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Effects of some physical conditions on leaching rate of radon from radioactive minerals originating from some hot springs

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Abstract

In order to determine the best physical conditions for leaching more radon from minerals into water, we measured the leaching rate of radon from radioactive minerals under the conditions of some different grain sizes and water temperatures. Water temperature affected the leaching rate of radon although grain size did not significantly affect it. Furthermore, we proposed ultrasonic irradiation to the mixture of a mineral and water as the method of leaching more radon. Ultrasonic irradiation was efficient to leach more radon from the mineral soaked in water because of ultrasonic cavitation.

Keywords: Radon hot spring; Radioactive mineral; Leaching; Grain size; Water temperature; Ultrasonic irradiation

1. Introduction

The radon hot spring therapy at Misasa in Japan and Badgastein in Austria is widely employed in medical treatment for various diseases such as osteoarthritis, bronchial asthma (Deetjen, 1997; Mitsunobu et al, 2005). Radon (^{222}Rn and ^{220}Rn) produced by alpha decay of its parent radium (^{226}Ra and ^{224}Ra) is a radioactive gas and exists within radioactive minerals. The huge radioactive minerals in underground constantly provide radon to water of the natural radon hot spring. High radon concentration in the hot spring water is stably maintained. On the other hand, in order to receive the benefits of the radon therapy in the region where there is no radon hot spring, artificial radon hot springs have been constructed by blending a radioactive mineral powder and heated water in a storage tank. The mixed water in the tank is used as water of the artificial radon hot springs. However, it is not easy to increase and keep the radon concentration to the objective concentration in the hot spring water because the leaching rate of

radon from a mineral into water is low in open system. Therefore, the study on the method of increasing the leaching rate of radon under the regulation value would provide useful information for the construction of an artificial radon hot spring. In a previous paper, we have shown that more acid solution increases the leaching rate of radon from minerals (Sakoda et al., 2007).

In this study, we measured the leaching rate of radon from some radioactive minerals under the conditions of some different grain sizes and water temperatures in order to determine the best physical conditions for leaching more radon. Furthermore, we proposed the ultrasonic irradiation to the mixture of the mineral and water as the method of increasing the leaching rate of radon. We simply simulated the radon hot spring by soaking the minerals that exist in the famous region for the radon hot spring and determined the leaching rates of radon by measuring gamma rays of radon's daughter nuclides in leached water.

2. Materials and methods

2. 1 Mineral samples

The radioactive sludge was obtained from the hot bathroom with a high concentration of radon at Misasa Medical Center of Okayama University Medical School and the radioactive rock was collected at Badgastein in Austria. Furthermore, we prepared also the mineral utilized at the artificial thoron hot spring in Japan. All the mineral samples were sieved to the five ranges of grain size d (μm): (1) $d \leq 63$ (–63), (2) $63 < d \leq 250$ (63–250), (3) $250 < d \leq 500$ (250–500), (4) $500 < d \leq 1000$ (500–1000) and (5) $1000 < d \leq 2000$ (1000–2000). The characteristics of these samples are shown in Table 1.

2. 2 Procedure of radon leaching under some physical conditions

2. 2. 1 Grain size

Each sample of the sludge of Misasa (50 g) and the rock of Badgastein (50 g) in all the grain sizes was blent with distilled water (200 g) in a beaker. Subsequently, the mixture stood for 30 d at room temperature (25˚C) because the radioactive equilibrium between ^{226}Ra (half life 1.600×10^{3} y) and ^{222}Rn (half life 3.824 d) is established within 30 d. After 30 d, the leached water (90 ml) was quickly drawn from the beaker and tightly sealed in a measuring container (inner diameter 46 mm, height 59 mm), which is called a U-8 container, to minimize the escape of radon. The container stood for 4 h in order to establish the radioactive equilibrium between ^{222}Rn and ^{214}Pb (half life 26.8 min) in the leached water.

2. 2. 2 Water temperature

Each sample of the sludge of Misasa (grain size 250–500 μm, 50 g), the rock of Badgastein (grain size 250–500 μm, 50 g) and the mineral of the artificial thoron hot spring (grain size 63–250 μm, 100 g) was blent with distilled water in a beaker. The ratio of the sample to water was 20% by weight. Subsequently, the mixture stood for 3 d at 5, 25, 40 or 55˚C. We confirmed that the radon concentration in the leached water was approximately saturated in 3 d. The temperature of the mixture was adjusted to 5˚C with a refrigerator, to 25˚C at room temperature, to 40 and 50˚C with a thermostatic bath. After 3 d, the leached water (90 ml) drawn from the beaker was sealed in the U-8 container and then stood for 4 h.

2. 2. 3 Ultrasonic irradiation

Each of the sludge of Misasa (grain size 250–500 μm, 50 g), the rock of Badgastein (grain size 250–500 μm, 50 g) and the mineral of the artificial thoron hot spring (grain

size 63–250 μm, 100 g) was blent with distilled water in a beaker. The ratio of the sample to water was 20% by weight. Subsequently, the mixture was irradiated with ultrasonic waves (40 kHz) for 5, 30 or 60 min. The water temperature was adjusted to 40˚C because the temperature of water utilized at common artificial radon hot springs is generally about 40˚C. In contrast, without the ultrasonic irradiation, the mixture of the sample and water stood for 180 min. After those processes, the leached water (90 ml) drawn from the beaker was sealed in the U-8 container and then stood for 4 h.

In all experiments as mentioned above, the gamma rays from ^{214}Pb (352 keV) and ^{212}Pb (239 keV) in the leached water were measured using the high-purity germanium detector (GMX-15200, SEIKO EG&G, Japan) for 24 h.

2. 3 Estimation of the radon concentration in the leached water

The peak area (S) of ^{214}Pb and ^{212}Pb in the gamma-ray spectra was calculated using a nonlinear least-squares fitting program adopting Gaussian peak shapes. The specific activity (A) of ^{214}Pb and ^{212}Pb at a certain time during the gamma-ray measurements are expressed as

$$A = \left(\frac{S}{T} - \frac{S_b}{T_b} \right) \frac{1}{\varepsilon_{gamma}} \cdot \frac{1}{\varepsilon_{effi}} \cdot \frac{1}{W}, \qquad (1)$$

where S_b is the peak area of the gamma ray of background, T the measurement time of the leached water, T_b the measurement time of background, ε_{gamma} the branching ratio of the gamma ray (Firestone and Shirley, 1996), ε_{effi} the detection efficiency of the detector and W the weight of the leached water used for the measurements. In order to obtain the radioactivity at the time t=0 when the leached water was drawn from the beaker, the radioactivities of ^{214}Pb and ^{212}Pb calculated from Eq. (1) must be corrected. After performing the decay corrections, the concentrations of ^{222}Rn and ^{220}Rn in the

leached water can be estimated. The decay correction of ^{214}Pb and estimation of ^{222}Rn were carried out according to a previous paper (Sakoda et al., 2007). On the other hand, the decay correction of ^{212}Pb and estimation of ^{220}Rn were carried out in basically the same way of the decay correction of ^{214}Pb and estimation of ^{222}Rn as described below.

The radioactive equilibrium among ^{224}Ra (half life 3.66 d) in the mineral grains which can emanate ^{220}Rn (half life 55.6 s), ^{220}Rn emanated from the grain into water and its progeny (^{216}Po (half life 0.145 s) and ^{212}Pb (half life 10.64 h)) may be established in the leached water of the beaker in all the experiments. Therefore, assuming that the radioactivities of ^{220}Rn, ^{216}Po and ^{212}Pb at t=0 were unity, we calculated the radioactivities of these nuclides at any time t from the Bateman equations (Bateman, 1910). The detail of the calculation process is omitted because it was previously reported (Sakoda et al., 2007). Fig. 1 shows the calculation result of ^{212}Pb.

It was needed to determine the time which the radioactivity of ^{212}Pb obtained from Eq. (1) corresponded to the radioactivity at. We calculated the area below the curve of ^{212}Pb from t=0 (drawing the leached water from the beaker) to t=28.25 h (finishing the gamma-ray measurement) in Fig. 1 and obtained the average of the area an hour. It took about 0.25 h to start the gamma-ray measurement after drawing the leached water from the beaker. The elapsed time for the value of the average area was determined from the curve of ^{212}Pb in Fig. 1.Consequently, the radioactivity of ^{212}Pb obtained from Eq. (1) corresponded to the radioactivity at t=14.72.

The radioactivity of ^{212}Pb at t=14.72 was 0.3834 times larger than that of ^{212}Pb at t=0. The decay correction of ^{212}Pb is finished by dividing the radioactivity of ^{212}Pb calculated from Eq. (1) by 0.3834. The corrected radioactivity of ^{212}Pb is equivalent to the ^{222}Rn concentration in the leached water of the beaker because it was assumed that

the radioactive equilibrium between ^{220}Rn and ^{212}Pb may be established.

3. Results and discussion

3. 1 Effect of grain size on leaching of radon from the mineral samples

Fig. 2 shows the leaching rate of radon from the mineral samples of the different grain sizes. Only ^{222}Rn was leached from the sludge of Misasa and the rock of Badgastein while ^{220}Rn as well as ^{222}Rn was leached from the mineral of the artificial thoron hot spring. There was no significant difference in the leaching rates of ^{222}Rn from the sludge of Misasa and the rock of Badgastein among all grain sizes.

In theory, the emanation of radon must be proportional to the surface area of a mineral grain. The leaching rates of ^{222}Rn from the sludge of Misasa and the rock of Badgastein did not depend on the grain size and this trend is consistent with the results reported by several researchers (Rama and Moore, 1984; Chau et al, 2005). Rama and Moore (1984) deduced the reason as follows. It is obvious that radon is released from inner grains as well as from the surface of the grains because the radon emanation rate are independent from the grain size. The solids used in their study had the pores with the very large wall areas, but the very small volumes, namely with the opening of the width of 10–20 nm, which was termed ‘nanopores’. Radon and other isotopes are introduced into nanopore water as a result of aloha recoil from the walls of the nanopores in solids. Radon, an inert gas, can diffuse out into intergranular water while the reactive isotopes are adsorbed within the nanopores. Our results in the present study supported their deduction.

3. 2 Effect of water temperature on leaching of radon from the mineral samples

Fig. 3 shows the leaching rate of radon from the mineral samples into water of the

different temperatures. Only ^{222}Rn was leached from the sludge of Misasa and the rock of Badgastein into water while ^{220}Rn as well as ^{222}Rn was leached from the mineral of the artificial thoron hot spring. Higher water temperature reduced the leaching rates of ^{222}Rn from all the mineral samples. The concentrations of ^{222}Rn leached from the sludge of Misasa and the mineral of the artificial thoron hot spring were below the detection limits when the water temperature was 55°C. In contrast, higher water temperature slightly increased the leaching rates of ^{220}Rn from the mineral of the artificial thoron hot spring.

It was previously reported that the higher temperature increases the radon emanation rate (Stranden et al., 1984; Iskandar et al, 2004). The results of the leaching rates of ^{222}Rn in this study did not agree with their reports while those of the leaching rates of ^{220}Rn agree with their reports. The difference of leaching between ^{222}Rn and ^{220}Rn can be reasonably explained as follows. Higher-temperature water allows radon to diffuse from the leached water to the atmosphere because higher water temperature decreases the solubility of a gas. These facts imply that a larger amount of radon emanated from the leached water to the atmosphere while the number of radon atom leached from the mineral into water increased with increasing water temperature. Therefore, the leaching rate of ^{222}Rn should reduce when the water temperature is increased. On the other hand, after ^{220}Rn is leached from the mineral, ^{220}Rn decays immediately due to its short half life and the ^{220}Rn progeny is produced. ^{220}Rn leached from the minerals is less subject to diffusing from the leached water to the atmosphere than ^{222}Rn because higher-temperature water dissolves more radon progeny of solid metals. Therefore, the leaching rate of ^{220}Rn may not reduce when the water temperatures is increased. Consequently, increasing water temperature may have a negative effect on leaching radon from the mineral.

3. 3 Effect of ultrasonic irradiation on leaching of radon from the mineral samples

Fig. 4 shows the leaching rate of radon from the mineral samples with the ultrasonic irradiation. Without the ultrasonic irradiation, ^{222}Rn leached from all the mineral samples was not detected. With the ultrasonic irradiation, only ^{222}Rn was leached from the sludge of Misasa and the rock of Badgastein while ^{220}Rn as well as ^{222}Rn was leached from the mineral of the artificial thoron hot spring. The leaching rates of ^{222}Rn and ^{220}Rn drastically increased with the ultrasonic irradiation for 5 min except ^{222}Rn leached from the mineral of the artificial thoron hot spring and then gradually increased as the irradiation time increased. Our result suggests that ultrasonic irradiation to the mineral soaked in water in order to leach more radon is very effective. One of important phenomena caused by ultrasonic waves is ultrasonic cavitation. Ultrasonic cavitation is the phenomenon that huge microscale bubbles in liquid break when ultrasonic waves passe through liquid. The ultrasonic irradiation increased the leaching rate of radon from the mineral samples because the ultrasonic cavitation may increase wetted surface area of the mineral grains as a whole, which caused the stirring effect.

4. Conclusions

Grain size may not affect the leaching rate of radon because there was no significant difference in the leaching rates of radon from the mineral of different grain sizes. As water temperature was increased, a larger amount of radon, especially ^{222}Rn, diffused from the leached water to the atmosphere while the number of radon atom leached from the mineral into water increased. Therefore, increasing water temperature may have a negative effect on leaching radon from the mineral. Ultrasonic irradiation to the

mineral is efficient in order to leach more radon from the mineral. The ultrasonic irradiation increased the leaching rates of radon because the ultrasonic cavitation caused the stirring effect.

A set of our results in this study clarified the trends of leaching of radon on some physical conditions from radioactive minerals into water. The present study would serve to construct an artificial radon hot spring where a radioactive mineral is used as a radon source. Moreover, we think that there is a possibility of its application to radon therapy in medicine all over the world.

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