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Radioactivity and Weathering of Granite from Mt. Tanakami, Shiga Prefecture

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ABSTRACT

Measurements of radioactive abundance were made for granite rock samples from Mt. Tanakami, Shiga Prefecture. Weathering degrees of those samples were also estimated, and a relationship between the radioactive abundance and the weathering degree was discussed from the view point of an outflux of radioactivity to the environment.

1. Introduction

Mt. Tanakami is located in the south-east part of Ohtsu City, Shiga Prefecture. The granite of this mountain is mainly a coarse-grained granitite. Moreover in several areas of this mountain a pegmatite is yielded, which makes this mountain famous in Japan. The radioactivity in the granite is stronger than that in other kinds of rock, and its mechanical strength becomes easily lower by weathering. Thus it is important to investigate how radioactive nuclides in the granite flow out to the environment. The granite samples were collected from three places on this mountain. The radioactivity of these samples were measured by a high purity germanium detector and the weathering degrees were estimated from area ratios of Fe^{2+} to Fe^{3+} in Mössbauer spectra.

2. Experimental Procedures

The samples were washed in water, and after drying they were crushed. They, weighting between 60 and 120g, were packed in plastic containers. The radioactive measurements were carried out with the high purity germanium detector (1113-16195-S, ORTEC). The sample in the container was placed on the end cap of the detector, and they were enclosed with lead blocks(10 cm thick) for reducing the background which were contributed by cosmic rays and radioactive nuclides in surrounding materials. A signal from the detector is transferred to a multi-channel analyser (MCA/PC98B, Labo Equipment Co.) through a pre- and a main amplifier. A γ -ray spectrum accumulated in the multi-channel analyser was transferred to a personal computer and stored in a floppy disk. The γ -ray spectrum was standardized by using the reference γ -ray sources (⁶⁰Co, ¹³⁷Cs and ²⁴Na).

Mössbauer spectroscopy has been extensively applied to minerals for characterizing oxidation states and co-ordination numbers of iron atoms [1, 2]. The Mössbauer spectra were obtained using a conventional constant acceleration spectrometer in a transmission geometry. A ⁵⁷ Co

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radioactive source in Rh matrix was used with a krypton-carbon dioxide proportional counter as γ -ray detector. Velocity calibration was carried out with an enriched ⁵⁷Fe foil. The Mössbauer absorber was prepared from the powdered sample which passed through a 200-mesh sieve.

3. Results and Discussions

A typical γ -ray spectrum is shown in Fig.1. The U/Th ratio in the granite was estimated by calculating the peak-area counts of the 351.9 keV γ -ray of ²¹⁴Pb(U series) and the 238.6 keV γ -ray of ²¹²Pb(Th series).

Figure 2 shows the Mössbauer spectra. It is easy to notice that there are two doublets due to iron atoms in the ferrous state (Fe^{2+}) and in the ferric state (Fe^{3+}). The spectrum was fitted to a sum of two doublets using a computer program with non-linear least square method. The lines in each doublet were assumed to have equal area and width, and Lorentzian line shapes were used for the fitting. The Fe^{3+}/Fe^{2+} radio was evaluted from the both peak areas. Iron atoms in the granite are mainly contained in a biotite. The obtained Mössbauer spectra are simillar to a spectrum of the biotite which was priviously reported [3].

The Fe³⁺ /Fe²⁺ radio is a measure of the weathering degree of the granite since the low strength of the granite is related to the alteration of biotite [4] and the Fe³⁺ peak increases with the oxidation of biotite by weathering [3]. Figure 3 shows a plot of 214 Pb/ 212 Pb against Fe³⁺/Fe²⁺. In this figure there is a peak around 2.0 in Fe³⁺/Fe²⁺. Considering a non-uniform distribution of radioactivity in the granite, following conclusion is deduced;

In the early stage of weathering, the part which contains Th series nuclides more than U series nuclides begins to flow away to the environment selectively. In the advanced stage of weathering, the part containing the U series nuclides also flows away as well as the part containing the Th series nuclides.

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Mössbauer spectra of granites. The weathering degree advances from top to buttom.



A plot of ²¹⁴Pb/²¹²Pb against Fe³⁺/Fe²⁺.

References

- [1] G. M. Bancroft, A. G. Maddock and R. G. Burns, Geochem. Cosmochem. Acta 31 (1961) 2219.
- G. M. Bancroft, Mössbauer Spectroscopy, an Introduction for Inorganic Chemists and Geochemists, McGraw-Hill Inc., London(1973).
- [3] B. A. Goodman and M. J. Wilson, Mineral. Mag. 39 (1973) 448.
- [4] D. J. Stierman and J. H. Healy, Pure Appl. Geophys. 122 (1984) 425.

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