RADIOACTIVITY IN HOKUTOLITE FROM PEITO HOT SPRING, TAIWAN

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INTRODUCTION

Hokutolite is a radioactive rock discovered at Peito hot spring, Taiwan and is famous for high content of Ra and daughter nuclides (1,2). Hokutolite is found along the river stream flowing from the source of spring water and the main chemical composition is recognized to be the sulfate of lead and barium. Radioactive elements containing in the hot spring water are incorporated into lattice structures of hokutolite during its formation. The radioactive analysis of hokutolite is necessary to understand the relationship between the growing condition of hokutolite and radioactivity and also important for radiation protection.

EXPERIMENTAL

Sampling and sample treatment

Survey of hokutolite was carried out in October, 1993 with a NaI scintillation survey meter along the river flowing from the orifice of the spring water. Hokutolite was found at about 300 m downstream from the source and high radiation was distributed as spots on the surface of rocks exposed in the river. The high radiation spots were sampled using a burin together with the underlying rock that supports the hokutolite. The mineral specimen was cut perpendicular to the surface, along the growing direction of hokutolite, into pieces with about 2 cm in width.

Hot spring water was also collected at the source and brought back to a laboratory. After addition of Ba and Pb to the water, concentrated sulfuric acid was added to make precipitate of $BaSO_4$ and $PbSO_4$ and the precipitate was recovered by filtration.

Measurement of radioactivity with an imaging analyzer

A two-dimensional radioactive distribution of hokutolite along the growing direction was measured with an imaging analyzer, BAS 1000 (Fuji Photo Film Corp.). The cut surface including the underlying rock was contacted with an imaging plate placed on a thick lead block and the sample was covered in lead grains during exposure.

Measurement of gamma spectra of hokutolite

To examine radionuclide concentrations in hokutolite along the growing direction, hokutolite was scraped off successively from the surface to the inner part with a chisel and the activity was measured with a Ge detector (ORTEC). Activities of ^{226}Ra and ^{228}Ra were determined by ^{214}Bi (609 keV) and ^{228}Ac (911 keV).

Chemical composition analysis

Hokutolite of the whole was ground to a powder and subjected to X-ray diffraction analysis to determine a ratio between $BaSO_4$ and $PbSO_4$ from lattice constants. The powder was then dissolved in HI solution with heating at 210 °C for 3 h in a sealed decomposition vessel and concentrations of Ba and Pb were determined by ICP-AES and ^{232}Th by ICP-MS.

RESULTS AND DISCUSSION

An illustration of the cut surface of hokutolite along the growing direction is shown in

Fig. 1. The hokutolite had a whitish color and was clearly distinguished from the underlying rock. The thickness of growth was about 6 mm at the maximum. Three-dimensional distribution of the radioactivity is also shown in Fig. 1. A part of the imaging picture of the hokutolite was divided into 50 x 50 mesh screen (0.16-0.64 mm^2/mesh) and the intensity of the activity in each mesh was obtained as PSL (Photo-Stimulated Luminescence) per unit area. The resolution of the imaging analyzer is 0.2 mm and the PSL is proportional to the intensity of radioactivity. The mountain located in the center of the three-dimensional distribution is hokutolite and left side of the mountain is the surface of hokutolite contacted with river water and right side towards the underlying rock. An intrusion of hokutolite into cracks of the underlying rock was observed and the mountain showed several peaks. The imaging pictures show that non-radioactive fine particles are incorporated into hokutolite during its growth (3). The mole ratio of Ba/Pb = 1.9 was determined by X-ray diffraction analysis and 2.4 by ICP-AES, and are similar to values previously reported (4,5).

The activity ratios of $^{228}Ra/^{226}Ra$ in the hot spring water and the whole hokutolite were listed in Table 1. The hokutolite (A) is the same sample shown in Fig. 1 and was located out of the river water, while the hokutolite (B) in the water. Lower activity ratio in the whole hokutolite than the spring water is reasonable because the activity ratio decreases with time: the hokutolite near the underlying rock has lower activity ratio compared to the surface where the activity ratio would be the same as that of the spring water. The faster decay of ^{228}Ra than ^{226}Ra decreases the activity ratio in the whole hokutolite. An equilibrium $^{228}Ra/^{226}Ra$ activity ratio of 0.095 was expected for hokutolite that had been formed at a constant growth rate for a long time within the river water having $^{228}R_a/^{226}Ra$ activity ratio of 26.3. However, the observed activity ratio for the hokutolite (A) was significantly lower than the expected one and even for the hokutolite (B)

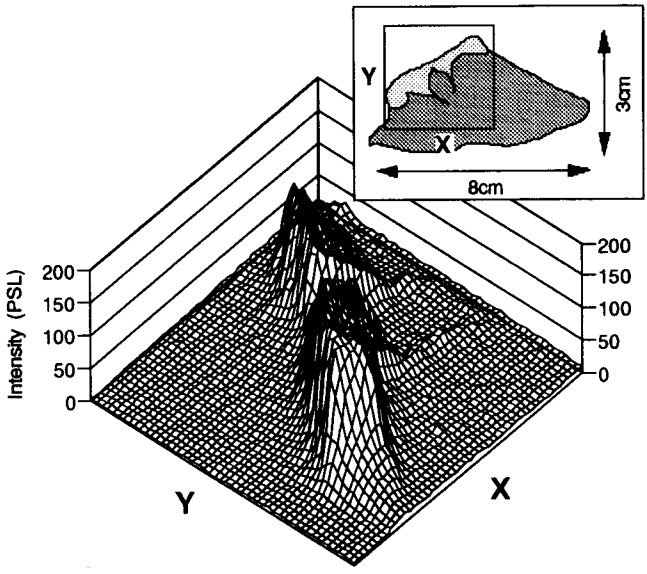

Figure 1. An illustration of the cut surface of hokutolite and its three-dimensional distribution of radioactivity measured with an imaging analyzer.

Table 1. Activity ratio in the spring water and hokutolite.

	Spring water	Hokutolite (A)	Hokutolite (B)
Activity Ratio $^{228}Ra/^{226}Ra$	26.3	0.027	0.178

that gives an unrealistic growing period within the river water, about 2,000 years. This would suggests that these hokutolite had been out of the river water for a certain period.

A change of $^{228}Ra/^{226}Ra$ activity ratio from the surface along the growing direction is shown in Fig. 2. The activity ratios were seemed to become constant at distance over 2.5 mm from the surface, suggesting an existence of ^{232}Th in the hokutolite and a radioactive equilibrium between ^{232}Th and ^{228}Ra. The average activity of ^{228}Ra from 2.5 to 5.5 mm is 1.52±0.34 Bq g^{-1} and the activity of ^{232}Th determined by ICP-MS was 1.55 Bq g^{-1}, supporting the radioactive equilibrium between ^{232}Th and ^{228}Ra over 2.5 mm. The decline in activity ratio from the surface to 2.5 mm could be attributed to decay of ^{228}Ra incorporated into hokutolite and the activity ratio at the surface would suggest an elapsed time after the hokutolite had isolated from the river water. A growth rate of the hokutolite was simulated and 0.17 mm y^{-1} gives a best fit as shown in Fig. 2. The elapsed time for the hokutolite after isolation was calculated to be about 60 years. The activities of ^{226}Ra and ^{228}Ra in hokutolite are summarized in Table 2. Higher activity ratio than the present work was reported for thin hokutolite deposited on the surface of sands and pebbles (1), indicating the older age of the present hokutolite. The concentration of ^{226}Ra in hokutolite is a few orders of magnitude higher than that in usual rocks.

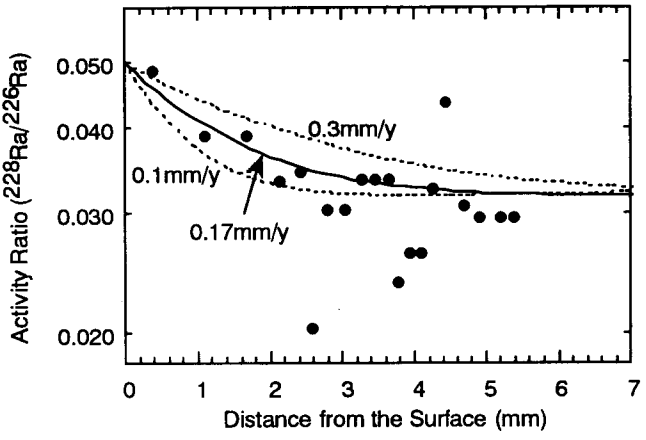

Figure 2. Activity ratios of ^{228}Ra to ^{226}Ra in hokutolite along the growing direction and results on simulation of growth rate.

Table 2. Activities* of radium isotopes and activity ratio in hokutolite.

^{228}Ra	^{226}Ra	$^{228}Ra/^{226}Ra$	Ref.
0.10	0.052	1.92	sands & pebbles (1)
0.48	0.059	8.14	sands & pebbles (1)
9.25	36.26	0.255	crystal (2)
3.46	71.3	0.0485	Present Work**

*Bq g^{-1}, ** The surface of the hokutolite

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