Review (Invited)

Mechanisms and Modeling Approaches of Radon Emanation for Natural Materials

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Radon emanation means the escape of radon atoms from solid grains into pore space; it is the very first process that may lead to radon exposure in the environment. Experimental and numerical studies of radon emanation have been diligently carried out since its recognition as a carcinogen. Our previous review of the measured data showed a wide range of radon emanation fractions from natural substances, and then we discussed the effects of environmental factors such as pore water. The present paper provides an overview of the approaches and progress of radon emanation modeling that may be useful for the interpretation of measured data. Recoil and/or diffusion of radon in solid following alpha decay of radium, which underlies the mechanisms of radon emanation, have been incorporated into numerical models. In the calculation based on recoil-based emanation, radium distribution and pore size were the most important parameters, which govern the magnitudes of radon ejections from the birth grain and of radon embedding into another solid surface, respectively. The solid diffusion appeared significant only at a temperature higher than several hundred degrees Celsius. A model is now desired to be developed that incorporates the transport process of radon atoms that are still settled in solid after alpha recoil, considering radiation damage and its resulting inner path network.

KEY WORDS: radon emanation, modeling, alpha recoil, solid diffusion, grain, pore, environmental factors.

I INTRODUCTION

Before radon, a radioactive inert gas, migrates through soil and disperses in the atmosphere, it must first escape from radium-bearing solid grains. Escape of radon atoms from grains into pore space is called as *emanation*, which should be distinguished from *exhalation* characterized by the emanation of radon and subsequent migration through pore fluid. A number of radon emanation measurements have been carried out from the viewpoint of health physics as well as geoscience, because emanation is the very first process in context of radon exposure.

Of radon isotopes, ^{222}Rn and ^{220}Rn are dominant in the environment. Rn-222 with a half-life of 3.8 days, formed by the alpha decay of ^{226}Ra belonging to uranium series, has been of great concern in several decades from the aspect of radiation protection. Recently, interest has also been growing in ^{220}Rn with a half-life of 56 s, as a decay product of ^{224}Ra in thorium series. Uranium, thorium, and radium are ubiquitous

elements in natural materials such as rock and soil, implying that radon is always produced in them. A fraction of radon atoms generated in such materials has a possibility to be emanated into inter- or intra-particle pores. **Table 1** indicates the representative values of radon emanation fractions for mineral, rock, soil, mill tailing, and fly-ash samples.^{1, 2)} The radon emanation fraction is defined as the number of radon atoms emanated per number of radon atoms generated; other terms such as emanation coefficient and emanating power have also been used alternatively. This fraction must be in the range 0–1. Among mineral, rock, and soil samples, mineral and soil generally tend to have the lowest and highest radon emanation fractions, respectively, which could be explained

Table 1 Representative radon emanation fractions for various materials.

Material	Emanation fraction (–)	
	^{222}Rn ¹⁾	^{220}Rn ²⁾
Mineral	0.03	0.01
Rock	0.13	0.11
Soil	0.20	0.14
Mill tailing	0.17	NA
Fly ash	0.03	NA

Note: All emanation tests were not necessarily performed under the same environmental conditions.

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by the different solid structures because of the formation and weathering processes. However, even based on the current knowledge, it is often difficult to predict how or understand why a given sample exhibits large or small radon emanation.

Many experimental and numerical studies have been carried out to facilitate the understanding of radon emanation phenomenon. A few reviews summarized measured data,^{1, 3, 4)} but no reviews overviewed numerical modeling studies (except for one Japanese article, which was dedicated only to specific topics of recoil-based emanation⁵⁾). The present paper reviews the mechanisms and modeling approaches and progress of radon emanation as well as discusses environmental factors affecting radon emanation with typical calculated data. Note that, in this paper, the term *radon* refers to ²²²Rn and ²²⁰Rn and *radium* refers to ²²⁶Ra and ²²⁴Ra.

II MECHANISMS OF RADON EMANATION

Figure 1 shows the main pathways of radon emanation. Possible driving forces for emanation are recoil because of the alpha decay of radium and subsequent diffusion in solid. This diffusion process should be considered, only if alpha recoil allowed radon atoms to be settled not in pore air/water but in grains (points (C), (D), (E), (F), and (I) in **Fig. 1**). For the detection in an emanation test, radon in point (H) would be necessary to move to inter-particle space. Generally, the radon emanation fraction, E (–), can be classified into two or three components:⁴⁾

$$E = E_{\text{Recoil}} + E_{\text{Diffusion}} = E_{\text{Direct}} + E_{\text{Indirect}} + E_{\text{Diffusion}}, \quad (1)$$

where E_{Direct} is the direct-recoil fraction (points (A), (B), (G), and (H) in **Fig. 1**), E_{Indirect} is the indirect-recoil fraction (point (E)), $E_{\text{Diffusion}}$ is the solid-diffusion fraction (point (F)), and E_{Recoil} is the sum of E_{Direct} and E_{Indirect} .

1. Alpha recoil

The transformation of ²²⁶Ra or ²²⁴Ra by alpha decay creates certain energy, which can be assigned to an alpha particle and its decay product. Subsequently, the decay product (i.e., ²²²Rn or ²²⁰Rn) can recoil, and thereby be emitted in the opposite direction to the alpha particle. The energy imparted to radon, as exhibited in **Table 2**,⁶⁾ can contribute to emanation. This table also indicates the ranges of recoiling radon in materials, calculated by SRIM-2013.⁷⁾ Only a recoil energy of 86.26 keV for ²²²Rn or 103.4 keV for ²²⁰Rn has been used in previous emanation modeling studies, because of high intensity. Even

Fig. 1 Scheme of radon emanation phenomenon. Radon atoms in points (A), (B), (E), (F), (G), and (H) are regarded as being emanated and in points (C), (D), and (I) as not emanated. Radon in point (H) is emanated to the intra-particle pore but would be necessary to move to inter-particle space for detection. The arrow following the terminal point of recoil represents the diffusion in solid. All arrows are not to scale.

if other energy levels of 82.96 keV for ²²²Rn or 99.13 keV for ²²⁰Rn have been considered for the calculation, the results would barely change.

Alpha recoil not only is specific to the decay of radium isotopes but also accompanies the decay of any alpha emitter. For example, successive alpha recoils in uranium and thorium series have been investigated to argue formations of radiation damage and inhomogeneous distributions of members belonging to those series in minerals.^{8–10)}

2. Solid diffusion

Diffusion of radon in solid at room temperature has been thought to make no contribution to emanation ($E_{\text{Diffusion}} = 0$) because of substantially low diffusion coefficients of radon (**Table 3**).^{2, 4, 11–18)} The diffusion length, L (m), is expressed as $L = \sqrt{\frac{D}{\lambda}}$, where D is the diffusion coefficient ($\text{m}^2 \text{s}^{-1}$) and λ is the decay constant (s^{-1}). A diffusion coefficient of $10^{-24} \text{ m}^2 \text{s}^{-1}$ corresponds to a diffusion length of 0.7 nm for ²²²Rn and $9 \times$

Table 2 Kinetic energies assigned to alpha-recoiling radon atoms and their ranges in materials.

Material	Density (g cm^{-3})	²²² Rn		²²⁰ Rn	
		82.96 keV (5.55%)	86.26 keV (94.4%)	99.13 keV (5.06%)	103.4 keV (94.9%)
Air	1.205×10^{-3}	$68.0 \pm 10.3 \mu\text{m}$	$69.7 \pm 10.5 \mu\text{m}$	$75.8 \pm 11.3 \mu\text{m}$	$77.9 \pm 11.5 \mu\text{m}$
Water	1	$73.9 \pm 10.5 \text{ nm}$	$75.7 \pm 10.7 \text{ nm}$	$82.3 \pm 11.4 \text{ nm}$	$84.5 \pm 11.7 \text{ nm}$
Quartz	2.65	$33.1 \pm 6.1 \text{ nm}$	$33.9 \pm 6.2 \text{ nm}$	$36.9 \pm 6.7 \text{ nm}$	$37.9 \pm 6.9 \text{ nm}$
Zircon	4.65	$22.5 \pm 6.1 \text{ nm}$	$23.0 \pm 6.2 \text{ nm}$	$25.0 \pm 6.7 \text{ nm}$	$25.7 \pm 6.9 \text{ nm}$

Note: The projected recoil ranges and the longitudinal straggling (standard deviation) were calculated by SRIM-2013.⁷⁾ Values of energy and intensity in parentheses were taken from ICRP Publ. 107.⁶⁾

Table 3 Estimated diffusion coefficients of radon and corresponding diffusion lengths in solids at ambient temperature.

Solid	D ($\text{m}^2 \text{s}^{-1}$)	L (m)		Reference
		^{222}Rn	^{220}Rn	
General ionic crystal	$\leq 10^{-24}$	$\leq 7 \times 10^{-10}$	$\leq 9 \times 10^{-12}$	TANNER (1980) ⁴⁾
Mineral	$10^{-25} - 10^{-69}$	$2 \times (10^{-10} - 10^{-32})$	$3 \times (10^{-12} - 10^{-34})$	LEACH et al. (1982) ¹¹⁾
Witherite (BaCO_3)	1.6×10^{-22}	8.7×10^{-9}	1.1×10^{-10}	FLÜGGE and ZIMENS (1939) ¹²⁾
Potassium feldspar ^a	$\approx 10^{-31}$	$\approx 10^{-13}$	$\approx 10^{-15}$	TOKUMORI et al. (1992) ¹³⁾
Zircon (ZrSiO_4) ^a	$\approx 10^{-46}$	$\approx 10^{-20}$	$\approx 10^{-22}$	EAKIN et al. (2016) ¹⁴⁾
Coal fly ash	$(1.05 - 9.17) \times 10^{-21}$	$(2.24 - 6.61) \times 10^{-8}$	$(2.90 - 8.58) \times 10^{-10}$	MARAZIOTIS (1996), ^{15, 16)} BARTON and ZIEMER (1986), ¹⁷⁾ KALKWARF et al. (1985) ¹⁸⁾
For reference				
Air	1×10^{-5}	2	3×10^{-2}	IAEA (2013) ²⁾
Water	1×10^{-9}	2×10^{-2}	3×10^{-4}	IAEA (2013) ²⁾

^a Re-evaluated by the present authors, with extrapolation from high temperature to 20°C (see Fig. 2).

10^{-3} nm for ^{220}Rn . Short distances, as seen in Table 3, can lead us to a conclusion that in natural conditions, $E_{\text{Diffusion}}$ is negligibly smaller than E_{Recoil} in Eq. (1).

However, the situation is different when the temperature is above several hundred degrees Celsius. The diffusivity is temperature-dependent in nature; $E_{\text{Diffusion}}$ can be enhanced with increasing temperature. BAXTER¹⁹⁾ listed enormous data on diffusion coefficients of noble gases (especially helium and argon) in minerals, but no data were reported on radon. Figure 2 shows the dependence of D on temperature.^{13, 14)} The values of D_0 and E_a for ^{222}Rn , which the present authors analyzed using the Arrhenius equation, were found to be similar to those for helium, argon, and neon.¹⁹⁾ The D values extrapolated from high temperature to 20°C in Fig. 2 are listed in Table 3.

The diffusion might contribute to E_{Indirect} as well.⁴⁾ If an energetic recoiling radon atom crosses pores and breaks the surface of an adjacent solid, one might model the formation of a path in the solid. The depth of this path might be compatible to the range, reduced by the fraction of energy already expended. A fraction of embedded radon atoms could move by diffusion to eventually escape from the grain through the path. On the other hand, radiation damages are created by not only recoils but also alpha particles and fission fragments, which can be observed in common minerals, rocks, and soils. Such damages could influence not only E_{Indirect} but also $E_{\text{Diffusion}}$ if they are interconnected and linked to open surface, as discussed later.

III MODELING APPROACHES OF RADON EMANATION

To our knowledge, a German paper written by FLÜGGE and ZIMENS¹²⁾ was the first report about radon emanation modeling. Since then, this research theme had not been advanced for a while. The radon emanation modeling study was resumed after radon was recognized as a carcinogen. This section presents an overview of literatures published from 1980s to date.

1. Recoil-based emanation

As summarized in Table 4, many studies have theoretically addressed the movement of recoiling radon in solid and pore fluid (air and/or water).^{10, 20–37)} The movement and resulting emanation fraction can be computed using information on

Fig. 2 Diffusion coefficients (D) of ^{222}Rn in minerals as a function of temperature (T). The data were fitted using the Arrhenius equation, $D(T) = D_0 \exp(-E_a/RT)$, where D_0 is the frequency factor, E_a is the activation energy, R is the gas constant. D was obtained in the present study by analyzing original data (^{222}Rn emanation fractions) reported by EAKIN et al.¹⁴⁾ and TOKUMORI et al.¹³⁾ with the following equation:

$$E_{\text{Diffusion}} = \frac{6}{d} (1 - E_{\text{Recoil}}) \sqrt{\frac{D}{\lambda}}.$$
 Such equation is the asymptotic solution of the diffusion equation based on an assumption that E yielded at ambient temperature corresponded to E_{Recoil} at any temperature.^{15, 16)}

recoil ranges in substances or stopping powers as a function of kinetic energy of radon. For simplicity, most researches adopted simple geometric models as displayed in Fig. 3 and assumed uniform radium distribution from solid surface to a certain depth (T , defined in Fig. 4). Furthermore, the embedment of radon has been commonly supposed to occur whenever radon ejected from a grain reaches another solid surface. Referring to previous studies,^{38, 39)} only SEMKOW^{22, 23)}

Table 4 Overview of recoil-based radon emanation modeling studies.

Reference	Model (Fig. 3)	Main achievement
FLEISCHER (1983) ²⁰⁾	Single grain; Planar pore	E_{Direct} was calculated using the single-grain or planar-pore model with a given grain- or pore-size distribution. The effect of pore water on the emanation was also presented.
BOSSUS (1984) ²¹⁾	Planar pore	The planar-pore model with infinite pore size (i.e., no embedding effect of the recoiling radon) was addressed to derive a simple formula for E_{Direct} . The calculation of $E_{\text{Diffusion}}$ was also presented.
SEMKOW and PAREKH (1990) ¹⁰⁾	Single grain	Successive alpha recoils in uranium series (from ^{238}U to ^{222}Rn) were simulated with an assumption of the surface or whole-volume distribution of ^{238}U in the single grain, indicating the resulting distribution of radium. E_{Direct} was then calculated using this radium distribution in the grain. An example of E_{Direct} calculated as a function of surface porosity was represented for different radium-distribution conditions. In addition, a brief description was provided about the basis of the multiple-grain model.
SEMKOW (1990) ²²⁾	Single grain; Cylindrical pore; Spherical pore; Uneven surface	First, the analytical solution of E_{Direct} (in fact, just ejection probabilities from the grain into another solid surface) was provided for a spherical or cylindrical grain or for spherical or cylindrical pore. The effect of surface shape was then examined to discuss radon emanation from a variety of uneven surfaces. Furthermore, other issues were also argued, such as the effects of radon embedding (and its threshold energy), pore water, grain size, and solid edge.
SEMKOW (1991) ²³⁾	Uneven surface	The fractal model of radon emanation from solid surface was developed, and the relation was derived between E_{Direct} and specific surface area. The result suggested that emanation measurements can be used to determine the fractal dimensions of surface on the scale from tens to hundreds of nanometers.
MORAWSKA and PHILLIP (1993) ²⁴⁾	Single grain; Cylindrical pore	The single-grain model or the combination model of single grain and cylindrical pore were employed, considering the surface or homogeneous radium distribution in the grain. The combination model was developed to study each contribution of outer and inner surface to E_{Direct} . The result explained the importance of the internal structure (inner pore network), which may be related to porosity, pore size, and pore water. Furthermore, the calculation of $E_{\text{Diffusion}}$ was presented.
SUN and FURBISH (1995) ²⁵⁾ ; SUN (1995) ²⁶⁾	Uneven surface	The recoil surface area of porous media was determined by quantifying the fractal of the simulated synthetic porous media. The effects of porosity and moisture content were also indicated.
SASAKI et al. (2004) ²⁷⁾	Multiple grains	The multiple-grain model, where the number of grains in a single unit was only four in this study, was developed for the first time. The effect of pore water on E_{Direct} was examined for different grain size ranges. It was found to be significantly given only for grains with less than a few hundred micrometers.
SASAKI et al. (2004) ²⁸⁾	Cylindrical pore; Planar pore	Two pore models (pore size 10 nm; filled with water) were used for the calculation of E_{Direct} to simulate the so-called nanopore proposed by RAMA and MOORE. ⁶¹⁾ These models could partially explain a role of nanopore, but were not able to yield E_{Direct} higher than 0.5, which had sometimes been observed experimentally. The result showed that the cylindrical-planar-pore model gives higher E_{Direct} than the cylindrical-pore model because of less embedding of radon.
SASAKI et al. (2005) ²⁹⁾	Adhering grain	Assuming that very fine grains on the base grain can exclusively include radium and emanate radon, the authors demonstrated the calculated E_{Direct} value higher than 0.5, which had sometimes been observed experimentally.
BARILLON et al. (2005) ³⁰⁾	Planar pore	The effects of radium distribution, moisture content, and pore size on E_{Direct} were illustrated in detail.
SASAKI et al. (2007) ³¹⁾	Single grain	The emanation calculations of E_{Direct} were performed with a cubic-grain model and a spherical-grain model. The spherical-grain model gave higher E_{Direct} by 35% for a grain size of 0.1 μm and by 15% for 1.5 μm .
SASAKI and GUNJI (2008) ³²⁾	Planar pore	E_{Direct} was calculated considering two types of travels of recoiling radon, i.e., straight and zigzag. The E_{Direct} values were found to be different between the two movements, when pore size was reduced. However, correction because of this difference may be unnecessary for other general emanation models that supposed the straight track.
ADLER and PERRIER (2009) ³³⁾	Multiple grains	The complex geometry with randomly packed grains and unsaturated porous media was implemented for the estimation of E_{Direct} .
SAKODA et al. (2010) ^{34, 35)}	Single grain; Multiple grains	The effects of grain size, moisture content, and grain-packing structure on E_{Direct} were shown for different radium-distribution conditions. It was demonstrated that the radon emanation fraction is not necessarily inversely proportional to grain size.
SAKODA and ISHIMORI (2014) ³⁶⁾	Multiple grains	The temperature between -20 and 40°C was found to have little impact on E_{Direct} , regardless of grain size.
STAJIC and NIKEZIC (2014) ³⁷⁾	Single grain; Multiple grains	The application of Monte Carlo simulation to the calculation of E_{Direct} was validated by comparison with the analytical solution. The effects of grain-packing structure and solid density on the emanation were shown.

Fig. 3 Grain and pore models used for calculation of radon emanation fraction.

introduced threshold energies for the embedding; the embedding probability ranges from $\sim 10^{-5}$ at eV energies to ~ 0.6 at 3 keV, which is equal to 1 at 5 keV and above. The above modeling design can simplify the complicated phenomenon by nature, as seen in **Fig. 4**.

Radon emanation modeling techniques have been advanced with time; here, we present an overview of these techniques in chronological order. Earlier studies dealt with simple geometric models (cylindrical-, spherical-, or planar-pore models as well as single-grain model, shown in **Fig. 3**), wherein the *ejection* probability of radon from a grain was determined as a function of diameter (d) or radium-existing thickness (T).^{10, 20–22, 24} Note that ejection probability is not necessarily equivalent to E_{Direct} because the concept of embedding effect was not considered in the model. That is, this corresponds with a theoretically maximum value of E_{Direct} for a given T . To evaluate the impact of pore water on E_{Direct} , FLEISCHER²⁰ and SEMKOW²² attempted to incorporate the embedding effect into emanation models. These studies clearly represented that the presence of water enhances emanation, while mathematical solutions of E_{Direct} were yielded only for two conditions that the pore space is filled by either air or water.

Thereafter, more complex geometric models were implemented, such as the adhering-grain model and uneven-surface model, shown in **Fig. 3**.^{22, 23, 25–33} SEMKOW,²³ SUN,²⁵ and SUN

and FURBISH²⁶) addressed the emanation computation for simulated solid surfaces with self-similarity to determine surface areas and demonstrate that E_{Direct} is increased with increasing porosity or moisture content. BARILLON et al.³⁰) worked on the prediction of E_{Direct} as a function of moisture content by using the planar-pore model into which a pore size distribution was incorporated; they concluded that the calculation successfully illustrated the results of radon emanation fractions measured for uranium mill tailing samples. A series of studies by SASAKI et al.^{28, 29, 31, 32}) were carried out with a variety of motivations and purposes. One of the most notable achievements of their studies was the development of an adhering-grain model that can provide a radon emanation fraction of more than 0.5,²⁹) which can be recognized as the expansion of uneven-surface model. When this model is employed, it is necessary to know the ratio of activity (not activity concentration) of radium in adhering fine grains to that in the matrix (base grain). Even with this model, it seems difficult to provide a radon emanation fraction of more than 0.5, if the matrix accounts for the most part of the total radium activity.

In more recent works, the movement of radon in solid and pore fluid was tracked by Monte Carlo simulation by using information on stopping powers of individual materials of interest as a function of kinetic energy of radon. The simulation can work for any complex geometry of grains and pores, given accurate stopping powers. The present authors^{34–36}) and STAJIC and NIKEZIC³⁷) have computed radon emanation fractions by Monte Carlo simulation as a function of grain size in a certain regular array structure of spherical grains with the same diameter, which is called the multiple-grain model (**Fig. 3**). To estimate stopping powers of any materials to radon as well as ranges, previous studies have often used a known code, SRIM.⁷)

2. Diffusion-based emanation

The general mathematical analysis of the fractional gas loss for other stable inert gases, as explained by FULDA and LIPPOLT,⁴⁰) is not applicable to $E_{\text{Diffusion}}$ for radioactive nuclides, including radon. MARAZIOTIS^{15, 16}) and RIBEIRO^{41–43}) addressed to derive general solutions for the diffusion of a radionuclide

Fig. 4 Simplified designs of radon emanation phenomenon. (a) Single-grain model or planar-pore model without the opposite grain surface. (b) and (c) Reduced size of pore ($d_f R_g / R_f$) filled with fluid (either air or water).^{20, 30}) The parameter T is the radium-existing depth from the surface, d_f is the actual pore size, R_g and R_f are the ranges of radon in grain and pore fluid, respectively. The simple pore-size reduction is not possible if the pore is composed of the mixture of air and water.

produced by the decay of an immobilized parent nuclide in a spherical, homogeneous, and isotropic grain. In particular, the motivation of MARAZIOTIS^{15, 16} was directed to radon, and a simple numerical expression of $E_{\text{Diffusion}}$ for a sphere was presented based on the diffusion theory (Fig. 2). Unlike the recoil-based emanation modeling, the assumption of T has not been introduced to the diffusion-based modeling. The diffusion calculation with such assumption should be performed in the future. At the same time, there is another issue on how to numerically treat the diffusion of radon near solid surface. A unified model of the recoil- and diffusion-based emanation could be interesting and challenging.

IV ENVIRONMENTAL FACTORS AFFECTING RADON EMANATION FRACTION

Figures 5–7 show the calculated results of E_{Direct} as a function of grain size or pore size, assuming various moisture contents or radium thicknesses from solid surfaces. The calculations were based on the single-spherical-grain model, the planar-pore model and the multiple-spherical-grain model (Fig. 3). Only for the single-spherical-grain model, $E_{\text{Diffusion}}$ was also determined with various diffusion coefficients (Fig. 5). Moreover, Fig. 8 shows the ejection probability of radon as a function of T/R_g . The following arguments are based on those data.

1. Radium distribution

The model calculations revealed that the radium distribution plays a very important role in determining the ejection probability and emanation fraction of radon from the birth grain. Obviously, radium existing at a shallower position can promote radon emanation. This may be consistent with experimental findings about the magnitude of E : soil $\approx$ mill tailing $>$ rock $>$ fly ash $\approx$ mineral.¹⁾ Intact minerals are expected to uniformly include radium in solid, leading to lower radon emanation fractions (Table 1). In soils, in contrast, more radium is distributed near solid surface because of factors such as weathering.^{44–46)} Quantitative and precise information on the location of radium atoms in grains would improve emanation modeling, which, however, is quite difficult to be acquired even using cutting-edge techniques.

2. Moisture content

A number of experimental studies have examined the effect of pore water on radon emanation.¹⁾ The modeling studies with a focus on E_{Direct} supported the interpretation of empirical data that indicated the elevation of radon emanation with increasing moisture content, as follows. The stopping power of water is much greater than that of air (see the ranges shown in Table 2), which makes recoiling radon very easily settled in pore space before reaching and being embedded into another solid surface. Table 5 indicates the ratios of ^{222}Rn emanation fractions in moist conditions to those in dry conditions. The calculated ratios covered the measured results. This water effect occurs, in theory, under the condition where air-pore size is not large enough to absorb residual energy of ejected recoiling radon.

Fig. 5 Radon emanation fractions as a function of diameter (d) in the single-spherical-grain model. The solid curves represent E_{Direct} for different radium distributions (T), i.e., uniform in or surficial on the grain. E_{Direct} was calculated using

$$E_{\text{Direct}} = \frac{3R_g}{2d} - \frac{1}{16} \left(\frac{2R_g}{d} \right)^3 \text{ for } T = d/2 \text{ (uniform radium)}^{12, 20)}$$

and Eqs. 4 (a)–(d) of SEMKOW²²⁾ for the others (surficial radium). The values of R_g for ^{222}Rn and ^{220}Rn were set to be 34 and 38 nm, respectively, assuming a common mineral of quartz (SiO_2) (Table 1). The dashed curves represent $E_{\text{Diffusion}}$ for various D ($\text{m}^2 \text{s}^{-1}$), which was calculated, in the same manner as Fig. 2, using the equation^{15, 16)}

$$E_{\text{Diffusion}} = \frac{6}{d} (1 - E_{\text{Recoil}}) \sqrt{\frac{D}{\lambda}}; E_{\text{Recoil}} = 0 \text{ was assumed.}$$

3. Grain size

The modeling studies have also examined the impact of grain size on radon emanation. Both single-grain and multiple-grain models were implemented to simulate more practical grain-packing structures. The two models appeared to result in different trends of E_{Direct} (Figs. 5 and 7). This difference can be understood from both inter-particle pore size and embedding effect. With the increase in pore size, grain size also increases in the multiple-grain model,^{34, 35)} but no concept of inter-particle pore (and its size) is incorporated into the

Fig. 6 ^{222}Rn emanation fraction as a function of pore size in the planar-pore model. The calculation was performed using Eqs. 7–9 of BARILLON³⁰⁾; pore space is assumed to be completely filled with either air or water. Three kinds of ^{226}Ra thicknesses from the solid surface were set: $T=0$, R_g , and $2R_g$.

Fig. 7 Radon emanation fraction as a function of diameter in the multiple-spherical-grain model. The computation was carried out by Monte Carlo simulation under various moisture contents in pore (0–100%).³⁵⁾ The radium distribution from the solid surface was assumed to be $T=R_g$.

single-grain model. On the other hand, experimental studies also represented approximately two types of trends of radon emanation fractions, i.e., inversely proportional to or stable independently of grain size.¹⁾ The measured trends have been

Fig. 8 Ejection probability of radon as a function of the ratio of radium thickness from solid surface to recoil range of radon in solid. *Ejection* is defined as a radon emanation fraction calculated without embedding effect **Fig. 4 (a)**. That is, this can be given by $\frac{R_g}{4T}$, which is a possible maximum value of predicted E_{Direct} and is equivalent to E_{Direct} in the single-grain model.^{30, 35)}

Table 5 Impacts of pore water and temperature on ^{222}Rn emanation fraction.

Factor	Ratio of ^{222}Rn emanation fractions	
	Calculation (E_{Direct}) ^a	Experiment ^b
Water (Wet/Dry)	74 (at $d=1\ \mu\text{m}$)	4.1 ± 3.8
	5.7 (at $d=10\ \mu\text{m}$)	
	1.0 (at $d=100$ and $1000\ \mu\text{m}$)	
Temperature (40°C/0°C)	0.9–1.0 (at $d=1$ – $1000\ \mu\text{m}$)	1.7 ± 0.9

^a Water: taken from radon emanation fractions at 0% and 100% in **Fig. 7**. Temperature: taken from **Fig. 4** of SAKODA and ISHIMORI.³⁶⁾

^b Water: averaged with data shown in **Fig. 5** of SAKODA et al.¹⁾ Temperature: averaged with data shown in **Fig. 4** of SAKODA et al.³⁶⁾

interpreted with the calculated data on a case-by-case basis, according to specific information and test conditions. Here, it is stated that if the chemical components are not guaranteed to be the same among different grain-size classifications, the grain-size dependence must be carefully examined for a mixture sample as soil.⁴⁷⁾ In addition, it should be noted that a part of earlier experiments with soil samples might measure not separate (individual) but aggregate grain sizes. The multiple-grain model could be used to express the structure of an aggregate grain.

4. Temperature

It can be predicted that if radon emanation tests are

performed using the same sample in a range of room temperature plus or minus a few tens of degrees Celsius, the result would be almost independent of temperature. This is because diffusion coefficients of radon in solid (as well as stopping powers) would not change in such temperature range. As seen from **Table 5**, in fact, with the decrease in air density, which is equivalent to the increase in temperature, E_{Direct} was slightly low because of the embedding effect.³⁶⁾ Nevertheless, several experiments showed that ^{222}Rn emanation fractions were proportionally increased with increasing temperature (summarized by the present authors³⁶⁾; the emanation fractions at 40°C were on average 1.7 times higher than those at 0°C (**Table 5**). Regarding this issue, MESLIN et al.⁴⁸⁾ pointed that the actually measured data in those experiments were *apparent* radon emanation fractions, since no adsorption correction onto sample grains was carried out. In particular, for dry samples, radon emanation fractions were likely to be underestimated with decreasing temperature. With this viewpoint, the discrepancy between calculated and measured data may be qualitatively understood, and then the methodology for radon emanation tests must be improved.

As temperature was increased by several hundred degrees Celsius, $E_{\text{Diffusion}}$ was elevated by the solid diffusion process (**Fig. 2**). In the field of geoscience, heating approaches are commonly used to evaluate the diffusivity of noble gases (in particular, helium and argon) for the characterization of mineral samples.^{19, 49)} It was sometimes also used for material analysis (called emanation thermal analysis).⁵⁰⁾ Naturally, high radon emanation fractions ($E_{\text{Diffusion}}$) can be observed only during heating. From **Fig. 5**, it is obvious that for ^{222}Rn , D should be at least 10^{-22} to $10^{-20} \text{ m}^2 \text{ s}^{-1}$ for silt and 10^{-20} to $10^{-18} \text{ m}^2 \text{ s}^{-1}$ for sand to yield significant values of $E_{\text{Diffusion}}$ in comparison to E_{Direct} . In the case of ^{220}Rn , large D is required because of its short half-life: 10^{-18} to $10^{-16} \text{ m}^2 \text{ s}^{-1}$ for silt and 10^{-16} to $10^{-14} \text{ m}^2 \text{ s}^{-1}$ for sand. This is similar to the results of BOSSUS,²¹⁾ where E_{Direct} and $E_{\text{Diffusion}}$ for ^{222}Rn and ^{220}Rn were calculated using a planar-pore model. On the other hand, several studies demonstrated a decrease in ^{222}Rn emanation fractions after heating (up to 1,000°C) and subsequent cooling.^{51–55)} This result may be ascribed to closure of intra- and inter-particle pores and annealing of lattice damages. In terms of radiation protection, this finding could be applied to building materials to suppress indoor radon concentration. For this purpose, however, further investigations are necessary to determine the recovery of radon emanation power over time. Its implication was reported earlier.⁵⁵⁾

5. Inner network in solid

Mineral grains have been considered to include an inner network of paths that are created by nature, radiation damage, or cracks in crystal lattice.^{8, 56, 57)} If this network is opened to atmosphere, all three components of radon emanation, i.e., E_{Direct} , E_{Indirect} , and $E_{\text{Diffusion}}$, can be affected. MORAWSKA and PHILLIPS²⁴⁾ illustrated a mathematical formulation to calculate E_{Direct} by considering inner porosities and inner pore diameters in grains, which implied the effect of inner paths. Rock-core deformation tests demonstrated that the collapse of original

micro-cracks first declined radon emanation fractions, before they were enhanced by the development connection of isolated micro-cracks.^{58–60)}

On the other hand, RAMA and MOORE⁶¹⁾ postulated the existence of interconnected network, so-called *nanopore*, to explain commonly observed values of radon emanation fractions in minerals and rocks, which were much higher than expected (for example, see the curve assigned with $T = d/2$ in **Fig. 5**). Radon was supposed to recoil into the nanopore and subsequently diffuse to the atmosphere. Later, KRISHNASWAMI and SEIDEMANN⁶²⁾ alternatively advocated that not nanopore but radium concentrated in thin layers on the solid surface, in secondary minerals, or in U-Th accessory minerals is responsible for radon emanation. This finding was based on their observation that the emanation fractions of noble gasses ^{39}Ar and ^{37}Ar , created by the reactions $^{39}\text{K}(n, p)^{39}\text{Ar}$ and $^{40}\text{Ca}(n, \alpha)^{37}\text{Ar}$ following fast neutron irradiation, were much lower than the ^{222}Rn emanation fractions from mineral and rock samples. The result seemed unreasonable under the assumption that radium is uniformly distributed like other major elements. NAZAROFF et al.⁶³⁾ mentioned that these two hypotheses were not mutually exclusive, and both are substantiated by experimental data.

Radiation damage has not yet been incorporated into radon emanation modeling, which could be a factor influencing the migration process in solid. In general, origins of radiation damage in U-Th-rich minerals (e.g., zircon) are alpha-recoiling atoms (mostly major factor), alpha particles, and nuclear fission fragments. In a previous work, the alpha-recoil damage zone in zircon was modeled as a cylinder with a length of 25 nm and a diameter of 2.5 nm.⁸⁾ At a low level of radiation damage, alpha-recoil damage zone behaves similar to a *sink*, lowering overall diffusivity of noble gas with its resulting high tortuosity. As the level of radioactivity rises and an increasing fraction of crystal structure is damaged, the damaged zones begin to be interconnected. When the damaged channels are connected through the solid, the state of percolation is established.

MALCZEWSKI and DZIURWICZ⁶⁴⁾ recently measured radon emanation fractions as a function of alpha dose in metamict minerals (oxides, silicates, and phosphates). The ^{222}Rn emanation fractions generally decreased with increasing alpha dose, which could imply the function of radiation damage as a sink. In contrast, the ^{220}Rn emanation fractions were apparently independent of alpha dose. The conclusion referred to the necessity to develop a comprehensive model of ^{222}Rn and ^{220}Rn emanation considering their transport through the inner path network in minerals.

6. Others

Emanation fractions of ^{222}Rn from mineral and rock samples in vacuum and ambient pressure (helium atmosphere) were measured.⁶¹⁾ The vacuum condition lowered the emanation fractions by 45% to 79% in comparison with those in the ambient pressure condition, which might be attributed to the embedding of more recoiling ^{222}Rn in vacuum. The aforementioned models may possibly replicate this result,

while no computational studies are reported so far.

In contrast, following factors have been studied numerically, but not yet validated experimentally. The multiple-grain models with different porosities were employed in the calculation of E_{Direct} ; porosity is 48% for simple cubic packing and 26% for face-centered cubic packing.^{34, 35, 37)} The tight-packing structure with low porosity has small pore sizes on average and thus decreases the emanation fraction only in a certain range of grain sizes (i.e., slope zone before reaching the saturated emanation value at a moisture content of 0% shown in Fig. 7). Moreover, E_{Direct} was calculated in relation to solid density.³⁷⁾ Eight major minerals were supposed in a wide range of solid densities (e.g., 2.65 g cm⁻³ for quartz and 4.65 g cm⁻³ for zircon). The result was evident, as expected, to depend on ejection probabilities of radon from the birth grain, because of the difference of recoil ranges in the samples. The range was short in heavy minerals, resulting in small E_{Direct} . The density could be one of the factors affecting radon emanation to some extent, but its importance cannot be experimentally validated because different minerals have own physical and chemical characteristics (other than density) that would strongly influence radon emanation.

V CONCLUSIONS

Many radon emanation measurements have been conducted in terms of radiation protection, with most emanation modeling studies focusing on E_{Direct} , which was considered to play a dominant role in the natural environment. A series of calculation results of E_{Direct} provided interpretations for existing experimental results (e.g., water effect), or implications on the impacts of environmental factors (e.g., radium distribution), which were difficult to be controlled as parameters in emanation measurements. A model that can simultaneously account for E_{Indirect} and $E_{\text{Diffusion}}$ as well as E_{Direct} is desired to be developed in the future, considering the transport of ²²²Rn and ²²⁰Rn through the inner path network in minerals. In this new model, radium distribution would still be a key factor governing radon emanation, and thereby more experimental evaluation should be conducted for the sake of more realistic modeling.

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