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原　著

Determination of Radon Concentration in Air Using a Liquid Scintillation Counter and an Activated Charcoal Detector

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活性炭型検出器と液体シンチレーションカウンタを用いた空気中ラドンの測定

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要　旨

温泉法および鉱泉分析法指針（改訂）では，地下水中ラドン濃度が74 Bq/kg以上で温泉，111 Bq/kg以上で療養泉に該当すると規定されている．ラドン（^{222}Rn）は希ガスであり，容易に空気中に揮散することから，放射能泉の利用による被ばく線量評価のためには，簡便で正確な空気中ラドン濃度の測定手法の確立が必須である．鉱泉分析法指針では，水中ラドンの分析方法として，液体シンチレーションカウンタを用いた手法が例示されている．そこで，測定法の普及を図るため，液体シンチレーションカウンタおよび取扱が簡便な活性炭型検出器を用いた空気中ラドンの測定法を検討した．測定条件として，20～2,000 keV，40～2,000 keV，60～2,000 keVの3ウインドウ設定とし，これらのカウント数から0～2,000 keVを外挿して求めた．また，液体シンチレータとしてDPO（2,5-diphenyloxazole）＋POPOP（1,4-bis（5-phenyl-2-oxazolyl）benzene）トルエン溶液とInsta-Fluor Plusを用い，その測定スペクトルから，適正なウインドウ幅設定，液体シンチレータの妥当性を確認するとともに，変換係数や定量限界を算出することが可能となった．計算の結果，液体シンチレータとしてDPO＋POPOPトルエン溶液を

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用いた際の変換係数が1.55，Insta-Fluor Plusを用いた際の変換係数が1.70であった．

キーワード：ラドン，液体シンチレーションカウンタ，活性炭型検出器，放射能泉

Abstract

According to the Japanese Hot Springs Law and the Guideline of Analytical Methods of Mineral Springs (revised), a spring containing more than 74 Bq/kg of radon is regarded as a hot spring and that with a radon level more than 111 Bq/kg is a medical spring. Radon (^{222}Rn) is a rare gas that easily diffuses in air. Therefore, to evaluate the exposure dose due to radioactive spring usage, it is important to establish an easy and accurate method to measure radon concentration in air. The Guideline of Analytical Methods of Mineral Springs requires that a liquid scintillation counter be used to measure radon in water. In this study, the concentration of radon in air was analyzed using an activated charcoal detector (PICO-RAD) and a liquid scintillation counter (ALOKA LSC LB-5) in order to validate and promote the widespread use of the measurement method. The measurements were done at three window settings of 20–2,000, 40–2,000 and 60–2,000 keV and a counting rate of 0–2,000 keV was extrapolated from these values. This study used DPO (2,5-diphenyloxazole) + POPOP (1,4-bis(5-phenyl-2-oxazolyl)benzene) toluene solution and Insta-Fluor Plus as a liquid scintillator. Based on the measured spectrum, we determined that it was possible to set the appropriate window width for validation of the liquid scintillator and for calculation of the conversion coefficient and detection limit. Our results indicated that both liquid scintillators provided quantitatively satisfactory results, and the conversion coefficients were 1.55 for DPO + POPOP toluene solution and 1.70 for Insta-Fluor Plus.

Key words : radon, liquid scintillation counter, activated charcoal detector, radioactive springs

1. Introduction

Radon (^{222}Rn) is a radioactive element derived from the earth and is a chemically inert rare gas. Based on the Japanese Hot Springs Law (HSL) (Government of Japan, 1948) and the Guideline of Analytical Methods of Mineral Springs of Japan (GAMM) (revised) (Ministry of the Environment of Japan, 2002), a spring is considered a "hot spring" when 1 kg of the spring water has a radon level ≧74 Bq (20×10^{-10} Ci, 5.5 Mache). Similarly, if 1 kg of spring water has a radon level ≧111 Bq (30×10^{-10} Ci, 8.25 Mache), the spring is considered to be a "medical hot spring" with "radioactive hot spring" properties. According to the notification article (Ministry of the Environment, Nature Conservation Bureau of Japan, 1982), bathing in a radioactive spring usually helps to alleviate the effects of gout, arteriosclerosis, hypertension, chronic cholecystitis, gallstones, chronic skin disease and chronic gynecologic diseases. Drinking such spring water usually helps in treating gout, chronic digestive disorders, chronic cholecystitis, gallstones, neuralgia, muscle pain and arthralgia.

Even since the Fukushima Dai-ichi Nuclear Power Plant accident of the Tokyo Electric Power Company, there have been growing concerns over low radioactive exposure from the environment. These concerns have led to a demand for reports ensuring the safe use of radioactive springs. Some reports have appeared on the exposure dose and safety in usage of Rousoku Hot Springs in Gifu Prefecture, Masutomi Hot Springs in Yamanashi Prefecture and Toriido Hot Springs near Yunoyama in Mie Prefecture (Shimo *et al.*, 2006 ; Horiuchi *et al.*, 2007 ; Mori *et al.*, 2012). For example, Mori *et al.* (2012) performed a case study at a hot spring facility using a radioactive hot spring. According to their results, only 5.2–18.0% of radon was retained in bath water after it was transported via a pipe from hot spring storage tanks to a bathtub. Furthermore, only 0.25% of radon remained in the bath water after two days, likely because radioactive decay and diffusion into the air increases as a result of bathing and recirculation filtering.

Furthermore, as mentioned above, as soon as water containing radon flows from the ground, the radon is diffused into the air. Therefore, it is necessary to measure radon in the air at radioactive spring facilities to estimate the exposure dose. Hot spring water normally has much higher concentrations of radon than tap water, which is why radon in hot spring water is one of the factors contributing to increased radon concentration in air (Yasuoka *et al.*, 2008). The method for measuring radon in water is well established in the GAMM ; however, currently there is no official method for measuring radon in air. Several studies have reported on the use of activated charcoal detectors and liquid scintillation counters for a concentration analysis of radon in air (Tokuyama and Izumi, 1992 ; Passo and Cook., 1994 ; Koga *et al.*, 1996 ; Iimoto *et al.*, 2005) ; however, this method is highly dependent on the equipment, the model, and manufacturer of the liquid scintillation counter.

The present study focuses on these issues and investigates a suitable analytical method for measuring the concentration of radon in air using an activated charcoal detector and a liquid scintillation counter.

2. Methods

2.1 Exposure conditions in a radon chamber

At the National Institute of Radiological Sciences (NIRS), investigations regarding the crosscheck measurements of radon and thoron are regularly carried out (e.g., Janik *et al.*, 2009). The crosscheck investigation conducted in 2011 had participants from 21 countries and 31 organizations. This investigation was the 4th International Intercomparison Exercise of Discriminative Radon/Thoron Detectors 2011, hereafter referred to as the "Intercomparison NIRS 2011." The results of that investigation have been explained in detail elsewhere (Janik *et al.*, 2012).

For the present study, exposure of the detector in the radon chamber at NIRS was carried out in the same manner as was done for Intercomparison NIRS 2011. The exposure conditions in the radon chamber are listed in Table 1. An experiment on the window width setting was carried out in a simplified radon chamber at the Kobe Pharmaceutical University.

2.2 Measurement conditions and reagents

This study used a PICO-RAD™ radon detector (manufactured by AccuStar Labs) (hereafter referred to as "detector") to collect the radon in the air. The detector is a type that adsorbs radon onto activated charcoal (Fig. 1), providing convenient handling, and high versatility. Furthermore, a liquid scintillation counter (Hitachi Aloka Medical, Ltd., ALOKA LSC LB-5, hereafter referred to as LSC) was also used. The measurement conditions and reagents used in this study are listed as follows :

Table 1　Exposure conditions in the radon chamber and measurement data using the liquid scintillation counter.

Items	Value
Exposure start (t_1)	18/Aug/2011
Exposure stop (t_2)	22/Aug/2011
Exposure time (h)	100
Radon concentration (Bq/m^3)	1,022 ± 59
Temperature (℃)	21.6 ± 0.1
Relative humidity (%)	57.8 ± 0.3
Absolute humidity (g/m^3)	11.0 ± 0.1
Air pressure (hPa)	1,007 ± 3
Adding liquid scintillator (t_3)	24/Aug/2011
Measuring at random (t_4)	24/Aug―13/Sep/2011
Netcps (maximum)	8.06[1]
	6.38[2]
Netcps (minimum)	0.0509[3]
	0.139[4]

[1] DPO + POPOP solution, $t_4 - t_3 = 0.52$d

[2] Insta-Fluor Plus, $t_4 - t_3 = 1.01$d

[3] DPO + POPOP solution, $t_4 - t_3 = 19.95$d

[4] Insta-Fluor Plus, $t_4 - t_3 = 19.10$d

(1) LSC measurement conditions

Measuring time : 10 min
Background correction : yes
Temperature : 15℃
Calculation method : integral bias method
TRIPLE
(20-2,000 keV, 40-2,000 keV, 60-2,000 keV)

(2) Reagents and liquid scintillators

Reagents :
DPO : 2,5-diphenyloxazole
(Wako Pure Chemical Industries, Ltd.)
POPOP : 1,4-bis(5-phenyl-2-oxazolyl)benzene
(Wako Pure Chemical Industries, Ltd.)
Toluene : for the scintillator
(Wako Pure Chemical Industries, Ltd.)

Liquid Scintillators :

① DPO + POPOP toluene solution : DPO (20 g) and POPOP (0.5 g) were dissolved in 1,000 mL of toluene to prepare the scintillator ; after adsorption of the detector, 15 mL of the liquid scintillator was added, as soon as possible, to the detector.

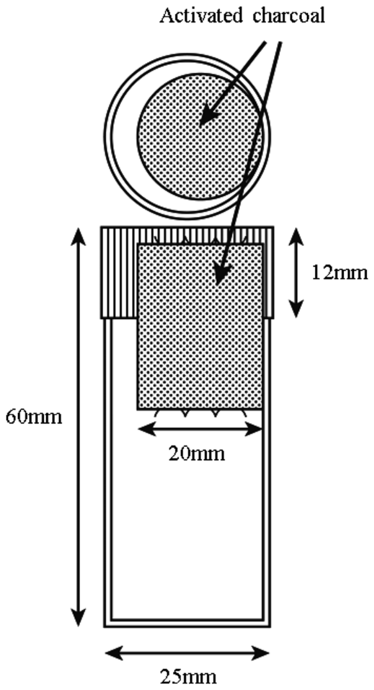

Fig. 1　Schematic drawing and cross-sectional views of the PICO-RAD, activated charcoal radon detector (Iimoto *et al.*, 2005).

② Insta-Fluor Plus : for the scintillator (PerkinElmer Japan Co., Ltd.) ; after adsorption of the detector, 15 mL of the liquid scintillator was added, as soon as possible, to the detector.

Measurements were performed using the LSC while repeatedly changing the type of liquid scintillator in order to determine the effect of different measurement conditions on the measurement values and to determine the attenuation of the measurement values.

2. 3　Calculation of radon concentration

t_1 was the starting time of exposure and t_2 was the ending time of exposure in the radon chamber at NIRS. After the exposure, the detector was brought in as soon as possible, added to the liquid scintillator, and measurements were taken using the LSC. t_3 was the time at which the liquid scintillator was added, and the measurement time was t_4.

With the results obtained from the measurement using the LSC, the net count per second (netcps) was converted into radon concentration according to Eq. (1) (Passo and Floeckher, 1991 ; Shefsky, 1998).

$$Rn(Bq/m^3)=netcps\times\frac{1}{0.7264}\times\exp\left(\frac{0.693\times(t_4-t_2)}{3.825}\right)\times\frac{1}{1-\exp\left(\frac{-(t_2-t_1)}{18/24}\right)}\times\frac{1}{1-\exp\left(\frac{-(t_4-t_3)}{2/24}\right)}\times1\times37\times A \quad\cdots(1)$$

$netcps$: net count per second

t_1 : starting time of exposure in the radon chamber

t_2 : ending time of exposure in the radon chamber

t_3 : Adding time of the liquid scintillator

t_4 : measurement time using the liquid scintillation counter

A : conversion coefficient

The constants in Eq. (1) are as follows : The unit of time is d (for day). The main constant is 0.7264 ; this constant should be expressed in units of counts per second per picocurie per liter [cps/(pCi/L)]. The main constant should be changed because the method in this study was different from the method that was required by the PICO-RAD detector system (Passo and Floeckher, 1991). The conversion coefficient "A" was determined in this study. 1 pCi/L is equivalent to 37 Bq/m^3. The decay constant is the half-life of ^{222}Rn (3.825 d). The absorb constant (18 h) is inversely proportional to the speed with which radon diffuses into the activated charcoal in the vial. The elute constant (2 h) is inversely proportional to the speed with which the scintillator elutes radon from the activated charcoal and the speed with which radon in the scintillator establishes radioactive equilibrium with the decay products of ^{222}Rn.

3.　Results and Discussion

3. 1　Validation of window width setting

A wide-range measurement spectrum from the low-energy region to the high-energy region, i.e., from 0 to 700 keV, is shown in Fig. 2. The measurement spectrum from 0 to 100 keV, which considers only the low-energy region with a noise area, is shown in Fig. 3. The measurement conditions were as follows : liquid scintillator of DPO＋POPOP toluene solution and a measurement time of 10 minutes.

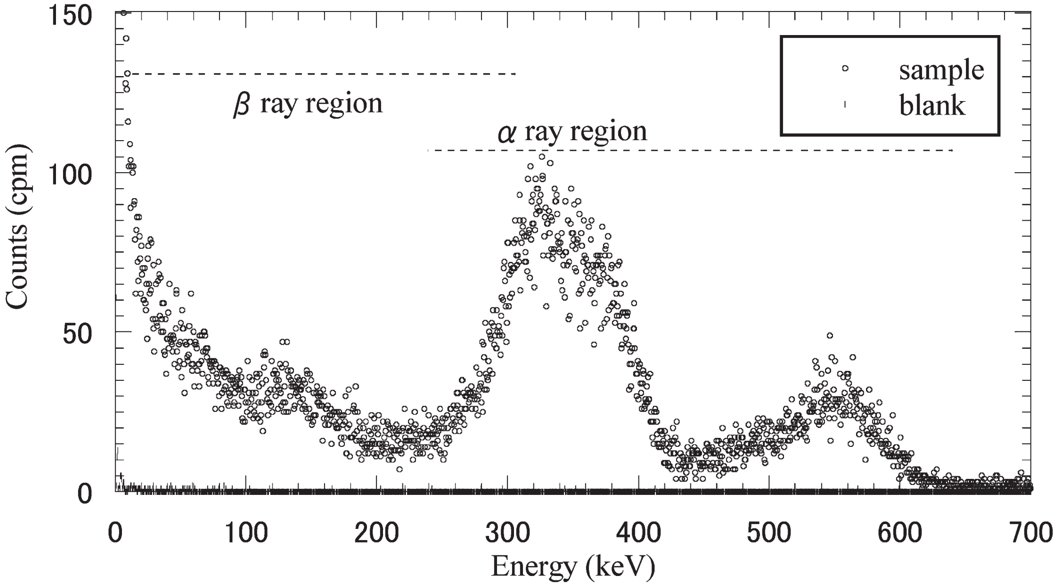

Fig. 2　PICO-RAD measurement spectrum (0–700 keV) for radon of a liquid scintillation counter using a toluene scintillator (DPO+POPOP).

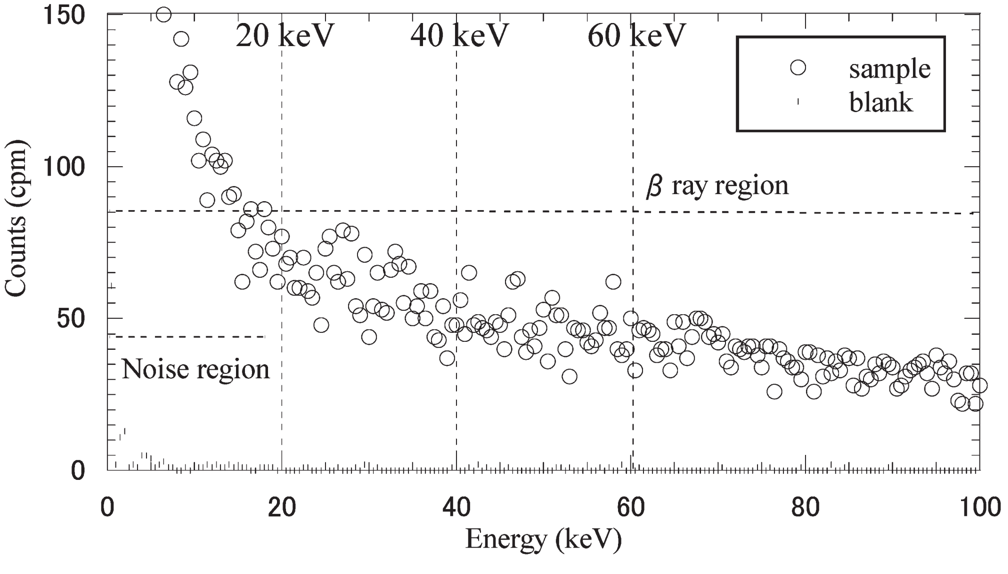

Fig. 3　PICO-RAD measurement spectrum (0–100 keV) for radon of a liquid scintillation counter using a toluene scintillator (DPO+POPOP).

As shown in Fig. 2, in the high-energy region, the region from about 250 keV to about 620 keV was named the α ray region, and this region showed a peak for radon attributable to its daughter nuclides including ^{218}Po and ^{214}Po. On the other hand, as shown in Fig. 3, in the low-energy region, the noise region was observed to be below about 18 keV.

It is important to avoid the low-energy noise region and the high-energy α ray region in each window width setting when using the integral bias method. The measurement conditions in this study involved three window settings at 20–2,000, 40–2,000 and 60–2,000 keV. With these window settings, there was no overlap with the noise region or the α ray region, successfully confirming that the settings were appropriate.

3.2 Effect of time lapse on measurement values

Radon is a radioactive material with a half-life of 3.8 days. Thus, it is desirable to perform a rapid measurement after the radon is absorbed onto the detector. However, since there are various factors involved, such as the transfer from the sampling site to the analysis laboratory, the timing of adding the liquid scintillator, and radioactive equilibrium, it is necessary to investigate any consequent changes in the counting rate.

Thus, each coefficient of variation (CV% ; n = 5) of measurement values at the times that elapsed ($t_4 - t_3$) between adding the liquid scintillator (t_3) and performing the measurement using the LSC (t_4) was determined. The results are shown in Fig. 4, and they demonstrate that a measurement performed immediately after adding the liquid scintillator resulted in an unstable quantitative value. The trend continued for about 20 h after adding the liquid scintillator. Afterward, the quantitative value was stable and the coefficient of variation was also reduced.

In the GAMM, there are no specific values at which the measurement accuracy needs to be maintained ; however, most values of the coefficient of variation in tap water analysis are set to be within 10%, based on government recommendations (Ministry of Health, Labor and Welfare of Japan, 2003). In the present experiment, a coefficient of variation of more than 10% was due to one of the following conditions : (1) the analysis was carried out within approximately 20 h

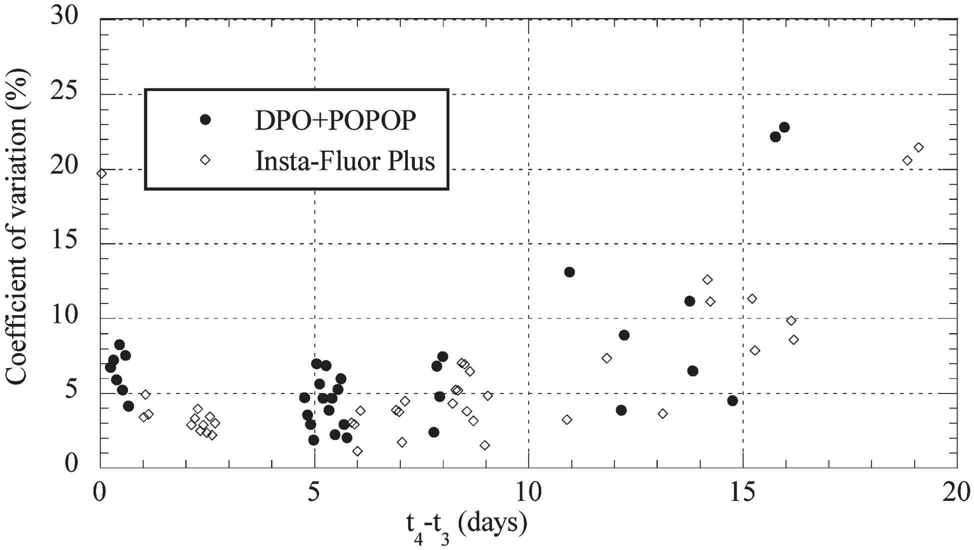

Fig. 4 Correlation between $t_4 - t_3$ and the coefficient of variation (n=5). The X-axis shows $t_4 - t_3$ and the Y-axis shows the coefficient of variation (CV %). $t_4 - t_3$ indicates the period between the time when the liquid scintillator was added to the time of measurement for radon using the liquid scintillation counter.

after adding the liquid scintillator ; (2) t_4-t_3 was more than 10 d (liquid scintillator : Insta-Fluor Plus), or (3) t_4-t_3 was more than 14 d (liquid scintillator : DPO + POPOP toluene solution). Thus, it was concluded that measurements needed to be started 20 h after adding the liquid scintillator and that they needed to be finished within a week of the addition.

3. 3 Detection limit from the background counting rate

Background measurements were carried out by the new detector, which was not exposed in a radon chamber. The measurements using DPO + POPOP toluene solution and Insta-Fluor Plus as the liquid scintillator were carried out for 10 and 60 min, respectively.

The detection limit was calculated using Eq. (2) (American Society for Testing Materials, 1999).

$$N_{DL}=\left(\frac{2.71}{t}+4.65\sqrt{\frac{B}{t}}\right) \cdots\cdots (2)$$

N_{DL} : detection limit (cps)

t : measurement time (s)

B : background counting rate (BG cpm), which is converted to cps (cps)

The results obtained using DPO + POPOP toluene solution are listed in Table 2, and those obtained using Insta-Fluor Plus are given in Table 3.

If the measurement condition is the same as Intercomparison NIRS 2011, then the calculated results reveal that with DPO + POPOP toluene solution, the N_{DL} for 10 min (600 s) of measurement was 21 Bq/m^3 at 0.0593 cps (3.55 cpm), and that for 60 min (3,600 s) was 8 Bq/m^3 at 0.0237 cps (1.42 cpm). In contrast, when Insta-Fluor Plus was used, the N_{DL} for 10 min (600 s) of measurement was 23 Bq/m^3 at 0.0590 cps (3.54 cpm), and that for 60 min (3,600 s) was 9 Bq/m^3 at 0.0231 cps (1.38 cpm).

3. 4 Calculation of the conversion coefficient from Intercomparison NIRS 2011

Exposure of the detector used in this study was carried out in the same manner as in Intercomparison NIRS 2011. For Intercomparison NIRS 2011, the reference value of the radon concentration was obtained from the NIRS standard device, which was calibrated at an

Table 2 Quantification limit of radon calculated by quantitative analysis using DPO+POPOP in toluene solution as the liquid scintillator.

t (s)	600	t (s)	3,600
BG (cps)	0.0831	BG (cps)	0.0878
BG (cpm)	4.99	BG (cpm)	5.27
N_{DL} (cps)	0.0593	N_{DL} (cps)	0.0237
N_{DL} (cpm)	3.55	N_{DL} (cpm)	1.42
N_{DL} (Bq/m^3)	21	N_{DL} (Bq/m^3)	8

t : Measurement time using a liquid scintillation counter
BG : Background measurement results
N_{DL} : Detection limit by the calculation based on Eq (2)

Table 3 Quantification limit of radon calculated by quantitative analysis using Insta-Fluor Plus as the liquid scintillator.

t (s)	600	t (s)	3,600
BG (cps)	0.0825	BG (cps)	0.0828
BG (cpm)	4.95	BG (cpm)	4.97
N_{DL} (cps)	0.0590	N_{DL} (cps)	0.0231
N_{DL} (cpm)	3.54	N_{DL} (cpm)	1.38
N_{DL} (Bq/m^3)	23	N_{DL} (Bq/m^3)	9

t : Measurement time using a liquid scintillation counter
BG : Background measurement results
N_{DL} : Detection limit by the calculation based on Eq (2)

international standard radon facility (Ishimori *et al.*, 2004). To convert the estimated value obtained by Eq. (1) to the reference value, a conversion coefficient A was considered for each measurement condition.

Calculation results showed a difference between the reference value obtained by Intercomparison NIRS 2011 and the measured value obtained for the measurement conditions used in this study. The changing state of the time lapse (t_4-t_3) starting after adding the liquid scintillator to the time of measurement using the LSC is shown in Fig. 5. Calculation results showed that the value more than doubled after 8 days had elapsed for the DPO + POPOP toluene solution, and after 9 days had elapsed for Insta-Fluor Plus. This result showed that it was best for measurements to be completed by the eighth day.

Furthermore, the lowest values of the conversion coefficient A out of the changing numbers were 1.55 (DPO + POPOP toluene solution) and 1.70 (Insta-Fluor Plus). Thus, using the DPO + POPOP toluene solution resulted in a slightly better retention of radon, but both liquid scintillators provide quantitatively satisfactory results. The conversion coefficient applied in this study could serve as a reference for the approximate value for the measurement conditions.

3.5 Comparison of this study and the previous method

Table 4 presents a comparison of the analytical items used in this study and those used in the previous method (Passo and Cook, 1994). This study used the liquid scintillation counter ALOKA LB-5, which had three photomultiplier tubes. The detection limit obtained from this study was lower than that of the previous method. Therefore, when measurements using an activated charcoal detector are performed under the conditions of this study, the observation of radon concentration in air can be carried out more properly.

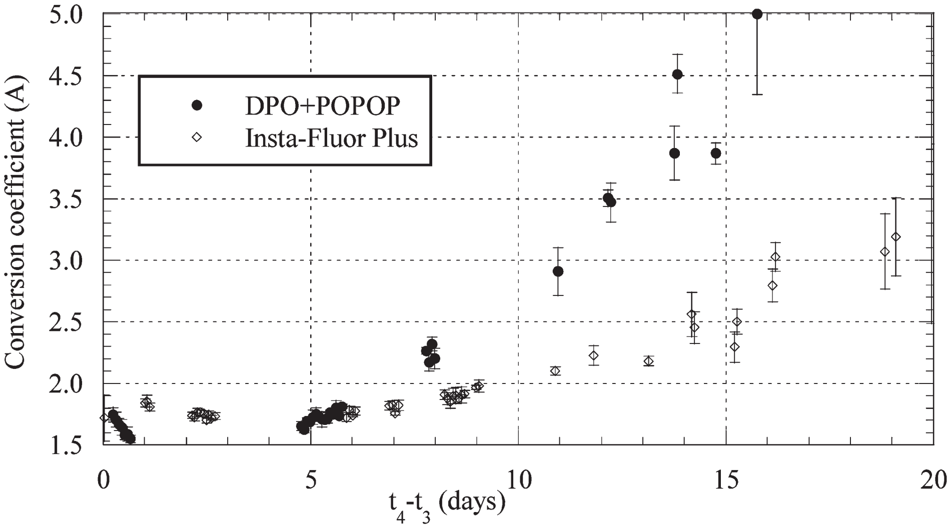

Fig. 5　Correlation between t_4-t_3 and the conversion coefficient for radon measurement.

Table 4 Comparison of the analytical items for radon concentration in the air in this study and the previous method.

Items	Previous method[1]	This study	
		DPO + POPOP toluene solution	Insta-Fluor Plus
Time of exposure in radon chamber $(t_2 - t_1)$	24–48 h	100 h (at NIRS radon chamber)	
Liquid scintillator	Insta-Fluor (discontinued)	DPO + POPOP toluene solution	Insta-Fluor Plus
Quantity of liquid scintillator	10 mL	15 mL[2]	
Time of radon elution $(t_4 - t_3)$	8 h–12d	20 h[3]–10d	20 h[3]–10d
LSC manufacturer ; model	PerkinElmer Life and Analytical Sciences Inc. Tri-Carb Liquid Scintillation Counters	Hitachi Aloka Medical, Ltd. ALOKA LB-5	
LSC measurement range	25–900 keV	Integral bias method[4]	
Conversion coefficient (A)	1	1.55	1.70
Detection limit (Bq/m^3)	below 11.1 Bq/m^3	8 Bq/m^3	9 Bq/m^3

[1] Passo and Cook (1994).

[2] 15 mL of liquid scintillator was the same quantity as used by Iimoto *et al.* (2005).

[3] Koga *et al.* (1996) reported that the time from days of adding the liquid scintillator to measurement days using the liquid scintillation counter $(t_4 - t_3)$ should be more than 20 hours and the finding of this study was in agreement with that.

[4] Integral bias method is the radon-in-water analytical method based on the Guideline of Analytical Methods of Mineral Springs (Ministry of the Environment of Japan, 2002).

4. Conclusion

In this study, exposure of the activated charcoal detector (PICO-RAD) in the radon chamber at the National Institute of Radiological Sciences was carried out. Liquid scintillators, DPO + POPOP toluene solution and Insta-Fluor Plus, were added and an analysis was carried out in each case using the ALOKA LB-5 liquid scintillation counter. The results are summarized as follows :

- Analysis of spectrum measurement values revealed that there was a noise region in the low-energy region that was below 18 keV, and there was an α ray region from about 250 keV to about 620 keV. Based on this finding, three points of energy from 20 to 100 keV should be considered when using the integral bias method with three windows settings, with an extrapolation to 0 keV.
- From a validation experiment for the liquid scintillators (DPO + POPOP toluene solution and Insta-Fluor Plus), a slight difference was found in radon retention ; however, both liquid scintillators were found to provide quantitatively satisfactory results.
- By assuming the reference value from the crosscheck project of the National Institute of Radiological Sciences to be the true value, the conversion coefficient was determined under the measurement conditions (time of exposure in the radon chamber : 100 h, quantity of liquid scintillator : 15 mL). The conversion coefficients were 1.55 for DPO + POPOP toluene solution and 1.70 for Insta-Fluor Plus.
- The most suitable timing of measurement was 1–5 days after adding the liquid scintillator. Furthermore, measurements performed 10 days after adding the liquid scintillator resulted in more than a 10% upward coefficient variation, which was not suitable for measurement.

Even since the nuclear accident at the Fukushima Dai-ichi Nuclear Power Plant of the Tokyo Electric Power Company in March, 2011, there has been an unprecedented demand from the general public for data on environmental radioactivity. Radiation exposure is currently a focus of national attention, and the behavior of radon in air is considered one of the major change factors of natural radiation exposure. As a result, establishing a highly convenient and versatile analytical method for detecting radon in air is necessary to facilitate the spread of reliable information on low exposure dose rates in the environment.

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