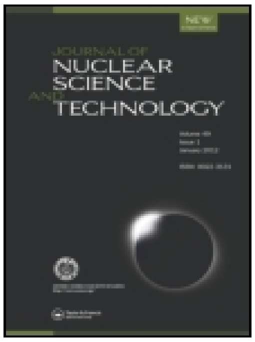

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ORIGINAL PAPER

Mathematical Modeling of Radon Emanation

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It is well known that radon emanation is affected by moisture. The models which explain radon emanation can be considered to come in three types. One is to predict the maximum possibility of radon emanation assuming uniform distribution of radium in a solid grain independent of moisture. The other two are to explain moisture effect on radon emanation, but their aims are different from each other. The first of them aims to demonstrate a theory explaining microscopic phenomena of radon emanation employing observed macroscopic phenomena of radon emanation under moist condition. The second aims to predict the leveling-off value of radon emanation under moist conditions employing an observed data of radon emanation under dry condition. The present paper develops a close-packed spherical grain model which belongs to the last type of model just mentioned. This model is oriented to contribute to the safety assessment of uranium-bearing waste disposal. Uranium-bearing waste generates radon as one of the uranium decay chain daughter nuclides. They will build up until about 200,000 years after disposal. At that far future, rainwater may well come into contact with the disposed waste and increase radon emanation. Therefore, leveling-off radon emanation has to be employed for the safety assessment. The model by the present paper is best oriented for this purpose.

KEYWORDS: radon, emanation coefficient, uranium ore, uranium-bearing waste, NORM, radon recoil, moisture effect on radon emanation, moisture saturation, close-packed spherical grain model

I. Introduction

1. Purpose

Radon is located in decay chains starting from uranium or thorium. It comes out as a daughter of radium. Radon exists in all environments. It gives the largest radiation exposure to human being among the naturally existing radiation sources. That is the reason why many research institutions study about radon. Their fields of study cover large area like the radon in groundwater, atmosphere or building.

An established theory gives explanation[1] of radon generation in solid substances. According to this theory, some of the radon atoms born in the vicinity of grain edge of solid substances get recoil energy high enough to jump out of that grain. The recoil energy comes from α-decay of its parent nuclide ‘radium’. There is pore volume between adjacent grains where some part of it is occupied with air or water. The radon atoms that have jumped out of the grains lose their kinetic energy in air or water finally stopping in the pore volume or reaching a grain that exists in its trajectory. The radon atoms that stopped in air or water diffuse through the pore volume and out of the substance, then finally becoming the source of radiation exposure. Diffusion of radon atoms within a grain of solid substance usually can be neglected.

In order to give quantitative explanation of the radon atom behavior mentioned above, a parameter called ‘emanation coefficient’ is defined as follows.

- Define A (Bq/g) as total radon atoms that are born in grains of solid substance.
- Define B (Bq/g) as those radon atoms that are born in grains of solid substance, jump out of the grains into pore volume regions between adjacent grains and remain in air or water in these pores, and not embedded in another grain traveling too far.

Then, emanation coefficient is defined by B/A. Hereafter, the word ‘emanation’ has a special meaning defined here. To achieve precise description, ‘emit’ or ‘emission’ is used instead of ‘emanate’ or ‘emanation’, when the destiny of radon after jumping out of the grains need not be considered.

Some papers give measured radon emanation coefficients of various soils and uranium ores. They state emanation coefficient is affected by moisture content. Among them, the one[2] from Bureau of Mines, U.S.A. gives clear illustrations how the moisture content affects the emanation factor. One of the illustrations is shown in **Fig. 1**. With increase of moisture, emanation coefficient shows steep increase. After that, it levels off at a certain value. When developing a mathematical model, these observed data of radon emanation coefficients are quite helpful.

The purpose of this paper is to develop a close-packed spherical grain model and study quantitatively if it can well predict the leveling-off value of radon emanation coefficient.

2. Quick Overview of Models Explaining Radon Emanation

There are two types of explanation for radon emanation phenomenon. One mainly consists of mathematical calculation employing geometrical models. The other mainly consists of qualitative interpretation of experimental results with regard to radon emanation. It has been a worldwide question why radon emanation is so large. Therefore, the purpose of the explanations until today is oriented to account for large radon emanation. Different from these explanations, the purpose of the proposed model in this paper is to predict the lev-

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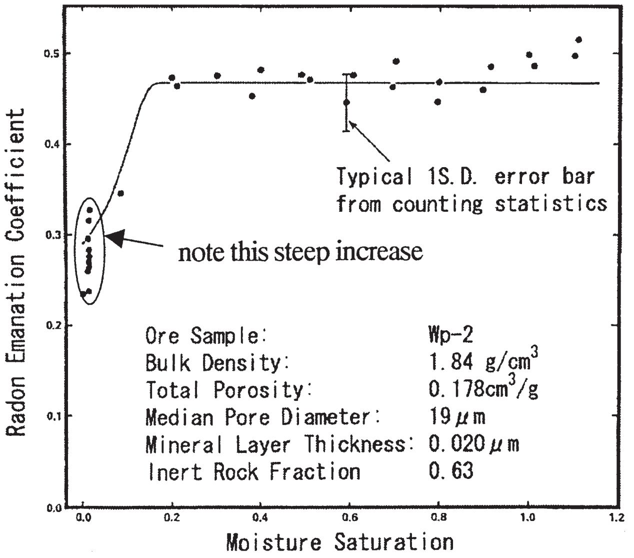

Fig. 1 Comparison of measured emanation coefficients with calculated emanation *vs.* moisture curve for ore sample WP-2 (Reproduced from Ref. 2))

eling-off value of radon emanation coefficient by mathematical calculation employing geometrical model.

(1) Mathematical Model

The models which explain radon emanation mathematically can be classified into three types. One is to predict the maximum possible value of radon emanation assuming uniform distribution of radium in a solid grain independent of moisture. In other words, this model deals with radon behavior from its birthplace until its departure place out of the surface of the grain, and not after that. Therefore it has nothing to do with the moisture and other adjacent grains surrounding the grain from where radon is emitted. A typical model is shown by Bossus.[3)] According to it, the maximum possible value of radon emanation coefficient is 0.25 for radon near the edge of the grain. But, Nazaroff[1)] states that even a spherical grain with a diameter so small as 20 μm has only 0.005 of possible value of radon emanation coefficient as a whole. This means radon emanation depends much on the radium distribution in the grain. As one application of this type of model, it was employed in revealing the process of rock formation.

The second type of model is to demonstrate a theory explaining microscopic phenomena of radon emanation, employing observed macroscopic phenomena of radon emanation under moist conditions. This model deals with radon behavior from its birthplace until its halt place outside the grain. Therefore it has much to do with the moisture surrounding the grain. One typical model is shown by Thamer *et al.*[2)] According to it, pore regions are dealt as parallel cylinders. Radium is assumed to be uniformly distributed on the surface of the cylinder. The reason why cylindrical model was adopted is to explain relatively large value of radon emanation coefficient. If spherical model is tried, the grain size has to be unrealistically small like 0.056 μm for emanation coefficient 0.5. By the cylindrical model, the thickness of radium and a normalization factor were adjusted using measured radon emanation coefficient (macroscopic phenomena) to explain the variation and the magnitude of emanation coefficient with moisture increase. The feature of this model is that the radium thickness and the normalization factor become nearly constant for several uranium ores and the established theory[1)] about microscopic behavior of radon is adopted to calculate radon emanation.

The purpose of mathematical models developed until today in the world mentioned above as the first and second types of model is mainly to account for the observed large emanation coefficients as have been considered to be unlikely and the adequacy of the assumed radon behaviors in the explanation such as being embedded in other adjacent grain. Different from these models, a model to predict the maximum or leveling-off radon emanation coefficient under moist conditions has been newly developed as the third type of model by the authors aiming at being employed in the safety assessment for uranium-bearing wastes disposal and the development of technology to suppress radon emanation. This model consists of a close-packed spherical grain. This model deals with radon behavior from its emitted place on the surface until its halt place outside the grain, and uses radon emanation coefficient under dry condition as a particular observed data to predict its leveling-off value under moist conditions. It does not matter whether the magnitude of radon emanation coefficient is large or not. It is how to predict the maximum or leveling-off value of radon emanation coefficient that counts.

Figure 2 illustrates the targeted research spectrum by each type of the model. In the first type of model, the radon born in the grain can come out of the grain only if the distance from the surface of the grain to where radon has been born is less than the recoil range in the grain for each travel direction. The ratio of radon atoms that can come out of the grain to those that cannot depends on the vertical depth of radon birthplace from the surface. This ratio can be calculated assuming uniform radium distribution in the grain. But, the model does not deal with radon behavior after it has left the grain. Therefore, those radon atoms that might be embedded in other adjacent grains are not accounted for and the calculated ratio becomes maximum possible emanation coefficient.

In the second type of model, radium layer and water film are assumed to exist around the cylindrical pore. The radon atoms born in the radium layer travels toward inside the ore, radium layer or water film. Those radon atoms toward inside the ore or radium layer never come out of the grain. Some of other radon atoms toward water film may be stopped in the radium layer, the water film or the pore, but the rest of them travels so far ending in embedded in the grain on the other side. The ratio of radon atoms that can come out of the grain to those that cannot depends on the geometry and can be calculated accounting for whole spectrum radon behavior just mentioned. But, assuming isotropic radon travel from its birthplace, this geometry never leads to emanation coefficient larger than 0.5. Therefore normalization factor was introduced to account for the magnitude of observed high radon emanation coefficients. In addition, the variation of radon emanation coefficient with water content was simulated

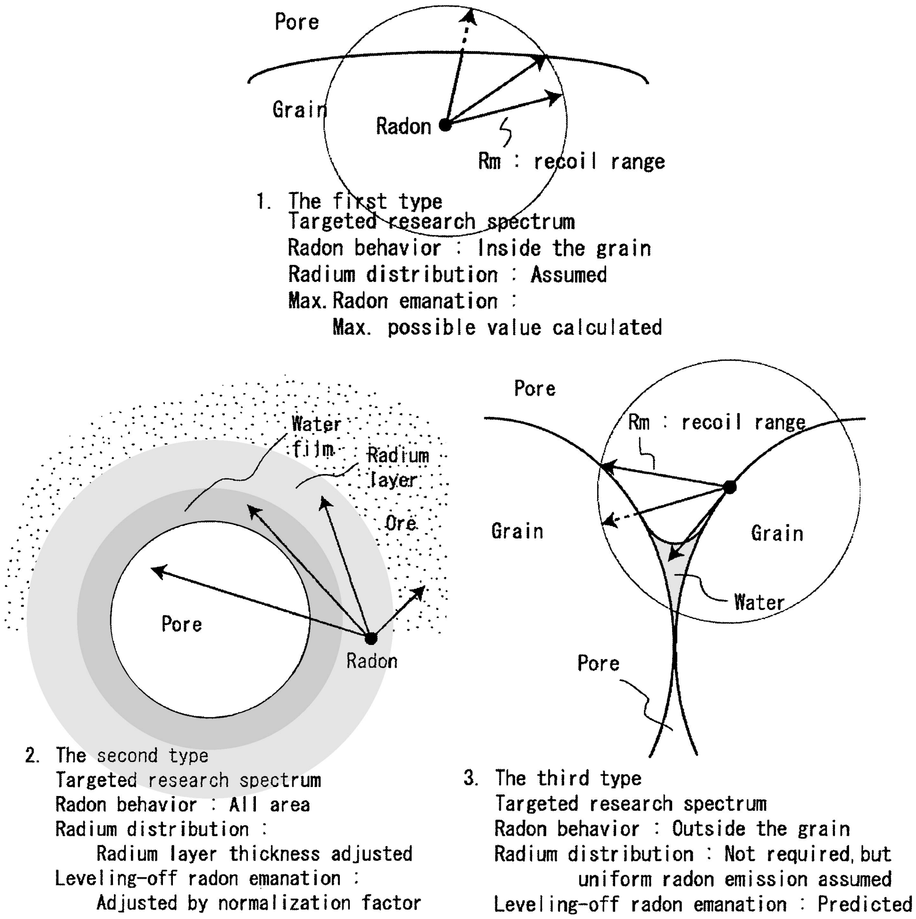

Fig. 2 Targeted research spectrum of each theoretical model for radon emanation

by adjusting the thickness of radium layer. Different from the first type of model, this model covers whole spectrum of radon behavior to reach emanation coefficient.

In the third type of model proposed in this paper, it does not matter how the radon atoms reached the grain surface from inside the grain. It deals with the radon behavior after leaving the surface of the grain. If there is not water in the travel direction of radon atoms, recoil range of radon in air is by far larger than that in water and some of the radon atoms may crash into the other adjacent grain to be embedded in it. On the other hand, if there is water in the pore, more radon atoms will halt in the pore, because small range of radon recoil in water makes radon crash decrease. The ratio of radon atoms that halt in the pore for these two cases can be calculated from the geometry. Combining this ratio with the observed radon emanation coefficient at dry condition leads to the leveling-off value at wet condition. Different from the second type of model, this model does not need normalization factor or radium layer adjustment.

(2) Qualitative Explanation

Qualitative explanation of high radon emanation in the world until today has been oriented to account for grain structure and radium distribution within the grain. As mentioned earlier, if try to explain high radon emanation employing spherical model, the grain size has to be quite small. In order to fill the gap between actual grain size and theoretical one, pore size of nanometer order within grain and concentrated radium along grain boundaries or in accessory mineral assemblages in crevices are proposed to account for high radon emanation.[1]

II. Mathematical Modeling of Close-Packed Spherical Grain Model

Uranium or radium distribution in a substance depends on the formation process of that substance, whichever natural or artificial. This means radon emanation varies from one substance to another. Therefore, at least single measured data of radon emanation has to be needed to grasp the property of the substance. Then using this data, there must be some method by which it is possible to predict radon emanation behavior with moisture saturation. Different from the first or second type of model mentioned above, the third type makes it come true by measuring the particular radon emanation coefficient under dry condition (zero moisture saturation). This data includes the information on the amount of radon that has left the grain of the substance, not the one on the amount of radon that is going to leave the substance from inside of it. It already includes the result of escaping process of radon out of the substance. Therefore there is no need to calculate the quantity of radon leaving the substance and the thickness of radium layer. Also it already includes the magnitude of radon emanation. Therefore there is no need for normalization. Then in the next step, the information necessary for the prediction of the destiny of radon that has left the substance are water thickness and grain geometry around radon, because radon is slowed down as it moves in the water and both the water thickness and grain geometry determines if radon stops in the water or crash into another grain and embedded in it that stands in the trajectory of the radon. The radon that has stopped in the water then diffuses finally into the air in the pore space.

Before describing this model, basic parameter values are defined for preparation according to Nazaroff[1] and the radon behavior in pore region is overviewed.

(1) Definition of Basic Parameter Values for ^{222}Rn

- Recoil range for common minerals 0.02–0.07 μm (use 0.02 μm in the present paper)
- Recoil range for water 0.1 μm
- Recoil range in air 63 μm

The present model is intended to deal with uranium-bearing wastes. Therefore ^{222}Rn is considered here.

(2) Radon Behavior in Pore Region

In order to overview radon behavior in pore region, geometrical relationship between water and grain of substance has to be grasped. **Figure 3**[4] shows water and gas in soil. Water grows as shown in **Fig. 4**. The front surface of the growing water may well be hollow-shaped. If the grain comes from sludge generated as one of the uranium-bearing wastes, the same relationship with water and gas would hold because its shape can be considered to somewhat resemble soil grains. Hereafter, the word 'grain' used in this paper means sludge grain, but it is the geometrical shape that counts, not the property of substance, because only radon atoms that have left the substance are targeted to be built into the close-packed spherical grain model, not those stopping inside the substance. Therefore it is apparent that this model can be applied to any substance that assumes grain-like shape.

The radon atom that has recoiled out of a grain travels in the pore region surrounded by adjacent grains. The recoil ranges are 63 μm in air and 0.1 μm in water. Taking this into account, radon atom that has come into pore region has one of the three destinies; stops in water, stops in air or embedded in another grain lying in its trajectory. Therefore, radon atoms that contribute to the increase of emanation coefficient are those that stop in air or water. Especially in the narrow pore regions shown in **Fig. 5** where the distance between adjacent soil grains is below 63 μm, some of the radon atoms that were to be imbedded in another grain will escape that destiny through stopping in water if there is water in that pore region. This is the reason why the moisture has a large effect on radon emanation coefficients and especially the water in the narrow pore region accounts for most of it. This effect depends on the grain size because the volumetric fraction of the narrow pore region varies with the grain size and is the sole geometrical factor that affects the moisture saturation in this region.

(3) Mathematical Description of Close-Packed Spherical Grain Model

In the present study, a close-packed spherical grain model is developed to explain observed variation of radon emanation coefficient with moisture increase.

The close-packed spherical grain model consists of four spheres contacting each other as shown in **Fig. 6**. Grains are regarded as spheres. These four spheres make a single unit. In order to develop mathematical equation about radon behavior, uniform radon emission is assumed. This assump-

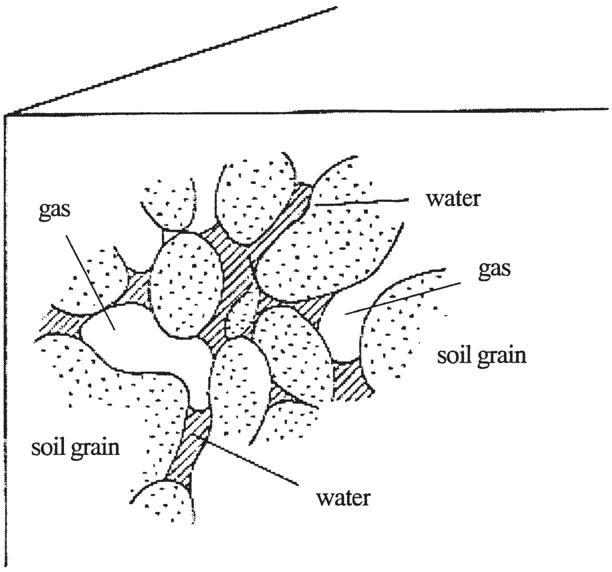

Fig. 3 Water and gas in soil (Reproduced from Ref. 4))

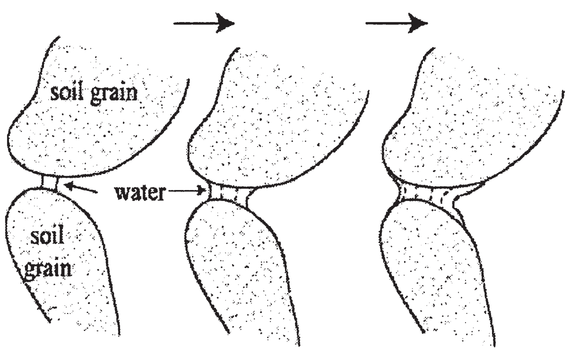

Fig. 4 Water growth on surface of soil grain

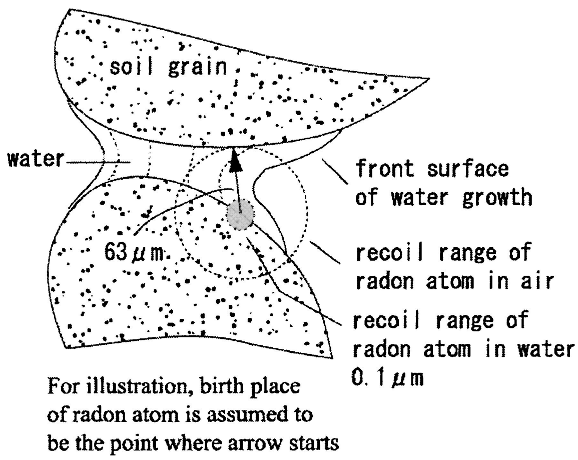

Fig. 5 Destiny of radon atom

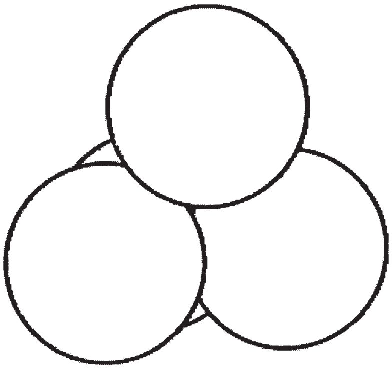

Fig. 6 Close-packed spherical grain model

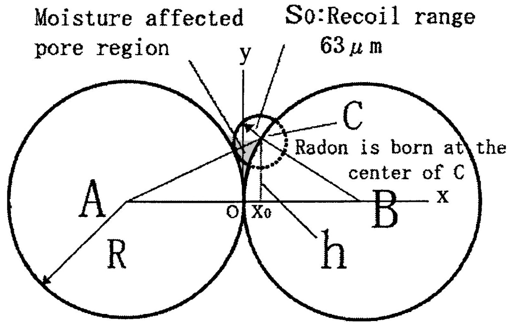

Fig. 7 Limiting condition for a radon atom to be imbedded in another grain

tion can be broken down into more concrete ones below.

Assumption 1: All radon atoms come out toward all directions in isotropic manner. Therefore, the amount of radon atoms coming out of small area on the surface of the grain is proportional to the solid angle formed on that small area and independent of the surface location.

Assumption 2: Radon atoms coming out of the grain lose their recoil energy in accordance with the distance they traveled in the grain. The remaining recoil energy is distributed uniformly. Therefore the amount of radon atoms traveling certain distance from the surface of the grain is constant between 0-recoil range in the pore region with neither moisture nor other grain interfering with radon travel around it.

Based on these assumptions, define 'q' as radon atoms emitted from a grain and traveling certain distance x ($x<$ recoil range in the pore region) per unit solid angle with neither moisture nor other grain interfering with the travel of these radon atoms. This q is independent of distance x.

The mathematical description of this model is illustrated both qualitatively and quantitatively.

(a) Qualitative Illustration of the Model

Radon emanation coefficient shows steep increase at low moisture saturation as shown in Fig. 1. This phenomenon is explained qualitatively by the present model as follows.

Recoil range of radon in water is 0.1 μm. Therefore very small amount of moisture should suffice to prevent radon atoms coming out into pore region from being embedded through crash into another grain. In order to know this amount of moisture, narrow pore region volume affecting radon emanation through moisture occupation has to be calculated. To avoid lengthy expression, 'moisture-affected pore region' hereafter is used to mean 'narrow pore region affecting radon emanation coefficients through moisture occupation'.

As mentioned before, water in the soil comes out at first in the narrow pore region between adjacent grains. Then it grows larger along the surface of the grains with increase of moisture. The front surface of the growing water may well be hollow-shaped. The depth of the hollow depends on the property of the grain. But in the present model, flat growing surface is assumed. With this assumption, the volume of the moisture-affected pore region is calculated.

In order to proceed with the calculation, symbols are assigned as in **Fig. 7**:

s_0: Radon recoil range in air (63 μm)

A, B: grain

C: sphere whose center is an emitting location of recoiled radon atom into pore region

R: Radius of grain A or B.

In Fig. 7, a radon atom is supposed to be emitted from the surface of grain B. Figure 7 shows the limiting condition for a radon atom to be embedded in another grain in its trajectory. If the surface location of an emitted radon atom moves away further from grain A, this radon atom never reaches A whether there is water or not in its trajectory. The moisture-affected pore region is shown as slightly grayed zone. In order to calculate the volume of this region, it is helpful to use the well-known integral formula for solid of revolution. Therefore, cut both the spheres by a plane passing through the two centers of them and this plain can give two-dimensional coordinate on it. The coordinate axes are determined as follows:

Origin: O, contacting point of A and B

Abscissa: The line connecting the centers of A and B

Ordinate: Vertical line through O.

In order to determine the integration range, h at the limiting condition is calculated at first. Then x_0 corresponding to h is calculated using Pythagorean theorem. The integration is done between 0–x_0.

a) Calculation of h in the Limiting Condition

According to Fig. 7 which shows limiting condition, the center of circle C sits on the periphery of circle B. Circles A and B contact each other at origin O. These conditions give the following equation

$$\sqrt{(R+s_0)^2-h^2}+\sqrt{R^2-h^2}=2R. \tag{1}$$

The radius R of the grain can vary. According to the information on measurements of radon emanation coefficients in U.S.A.[2] uranium ore was crushed to small particles on the order of 2–3 mm. This particle size is taken for comparison between the experiment done in U.S.A. and the present model. When calculating h from Eq. (1), the particle size is by far larger than radon recoil range in air, 63 μm. If R is larger than 800 μm, h becomes less than a third of R. Below this value of h, the fourth power of h/R becomes so small compared with 1. This leads to the well-known approximation shown below:

$$\sqrt{R^2 - h^2} \approx R\left\{1 - \frac{1}{2}\left(\frac{h}{R}\right)^2\right\}. \tag{2}$$

Using this kind of approximation leads to the solution of Eq. (1):

$$h = \sqrt{s_0 R}. \tag{3}$$

In the range of R larger than 800 μm, this approximation agrees with accurate solution within 1%.

b) Calculation of x_0 in the Limiting Condition

Using Pythagorean theorem, x_0 is calculated as follows:

$$x_0 = R - \sqrt{R^2 - h^2} \approx \frac{s_0}{2}. \tag{4}$$

c) Calculation of the Volume of the Moisture-Affected Pore Region

Using the well-known integral formula for solid of revolution, the volume of the moisture-affected pore region shown in Fig. 7 is calculated as follows. Factor '2' accounts for the symmetry with respect to y axis:

$$(\pi {s_0}^2 R/2 - \pi {s_0}^2 R/4) \cdot 2 = \pi {s_0}^2 R/2. \tag{5}$$

Hereafter the region corresponding to this volume is called as the complete moisture-affected pore region. The single unit of the close-packed spherical grain model consists of four spheres. To be more precise for the purpose of volume calculation, the unit consists of incomplete-shaped spheres cut by four planes each of which runs through three centers of the spheres. Therefore, there are fragmented moisture-affected pore regions and recombining them leads to two complete moisture-affected pore regions in the single unit. Thus, the total volume of them is

$$\text{Total volume of the moisture-affected pore region}$$
$$= 2 \cdot (\pi {s_0}^2 R/2) = \pi {s_0}^2 R. \tag{6}$$

Now the targeted volume has been obtained. But in order to show the effect of moisture on radon emanation coefficient in a more intelligible way, a parameter called 'moisture saturation' has been introduced. In order to avoid confusion with humidity, the definition of 'moisture saturation' is shown below as a precaution.

d) Radon Behavior with Moisture Saturation

In the course of studying the effect of moisture on radon emanation coefficient, it is appropriate to use a non-dimensional parameter called 'moisture saturation'. It is defined as a quotient of moisture volume in pore region divided by that pore volume. By introducing this non-dimensional parameter, generalized study can be done. Then, moisture saturation that affects the radon emanation coefficient can be written as

(Total volume of the moisture-affected pore regions)/

(Total pore volume).

Written in a mathematical equation with respect to a single unit of close-packed grain model, this becomes as

$$= \frac{\pi {s_0}^2 R}{0.734 R^3} = 4.3\left(\frac{s_0}{R}\right)^2. \tag{7}$$

Water growth in the pore region begins from moisture-affected pore region and the region is gradually occupied with water whose amount is calculated by Eq. (7). If this region is occupied with water, radon atoms stop in the water and are counted as emanated ones. Therefore, radon emanation coefficient starts to increase at an extremely low moisture saturation. If R is much larger than s_0 (63 μm), then this region is occupied soon with water in which case emanation coefficient shows steep increase. But, of course the slope of increase depends on the water adhesion property of the grain. After this region is completely occupied with water, the emanation coefficient levels off at some value, even if moisture keeps on increasing. Without water, radon atoms coming into this region crash into the opposite grain and are embedded in it. This is the reason why radon emanation coefficient shows low value at low moisture saturation.

(b) Quantitative Illustration of the Model

Radon emanation coefficient levels off at some value even if moisture keeps on increasing. If this value can be predicted, it is very helpful to secure radiological safety with respect to uranium-bearing wastes disposal. The present model is intended to make it possible to predict the leveling-off value of radon emanation coefficient.

As mentioned before, the present model employs measured emanation coefficient with dry condition. The leveling-off emanation coefficient with moisture is calculated using this measured data, information on the geometry of the grain and the assumption of uniform radon emission. Hereafter to avoid lengthy expression, 'moisture-affected radon' is used. This radon means the one that might have been embedded in another grain, if it had not been for water in its trajectory, but actually stopped in the water that happened to be around. This radon is counted as emanated.

The leveling-off radon emanation coefficient is calculated as shown below:

Step 1: Calculate the ratio of the area that emits moisture-affected radon to the whole area of the grain.

Step 2: Calculate the ratio of the moisture-affected radon to the whole emitted radon with regard to a particular surface area on the grain. Then, calculate the average value of this ratio over the area that emits the moisture-affected radon.

Step 3: Calculate the leveling-off radon emanation using the results calculated in Steps 1 and 2.

a) Calculation of Step 1

As previously mentioned, the close-packed spherical grain model consists of four spherical grains as a unit and two of them contacting each other are taken out for the calculation of microscopic radon behavior. Since these two spheres are rotationally symmetric with regard to one axis that runs through the centers of them, they can be treated as two circles on the two-dimensional plane and then translated into three-dimensional space using theorems in terms of rotational symmetry. Therefore the grain of the present model is drawn as a circle. Then, the area that emits moisture-affected

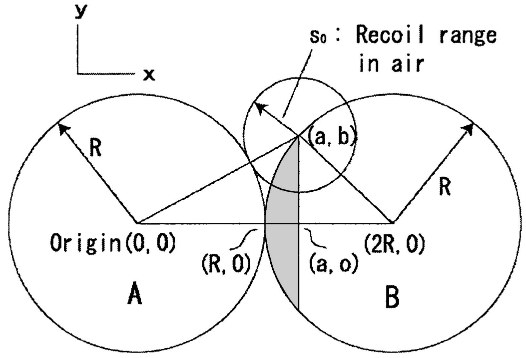

Fig. 8 Area emitting moisture-affected radon

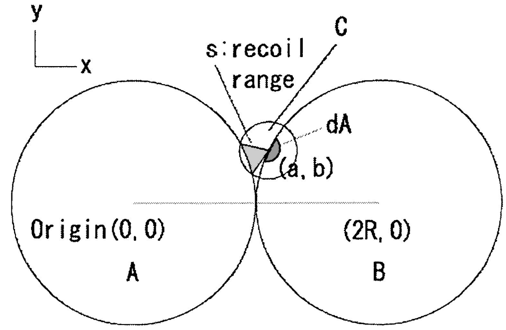

Fig. 9 Travel region of moisture-affected radon

radon is shown as a gray region in **Fig. 8**. In this region, maximum length between two spheres is less than the radon recoil range in air (63 μm). The radon traveling in this region crashes into the opposite sphere if there is no water in there. The corresponding end points of this gray area on the x-coordinate is (R, 0) and (a, 0). The value of 'a' is calculated solving the following equations

$$a^2 + b^2 = (R + s_0)^2 \tag{8}$$

$$(a - 2R)^2 + b^2 = R^2. \tag{9}$$

Solving this equation leads to

$$a = \frac{4R^2 + 2Rs_0 + {s_0}^2}{4R}. \tag{10}$$

The gray surface area of the sphere 'B' is calculated as $2\pi(a - R)R$ which leads to the following equation

$$\text{The area that emits moisture-affected radon} = \frac{\pi(2Rs_0 + {s_0}^2)}{2}. \tag{11}$$

The surface area of the whole sphere is $4\pi R^2$. There are twelve contacting locations with other grains on a single grain surface. Therefore the ratio 'RA1' of the area that emits moisture-affected radon to the whole area of the grain becomes

$$\mathrm{RA1} = \frac{3(2Rs_0 + {s_0}^2)}{2R^2}. \tag{12}$$

b) Calculation of Step 2

At first, the amount of moisture-affected radon is calculated. The gray region in **Fig. 9** shows the space where moisture-affected radon travels. The recoil range 's' of moisture-affected radon has to be greater than the radius of circle C just as it contacts circle B. This is denoted as $s_{\min}$ and shown in **Fig. 10**.

It is calculated employing Pythagorean theorem as follows:

$$s_{\min} = \sqrt{a^2 + b^2} - R. \tag{13}$$

The 's' varies from $s_{\min}$ to s_0. Therefore, the amount of radon affected by moisture has to be integrated over $s_{\min}$–s_0 for a small area dA around each particular position (a,b) on sphere B. This is denoted as 'Q'. The solid angle shown as a gray region in **Fig. 9** is denoted as $\Omega(s)$. As previously defined,

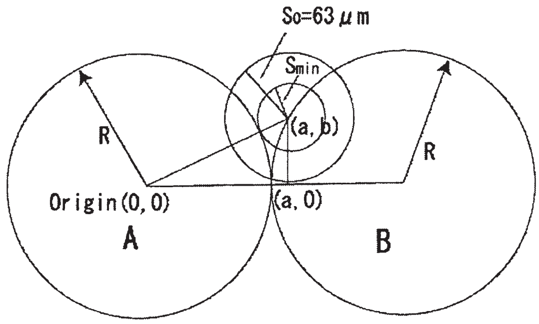

Fig. 10 Limits of recoil range s_0, $s_{\min}$ of moisture-affected radon

'q' is the number of radon atoms traveling distance 's' per unit solid angle. Then, $\Omega(s)$ and Q are calculated as follows:

$$\Omega(s) = \frac{\pi\left\{R^2 - \left(\sqrt{a^2 + b^2} - s\right)^2\right\}}{s\sqrt{a^2 + b^2}} \tag{14}$$

$$Q = \int_{s_{\min}}^{s_0} q\Omega(s)dsdA = \int_{\sqrt{a^2+b^2}-R}^{s_0} q\Omega(s)dsdA, \tag{15}$$

$\Omega(s)$ becomes maximum 'Ω_0' at s=s_0. This leads to Ω_0 as follows:

$$\Omega_0 = \frac{\pi\left\{R^2 - \left(\sqrt{a^2 + b^2} - s_0\right)^2\right\}}{s_0\sqrt{a^2 + b^2}}. \tag{16}$$

The total amount of radon 'Q_0' in the solid angle Ω_0 is

$$Q_0 = \int_0^{s_0} q\Omega_0 dsdA = qs_0\Omega_0 dA. \tag{17}$$

The total amount of radon emitted from small area dA equals $2\pi qs_0(dA)$

Therefore, the ratio 'W' of the moisture-affected radon to the whole emitted radon with regard to a particular surface area on sphere B is

$$W = \frac{\int_{\sqrt{a^2+b^2}-R}^{s_0} \Omega(s)ds}{2\pi s_0}. \tag{18}$$

Substituting Eq. (14) for $\Omega(s)$ gives

$$W = \frac{\left[\left\{R^2 - \left(a^2+b^2\right)\right\}[\ln(s)]^{s_0}_{\sqrt{a^2+b^2}-r} + 2\sqrt{a^2+b^2}[s]^{s_0}_{\sqrt{a^2+b^2}-R} - \frac{1}{2}\left[s^2\right]^{s_0}_{\sqrt{a^2+b^2}-R}\right]}{2s_0\sqrt{a^2+b^2}}. \tag{19}$$

This W is obtained with regard to the particular small area around point (a,b) on sphere B. Then W has to be averaged over the area that emits moisture-affected radon. In order to do this calculation, making use of rotational symmetry, the small area on sphere B is taken as the gray region in **Fig. 11**. This small surface of sliced part of sphere B is $2\pi R dx$. One of the end point of the range for W to be averaged is already calculated in Eq. (10). Another is the contacting point of circles A and B. Then the range is from $x = R$ to $x = (4R^2 + 2Rs_0 + s_0{}^2)/(4R)$ and the latter is denoted as a_{lim}. Average W is calculated as follows:

$$\bar{W} = \frac{\int_R^{a_{\text{lim}}} W \cdot 2\pi R dx}{\int_R^{a_{\text{lim}}} 2\pi R dx} = \frac{\int_R^{a_{\text{lim}}} W dx}{\int_R^{a_{\text{lim}}} dx} \tag{20}$$

$$a_{\text{lim}} = \frac{4R^2 + 2Rs_0 + s_0{}^2}{4R}. \tag{21}$$

c) Calculation of Step 3

The total amount of moisture-affected radon can be translated as 'the amount of radon that would crash into other grain to be embedded in it and not be counted as emanated, if it were not for moisture in the pore region'. And $\bar{W}$ has been calculated with regard to the two contacting spheres, A and B. But, sphere B contacts twelve other spheres likewise. Therefore this has to be taken into account, when emanation over the whole sphere is calculated. Here, strictly defined wording is needed. When the word 'total' is used, it is related to the contacting region of two spheres. When the word 'whole' is used, it is related to all the region of the targeted sphere. Then, the ratio of radon emanation coefficient with moisture 'Em' to radon emanation coefficient with no moisture 'Enm' is

$$\frac{\text{Em}}{\text{Enm}} = \frac{\text{Whole amount of radon emitted from a grain}}{(\text{Whole amount of radon emitted from a grain}) - (\text{Whole amount of moisture-affected radon})}$$
$$= \frac{1}{1 - \dfrac{\text{Whole amount of moisture-affected radon}}{\text{Whole amount of radon emitted from a grain}}}. \tag{22}$$

The ratio of whole amount of moisture-affected radon to whole amount of radon emitted from a grain can be calculated using $\bar{W}$. The average $\bar{W}$ is defined over the limited region as a grey part of sphere B shown in Fig. 8. In order to translate it with respect to the whole sphere taking 12 contacting points into account, $\bar{W}$ has to be multiplied by the ratio of twelve times the gray surface area to the whole surface area '$4\pi R^2$', using Eq. (16) to get

$$\frac{\text{Whole amount of moisture-affected radon}}{\text{Whole amount of radon emitted from a grain}} = 1.5(2Rs_0 + s_0{}^2)R^{-2}\bar{W}. \tag{23}$$

Using Eq. (23), Em/Enm becomes

$$\frac{\text{Em}}{\text{Enm}} = \frac{1}{1 - 1.5(2Rs_0 + s_0{}^2)R^{-2}\bar{W}}. \tag{24}$$

The leveling-off radon emanation coefficient Em can be calculated employing Eq. (24) and the observed emanation coefficient with no moisture Enm.

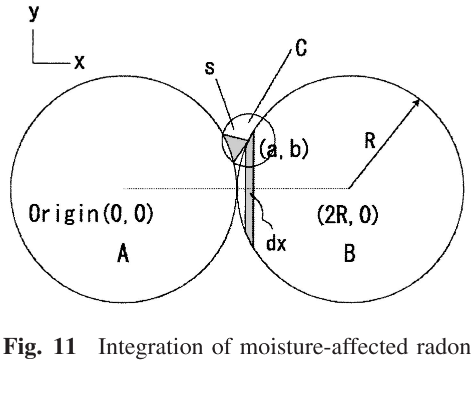

Fig. 11 Integration of moisture-affected radon

III. Comparison with Experiments and Discussion

(1) Qualitative Comparison and Discussion

As mentioned before, the experiment done in U.S.A.[2)] gives clear illustrations which show how moisture saturation affects emanation of radon. In the experiment, crushed uranium ores on the order of 2–3 mm were used. The present model has been compared with this experiment. For comparison, Eq. (7) of the present model has been employed substituting 2 mm for R and 63 μm for s_0. Then moisture saturation affecting radon emanation coefficient has been calculated as 0.004. Slight amount of moisture may well be needed for water in narrow pore region before starting to grow. After that, the present model predicts theoretically that only 0.004 of moisture saturation increase should account for almost all part of radon emanation coefficient increase. This means steep increase of radon emanation coefficient should be observed at very low moisture saturation. Figure 1 shows this steep increase as almost perpendicular to the abscissa. The increase of radon emanation coefficient still goes on and settles at around 0.47. The reason of this increase can be explained as follows. The front surface of growing water actually is skewed concave rather than flat depending on the water adhesion property of the grain. Therefore a good amount of moisture is needed for water to grow on the surface of grain before moisture increase does not affect radon emanation any further where the bottom of the concave reaches h in the limiting condition in Fig. 7. In the present paper, only one illustration is shown in Fig. 1, but there are eighteen of them in the paper[2] from U.S.A. and most of them show just the same variation of radon emanation coefficient as mentioned above.

As a conclusion, it has been proved that the close-packed spherical grain model offers a good explanation of the variation of radon emanation coefficient qualitatively. One of the applications of this model could be directed toward the development of the technology to reduce environmental radioactivity brought about by naturally occurring radioactive materials (NORM), especially those containing radium, the parent nuclide of radon. According to the present model, deleting narrow pore regions means deleting moisture that contributes radon emanation. Perhaps heating is one of the measures to delete narrow pore regions. Decrease of radon emanation by heating is reported[5] for some materials. The effect of heating is called annealing in the report.

According to the present model, the best way to lower radon emanation would be melting material, thereby eliminating pore region completely. Melting may well have an advantage of relocating radium atoms uniformly in material. Radon recoil range for common minerals is only 0.02–0.07 µm. Therefore if radium atoms are uniformly relocated in material, almost all of the radon atoms stop within that material, which means very low emanation.

(2) Quantitative Comparison and Discussion

The present model needs information on grain size. The only experimental data at hand that includes this information comes from an experiment[6] done under the sponsorship of Ministry of Economy, Trade and Industry (METI), Japan and is shown in **Fig. 12**. The present model has been compared with this experiment.

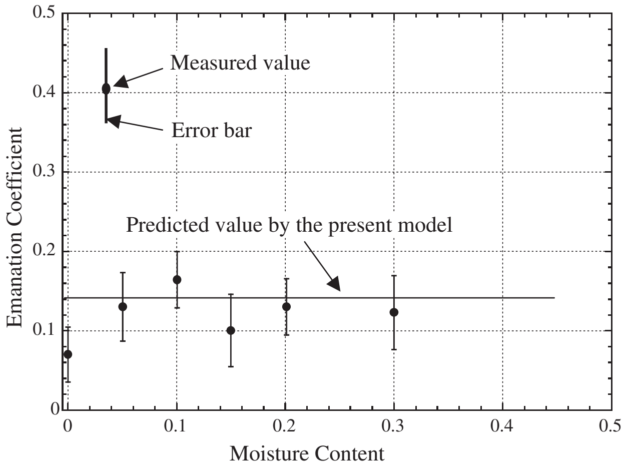

Fig. 12 Emanation coefficients measured and predicted for calcium superphosphate

Table 1 Grain size distribution of calcium superphosphate

Grain size (sieve mesh) (µm)	Average grain radius (µm)	Amount (g)	Weighting factor (Relative number of grains)
1,000–2,000	750	9.42	0.065
500–1,000	375	29.41	0.407
300–500	200	42.88	1.112
125–300	106.25	44.99	2.197
63–125	47	9.06	1

According to the experiment, calcium superphosphate was employed to simulate uranium-bearing wastes, because it resembles sludge or ash in light of both uranium (radium) adhesion on the surface of grain (surface contamination) and the geometric shape of the grain. The grain size distribution of calcium superphosphate is shown in **Table 1**. On the other hand, the grain size of sludge ranges from several tens of µm to several thousands of µm and that of ash from several tens of µm to several hundreds of µm. They overlap that of calcium superphosphate. Using the grain size distribution, the ratio of radon emanation coefficient with moisture 'Em' to radon emanation coefficient with no moisture 'Enm' has been calculated by Eq. (24) for each grain size and this ratio has been weight-averaged by the relative number of grains which belong to some specific grain interval. The relative number of grains has been calculated assuming the grain as a sphere of average radius in that interval and specific weight as '1'. Thus calculated as shown in **Table 2**, weighted mean Em/Enm becomes 1.995. The measured Enm(0.07) multiplied by this value becomes predicted Em(0.14) and is shown as a horizontal line in Fig. 12, where the abscissa is shown as moisture content instead of moisture saturation for convenience sake. On the other hand, the paper from Bu-

Table 2 Ratio of radon emanation coefficients, Em/Enm

Grain size (sieve mesh) (µm)	Em/Enm	Weighting factor (Relative number of grains)	Weighted mean Em/Enm
1,000–2,000	1.069	0.065	
500–1,000	1.145	0.407	
300–500	1.279	1.112	1.995
125–300	1.677	2.197	
63–125	3.874	1	

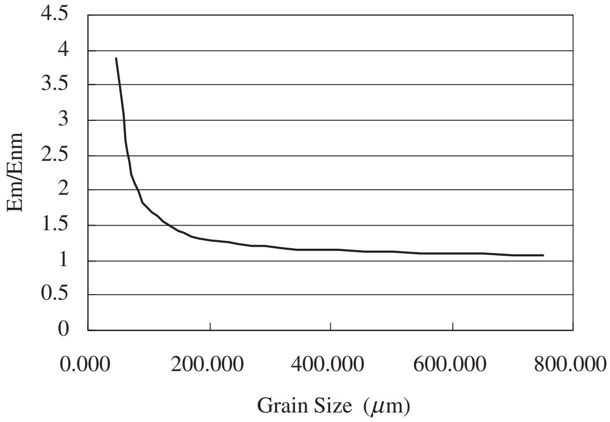

Fig. 13 Ratio of moist to dry emanation coefficient Em/Enm

reau of Mines, U.S.A.[2)] states 'The ratios of the moist to dry emanation coefficients varied from 1.38 to 4.19, with a mean ratio of 2.3. Radon emanation was thus increased, on the average, by more than a factor of two by the presence of moisture'. Since no information on grain size distribution is available with respect to the paper from U.S.A., direct comparison cannot be done between the predicted values from present model and the data in the U.S.A. paper with respect to Em/Enm. But, both deal with the radon emanation phenomenon of the order of μm. Therefore it is no wonder that the predicted Em/Enm values of the present model shown in Table 2 have overlapped the ones in the U.S.A. paper. For reference, Em/Enm versus grain size is given in **Fig. 13**.

IV. Concluding Remarks

In the present study, close-packed spherical grain model has been developed. So far it seems to give a good explanation of radon emanation coefficient affected by moisture. But this is nothing more than or less than a model. Therefore it is not almighty, which means the applicable area is obviously limited. But, one of the most useful applications of the present model in the field of reducing environmental radioactivity has already been referred to in the present paper. It lies in the field of safety assessment of uranium-bearing waste and naturally occurring radioactive material (NORM) disposal. The next development of this model can be thought of as the one directed toward academic elaboration employing more detailed structure of grain shape, but the authors think it can wait. The first thing to be done now is the application of this model to develop adequate processing technologies for uranium-bearing waste and NORM, thereby reducing radiation exposure to the human being. One of such technologies off the top of one's head is to melt or heat to eliminate small pores which contribute to radon emanation increase. For that purpose, the authors are now proceeding with experiments to reveal radon behavior and the results will be open to the public in the near future.

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