การตรวจสอบความถูกต้องของเทคนิคทางนิวเคลียร์บนแผ่นฟิล์ม CR-39 สำหรับวัดการกระจายขนาดของฝุ่นรังสีของนิวไคลด์ลูกหลานเรดอน

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บทคัดย่อ

เทคนิคใหม่สำหรับการตรวจวัดการกระจายขนาดของฝุ่นรังสีของนิวไคลด์ลูกหลานเรดอน และ โทรอน สำหรับการประเมินปริมาณรังสี ได้เลือกและปรับปรุง impactor สำหรับเก็บตัวอย่างฝุ่นขนาดพกพา ที่ประกอบด้วย ชั้นเก็บตัวอย่างจำนวน 4 ชั้น โดยสามารถกัดแยกขนาดอนุภาคของฝุ่นได้ในช่วง 0.5-10 ไมโครเมตร ที่ อัตราการ ไหล4 ลิตรต่อนาที และได้ใช้ CR-39 เป็นหัววัดรังสีแอลฟาจากฝุ่นรังสีของนิวไคลด์ลูกหลานเรดอน และในการ ตรวจวัดเพื่อกัดแยกนิวไคลด์ลูกหลานเรดอนนั้นจะใช้ แผ่นไมลาอะลูมิเนียมตามความหนาที่เหมาะสมกับพลังงาน ของแอลฟาที่ถูกปลดปล่อยจากนิวไคลด์ลูกหลานเรดอน ให้ทำอันตรกิริยาบน CR-39 และได้ติดตั้ง metal wire screen ขนาด 400 mesh ไว้ตรงทางเข้าของอากาศด้านบน impactor เพื่อป้องกัน unattached fraction ของนิวไคลด์ ลูกหลานเรดอน และโทรอน

จากการตรวจสอบความถูกด้องของเทคนิกที่พัฒนาขึ้นนี้กับเครื่องมือที่นิยมใช้ในการตรวจวัดการกระจาย ขนาดของฝุ่นรังสี และผลจากการตรวจสอบทำให้มั่นใจได้ว่าก่าการกระจายตัวของฝุ่นรังสีของนิวไกลด์ลูกหลาน เรคอน และโทรอนที่ได้จากเทกนิกที่พัฒนาขึ้นสามารถนำไปใช้ในการประเมินปริมาณรังสีได้จริง

คำสำคัญ : การกระจายขนาด ลูกหลานเรดอน การประเมินปริมาณรังสี

Verification of CR-39 Technique for Attached Radon Progeny Size Distribution

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Abstract

A new type cascade impactor has been developed to determine the activity size distribution of radon and thoron progeny in a living environment more efficiently. The modified impactor consists of 4 stages for the collection of aerosol samples. The aerosol cut points in the impactor are set for 10, 2.5, 1 and 0.5 μ m at a flow rate of 4 L.min⁻¹. Five CR-39 chips were used as alpha detectors for each stage. In order to separate α particles emitted from radon and thoron progeny, CR-39 detectors were covered with aluminum-vaporized Mylar films. Thickness of film was properly adjusted to allow α particles emitted from radon and thoron progeny to reach the CR-39 detectors. In addition, a 400-mesh metal wire screen was mounted as diffusion collector at the air inlet of the impactor to remove the unattached fraction of radon and thoron decay products.

Validation of the technique was performed with the commercial devices. The results confirmed that the developed technique could provide us significant information to estimate the activity size distribution of attached radon and thoron progeny for dose assessment.

Keywords: activity size distribution, radon progeny, dose assessment

1. Introduction

It is well known that the dose to the public due to radon progeny is the most significant source of natural radiation when taking into consideration the total annual dose. In order to accurately assess the dose due to radon and thoron by dosimetric models, one of the important physical parameters is the activity median aerodynamic diameter (AMAD) derived from the activity-weighted size distribution, as well as the radon and thoron concentrations¹. Method of measuring this physical parameter is different from that for radon and thoron. The cascade impactors are widely used for measuring the size distribution of aerosol particles in environmental pollution and health physics². In order to easily determine the activity size distribution, a new instrument is being developed by modifying the 4-stage portable impactor sampler and applying the allyl diglycol carbonate (commercially known as CR-39) as the alpha detection system for radon and thoron progeny.

The purpose of this study is to verify the developed technique for measuring the activity size distribution of radon and thoron progeny with the commercial device.

2. Structure of impactor sampler

In this study, the 4-stage impactor (Tokyo Dylec Corp.) was selected as the air sampler. The impactor is a round four-jet type with 4 size fractionating stages and a backup filter holder. The cross sectional view of the impactor sampler is shown in Fig. 1(a). The aerodynamic cut-off sizes were calculated as 10, 2.5, 1 and 0.5 µm for the 1st to the 4th impactor stage at a flow rate of 4 L.min⁻¹. The particles smaller than 0.5 µm were collected by the backup filter on the last stage. In this study, the impactor has been slightly modified by inserting a CR-39 detector on the impactor plate as the collection substrate of stage 1 to 4 and on the ceiling of the backup filter holder, facing the glass microfiber filter. In addition, each CR-39 was covered with proper aluminum vaporized Mylar films to detect the target nuclides as follows; CH1 was for Po-218, Po-214, Bi-212 and Po-212, CH2 was for Po-214 and Po-212, CH3 was for Po-212 and CH4 was for all radionuclides. Fig. 1(b) shows arrangement of the detection channel on the impactor plate. Each thickness of film was properly adjusted to let alpha particles reached CR-39 detectors. The test of proper film area density has been reported elsewhere³. The film thickness of each channel is shown in Table 1. This technique provides information on the activity of attached radon and thoron progeny with size separation.



- Fig. 1 (a) Cross-section view of portable cascade impactor.
 - (b) Arrangement of the detection channel on the impaction plate.



Channel	Film Thickness (mg.cm ⁻²)		Cut-off of alpha energy	Radionuclide collection	
	(A)	(B)	(MeV)		
CH1	3.00	1.05	4.2	Po-218, Po-214, Bi-212, Po-212	
CH2	5.00	3.00	6.1	Po-214, Po-212	
CH3	7.10	5.00	7.7	Po-212	
CH4	0.54	0.29	-	Blank	

Table 1 Film area densities absorber for each channel

Remark: (A) Measuring the alpha particles emitted from aerosols and impinges on CR-39. (B) Measuring the alpha particles emitted from the surface of the glass fiber filter and impinges on the CR-39 through an air layer of 22.5 mm.

3. Validation of developed technique

In order to achieve our purpose, a radon chamber of National Institute of Radiological Sciences (NIRS) Japan, was used (about 25,000-L inner volume).⁴ Radon concentration in the chamber was continuously monitored with an interval of 10 min by a commercially available AlphaGUARD (Genitron GmbH, Germany) ionization chamber. The Alpha Guard chamber was calibrated by Physikalisch Technische Bundesanstalt (PTB), Germany.

In order to test the influence of the detection response of the impactor on the presence of ambient aerosols, a condensation monodisperse aerosol generator Model 3472S (TSI Inc., USA) was used. The aerosol particles were generated by the evaporation-condensation method and supplied into the chamber through the sampling port. Carnauba wax was applied as the aerosol material in this study. The continuous particle size distribution was monitored by an Electrical Low Pressure Impactor (ELPI). It was operated at a flow rate of 29.41 L.min⁻¹. The measured 50 % cut-off diameters for the thirteen stages were 0.029, 0.060, 0.105, 0.166, 0.255, 0.373, 0.637, 0.99, 1.61, 2.46, 3.98, 6.60 and 10.20 μ m.

The AMAD of attached radon progeny in this study was verified using the ELPI. The experimental conditions during the test were maintained at 5 kBq.m⁻³ radon concentration, 20 °C temperature, 60% relative humidity and aerosol particles with peak diameter of 0.5 and 1 μ m.

For this study, the impactor sampler and measuring technique described above were performed. It was operated at a flow rate of 4 L.min⁻¹ for 5 minutes. After sampling, it was kept for 4 hours to allow all Bi-214 atoms decay to Po-214.

The CR-39 detectors were etched for 24 hours at 60°C in 6.25 N NaOH solution. The etchpits were counted by a microscope. The Equilibrium Equivalent Radon Concentrations (EERC) were calculated theoretically from the etch-pit counts on the CR-39 detectors that divided into four channels by the equations (1).

$$EERC = \frac{\left(E_{Po-218} \times N_{Po-218 \text{ in } CH1}\right) + \left(\left(E_{Po-214} - E_{Po-218}\right) \times N_{Po-214 \text{ in } CH2}\right)}{\eta \times T \times V} \times K$$
(1)

where the counts of etch-pits on all channels are obtained with E_p Ni-ch, A, T, V, K and η ; in which E_i is alpha energy of i-nuclide in MeV, Ni-ch is number of alpha tracks due to i-nuclide in the detection area of i-channel, T is collecting period in min, V is flow rate in m³.min⁻¹, K is the conversion constant equal to 2.864 x 10⁻⁵ in (Bq.m⁻³.(MeV.m⁻³)⁻¹) and η is the geometrical efficiency, that was estimated to be 11.84% for measuring the alpha particles emitted from aerosols and impinge on CR-39 through the proper Al film and 10.01% for measuring the alpha particles emitted from the surface of the glass fiber filter and impinge on the CR-39 through an air layer of 22.5 mm and the proper Al film. N_{Po-218} in CH1 and N_{Po-214} in CH2 are obtained by the following equations (2) and (3), respectively;

$$N_{Po-218 \ in \ CH1} = N_{CH1} - \left(\frac{1}{0.6406}\right) N_{CH3}$$
(2)

$$N_{Po-214 \ in \ CH2} = N_{CH2} - N_{CH3}$$
 (3)

where N_{CH1} , N_{CH2} and N_{CH3} mean the alpha track counts on CH1, CH2 and CH3, respectively. Therefore, N_{Po-218} in CH1 and N_{Po-214} in CH2 are counts per unit area of alpha track by Po-218 + Po-214 and Po-214 only, respectively. Hence, 0.6406 is the branching ratio of Bi-212 decay to Po-212.

For ELPI, the attached radon progeny was collected on the aluminum foil as a material substance at a flow rate of 29.4 L.min⁻¹ for 5 minutes. After sampling, activity on the aluminum foil from stages 4-8 (for 0.5 μ m) and 6-10 (for 1 μ m) were simultaneously measured using ZnS(Ag) scintillation detectors. The activity concentrations of radon progeny for each stage were analyzed by using the decay method⁵.

During the sampling period of ELPI and 4-stages impactor, a 400-mesh metal wire screen was set at the inlet of each sampler to prevent invasion of unattached progeny, and the collection efficiency was estimated to be 89.1% and 100%, respectively, based on fan model filtration theory⁶. Two or three air samples were collected under each condition.

The particle size distributions were described in terms of a log-normal distribution, defined by the activity median aerodynamic diameter (AMAD) and geometric standard deviation (σ_g). In addition to this numerical evaluation, the impactor data were also evaluated by a graphical method⁷ (Cumulative method).

4. Results and Discussions

To validate the developed technique with the commercial devices e.g. ELPI and MOUDI were conducted to obtain activity size distribution of radon progeny of particular particle size. Results showed in Table 2 indicated that the activity median aerodynamic diameters (AMAD) calculated by developed technique were corresponding with commercial devices from the range of 0.5 to 1 μ m. In addition, the dose conversion factor was calculated using a dosimetric approach⁸ seemed to increase with σ_g (Table 3) but less than 30%. Thus, for the measurements with particle size 0.5 and 1 μ m, this technique (4-stage impactor) was an alternative to ELPI from the viewpoint of dosimeter.

Test ID	A	$MAD(\mu m)$		$\sigma_{_{g}}$	Average AMAD (μ m)	
	ELPI	4-stage impactor	ELPI 4-stage impacto		ELPI	4-stage impactor
0.5 µm;						
1	0.496	0.542	1.36	1.96	0 474+0 022	0.531±0.016
2	0.451	0.520	1.36	1.91	0.474±0.032	
1 µm;						
1	0.995	0.942	1.29	1.77		0.984±0.077
2	1.168	1.073	1.43	1.81	1.048 ± 0.104	
3	0.982	0.938	1.32	1.94		

Table 2 Verification results of developed technique.

Table 3 A comparison of dose conversion factor from dosimetric approach

ELPI		4-stage impactor		Dose conversion factor (nSv/(Bq h m ⁻³))		Difference value of dose
Average AMAD (μm)	$\sigma_{_g}$	Average AMAD (µm)	$\sigma_{_g}$	ELPI	4-stage impactor	(%)
0.474	1.36	0.531	1.94	7.82	10.12	26
1.048	1.35	0.984	1.84	14.73	16.43	11

5. Conclusions

In this study, we verified our technique with the commercial device. Therefore, the measurement technique that we developed in this study may provide significant information to estimate the activity size distribution of attached radon and thoron progeny for dose assessment.

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7. References

- International Commission on Radiological Protection (ICRP), 1994. Human respiratory tract model for radiological protection. ICRP Publication 66, Annals of the ICRP 24(1/4), Oxford: Pergamon Press.
- World Health Organization, 2009. WHO handbook on indoor radon. A public health perspective. Geneva : WHO.
- ชุติมา กรานรอด, 2553. การพัฒนาเทคนิคอย่างง่ายสำหรับการตรวจวัดการกระจายขนาดของ ฝุ่นกัมมันตรังสีของธาตุลูกเรดอนและ โทรอนเพื่อใช้ในการประเมินปริมาณรังสี, วิทยานิพนธ์ ปริญญาดุษฎีบัณฑิต, สาขาวิชาวิสวกรรมนิวเคลียร์ ภาควิชานิวเคลียร์เทคโนโลยี คณะ วิสวกรรมสาสตร์ จุฬาลงกรณ์มหาวิทยาลัย.
- Tokonami, S., Ishimori, Y., Ishikawa, T., Yamasaki, K., Yamada, Y., 2005. Intercomparison exercise of measurement techniques for radon, radon decay products and their particle size distributions at NIRS. Japan J. Health Phys. 40[2], 183-190.
- 5. Thomas, J.W., 1972. Measurement of radon daughters in air. Health Phys. 23, 783-789.
- Cheng, Y.S., Keating, J.A., Kanapilly, G.M., 1980. Theory and calibration of a screen-type diffusion battery. J. Aerosol Sci. 11, 549-556.
- 7. Martin, J.E., 2006. Physics for radiation protection, Wiley-VCH, Germany, p.630.
- Ishikawa, T., Tokonami, S., Nemeth, Cs., 2007. Calculation of dose conversion factors for thoron decay products. J. Radiol. Prot. 27, 447-456.