since the Odachi quarry is close to the Giin one, the lithosphere at the Giin quarry also includes Odachi type granite. Both types of stones seem to have been produced at Kurahashi Island before the 2nd World War.

Table 2 Rock samples exposed to the atomic bomb.

No.	Name #	Slant Distance (m)+	Content
1	Aji-Niitani	590	Aji stone in grave stone for Niitani family
2	Iyo-Niitani	590	Iyo stone in grave stone for Niitani family
3	Shirakami-R	763	Stone at Shirakami shrine including red minerals
4	Shirakami-Y	763	Stone at Shirakami shrine including yellow minerals
5	Shinkoji-4	1012	Grave stone at Sinkoji
6	Ganjyoji-1	1159	Grave stone No. 1 at Ganjyoji
7	Ganjyoji-2	1159	Grave stone No.2 at Ganjyoji
8	Tokueiji	1295	Grave stone at Tokueiji
9	Hosenji-1	1303	Grave stone No. 1 at Hosenji
10	Hosenji-2	1303	Grave stone No. 2 at Hosenji
11	Hosenji-3	1303	Grave stone No. 3 at Hosenji
12	E-buildingD1	1493	Ditch cover, Faculty of Science, Hirohsima Univ.
13	E-buildingD2	1493	Ditch cover, Faculty of Science, Hirohsima Univ.
14	E-buildingD3	1493	Ditch cover, Faculty of Science, Hirohsima Univ.
15	E-buildingD4	1494	Ditch cover, Faculty of Science, Hirohsima Univ.

Name is determined only to identify each sample.
+ Slant distance is measured from the epicenter of the Hiroshima A-bomb, which was used for the Dosimetry System 1986 at the Radiation Effects Research Foundation[22].

The rock samples shown in Table 2 were exposed to the atomic bomb and are a part of the samples for ^{36}Cl/Cl AMS, excluding the stones at Shirakami-shrine[4]. The samples were gravestones, except for two from Shirakami-shrine and four samples from ditch covers of the building formerly used for the Faculty of Science, Hiroshima University. Many measurements were performed for the sample pieces from the gravestone of the Niitani family (No. 1 and No. 2), as previously reported[1,2]. The Niitani gravestones were composed of typical Aji stone and Iyo stone. The ditch covers from the building used for the Faculty of Science of Hiroshima University are probably Kurahashi stones. The granites at Shirakami-shrine shown in Table 2 are rare large stones, which belong to a lithosphere in the city. The location is now at the center of Hiroshima City, but faced the Inland Sea in the past. Therefore, the surface was seriously damaged. The locations of lithospheres for No. 1,2,3 and 4 samples in Table 2 are well known, and hence are useful for examining how the contents varied amongst different lithospheres.

Results

The nine full energy peaks were analyzed for determining U, Th and K contents. The gamma-ray energies are shown in Figure 2. Four peaks were emitted from the nuclides in the uranium series, and the other four peaks were from the nuclides in the thorium series. The remainder is the peak of the gamma ray from ^{40}K. The detection efficiencies were obtained from the spectra for JG1a (Granodiorite) and JR1 (Rhyolite) reference samples, and the recommended values of U, Th and K contents are shown in Table 1[19]. The results for JR1 were coincident with those for JG1a. The detection efficiencies from the measurements of the ^{152}Eu standard source are compared with those from the GSJ geological reference samples, as shown in Figure 3. The detection efficiencies for the gamma rays from the GSJ geological reference samples

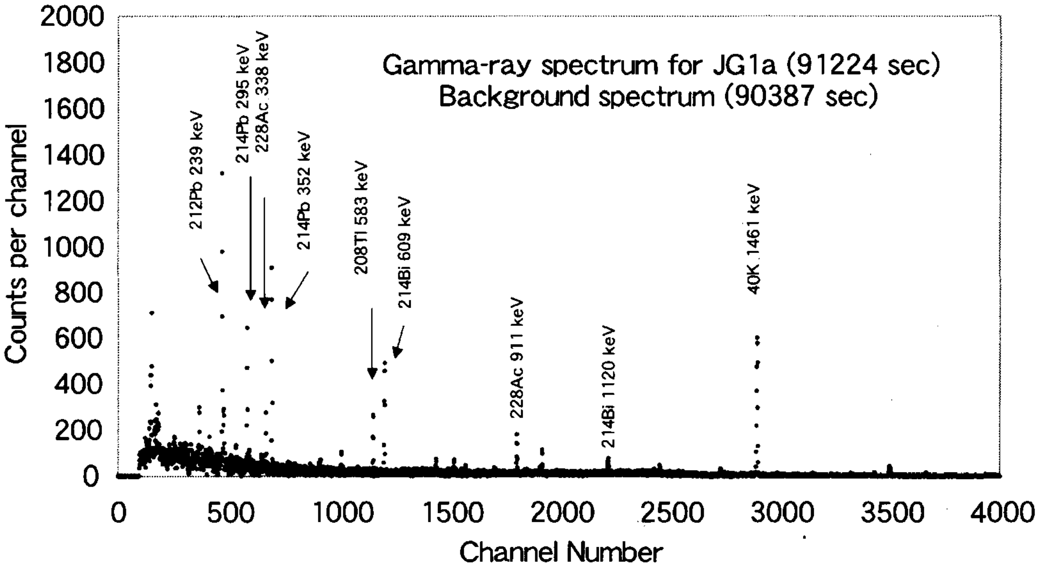

Figure 2 Gamma-ray spectrum for 25 g of GSJ reference sample (JG1a)
Background spectrum was measured without any sample and was subtracted from the spectrum for JG1a after adjusting the difference in elapsed times for measurements.

were determined assuming that the radioactive equilibrium states in the uranium and thorium series in a closed powdered rock sample. The comparison shows that the present detection efficiencies are almost completely consistent with those estimated from the measurements of a ^{152}Eu standard point-like source. The compositions of major elements in JR1, JG1a, Hiroshima granites and Aji-stone are similar to each other[19,20,21,23]. This is the reason why the detection efficiencies determined for JG1a and JR1 samples were used for the measurements of the other granite samples.

Figure 4 shows the count rates of background gamma rays. The lead shield is effective in suppressing the background. As shown in this figure, the background count rate is less than 10% of the count rate from the JR1 reference rock sample. However, background subtraction becomes more important as the K, U or Th content becomes lower. Frequent measurements of the background were therefore necessary.

Using the measured spectra, K, Th and U contents in the granites were estimated. The results are shown in Table 3. The relationships of K content to Th content and to U content are shown in Figure 5. The mean contents of these elements in 22 Giin stones were 4.8 (0.5) % K_2O, 4.8 (1.0) ppm U, and 24 (10) ppm Th, where the values in parentheses are standard deviations. The mean contents in 8 stones from Iyo-Ohshima Island in Ehime Prefecture were 3.3 (0.3) % K_2O, 2.2 (0.5) ppm U, and 9.6 (1.3) ppm Th. The K, U and Th contents in two Aji stones from Kagawa Prefecture were similar to those in the Iyo stones, as shown in Table 3. High contents of U and Th were observed in a large granite body near the top of the mountain for the Giin quarry. The U or Th contents relate to the depth, and the maximum values were 6.5 ppm of U and 55 ppm of Th at 2.08 m from the top surface of the granite body.

Kitagi Island is close to both Iyo-Ohshima Island and Aji. The contents of the three elements are also close to those in Iyo and Aji stones, as shown in Figure 5. Although Giin and Odachi granites are generally rich in U and Th, the graphical region of the data overlaps those for the other Hiroshima granites. For Hiroshima granites, it is difficult to find the quarry where each sample was produced.

Among the granite samples used in this study, the 11 granite samples (No. 5 - No. 15 in Table 2) were distantly exposed to the atomic bomb. The contents of K_2O, U and Th are determined as shown in Table 4. The results and the data shown in Figure 5 suggest that two of the gravestones (Shinkoji-4 and Hosenji-3) are either Iyo-stone or Aji-stone, and the others are Hiroshima granites (Giin stone, Odachi stone or the other Hiroshima granite). The results from the three samples from Ganjyoji-1 suggest that the granites used as gravestones are homogenous.

Discussion

Komura *et al.*[24] showed that the potassium, uranium and thorium contents in the GSJ reference rock samples could be determined by gamma-ray measurements with a Ge

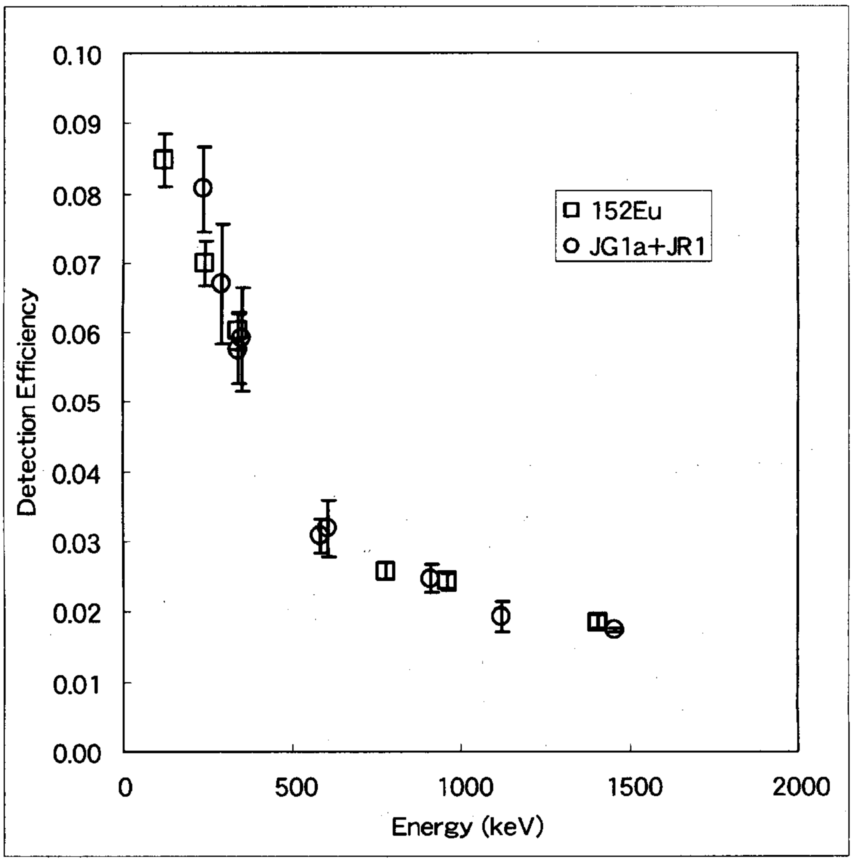

Figure 3 The detection efficiencies examined by the GSJ reference rock samples (open circles: JG1a + JR1), and estimated by the ^{152}Eu point-like standard source (open squares: 152Eu).

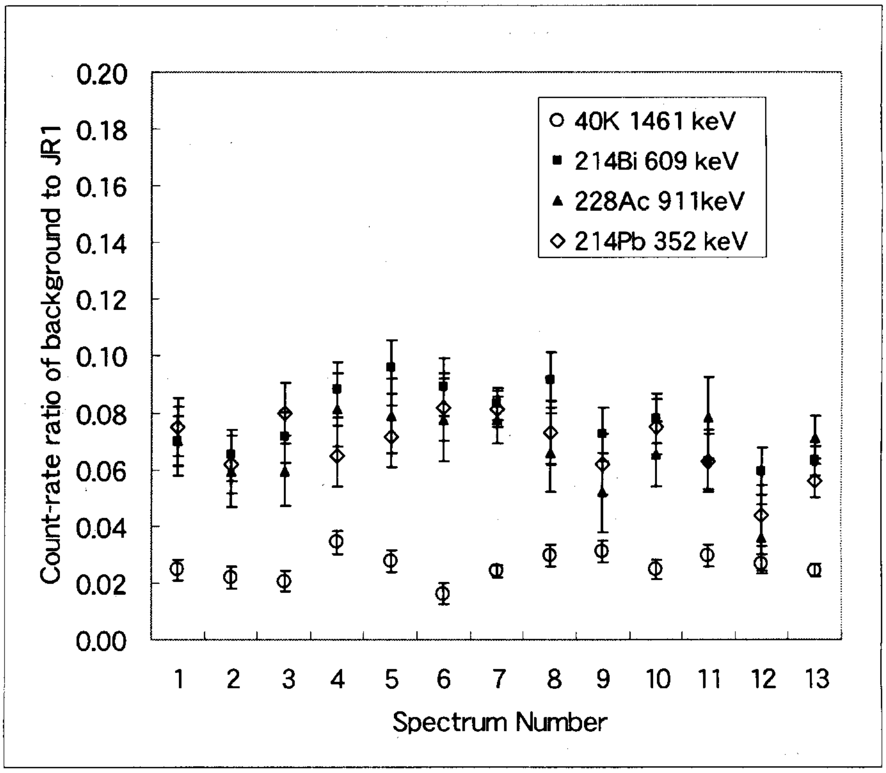

Figure 4 Ratios of gamma-ray count-rate in background to that of the JR1 reference sample
Count rates of full energy peaks were obtained from 13 background spectra and a spectrum for the JR1 reference rock sample. The vertical axis indicates the ratio of background count rate to that of the JR1 spectrum.

Table 3 Potassium, uranium and thorium contents in granite of which quarry is known.

Name #	Location +	Depth (m)♪	K_2O (%)	U (ppm)	Th (ppm)
Aji-1	Aji	-	3.29 ± 0.08	0.85 ± 0.13	5.87 ± 0.47
Aji-Niitani	Aji	-	3.27 ± 0.07	1.00 ± 0.14	5.68 ± 0.45
Kitagi	Kitagi	-	3.69 ± 0.06	3.45 ± 0.39	11.59 ± 0.77
Iyo-3	Iyo	2 ± 0.5	3.74 ± 0.07	1.19 ± 0.16	7.09 ± 0.53
yo-1a	Iyo	5 ± 0.5	3.20 ± 0.07	2.47 ± 0.29	12.03 ± 0.81
Iyo-1b	Iyo	5 ± 0.5	2.80 ± 0.07	2.18 ± 0.26	10.02 ± 0.69
Iyo-2	Iyo	5 ± 0.5	3.35 ± 0.08	2.60 ± 0.30	8.83 ± 0.64
Iyo-4	Iyo	30 ± 5	3.36 ± 0.08	2.37 ± 0.28	10.02 ± 0.71
Iyo1999	Iyo	-	3.26 ± 0.07	2.31 ± 0.27	9.80 ± 0.68
Iyo-5	Iyo	-	3.03 ± 0.07	1.82 ± 0.22	8.84 ± 0.64
Iyo-Niitani	Iyo	-	3.29 ± 0.07	2.77 ± 0.32	10.15 ± 0.70
GiinB1-1	Giin	0.05 ± 0.05	4.41 ± 0.06	5.70 ± 0.63	19.12 ± 1.22
GiinB1-2	Giin	0.05 ± 0.05	5.26 ± 0.07	3.25 ± 0.37	14.01 ± 0.92
GiinB1-3	Giin	0.05 ± 0.05	5.01 ± 0.09	4.26 ± 0.48	23.63 ± 1.53
GiinB1-4	Giin	0.05 ± 0.05	4.67 ± 0.07	3.58 ± 0.40	17.31 ± 1.12
GiinB2-1	Giin	1.20 ± 0.05	4.95 ± 0.09	3.20 ± 0.37	11.89 ± 0.81
GiinB2-2	Giin	1.20 ± 0.05	5.29 ± 0.08	3.96 ± 0.45	15.08 ± 0.99
GiinB2-3	Giin	1.25 ± 0.05	5.27 ± 0.09	4.15 ± 0.47	12.98 ± 0.87
GiinB3-1	Giin	2.01 ± 0.05	4.52 ± 0.09	5.77 ± 0.65	33.13 ± 2.13
GiinB3-2	Giin	2.05 ± 0.05	4.77 ± 0.09	6.04 ± 0.68	31.85 ± 2.05
GiinB3-3	Giin	2.05 ± 0.05	4.33 ± 0.08	5.83 ± 0.65	32.36 ± 2.08
GiinB3-4	Giin	2.08 ± 0.05	4.29 ± 0.09	6.49 ± 0.73	55.10 ± 3.51
GiinB3-5	Giin	2.09 ± 0.05	4.23 ± 0.06	5.77 ± 0.64	41.88 ± 2.66
GiinB3-6	Giin	2.20 ± 0.10	3.78 ± 0.08	5.75 ± 0.65	35.38 ± 2.27
GiinB4-1	Giin	2.85 ± 0.05	5.39 ± 0.07	5.14 ± 0.57	19.61 ± 1.26
GiinB4-2	Giin	3.11 ± 0.05	5.35 ± 0.09	3.85 ± 0.44	18.95 ± 1.24
GiinB4-3	Giin	3.19 ± 0.05	5.93 ± 0.08	4.00 ± 0.45	14.36 ± 0.94
GiinB4-4	Giin	3.25 ± 0.05	3.83 ± 0.05	4.38 ± 0.49	20.84 ± 1.33
GiinC-1	Giin	30 ± 4	4.88 ± 0.06	3.50 ± 0.39	19.82 ± 1.27
GiinC-2	Giin	30 ± 4	4.65 ± 0.09	5.44 ± 0.61	24.52 ± 1.59
GiinC-3	Giin	30 ± 4	4.71 ± 0.10	4.29 ± 0.49	22.34 ± 1.46
GiinC-4	Giin	30 ± 4	5.36 ± 0.10	5.49 ± 0.62	30.41 ± 1.96
Giin-SP	Giin	-	4.70 ± 0.09	5.14 ± 0.58	23.20 ± 1.51
Odachi	Odachi	-	4.40 ± 0.06	6.71 ± 0.75	18.40 ± 1.19
Miyajima	Miyajima	-	4.49 ± 0.09	1.46 ± 0.19	11.97 ± 0.82
Nabarakyo	Nabara	-	5.00 ± 0.09	3.72 ± 0.42	15.34 ± 1.03
Shirakami-R	Ohtemachi	0.05 ± 0.05	4.93 ± 0.09	3.36 ± 0.39	18.42 ± 1.22
Shirakami-Y	Ohtemachi	0.05 ± 0.05	4.36 ± 0.06	3.57 ± 0.40	14.16 ± 0.93
SUZUGC	Itsukaichi	-	4.49 ± 0.08	2.52 ± 0.29	14.71 ± 0.98
SUZUGCSD	Itsukaichi	-	4.85 ± 0.09	2.66 ± 0.31	17.56 ± 1.15
Ushirohata	Ushirohata	-	4.49 ± 0.08	2.77 ± 0.32	14.15 ± 0.94
Kawanishi	Kawanishi	-	4.35 ± 0.09	1.99 ± 0.24	17.17 ± 1.13

Name is determined only for identification of each sample. The results for Aji-Niitani, Iyo-Niitani, Shirakami-R and Shirakami-Y are shown in this table, because the locations of lithospheres are well known.

+ Location: quarry name or place name.

♪ "-" means that the depth has not been clearly determined.

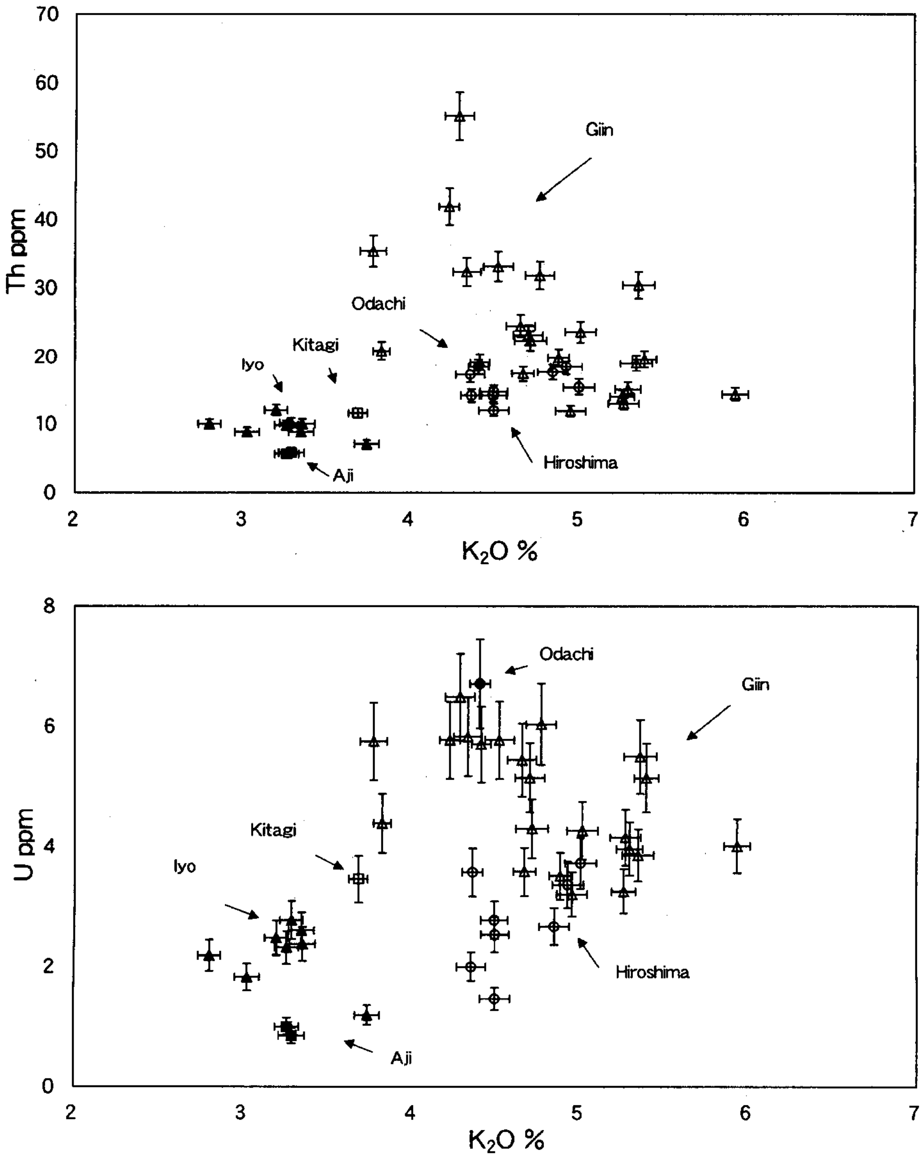

Figure 5 Potassium, thorium and uranium contents of granites around Hiroshima. Hiroshima indicates Hiroshima granite, except granites at Kurahashi Island (Giin or Odachi).

detector. The detector was calibrated using powdered rock samples mixed with the known amounts of K, U and Th. Owing to the numerous previous investigations up to the present time, the elemental compositions of the GSJ rock samples have been determined within small uncertainties[19]. Therefore, the GSJ reference samples are available for calibrating Ge detectors, as used in this study.

As shown in Table 3, Aji-stone used as the Niitani-gravestone (Aji-Niitani) includes 3.27 ± 0.07 % K_2O, 5.68 ± 0.45 ppm Th and 1.00 ± 0.14 ppm U. The chemical compositions of the Aji stone (Aji-Niitani) were previously examined by the XRAL Company in Canada, and the results for three sample pieces were 3.02 % K_2O, 4.4 ppm Th and 0.95 ppm U[23]. Although the Th content is slightly lower than the present value, few differences are seen concerning the U and K_2O contents. The Aji-stone used for Niitani gravestone was relatively homogeneous, and so, for example, europium and chlorine contents were approximately constant[17,18]. Eight samples were taken from the Iyo-Ohshima quarries. The Iyo-stones used in this study include the stones taken from the surface layer of the mountain, whose quality is low and which include pink minerals. Nevertheless, the K, U and Th contents are approximately constant in the granite samples from the Iyo-quarries, as shown in Table 3 and Figure 5. As noted in Figure 5, the position of a Kitagi sample in the K_2O vs. Th graph is clearly different from those of the Hiroshima granites. The Hiroshima granite is rich in K, U and Th.

Table 4 Potassium, uranium and thorium in granite samples exposed to the Hiroshima atomic bomb.

Name #	K_2O (%)	U (ppm)	Th (ppm)
Shinkoji-4	3.30 ± 0.07	3.12 ± 0.36	10.1 ± 0.7
Tokueiji	4.91 ± 0.09	5.73 ± 0.64	28.8 ± 1.9
Hosenji-1	4.76 ± 0.07	3.94 ± 0.44	17.8 ± 1.2
Hosenji-2	4.28 ± 0.09	3.41 ± 0.39	13.9 ± 0.9
Hosenji-3	3.82 ± 0.08	2.42 ± 0.29	9.8 ± 0.7
Ganjyoji-1a	4.54 ± 0.09	4.05 ± 0.46	21.3 ± 1.4
Ganjyoji-1b	4.59 ± 0.09	3.84 ± 0.44	20.1 ± 1.3
Ganjyoji-1c	4.53 ± 0.09	3.85 ± 0.44	19.9 ± 1.3
Ganjyoji-2	4.41 ± 0.09	3.99 ± 0.45	16.5 ± 1.1
E-buildingD1	4.52 ± 0.07	6.46 ± 0.72	28.6 ± 1.8
E-buildingD2	4.19 ± 0.09	6.25 ± 0.70	26.8 ± 1.7
E-buildingD3	5.31 ± 0.10	4.87 ± 0.55	26.0 ± 1.7
E-buildingD4	3.90 ± 0.08	4.63 ± 0.52	20.7 ± 1.3

Ganjyoji-1a, -1b and -1c were taken from Ganjyoji-1 shown in Table 2.

Measurements of these elements seem to be useful to distinguish the Hiroshima granites from the other samples. Excluding the granites from Kurahashi Island, the mean contents of eight Hiroshima granite samples from Miyajima to Kawanishi in Table 3 are 4.56±0.24 % K_2O, 2.69±0.71 ppm U, and15.05±1.96 ppm Th. These small standard deviations mean that it is difficult to identify the stones for each location of Hiroshima granite using only the data of K, U and Th contents. Identifications of the quarries for the Hiroshima granites require more precise examinations of the sizes of grains and colors. This will be performed in the near future.

Fujiwara *et al.*[25] estimated the erosion rates in the Japanese Islands based on the data of the sediment delivery rates to reservoirs. The study revealed the relationship between the erosion rate and the dispersion of the altitude of drainage basins, and that for example, the erosion rate in Kibi plateau near the Inland Sea was estimated to be 0.21 mm/y[25]. Using the data obtained by Fujiwara *et al.*[25] and the precise data of the altitudes[26], the erosion rates were estimated to be 0.15 - 0.25 [mm/y] in the present Aji, Iyo and Giin quarries. However, the application of the relationship to the lower lands may be inappropriate, because the surroundings around the mountains are flat land or close to the sea. Using the results obtained by Fujiwara *et al.*[25], the mean erosion rates around the dams are calculated to be 0.48 ± 0.12 [mm/y] in 16 regions covered with volcanic-rocks, 1.51 ± 0.68 [mm/y] in 6 regions covered with granite rocks, and 1.11 ± 0.27 [mm/y] in 32 regions covered with sedimentation rocks. Here, the regions are classified into one of the three groups, corresponding to the kind of rock that covered 70% or the more of the area. The erosion rate in granite regions is apparently much higher than those in volcanic rock regions. No trace of even weak weathering could be seen through careful petrographic inspection of the Aji stone surface of the gravestone for the Niitani family[27]. On the other hand, Giin stone is much softer than Aji stone. The mountains at Kurahashi Island are poor in plants. Kaizuka [28] showed that the erosion rate in Japan is 1 - 10 [mm/y] for rocky mountains poor in plants. Thus, erosion rate higher than 1 [mm/y] is plausible at the Giin quarry for a time interval longer than 3×10^5 [y]. More studies are necessary for estimating the erosion rate at the Giin quarry.

Feige *et al.*[14] determined the neutron production rate in 1 g of granite to be 1.91 [y^{-1} g^{-1}] per 1 ppm U, and 0.65 [y^{-1} g^{-1}] per 1 ppm Th. If the cosmogenic ^{36}Cl is negligible in granite not exposed to the atomic bomb, the $^{36}Cl/Cl$ ratio is approximately proportional to the neutron production rate. The Giin-SP and Odachi in Table 3 are identical to the Giin and Odachi samples in which $^{36}Cl/Cl$ ratios were previously reported by Rühm *et al.*[4] to have been $(1.1\pm0.4) \times 10^{-13}$ and $(1.3\pm 0.3) \times 10^{-13}$. Since the samples were given by stone shops, the depths of the quarries for the samples are not precisely determined. However, the locations are estimated to have been deep, because they were recently produced at the quarries of which the bottoms have already reached 30 m at Giin, and 100 m at Odachi. Assuming that the number of ^{36}Cl atoms produced by cosmic rays is negligible, the increase of $^{36}Cl/Cl$ ratio per 1 $[y^{-1}g^{-1}]$ neutron production rate is estimated to be $(4.5\pm1.7) \times 10^{-15}$ [yg], and $(5.4\pm1.3) \times 10^{-15}$ [yg] from the above mentioned neutron production rates and the $^{36}Cl/Cl$ ratios in Giin and Odachi samples. The mean value was $(5\pm1) \times 10^{-15}$ [yg]. Under the assumption that cosmogenic ^{36}Cl atoms are negligible, the natural $^{36}Cl/Cl$ in the Giin stone samples shown in Table 3 were estimated from Th and U contents by using the increase of $^{36}Cl/Cl$ ratio per 1 $[y^{-1}g^{-1}]$ neutron production rate. The maximum ratio was 2.4×10^{-13} in the sample of GiinB3-4 in Table 3. This ratio corresponds approximately to the $^{36}Cl/Cl$ ratios in the granite samples (No. 8 and No. 9 in Table 2) at 1.3 km slant range, and in the concrete samples at 1.7 km slant range[4,7]. Heisinger and Nolte[11] estimated the natural $^{36}Cl/Cl$ ratio to be in the range of 2×10^{-13} at the surface of Aji-quarry including the contribution by cosmic rays. The minimum $^{36}Cl/Cl$ ratio among those in the Giin stones shown in table 3 was estimated to be 0.7×10^{-13} in GiinB2-1. This was less than 1/3 of the maximum ratio in the same granite body at Giin quarry.

The present results in Table 4 and Figure 5 suggest that some Hiroshima granites were used for gravestones before the 2nd World War. The Hiroshima granite is generally weak against rainy effect, and hence it is not a good material for gravestones. However, various kinds of granites are classified into the Hiroshima granite group, as described above. Before the 2nd World War, there might have been quarries of Hiroshima granite from which stones were available for gravestones. A survey is in progress to examine whether such an old quarry existed near Hiroshima City. The results will be reported in the near future.

Conclusion

The K, U and Th contents in granites exposed to the Hiroshima atomic bomb were determined by measurements of gamma rays from natural radioactivities. The contents of K, Th and U in granites newly sampled from lithospheres in Hiroshima and the surrounding areas were also determined, and the results revealed that the K-U and K-Th relationships in the stones from Iyo and Aji quarries in the Shikoku area were clearly different from those in the Hiroshima granites. Especially the U and Th contents in a Giin stone from Kurahashi Island were remarkably high. The present results suggest that the $^{36}Cl/Cl$ background ratios are high in several local spots in the lithosphere of the Giin quarry near Hiroshima City.

Acknowledgement

The author is grateful to Dr. Thomas Huber and Dr. Eckehart Nolte (Technical University of Munich) and Dr. Werner Rühm (Ludwig Maximilians University of Munich) for their helpful discussions on the data of the $^{36}Cl/Cl$ in the granite samples exposed to the A-bomb. The author wishes to express his deep appreciation to Dr. Gensei Yoshino (Suzugamine Women's College, formerly Hiroshima University) and Dr. Takehiro Hayashi (Hiroshima University) for the cooperative discussions and helpful suggestions. The author wishes to express his special thanks to Mr. Akira Niitani, Dr. Gensei Yoshino, Amano Sekizai Co., Ltd and Ogasawara Sekizai Co., Ltd for contributing the granite samples. The author is grateful to Mr. Hidemi Nakagawa (Kure-Sekizai Goushi Co., Ltd.), Mr. Kouji Nakano (Kurahashi-Sekizai-Kogyo Co., Ltd.), Masanobu Oda (President of Yamanishi-Sekizai Co., Ltd, and Present Chairman of the Board of Directors for the Ohshima Stone-Maker's Cooperative), and Toshio Tanimura (Iwasaki Marble Co., Ltd.) for their contributing the granite samples, helpful suggestions and important information about the status of the quarries around Hiroshima. The author is also grateful to Dr. Kazuo Iwatani, Mr. Kiyoshi Ishiwaki and Miss. Yukari Inhira (Hiroshima Prefectural College) for their design of the lead shield for the Ge detector. The author deeply appreciates the cooperation of the staff of Shirakami Shrine, Ganjyoji, Tokueiji, and Housenbou for the rock samples and the information on the status of the samples at the time of the atomic bomb detonation. The author is also very thankful to Dr. Yasukazu Yoshizawa (formerly Hiroshima University) and Dr. Taizo Muta (President of Hiroshima University) for their cooperation in sampling of the former building for the Faulty of Science of Hiroshima University. The author is grateful to Misses Rina Kodama and Tomoe Anan (Suzugamine Women's College) for their cooperation in sampling the stones. The cooperation of Mr. Peter Howell in preparing the manuscript is deeply appreciated.

References

1) Kato, K., Habara, M., et al.: Accelerator mass spectrometry of ^{36}Cl produced by neutrons from the Hiroshima bomb. Int. J. Radiat. Biol., 58: 661-672, 1990
2) Kato, K., Habara, M., et al.: Gamma-ray measurement of ^{152}Eu produced by neutrons from the Hiroshima atomic bomb and evaluation of neutron fluence. Jap. J. Appl. Phys., 29: 1546-1549, 1990
3) Rühm, W., Kellerer, et al.: The dosimetry system DS86 and the neutron discrepancy in Hiroshima - historical review, present status, and future options. Radiat. Envion. Biophys, 37: 293-310,1998
4) Rühm, W., Huber, T., et al.: Measurement of ^{36}Cl at Munich - a status report. Radiobiological Institute, Ludwig Maximilians University of Munich: Technical Report SBI 212/11.2000; 2000
5) Shizuma, K., Iwatani, K., et al.: Residual ^{152}Eu and ^{60}Co activities induced by neutrons from the Hiroshima atomic bomb. Health Phys., 65: 272-282, 1993
6) Shizuma, K., Aden, K., et al.: Residual ^{60}Co Activity in Steel Samples Exposed to the Hiroshima Atomic-Bomb Neutrons. Health Phys., 75: 278-284, 1998
7) Straume,T., Egbert,S.D., et al.: Neutron discrepancies in the DS86 Hiroshima dosimetry system. Health Phys., 63:421-426,1992
8) Katayama, H., Matsuura, M., et al.: Reassessment of the cancer risk among Hiroshima atomic-bomb survivors using a new Dosimetry system, ABS2000D, compared with ABS93D. J. Radiat. Res., 43: 53-63, 2002
9) Shizuma, K., Endo, S., et al.: Measurement of residual ^{60}Co activity induced by atomic-bomb neutrons in Nagasaki and background contribution by environmental neutrons. J. Radiat. Res., 43: 387-396, 2002
10) Dockhorn, B., Neumaier, S., et al.: Determination of erosion rates with cosmic ray produced ^{36}Cl. Z. Phys. A - Hadrons and Nuclei, 341: 117-119, 1991
11) Heisinger, B. and Nolte, E.: Cosmogenic in situ production of radionuclides: Exposure age and erosion rates. Nuclear Instruments and Methods in Physics Research, B172: 790-795, 2000
12) Heisinger, B., Lal, D., et al.: Production of selected cosmogenic radionuclides by muons. 1. Fast muons. Earth Planet. Sci. Lett., 200: 345-355, 2002
13) Heisinger, B., Lal, D., et al.: Production of selected cosmogenic radionuclides by muons. 2. Capture of negative muons. Earth Planet. Sci. Lett., 200: 357-369, 2002
14) Feige, Y., Oltman, B.G., et al.: Production Rates of Neutrons in soils due to natural radioactivity. J. Geophys. Res., **73**: 3135-3142; 1968
15) Kaizuka, S.: Historical Geomorphology. University of Tokyo Press, 60-63, 1988 (in Japanese)
16) Yonehara, H., Aoyama, A., et al.: Radon concentration in residential housing in Hiroshima and Nagasaki. Health Phys., 68:683-688, 1995
17) Kato, K., Yamamoto, M., et al.: Chloride Isolation for Accelerator Mass Spectrometry of ^{36}Cl produced by atomic bomb neutrons. Anal. Sci., 3: 489-491, 1987
18) Kato, K., Yamamoto, M., et al.: Europium isolation from silicate rock samples exposed to Hiroshima atomic bomb neutrons. Anal. Sci., 3: 493-497, 1987
19) Imai, N., Terashima, S., et al.: 1994 Compilation of analytical data for minor and trace elements in seventeen GSJ geological reference samples, "Igneous Rock Series". Geostandards Newsl., 19: 135-213, 1995
20) Ando, A., Mita N., et al.: 1986 values for fifteen GSJ rock reference samples, "Igneous Rock Series"., Geostandards Newsl., 11: 159-166, 1987
21) Murakami, N.: Compositional variation of meozoic igneous rocks and their constituent minerals of the area between Masuda and Matsuyama, South West Japan, with special reference to the plutonic rocks from Masuda-Yanai area. Bulletin of Faculty of Liberal Arts, Yamaguchi University, 15: 33-74, 1981
22) US-Japan Joint reassessment of atomic bomb radiation dosimetry in Hiroshima and Nagasaki. Final report Volume 1 (edited by W. C. Roesch), Hiroshima, Radiation Effects Research Foundation, 1987
23) Rühm, W.: "Das Neutronenspektrum der Atombombe von Hiroshima und das Dosimetriesystem DS86", Doctor thesis of Technical University of Munich, 1993
24) Komura, K., Tan, K.L., et al.: Uranium, thorium and potassium contents in eighteen geochemical references. Geostandards newsletter, 12: 371-364, 1988
25) Fujiwara, O., Sanga, T., et al.: Regional distribution of erosion rates over the Japanese islands. JNC Technical Review, 5: 85-93, 1999 (in Japanese)
26) Digital Map 50 m Grid, NIPPON-III, Tsukuba , Geographical Survey Institute, Ministry of Land, Infrastructure and Transport, 2000
27) Blamart, D., Rühm, W., et al.: Oxygen stable isotope measurements on a gravestone exposed to the Hiroshima A-bomb explosion and the "Dosimetry System 1986". Chemical Geology (Isotope Geoscience Section), 101: 93-96, 1992
28) Kaizuka S.: Historical geomorphology. Tokyo, University Tokyo Press, 188-188, 1998 (in Japanese)

広島県立保健福祉大学誌　人間と科学　4(1)　9－20　2004

広島市とその周辺の花崗岩中に天然放射能によって生じた中性子

加藤　一生

広島県立保健福祉大学放射線学科

2003年９月10日受付
2003年12月３日受理

抄　録

広島原爆に遠距離で被爆した花崗岩中に自然発生した長寿命核種^{36}Cl(半減期　3×10^{5}　年)の量の正確な推定を行う上で必要となる情報を得るために，カリウム(K)，ウラニウム(U)ならびにトリウム(Th)含有率を天然放射性同位元素からのガンマ線測定を行い定量した。非被爆花崗岩中のU,ThならびにKも定量し，それらが広島周辺の岩盤ごとにどのように変わるか調べた。定量結果から，たとえば愛媛県の伊予大島からの伊予石におけるKの含有率は広島市に近い倉橋島の議院石の含有率に比べて低い，などいくつかの興味深い事柄が分かった。議院石採石場の山頂近くにあった大きな岩盤の様々な深さから採取した17個の測定結果から，UとTh含有率が岩盤のある小さな部分で極めて高いことが分かった。その最大値はUの6.5ppm，そしてThの55ppmである。このことから，UとThの含有率は議院石採石場の中の位置によってかなり変化することが推察された。

キーワード：ウラニウム，トリウム，カリウム，広島原爆，中性子