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Measurements of Natural Radioisotopes in Granites Around Hiroshima City

Kazuo KATO

Department of Radiological Sciences, Faculty of Health and Welfare,
Hiroshima Prefectural College of Health Sciences

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Abstract

Approximately 2 years ago, the contents of potassium (K), uranium (U) and thorium (Th) in granites around Hiroshima City were estimated by determining the natural gamma rays from powdered samples. The powdered samples were stored in plastic capsules, and the K, U and Th contents were estimated again. The K, U and Th contents in the stored samples were well coincident with those estimated 2 years ago. This suggests that the powdered granite sample sufficiently retained K and elements in the U and Th series. A granite from Kurahashi Island near Hiroshima City contained about 5% biotite. Removing the biotite from the granite, the K content decreased by 13%, but neither the U content nor Th content changed. The minerals of feldspar (mainly potassium feldspar) and quartz were extracted from a Kurahashi granite. The U and Th contents were 2.3 ppm and 8.6 ppm in the feldspar, and 1.7 ppm and 9.6 ppm in the quartz, respectively, while the Kurahashi stone contained 4.7 ppm U and 25 ppm Th. This suggests that small minerals, except potassium feldspar, quartz and biotite, contain a large part of the U and Th elements in the Kurahashi stones. In this study, four quarries around Hiroshima City were surveyed. The granite stones examined up to the present covered totally eight quarries and six rock bodies on the surface of the earth. Only the granites from Kurahashi Island and Akasaka quarry (Fukuyama City, Hiroshima Pref.) contained more than 5 ppm U. Th contents higher than 25 ppm were observed only in the granites from Kurahashi Island. In Kurokami Island (Syunan City, Yamaguchi Pref.), stones were taken from a large rock body since before the Second World War. The granite samples from the rock body contained about 4.3 ppm U, which were about twice the amount of those in the granites from a rock body recently developed. The black-and-white stones from Akasaka quarry and Kitagi (Kasaoka City, Okayama Pref.) quarry were visually similar to each other. The data set of K, U and Th contents for the stones from Akasaka and Kitagi will be useful to differentiate each one.

Key words : granite, potassium, uranium, thorium, Hiroshima

Introduction

The mean radon concentration in the residences of Hiroshima was approximately two times as high as that in Nagasaki [1]. One of the plausible reasons for the high concentration is that many granite stones were used in the surroundings of Hiroshima City. A large part of the land was reclaimed, and so few rock bodies can be seen in the city [2]. There are several rivers in the city. It was therefore easy to carry the stones into the center of the city from the quarries in the Inland Sea. In a previous investigation, potassium (K), thorium (Th) and uranium (U) contents in the granite samples from 11 locations around Hiroshima City were measured [3]. There are many quarries for granite materials around Hiroshima City developed before the Second World War. New surveys were therefore necessary to obtain the more precise data for K, U and Th contents in the granites around Hiroshima City. The data for K, U and Th contents in granites around Hiroshima City will be useful for estimating the locations where the old granite materials used in Hiroshima City were produced. The ^{40}K and the radioisotopes in U and Th series may give the natural radiation doses to the human body. The data for K, U and Th contents in granites will be useful for revealing how natural radioisotopes contribute to natural radiation doses in Hiroshima.

Material and Methods

Granite samples

The granites from the four quarries surveyed in this study are called Okimi-stone, Akasaka-stone, Kitagi-stone, and Tokuyama-stone, respectively. The locations are shown in Figure 1. The granite stones obtained in my previous study were also used in this study. There are two quarries, Odachi and Giin on Kurahashi Island. These are close to each other, and hence the granites from both are called "Kurahashi stone" in this report. "Hiroshima" does not mean a developed quarry but a rock body appearing on the surface of the earth around Hiroshima City.

Before the Second World War, the paving stones for constructing Ujina port in Hiroshima City were carried from the quarries at Okimi-cho, Saeki-gun, Hiroshima Prefecture [4]. Most of the quarries remain intact, and the rock walls are about 15 m in height. The typical Okimi stone is fine grain biotite granite. The stones from the top to about 5 m depth of the wall were soiled or strongly damaged by weathering. The quarries were used until 1941, and hence the present quarries are useful to know how the stones were produced at quarries before the Second World War.

The stones from the quarry at Akasaka-cho, Fukuyama City, Hiroshima Prefecture were probably used for constructing a part of the stone wall of Fukuyama castle, before 1830 [5]. The quarry was probably initiated in the former times. The typical stone from Akasaka quarry is fine grain biotite granite. Red-colored middle-size grain granites were also included in the rock body in the quarry.

Kitagi quarry is located in Kasaoka, Okayama Prefecture. One sample from there was used in my previous study [3]. Most of the stones are fine grain biotite granite. Pink granite is taken from deeper rock bodies. Recently, stones with stains were also used as gravestones. It is well known that

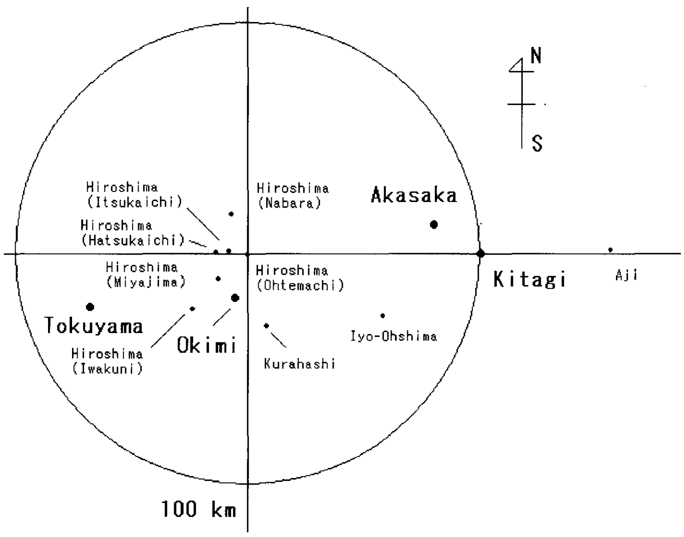

Fig. 1 Locations of quarries.
The four large circles show the locations of the quarries where the samples were obtained in this study. The small circles show the locations where the samples used in the previous study were obtained [3]. "Kurahashi" indicates both of the Odachi and Giin quarries in Kurahashi Island.

Kitagi stones were used as a base for the streetcar rails in Hiroshima City [6]. The stones taken in the former times were probably typical Kitagi stones without pink mineral or stained mineral.

Tokuyama stone is produced at Kurokami Island, Syunan City, Yamaguchi Prefecture. Most of the minerals are black-and-white, and the grains are large. Most of the stones used at Gokoku-shrine in Hiroshima City were Tokuyama stone [7]. Quarry on Kurokami Island started in 1878 [8]. Large stones from Ouzushima Island were used for constructing Osaka castle [8]. The island is in the vicinity of Kurokami Island, and stones from both islands resemble each other.

Gamma-ray measurements

The gamma rays from the rock samples were measured with a high purity co-axial type Ge-detector (Oxford CPVDS30-0190, volume of 160cc) shielded with lead blocks thicker than 15 cm. Each rock sample was powdered in an aluminum mortar, and 25 g of powdered sample in a cylindrical plastic capsule was put on the Ge-detector. Gamma-ray measurement for each sample took two or three days. The detection efficiency of the Ge-detector was 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 isotropic pressing machine (Mitsubishi, MCT 100) at Hiroshima City Industrial Technology Center. The K_2O, U and Th contents in the GSJ reference rock samples were precisely determined by numerous investigations, and hence they could be used for establishing the relationship between the count rates of gamma-ray full energy peaks and the K_2O, U and Th contents [3,9,10].

Firstly, two experiments were performed. The first experiment was conducted using some of the samples previously measured [3]. The purpose of the experiment is to reveal whether the radioactive equivalences in Th and U series were well preserved in the powdered samples. The second experiment was conducted using two stones taken from Giin quarry on Kurahashi Island [3]. The K, Th and U contents in the stones from Giin quarry were also previously determined. Almost all the biotite in 64.8 g of a Giin stone (Giin-B3) were taken out using a magnet and tweezers. The amount of biotite was 3.3 g. In order to estimate the K, Th and U contents, the gamma rays from the remainder were measured. In sequence, the feldspar and the quartz were extracted from a Kurahashi stone (Giin-B4) [3]. The minerals in the Giin stone were large, and hence it was easy to extract them. The gamma rays from the feldspar, and from the quartz were measured. The gamma rays from all minerals were also measured. Finally, in a third experiment, the gamma rays from 17 samples obtained at the four quarries mentioned above were measured.

Results

A typical gamma ray measured spectrum is shown in Figure 2. Using the measured spectra, K, Th and U contents in the granites were determined. The gamma rays

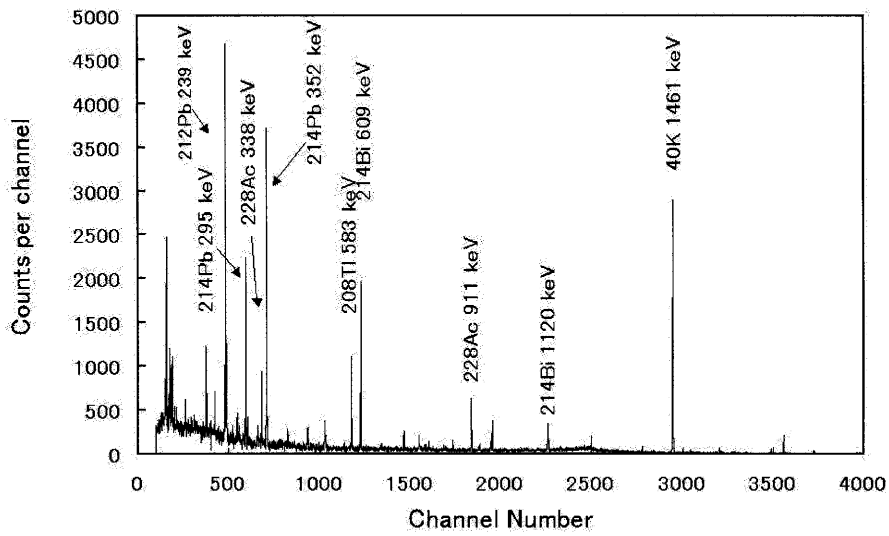

Fig. 2 Gamma-ray spectrum for 25 g of granite powdered sample from Akasaka quarry. Background spectrum was also measured using an empty plastic capsule and was subtracted from the spectrum for the sample after adjusting the difference in elapsed time for measurements.

from the four samples were measured twice. The results are shown in Table 1. The U and Th contents determined in this study were coincident to those determined 2 years ago. The biotites were excluded from the sample of Kurahashi stone (Giin). The K_2O, U and Th contents in the remainder and the whole sample are shown in Table 2. Here, the whole sample means the powdered Kurahashi granite without exclusion of biotite minerals. The K_2O content in the remainder was clearly less than that in the whole sample. The feldspar and quartz were extracted out from a Kurahashi stone. The K, U and Th contents in the minerals and in the whole sample are also shown in Table 2.

Gamma-ray measurements were performed for 17 granite samples taken from the four quarries around Hiroshima City. The K_2O, U and Th contents were determined as shown in Table 3. The relationships of K content to Th one

Table 1. K_2O, U and Th contents in the granite samples powdered about 2 years ago.

Sample+	K_2O %		U ppm		Th ppm	
Tokueiji						
827 days ago ++	4.91	±0.09	5.73	±0.64	28.80	±1.86
present	4.95	±0.09	5.60	±0.63	28.10	±1.80
Giin B3 - 4						
770 days ago ++	4.29	±0.09	6.49	±0.73	55.10	±3.51
present	4.28	±0.07	6.75	±0.75	55.21	±3.51
Iyo - 1b						
783 days ago ++	2.80	±0.07	2.18	±0.26	10.02	±0.69
Present	2.69	±0.05	2.46	±0.20	10.94	±0.73
Aji - 1						
782 days ago ++	3.29	±0.08	0.85	±0.13	5.87	±0.47
Present	3.43	±0.07	0.76	±0.12	6.17	±0.48

+ Samples obtained in the previous study [3].
++The contents quoted from my previous paper [3].

Table 2. Potassium, uranium and thorium in minerals from Kurahashi stones (Giin).

Sample	K_2O %	U ppm	Th ppm
Giin-B3	4.03 ±0.06	5.99 ±0.67	44.86 ±2.85
Giin-B3 without biotite	3.50 ±0.06	5.54 ±0.62	44.73 ±2.85
Giin-B4	3.79 ±0.08	4.71 ±0.53	25.28 ±1.64
Feldspar from Giin-B4	8.45 ±0.10	2.31 ±0.27	8.55 ±0.58
Quartz from Giin-B4	0.33 ±0.03	1.70 ±0.20	9.64 ±0.66

Table 3. Potassium, uranium and thorium in granite samples from four quarries.

Sample-No.	Name	Quarry	K_2O %		U ppm		Th ppm	
1	Kitagi +	Kitagi	3.64	±0.06	3.37	±0.38	11.28	±0.75
2	Nkitagi - 1	Kitagi	4.24	±0.06	3.28	±0.37	13.83	±0.89
3	Nkitagi - 2	Kitagi	3.85	±0.06	2.42	±0.27	10.63	±0.70
4	Nkitagi - 3	Kitagi	4.11	±0.06	3.83	±0.43	15.99	±1.03
5	Nkitagi - 4	Kitagi	3.81	±0.06	3.83	±0.43	21.65	±1.39
6	Okimi - 2	Okimi	4.87	±0.07	3.87	±0.43	16.70	±1.07
7	Okimi - 1	Okimi	4.74	±0.08	3.71	±0.42	16.78	±1.09
8	Okimi - 3	Okimi	4.76	±0.09	3.99	±0.45	18.66	±1.23
9	Akasaka 1	Akasaka	4.74	±0.07	5.50	±0.61	17.95	±1.15
10	Akasaka 1a	Akasaka	4.90	±0.08	5.50	±0.62	16.59	±1.08
11	Akasaka 3	Akasaka	4.92	±0.07	5.92	±0.66	14.60	±0.94
12	Akasaka 2	Akasaka	5.28	±0.07	3.48	±0.39	15.01	±0.96
13	Akasaka 4	Akasaka	4.64	±0.06	3.41	±0.38	15.28	±0.98
14	Akasaka 5	Akasaka	4.66	±0.06	4.52	±0.50	16.46	±1.06
15	Toku-TP	Tokuyama	4.30	±0.07	2.19	±0.26	16.18	±1.06
16	Toku-spgr	Tokuyama	3.90	±0.06	2.81	±0.32	15.66	±1.02
17	Toku-samp	Tokuyama	4.87	±0.06	4.28	±0.48	17.68	±1.14
18	Toku-UndGr	Tokuyama	4.08	±0.07	4.29	±0.35	17.29	±0.83

+ Sample used in the previous investigation [3].

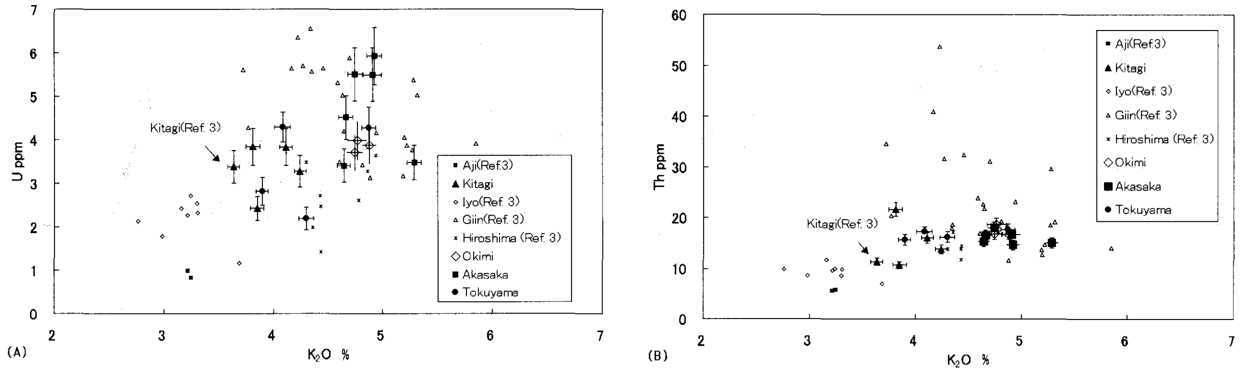

Fig. 3 Potassium, uranium and thorium contents in the granites around Hiroshima City.
The data for the granite from Kitagi, Akasaka, Okimi and Tokuyama are indicated by large symbols with error bars. Legend and label following "(Ref. 3)" mean that the potassium, uranium and thorium contents were determined in my previous study [3].

and to U one are shown in Figure 3, together with those obtained in my previous study. The U contents in the newly obtained granite stones were lower than the maximum value of U content in the Kurahashi granites.

Discussion

In this study, purchased plastic capsules were used for mounting the powdered samples on the Ge detector. The capsules were also used for preserving the samples used in my previous study. The plastic capsule is insufficiently air proof. The elements in U and Th series may have been released from the powdered samples and the capsule. However, as shown in Table 1, the U and Th contents were coincident with those determined two years ago [3]. This suggests that the powdered samples enclosed in the plastic capsules retained the radioisotopes in U and Th series.

As mentioned above, a Kurahashi stone (Giin B3 from Giin quarry) contain 5.1% biotite. The biotite content decreased as the altitude increased in the granites from Mt. Okue (1,643 m height) on the border between Miyazaki and Oita Prefectures in Kyusyu Island, and the mean content of biotite was about 5% in the granites beyond 1 km of the altitude [11]. In the granites from 1 km in altitude, the volume of potassium feldspar was larger than that of plagioclases [11]. Kurahashi stones also contain a lot of potassium feldspar. Removing the biotite, the content of U did not change, and hence the U content in the biotite was not so high. As shown in Table 2, the U content in the feldspar and quartz from the Kurahashi stone were low. This suggests that the high U content in Kurahashi stones is due to some other minor minerals, such as zircon. The age of granite can be estimated examining fission tracks in zircon, which is frequently found in pegmatite [11]. The minerals of feldspar extracted from Giin-B4 were mainly potassium feldspar, but a few plagioclase may have been included. A more precise investigation of the minerals in the granites from Giin quarry in Kurahashi Island will be carried out in the near future.

As shown in Table 3, the U content in Akasaka-2, -4 and -5 was lower than that in Akasaka-1 and -3. Akasaka-2 contained yellow minerals and Akasaka-4 and -5 contained red minerals. These were easily differentiated from Akasaka-1 and -3, which were black-and-white. The U content in Kitagi-2 was clearly lower than that in the other Kitagi stones. Kitagi-2 was taken from a rock body near the bottom (60-70 m from the top) of the quarry, which included light pink grains. This type of granite is infrequently produced at Kitagi quarry [6]. Among the Tokuyama stones, Toku-samp and Toku-UndGr shown in Table 3 were extracted from a large rock body from where stones were taken out before the Second World War. The other two Tokuyama stones were taken from a rock body newly developed in a higher position of the mountain. Thus, the U content in the old Tokuyama stones was estimated to be close to 4.3 ppm.

The data of the K_2O and U contents may be useful for differentiating the black-and-white Akasaka stone from the other stone, as the U content in the Akasaka stone was higher than those in the other granites, excluding Kurahashi granite. Kurahashi granites have large grains, and hence it is relatively easy to distinguish Akasaka stone and Kurahashi stone. It seems to be difficult to distinguish Tokuyama stone from Kitagi stone or Kurahashi stone using the data of K_2O, U and Th contents. Fortunately, those three samples of stone have different visible charac

Fig. 4 Photographs of Tokuyama, Kitagi and Kurahashi stones.
Panels (A): Tokuyama stone (Toku-samp), (B): Kitagi stone (Kitagi), (C): Kurahashi (GiinB4[3]).
White horizontal line shows 1 cm.

teristics from each other, as shown in Figure 4. Black-and-white granites from Kitagi and Akasaka quarries are visually similar to each other. The relationship between the U and K_2O contents can help differentiate the granite from each other. In this way, combined with the visual observations, the data of the K_2O, U and Th contents will be useful for identifying where granites originated from.

Several quarries around Hiroshima City have not been surveyed. Before the Second World War, granite stones were taken from the shallow part of the rock body removing the surface layer of soil and rocks disintegrated by weathering [12]. Moreover, ships were convenient for transporting the soils and heavy stones. Therefore, rocky mountains near the sea were selected for granite quarrying in the former times. All of the eight quarries surveyed until now are also located in the rocky mountains near the sea. There are several aged quarries which have not been surveyed yet. More surveys will be performed in order to establish more precisely the relationship between the data sets of K, U and Th contents and the locations where the granites were quarried out.

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広島市周辺の花崗岩中の天然放射能の測定

加藤　一生

広島県立保健福祉大学保健福祉学部放射線学科

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抄　録

約2年前に，広島市周辺の岩石粉末試料からのガンマ線を測定してカリウム（K），ウラニウム（U），そしてトリウム（Th）含有率を推定した。プラスチック容器に保管していた粉末試料からのガンマ線を再び測定して推定したK，Uならびに Th 含有率は2年前の測定値とよく一致した。このことはKならびにU系列と Th 系列に属する元素が粉末試料の中でよく保持されていることを示唆している。広島市に近い倉橋島の採石場で得られた花崗岩は約5％の黒雲母を含有していた。その黒雲母を除いたところ，K含有率は13％減少したがUと Th の含有率は変化しなかった。さらに，長石（主にカリ長石）と石英をそれぞれ倉橋島花崗岩から取り出して測定した結果 2.3 ppm U と 8.6 ppm Th（長石），そして 1.7 ppm U と 9.6 ppm Th（石英）であった。その倉橋花崗岩全体の測定結果は 4.7ppm U と 25ppm Th であった。このことは，倉橋島花崗岩中のUと Th の大部分がその黒雲母，カリ長石および石英以外の鉱物にあることを示唆している。本研究では広島市周辺の4つの採石場を新たに調査した。これまでに広島周辺の8採石場ならびに地表面の6岩盤からの花崗岩を測定した。その中で，5％以上のU含有率を示したものは倉橋島花崗岩と白黒の赤坂石（広島県福山市）だけであった。また，25ppm 以上の Th を含有していたのは倉橋島花崗岩だけであった。黒髪島（山口県周南市）からの花崗岩の中で戦前から切り出されていた岩盤からの石のU含有率は約 4.3ppm であり，近年になって切り出されている場所からの石におけるU含有率の約2倍であった。赤坂および北木（岡山県笠岡市）からの白黒花崗岩は見た目が相互に似ているが，K，Uならびに Th 含有率のデータはそれらを区別する上で役立つと思われる。

キーワード：花崗岩，カリウム，ウラン，トリウム，広島