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Abstract

The chemical composition was analyzed and the radioactivity, radon exhalation rate and emanation fraction were measured to investigate the characteristics of the granites sampled at Misasa and Badgastein world-famous for the radon therapy. The Misasa granite was probably composed of quartz, albite and microcline. The Badgastein granite was probably composed of quartz and muscovite. The radon exhalation rates and emanation fractions of the Misasa granite were much higher than those of the Badgastein granite, regardless of the ^{226}Ra activity concentrations.

Keyword: Radioactivity; Radon emanation fraction; Granite; Misasa; Badgastein

1. Introduction

A hot spring at Misasa in Japan and a gallery at Badgastein in Austria are world-famous for the radon therapy (Mitsunobu et al., 2005; Zötl, 1995). Patients with rheumatic diseases etc. inhale radon and its progenies for therapeutic purposes (Falkenbach et al., 2005). Many investigations have been made in order to clarify the mechanism of the radon therapy (Yamaoka, 2006). However, the mechanism has not yet been understood because the generation and behavior of radon and its progenies in environment and the internal movement of inhaled radon and its progenies are not entirely obvious (Porstendörfer, 1994; Ishikawa et al., 2003). We previously reported the leaching rate of radon from the rocks into water, which were sampled at the region where the radon therapy is practiced (Sakoda et al., 2007a; Sakoda et al., 2007b). On the other hand, to our knowledge, there is no information on the radon exhalation rate and emanation fraction of these rocks. Therefore, it is important to obtain the radon exhalation rate and emanation fraction of the radioactive rock as a radon source.

In this study, as part of the study to elucidate the mechanism of the radon therapy, the chemical composition was analyzed and the ^{226}Ra activity, radon exhalation rate and emanation fraction was measured in order to investigate the characteristics of two granites sampled at Misasa and Badgastein. The ^{232}Th and ^{40}K activity was also measured. In addition, the difference of the radon emanation fraction between the two granites was discussed. Here, it is noted that in this paper “radon” means ^{222}Rn, and “emanation fraction” means the quantity better known under “emanation power”.

2. Materials and methods

2. 1 Sample

The two granites were used in this study. One (Misasa granite) was collected at the hot bathing hall with the high concentration of radon at Misasa Medical Center of Okayama University Medical School. In the corner of the bathing hall, there is the well in which the granite is transported through underground and deposited. The granite of varied grain size was sampled from there. The other one (Badgastein granite) was collected near the gallery of the Gasteiner Heilstollen in Austria. A block of the granite was crushed and then part of the crushed granite which was originally inside the block was sampled. For this study, each sample was sieved to the grain size of 250–500 μm.

2. 2 Chemical composition analysis

The constituent compound of the samples was studied by X-ray diffraction (XRD). The XRD measurements were performed on an X-ray diffractometer (RINT-1000, Rigaku, Japan) using Cu K_α radiation at 40 kV and 200 mA.

The constituent element of the samples was studied by inductively coupled plasma atomic emission spectrometry (ICP-AES). The granite samples were appropriately

dissolved into solution before the ICP-AES measurement. The elements in the samples were measured by ICP-AES (ICPA-577, Nippon Jarrell Ash, Japan) under the following conditions: plasma gas flow 0.4 l min^{-1}; cooling gas flow 16 l min^{-1}; carrier gas flow 0.7 l min^{-1}.

2. 3 Measurement of radioactivity, radon exhalation rate and emanation fraction

The ^{226}Ra, ^{232}Th and ^{40}K activity of the samples was determined by gamma-ray spectrometry. Each sample of several tens of grams were put in a plastic container (inner diameter: 46 mm, height: 59 mm) called a U-8 container and sealed with an epoxy resin adhesive. After establishing the radioactive equilibrium among ^{226}Ra, radon and its progeny, gamma rays from the samples were measured using a high-purity germanium detector (GMX-15200, SEIKO EG&G, Japan). The gamma rays of ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 keV) were used for the ^{226}Ra activity analysis, those of ^{212}Pb (239 keV), ^{208}Tl (583 keV) and ^{228}Ac (911 keV) for the ^{232}Th activity analysis. For the ^{40}K activity analysis, the gamma ray of 1461 keV was used.

A chamber of 26 L (0.37×0.26×0.27 m^3), air filter (pore size: 0.1 μm), scintillation cell (300A, Pylon, Canada) and its monitor (AB-5, Pylon, Canada) were used in order to measure the radon exhalation rate and emanation fraction of the samples. The schematic of apparatus is shown in Fig. 1. A cylindrical vessel of 10 cm in diameter was first filled with the Misasa granite of 180 g or the Badgastein granite of 360 g up to a height of 2 cm and then the vessel was placed in the chamber. In the chamber and scintillation cell, air was continuously circulated by a built-in pump in the radiation monitor and the flow rate was 1 L min^{-1}. The temperature in the chamber was controlled to about 25˚C. Under these conditions, the concentrations of radon emanated from the sample in the chamber were measured at intervals of 1 h for 10 days.

Subsequently, the growth curve of the radon concentration was plotted.

In this study, the change of the radon concentration C (Bq m^{-3}) per unit time t (s) in the chamber is expressed as

$$\frac{dC}{dt} = \frac{EM}{V} - (\lambda + \alpha)C, \quad (1)$$

where E (μBq kg^{-1} s^{-1}) is the radon exhalation rate per unit mass (radon mass exhalation rate), M (kg) the mass of the sample, V (m^3) the volume of the chamber, λ (s^{-1}) the decay constant of radon and α (s^{-1}) the leakage rate from the chamber. Solving Eq. (1), the radon concentration growth as a function of time is given by the formula:

$$C(t) = \frac{EM\left\{1 - e^{-(\lambda+\alpha)t}\right\}}{(\lambda + \alpha)V} + C_0 e^{-(\lambda+\alpha)t}, \quad (2)$$

where C_0 (Bq m^{-3}) is the initial condition at t=0. Fitting a series of the measured radon concentrations to Eq. (2), E and α are obtained. Consequently, the radon emanation fraction F (%) is calculated by the following equation.

$$F = \frac{E \times 10^{-6}}{\lambda R} \times 100, \quad (3)$$

where R (Bq kg^{-1}) is the ^{226}Ra activity concentration of the sample.

3. Results and discussion

3. 1 Chemical composition

Fig. 2 shows XRD patterns of the samples. Most peaks of the Misasa granite corresponded to the diffraction patterns of quartz (SiO_2), albite ($NaAlSi_3O_8$) and microcline ($KAlSi_3O_8$) and most peaks of the Badgastein granite corresponded to the diffraction patterns of quartz (SiO_2) and muscovite ($KAl_2(Si_3Al)O_{10}(OH)_2$). Table 1 shows the major elements in the samples determined by ICP-AES. The Misasa granite

was composed of SiO_2, Al_2O_3, Fe_2O_3, Na_2O, K_2O and CaO and the Badgastein granite was composed of SiO_2, Al_2O_3, K_2O and Fe_2O_3 in order of the concentration. All the constituent elements of the Misasa granite consisting of SiO_2, $NaAlSi_3O_8$ and $KAlSi_3O_8$ and the Badgastein granite consisting of SiO_2 and $KAl_2(Si_3Al)O_{10}(OH)_2$ were detected by ICP-AES. Consequently, the Misasa granite was probably composed of quartz, albite and microcline. The Badgastein granite was probably composed of quartz and muscovite.

3. 2 Radioactivity, radon exhalation rate and emanation fraction

The radioactivity concentration of the samples is shown in Table 2. The ^{226}Ra activity concentrations of the Badgastein granite were 7.9 times higher than those of the Misasa granite. The ^{232}Th and ^{40}K activity concentrations of the Badgastein granite were 3.0 and 1.5 times higher than those of the Misasa granite, respectively

Fig. 3 shows the measured growth curves of radon exhaled from the samples. Both curves were very similar but this was a coincidence. If a radon source is set in a completely airtight chamber, radon concentration will saturate about 30 days after closing the chamber. However, in this study, the radon-saturated time is shorter than 30 days because of the leakage of radon from the chamber. The leakage rate ranged from 3.72×10^{-6} to 4.18×10^{-6} s^{-1} in this study. On the other hand, it also should be discussed whether thoron emanated from the sample and its progeny contributed to the measured radon concentration (alpha counts) because the scintillation cell used in this study is sensitive to not only radon but also thoron. The radioactivity concentrations of ^{232}Th, which is the parent nuclide of thoron, in both samples used in this study were much lower than those of ^{226}Ra as shown in Table 2. In addition, the diffusion length of thoron in air is only about 3 cm although that of radon is about 2 m (Søgaard-Hansen

and Damkjær, 1987) and the air-flow volume (1 L) per minute in this experiment was not too high compared with the chamber volume (26 L). Thus, little thoron would eventually reach the detector cell and the alpha particles from thoron and its first progeny ^{216}Po would not contribute much. Moreover, thoron's third progeny ^{212}Bi of alpha emitter also may not contribute to the alpha counts because most thoron's progenies may be deposited on the wall of the chamber or be filtrated by the air filter. Consequently, the influence of thoron and its progeny to the measured radon concentration was ignored.

Taking the leakage of radon into account, the fitting curves in Fig. 3 were plotted and then the radon mass exhalation rate and emanation fraction are shown in Table 2. The radon mass exhalation rates of the Misasa granite were 1.9 times higher than those of Badgastein granite although the ^{226}Ra activity concentrations of the Badgastein granite were 7.9 times higher than those of the Misasa granite. Moreover, the radon emanation fractions of the Misasa granite were 13.6 times higher than those of Badgastein granite. UNSCEAR (2000) reported that the typical radon emanation fractions for rocks and soils range from 5 to 70% and employed 20% as a representative value. In this study, the radon emanation fractions of the Badgastein granite were lower than the typical value although those of the Misasa granite were above the representative value.

The constituent mineral and the radon emanation fraction were much different between the Misasa and Badgastein granite, as described above. This result might suggest that the radon emanation fraction is subjected to the chemical composition of the rock sample. Sato and Nakamura (1993) studied the leaching of radon from weathered and fresh granites into water. Their result showed that there is a positive correlation between leaching of radon and the degree of weathering of the rock because

the weathering process increases surface area of grain. Their conclusion can be applied to the radon emanation from rock to air because the leaching of radon from rock into water corresponds to the maximum value of the radon emanation fraction. In the present study, the Misasa granite may be weathered because it was transported through underground and deposited in the well, as described in subsection 2. 1. In contrast, the Badgastein granite may be fresh because it was originally inside a block of the rock. Our result that the radon emanation fraction of the weathered granite sampled at Misasa was higher than that of the fresh granite sampled at Badgastein agrees with the conclusion shown by Sato and Nakamura (1993).

4. Conclusions

The chemical composition of the two granites was analyzed and the radioactivity, radon mass exhalation rate and emanation fraction were measured, which were sampled at Misasa in Japan and at Badgastein in Austria. The Misasa granite was probably composed of quartz, albite and microcline. The Badgastein granite was probably composed of quartz and muscovite. The radon mass exhalation rates of the Misasa granite were 1.9 times higher than those of the Badgastein granite although the ^{226}Ra activity concentrations of the Badgastein granite were 7.9 times higher than those of the Misasa granite. The radon emanation fraction of the Misasa granite (28.5%) was 13.6 times higher than that of the Badgastein granite (2.1%). Furthermore, the ^{232}Th and ^{40}K activity concentrations of the Badgastein granite were 3.0 and 1.5 times higher than those of the Misasa granite, respectively.

The radon emanation fraction of radioactive rock is sensitive to the environmental conditions such as water content and temperature. More detailed work is necessary to study how much such a condition affect the radon emanation using the granites

sampled at Misasa and Badgastein.

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