

Regular Article

Generation and Migration of ^{222}Rn in BaSO_4 Precipitate Samples and Implications for their Analysis for ^{226}Ra by Gamma Spectrometry

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Received 7 March 2016; revised 24 June 2016; accepted 30 June 2016

The generation and migration of ^{222}Rn in BaSO_4 precipitate samples obtained by a barium co-precipitation method were investigated using measurements of ^{226}Ra activity concentrations, ^{222}Rn emanation coefficients and published values of ^{222}Rn diffusion coefficients in this study. ^{226}Ra activities obtained using different radon-tight sealing methods were not observed to be significantly different compared with those obtained without sealing. The emanation coefficient and the diffusion coefficient in the precipitate – factors that influence the ^{222}Rn exhalation rate – were found to be much lower than those in other materials. As a result, the evaluated exhalation rate, which was found to be $1.8 \times 10^{-5} \text{ Bq m}^{-2} \text{ s}^{-1}$, was found to be lower than that from other materials. Thus, the sealing method is not a significant influencing factor on the ^{226}Ra activity concentrations determined using this barium co-precipitation method. Additionally, it was found that the uncertainty of ^{226}Ra activity was most strongly influenced by the chemical recovery of the ^{226}Ra using the barium co-precipitation method.

Key words: radon-222, radium-226, barium co-precipitation method, high purity germanium detector, sealing method, exhalation rate

1. Introduction

^{226}Ra is one of the decay products of the ^{238}U -series which is included in many materials such as soil, rocks, water and building materials. Furthermore, ^{226}Ra and other radioisotopes are often generated as byproducts in industrial waste referred to as Naturally Occurring

Radioactive Material¹. ^{226}Ra has a long half-life of approximately 1600 years. Gamma-rays emitted from ^{226}Ra in rocks, soils, and building materials can also give rise to external exposure to humans². If radium is taken into the human body by the consumption of drinking water and food it can be accumulated into bones, due to having a similar chemical behavior as calcium which is in the same group in the periodic table¹. Consequently, internal exposure by alpha-particles emitted from ^{226}Ra and its decay products may cause cancer. Furthermore, the dose coefficient of the activity to the committed effective dose for the intake of ^{226}Ra is $2.8 \times 10^{-7} \text{ Sv Bq}^{-1}$ for adults, and this value is relatively higher than that of

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other radionuclides³). Thus, the protection of external and internal exposures caused by ²²⁶Ra is significant from the viewpoint of health physics.

²²²Rn is generated by alpha decay of the ²²⁶Ra-containing materials. ²²²Rn is a radioactive noble gas that is taken into the body by inhalation and it is well-known as a hazardous material that can cause lung cancer through internal exposure⁴). Evaluation of ²²⁶Ra activity in materials is also important to predict the amount of the radon exhaled from materials⁵). Thus, many researchers report the ²²⁶Ra activities in soils, building materials, drinking waters, and food samples⁶⁻⁹).

²²⁶Ra activity in materials can be evaluated by gamma-ray spectrometry, alpha spectrometry, mass spectrometry, and liquid scintillation counting techniques^{1, 10}). ²²⁶Ra activities in rocks and soils are evaluated without chemical preparation by gamma-ray spectrometry which use high-purity germanium (HPGe) detectors. In contrast, chemical preparation, such as barium coprecipitation methods, are typically used for the analysis of ²²⁶Ra activity in drinking waters and sea water as the ²²⁶Ra activity concentrations in these waters are, in most cases, lower than the levels found in rocks and soils. Typically, the minimum detectable activity for gamma-ray spectrometry is higher than that for alpha spectrometry and liquid scintillation counting techniques¹⁰). However, the sample preparation for gamma-ray spectrometry is much easier than that for alpha spectrometry and liquid scintillation counting techniques. Additionally, the total cost of the measurement system for gamma-ray spectrometry is cheaper than that for alpha spectrometry and liquid scintillation counting techniques. Thus, many researchers conduct ²²⁶Ra analysis by gamma-ray spectrometry.

A gamma-ray of energy 186.1 keV is emitted from the inside of a nucleus following alpha decay of ²²⁶Ra. However, a gamma-ray of energy 185.7 keV is emitted from ²³⁵U, and the full energy absorbed peak made by this energy is well known as an interference peak for ²²⁶Ra analysis by gamma-ray spectrometry¹⁰). That is, for most samples, the full energy absorbed peak of 186.1 keV is not suitable to use for the analysis of ²²⁶Ra by gamma-ray spectrometry using HPGe. In general, ²²⁶Ra is determined by the 295 and 352 keV gamma energies for ²¹⁴Pb and the 609 keV gamma energy for ²¹⁴Bi, which are decay products of ²²⁶Ra after allowing for an ingrowth period of at least 30 days to allow these decay products reach equilibrium with ²²⁶Ra¹¹). In order to ensure the radioactive equilibrium of ²²⁶Ra, ²²²Rn and their decay products, it is important to ensure that no ²²²Rn is lost from the sample container.

In the present study, ²²⁶Ra activities in water obtained by a barium coprecipitation method were evaluated by a HPGe detector in order to compare the values obtained

using, respectively, a sealing method by an aluminum tape and a sealing method by an epoxy resin adhesive in the Office of Radiological Protection, Environmental Protection Agency, Ireland (EPA-ORP) and Hirosaki University.

The ²²⁶Ra activity concentration, the ²²²Rn emanation coefficient, and the ²²²Rn diffusion coefficient are the significant influence factors for the generation and migration of ²²²Rn in materials¹²). However, the ²²²Rn emanation coefficient and the ²²²Rn diffusion coefficient in BaSO₄ precipitate have not yet been published. Thus, a ²²²Rn emanation coefficient in the BaSO₄ precipitate was evaluated using the accumulation method in this study. Additionally, the ²²²Rn diffusion coefficient in the BaSO₄ precipitate was discussed using the published data on similar chemical components. An important parameter related to the exhaled ²²²Rn from the surface of the materials is the ²²²Rn exhalation rate which is often called the ²²²Rn flux density. The ²²²Rn exhalation rate from the BaSO₄ precipitate surface was also evaluated in this study.

2. Materials and methods

2.1. Chemical preparation of water samples by barium coprecipitation method

The chemical preparation of water samples were carried out according to a test procedure in the EPA-ORP. The test procedure has not been published elsewhere. 4 L of purified water which was not including ²²⁶Ra was used in this study. 1 mL of ²²⁶Ra standard solution (23.6 ± 0.7 Bq g⁻¹) was added into the purified water, and stirred. The water sample was acidified using 40 mL of concentrated nitric acid in order to adjust the pH to between 0 and 1. 30 mL of concentrated hydrochloric acid was added into the beaker, and then 5 mL of a barium carrier solution was added. The water sample was boiled for approximately 10 min. 30 mL of 9 M sulphuric acid was added to the boiling solution in order to form the precipitate and the boiling and stirring was continued for approximately 30 min. The beaker was stored overnight to allow the barium sulphate (BaSO₄) precipitate to settle. The precipitate was collected onto a glass microfiber filter (Whatman GF/C, 47 mmφ) using filtration equipment. The precipitate on the filter was washed with 10 mL of ethanol and then 10 mL of diethyl ether. The precipitate on the filter was dried overnight in a vacuum desiccator. Six samples were made in this study.

2.2. Calculation method of the chemical recovery of barium

The chemical recovery (*R*) of barium was calculated by Eq. (1).

$$R(\%) = \frac{m_{\text{BaSO}_4} \cdot m_{\text{Ba}}^{\text{atom}}}{m_{\text{Ba}} \cdot V_{\text{Ba}} \cdot m_{\text{BaSO}_4}^{\text{mol}}} \times 100 \quad (1)$$

Where, m_{BaSO_4} is the mass (g) of BaSO_4 precipitate on the filter, m_{Ba} is the mass of barium in 1 mL of barium carrier solution, V_{Ba} is the volume (5 mL) of barium carrier solution added to the sample. Thus, $m_{\text{Ba}} \times V_{\text{Ba}}$ is the mass (g) of added barium. $m_{\text{BaSO}_4}^{\text{mol}}$ is the molecular weight of BaSO_4 (233.40 g mol⁻¹), $m_{\text{Ba}}^{\text{atom}}$ is the atomic weight of barium (137.34 g mol⁻¹). Furthermore, m_{Ba} was calculated by Eq. (2).

$$m_{\text{Ba}} = \frac{m_{\text{Ba}}^{\text{atom}} \cdot m_{\text{BaCl}_2 \cdot \text{H}_2\text{O}}}{m_{\text{BaCl}_2 \cdot \text{H}_2\text{O}}^{\text{mol}} \cdot V_{\text{H}_2\text{O}}} \quad (2)$$

Where, $m_{\text{BaCl}_2 \cdot \text{H}_2\text{O}}^{\text{mol}}$ is the molecular weight of $\text{BaCl}_2 \cdot \text{H}_2\text{O}$ (244.358 g mol⁻¹), $m_{\text{BaCl}_2 \cdot \text{H}_2\text{O}}$ is the weight of $\text{BaCl}_2 \cdot \text{H}_2\text{O}$ (1.76 g), $V_{\text{H}_2\text{O}}$ is the volume (100 mL) of deionized water. Thus, m_{Ba} was calculated as 9.89×10^{-3} g.

2.3. Sealing methods

The filter sample was placed, precipitate side down, into the inverted lid of a counting container (52 mm ϕ) which was made of polypropylene. The bottom of the container was placed on top of the filter and precipitate (Fig. 1). The sample was then sealed using different methods and the prepared sample was stored for more than 30 days to allow radioactive equilibrium between ²²⁶Ra and its decay products to be reached. The different methods of sealing are shown in Fig. 1. The same six BaSO_4 precipitate samples were used for the comparison of results obtained by the different sealing methods.

Firstly, the six samples were placed inside the polypropylene container without sealing (this is the “Without seal” approach). After the “Without seal” sample measurement was completed, the polypropylene containers were sealed by aluminum tape (this is the “Seal-1” approach). This sealing method is currently used in the determination of radium in drinking water in the EPA-ORP. After the completion of the “Seal-1” sample measurement, the silver foil on the container was removed and the gap between bottom and the lid of the polypropylene container was filled using a plastic paraffin film (PARAFILM “M”[®]). Furthermore, the joint between the bottom container and the lid was covered carefully by an epoxy resin adhesive (Araldite[®]). This sealing method was called the “Seal-2” approach¹³. This method, used in Hirosaki University, was slightly modified for this study. Typically, a silicone rubber is used in order to fill the gap. However, the volume of the gap of bottom and lid used in this study was relatively small. Thus, a plastic paraffin film was used instead of silicone rubber.

2.4. Sample measurement

Samples were measured by gamma spectrometry using a p-type HPGe detector (GC7520/S, CANBERRA Industries Inc., U.S.A.). Spectral analysis was performed using Canberra ApexGamma (Genie 2k) software.

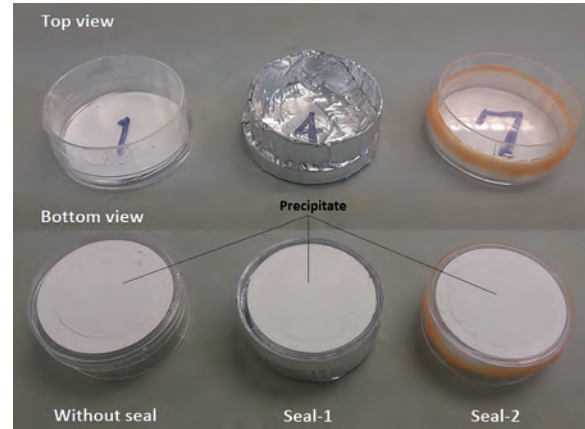


Fig. 1. Photo of the sealed samples by the different methods.

The measurement time was set as 86,400 s. The ²²⁶Ra activity was determined as the weighted average¹⁴ of the activities of ²¹⁴Pb (295 keV, 352 keV) and ²¹⁴Bi (609 keV) decay nuclides. Each activity (A_i) in the BaSO_4 precipitate can be calculated by Eq. (3).

$$A_i = \frac{\frac{N_t}{T_t} - \frac{N_b}{T_b}}{\eta_p \cdot \eta_f \cdot R \cdot f} \quad (3)$$

Where, N_t is the total counts of the sample (count), N_b is the background counts (count), T_t is the counting time (86,400 s) for sample, T_b is the background counting time (1,036,800 s). η_p is the peak efficiency of the peak of interest for the gamma-ray energy. η_f is the emission probability of the gamma-ray energy of interest, and R is the chemical recovery. f is the correction factor for the true coincidence summing. A geometry specific efficiency curve was determined using an Eckert & Ziegler Analytics custom-made multi-gamma standard. GESPECOR[®] Monte-Carlo software (CID Media GmbH, Germany) was used for correction for true coincidence summing was required. No self-attenuation or decay corrections were applied.

2.5. Measurement of ²²²Rn emanation coefficient in BaSO_4 precipitate

The fraction of ²²²Rn generated within the components of materials and escaping to the pore space is called as the emanation coefficient, emanation fraction, or emanating power¹⁵. The BaSO_4 precipitate sample was put on the bottom of a polypropylene container (525 mL volume) and the passive type ²²²Rn monitor used by the EPA-ORP based on a NRPB ²²²Rn monitor¹⁶ was set inside the lid of the container. The ²²²Rn monitor consists of a two-part polypropylene holder and a CR-39 plastic. According to a report by Hanley *et al.*¹⁷, the holder acts as a simple ²²²Rn diffusion chamber, excluding ²²²Rn decay products and

Table 1. The results of chemical recovery and ^{226}Ra activity in precipitate of each sample

Sample No.	Chemical recovery (%)	^{226}Ra activity in precipitate (Bq)		
		Without seal	Seal-1	Seal-2
Sample 1	89 ± 2	25.2 ± 0.5	25.3 ± 0.5	23.6 ± 0.5
Sample 2	75 ± 2	25.1 ± 0.5	25.3 ± 0.6	24.2 ± 0.5
Sample 3	82 ± 5	24.5 ± 0.9	24.8 ± 0.9	23.8 ± 0.9
Sample 4	102 ± 5	23.5 ± 0.8	23.5 ± 0.8	23.2 ± 0.8
Sample 5	102 ± 5	23.5 ± 0.8	23.4 ± 0.8	23.3 ± 0.8
Sample 6	94 ± 5	25.1 ± 0.9	25.0 ± 0.9	25.0 ± 0.9
Average ± s.u.*	91 ± 2	24.5 ± 0.3	24.5 ± 0.3	23.9 ± 0.3

*s.u.: standard uncertainty ($k = 1$)

dust, limiting access of moisture but allowing the entry of ^{222}Rn gas by molecular diffusion.

The container was sealed by silver foil, and the exhaled ^{222}Rn from the precipitate was accumulated for 16 days. After exposing the CR-39, it was chemically etched for 1 h in a 6.25 M NaOH solution at 98°C¹⁸. The number of alpha tracks were counted using an automatic track reading system (TASLIMAGE, Track Analysis System Ltd., UK). The ^{222}Rn activity concentration is calculated using the track densities for sample and control and the sensitivity factor¹⁷.

The ^{222}Rn emanation coefficient was calculated using the Eq. (4)⁵.

$$f = \frac{C_{\text{Rn}}^{\text{eq}} \cdot V}{A_{\text{Ra}} \cdot M} \quad (4)$$

Where, f is the ^{222}Rn emanation coefficient, $C_{\text{Rn}}^{\text{eq}}$ is the radioactive equilibrium ^{222}Rn activity concentration (Bq m⁻³), V is the volume of the polypropylene container (5.25×10^{-4} m³), A_{Ra} is the ^{226}Ra activity concentration in BaSO₄ precipitate (Bq kg⁻¹), and the M is the mass of BaSO₄ precipitate. $C_{\text{Rn}}^{\text{eq}}$ was estimated using the Eq. (5).

$$C_{\text{Rn}}^{\text{eq}} = \frac{C_{\text{Rn}}^{\text{t}}}{(1 - e^{-\lambda_{\text{Rn}} \cdot t})} \quad (5)$$

Where, λ_{Rn} is the decay constant of ^{222}Rn (2.1×10^{-6} s⁻¹), and t is the accumulation time (s).

3. Results and discussion

3.1. ^{226}Ra activities of the different sealing methods

^{226}Ra activities and standard uncertainties of “Without seal”, “Seal-1”, and “Seal-2” were 24.5 ± 0.3 , 24.5 ± 0.3 , and 23.9 ± 0.3 Bq, respectively (Table 1). Homogeneity of variances of the three different kinds of samples was evaluated using the Bartlett’s test by EZR, and the significance level was set as 0.05¹⁹. These statistic results were observed as homogeneous variances ($P = 0.853$). Thus, the differences of the average of the three different kind of samples were evaluated using the

one-way analysis of variance by EZR¹⁹. The statistical analysis of the results were not observed as significantly different ($p = 0.291$). The ^{226}Ra activities of “Without seal”, “Seal-1”, and “Seal-2” were compared with the value of the standard solution by a z-test. z-values of “Without seal”, “Seal-1”, and “Seal-2” were calculated as 1.17, 1.21, and 0.33, and these values were observed as less than 1.96 (95% confidence level). Thus, ^{226}Ra activity in each precipitate sample and standard solution was not observed as significantly different in the range of the 95% confidence level. According to a method by Tomita *et al.*, BaSO₄ precipitate was packed in a film from which radon escapes were negligibly small²⁰. However, the result obtained in this study suggested that the sealing of the BaSO₄ precipitate sample was not an influential factor in the measurement of ^{226}Ra in water samples.

Moore²¹ and Moore and Dymond²² reported that the leakage of ^{222}Rn was negligible when ^{226}Ra was measured as BaSO₄. Recently, Scholten *et al.* reported that the leakage of ^{222}Rn from BaSO₄ precipitate in an open plastic tube was observed as $0.1 \pm 0.02\%$ ²³. Furthermore, an additional reason for the results reported is that the ^{226}Ra is incorporated into the BaSO₄ lattice (barite) and because of the very dense nature of barite, ^{222}Rn emanation from the crystal is very low²³.

The ^{222}Rn emanation coefficient of BaSO₄ precipitate was evaluated as 0.0005 ± 0.0001 . The ^{222}Rn emanation coefficient of BaSO₄ precipitate has not been published. However, previous studies on the ^{222}Rn emanation coefficient of barite have been published²⁴⁻²⁶. These may provide useful information for the discussion of the results obtained in this study. ^{222}Rn emanation coefficients of radium-containing barite scale at petroleum exploration and production sites in U.S.A. were evaluated by White *et al.*, and these values ranged from 0.037 to 0.138 ($n = 48$)²⁴. Furthermore, White *et al.* reported that the reason for variation of these values may be the differences in the mean particle size of the samples²⁴. Additionally, according to the report by Paytan *et al.*, the $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratios of marine barite were found to

be from 0.04 to 0.15²⁵). If the ²²⁶Ra and ²¹⁰Pb has reached radioactive equilibrium in this study, the ²¹⁰Pb/²²⁶Ra activity ratios reported can be considered as the radon emanation coefficient. However, ²²²Rn emanation coefficients for rocks, soils, and building materials were reported from 0.021 to 0.32, from 0.0014 to 0.80, and from 0.0010 to 0.58, respectively²⁶). The ²²²Rn emanation coefficient of BaSO₄ precipitate obtained in this study was much lower than that reported by these previous studies.

In general, the transfer of radon from materials such as rock and soil into the atmosphere is considered to be primarily due to the molecular diffusion. The radon diffusion coefficient in many materials has been reported²⁶⁻³²). The ²²²Rn diffusion coefficient for BaSO₄ has not yet been published. However, radon diffusion coefficients for BaCO₃ and Ba(NO₃)₂ were reported as 1.6×10^{-22} and $\sim 10^{-24}$ m² s⁻¹, respectively^{27, 28}). Hirst and Harrison reported the ²²²Rn diffusion coefficient for free air at 15°C and 1013 hPa as 1.2×10^{-5} m² s⁻¹²⁹). A similar experimental value has been reported by some researchers^{30, 31}). Rogers and Nielson reported on the empirical formula for the ²²²Rn diffusion coefficient of soil. The ²²²Rn diffusion coefficient can be evaluated as 8.2×10^{-6} m² s⁻¹, if the porosity of soil is given as 0.67³²). This calculated result is in good agreement (8.2 - 9.6×10^{-6} m² s⁻¹) with the experimental value of soil samples (porosity: 0.67) reported by Parasad *et al.*³¹). So the ²²²Rn diffusion coefficient of solids such as BaCO₃ and Ba(NO₃)₂ are lower compared to the values of air and soil. Thus, the results reported by many researchers suggest that BaSO₄ precipitate plays a crucial role as a diffusion barrier for ²²²Rn migration. That is, the ²²²Rn emanation coefficient of BaSO₄ was observed as lower value than that of the other materials.

According to the report by Várhegyi *et al.*³³), ²²²Rn effective diffusion coefficient (D_e) can be described by the following Eq. (6).

$$D_e \leq (1-n) \cdot D_{\text{BaSO}_4} + v \cdot D_w + (n-v) \cdot D_{\text{air}} \quad (1 > n > v) \quad (6)$$

Where, n and v are the pore space and moisture volume ratio, respectively. D_{BaSO_4} , D_w , and D_{air} are respectively the ²²²Rn diffusion coefficient of BaSO₄, water, and air. In this study, the ²²²Rn diffusion coefficient of water could be considered as zero due to the fact that dried samples were used. Thus, the effective ²²²Rn diffusion coefficient of the BaSO₄ precipitate samples may be similar to the ²²²Rn diffusion coefficient of air.

These facts suggest that the ²²²Rn exhalation rate from the BaSO₄ precipitate surface might be lower than that from the other materials. Here, the ²²²Rn exhalation rate from the precipitate surface was evaluated using the following formula (Eq. 7)³⁴). The values obtained in the experiment of the ²²²Rn emanation coefficient were used

for this calculation.

$$F_{\text{Rn}} = \frac{C_{\text{int}} \cdot V_C \cdot \lambda_{\text{Rn}}}{\left\{ S_{\text{BaSO}_4} \cdot \left[t_e + \left(\frac{e^{-\lambda_{\text{Rn}} \cdot t_e} - 1}{\lambda_{\text{Rn}}} \right) \right] \right\}} \quad (7)$$

Where, F_{Rn} is the ²²²Rn exhalation rate (Bq m² s⁻¹), C_{int} is the integrated ²²²Rn activity concentration as measured by passive detectors (3.2×10^7 Bq m³ s), V_C is the effective volume of the accumulation chamber (m³), λ_{Rn} is the decay constant of ²²²Rn, t_e is the exposure time (s), S_{BaSO_4} is the area of the sample (1.5×10^{-3} m²). Thus, the ²²²Rn exhalation rate from the precipitate was evaluated as 1.8×10^{-5} Bq m⁻² s⁻¹.

According to the report by Iwaoka *et al.*, the median of the ²²²Rn exhalation rate from building materials made of tuff was found to be 4×10^{-5} Bq m² s⁻¹³⁴). Hassan *et al.* reported that the average value of ²²²Rn exhalation rate from the building materials made of granite was 4×10^{-4} Bq m² s⁻¹³⁵). Furthermore, the average value of the ²²²Rn exhalation rate from Japanese soil surfaces was reported to be approximately 9×10^{-3} Bq m² s⁻¹³⁶). Thus, the exhalation of ²²²Rn from the surface of the precipitate may be negligible. That is, radioactive equilibrium between ²²⁶Ra and ²²²Rn would be reached after 30 days in a BaSO₄ precipitate. This estimated result suggests that the sealing method is not significant influence factor on ²²⁶Ra activity analysis by the barium co-precipitation method.

3.2. Contribution of the uncertainty of each parameter to the uncertainty of ²²⁶Ra activity

The contribution of the uncertainty of each parameter to the uncertainty of ²²⁶Ra activity is shown in Table 2. These contributions were evaluated by dividing the values of residuals of each parameter which was calculated by the Kragten approach by the uncertainty of ²²⁶Ra activity³⁷). The uncertainty of ²²⁶Ra activity was significantly influenced by the chemical recovery and the peak efficiency of the HPGe detector. The contributions of these parameters to the uncertainty of ²²⁶Ra activity were found to be 35.9-79.9% and 19.1-62.1%, respectively. The chemical recovery of six samples ranged from 75 to 102% as shown in Table 1. That is, this value fluctuates depending on the sample preparation each time. On the other hand, the same HPGe detector was used for the measurement of each sample. Although the peak efficiency depends on the detector used for the sample measurement, it does not depend on the different samples. Furthermore, the uncertainty of the ²²⁶Ra activity was not influenced significantly by the sample counting time and the background counting time when these measurement times were set as 86,400 and 1,036,800 s, respectively.

Table 2. The summary of the contribution of the uncertainty of each parameter to the uncertainty of ^{226}Ra activity (a) The results obtained by “Without seal”. (b) The results obtained by “Seal-1”. (c) The results obtained by “Seal-2”. The definitions of these symbols in this table are same with those in the Eq. (4)

Sealing technique	Nuclide (Gamma-ray energy)	Contribution of the uncertainty of each parameter to the uncertainty of ^{226}Ra activity: (%)					
		N_t	N_b	η_p	η_f	R	f
(a)	^{214}Pb (295 keV)	0.6 – 1.9%	0.0%	21.0 – 62.1%	0.1 – 0.3%	35.9 – 78.3%	0.0%
	^{214}Pb (352 keV)	0.4 – 1.2%	0.0%	20.6 – 61.9%	0.1 – 0.3%	36.8 – 78.9%	0.0%
	^{214}Bi (609 keV)	0.6 – 1.8%	0.0%	19.1 – 58.8%	0.4 – 1.4%	38.2 – 79.9%	0.0%
(b)	^{214}Pb (295 keV)	0.6 – 1.9%	0.0%	21.0 – 62.1%	0.1 – 0.3%	36.0 – 78.3%	0.0%
	^{214}Pb (352 keV)	0.4 – 1.1%	0.0%	20.6 – 61.9%	0.1 – 0.3%	36.8 – 78.9%	0.0%
	^{214}Bi (609 keV)	0.6 – 1.8%	0.0%	19.1 – 58.8%	0.4 – 1.4%	38.2 – 79.9%	0.0%
(c)	^{214}Pb (295 keV)	0.6 – 2.0%	0.0%	21.0 – 62.0%	0.1 – 0.3%	35.9 – 78.2%	0.0%
	^{214}Pb (352 keV)	0.4 – 1.2%	0.0%	20.6 – 61.8%	0.1 – 0.3%	36.8 – 78.9%	0.0%
	^{214}Bi (609 keV)	0.6 – 1.9%	0.0%	19.1 – 58.8%	0.4 – 1.4%	38.2 – 79.9%	0.0%

4. Conclusions

The effect of the different sealing methods for ^{226}Ra activity analysis in BaSO_4 precipitate samples obtained by the barium co-precipitation method was discussed in this study. ^{226}Ra activities obtained by the sealing methods in EPA-ORP and Hirosaki University were not observed as significantly different compared with the values obtained without sealing. The ^{222}Rn exhalation rate from the surface of the BaSO_4 precipitate was evaluated using the values obtained in the experiment of ^{222}Rn emanation coefficient, and the value was found to be $1.8 \times 10^{-5} \text{ Bq m}^{-2} \text{ s}^{-1}$. Since this is one of the factors that influence the exhalation rate and was found to be much lower than that in other materials, the evaluated exhalation rate was considered to have been much lower than that from other materials. The results obtained by different approaches compared to previous research did not contradict the previous reports. Thus, the sealing method was not a significant influence factor to the ^{226}Ra activity analysis by the barium co-precipitation method. The ^{226}Ra activity in each precipitate sample and the standard solution was not observed as significantly different in the range of the 95% confidence level. The uncertainty of ^{226}Ra activity was influenced significantly by the chemical recovery as the chemical recovery fluctuated from 75% to 102% depending on sample preparation each time.

Conflict of Interest Disclosure

All the authors report no conflicts of interest.

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