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Performance Experiments of the Humidity Control System in the ^{220}Rn Chamber

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This paper describes performance experiments of a humidity control system in a thoron ^{220}Rn chamber which adjusts and controls the humidity inside the chamber. The easily constructed and used humidity control system has a perfluorosulfonic membrane (commercially known as Nafion®) tube as the air humidity conditioner. The tube includes tube moisture exchangers that allow the transfer of water vapor between a flowing sample gas stream and a flowing purge gas stream over the exterior surface of the tube. Thus the tube has an advantage of being able to adjust the sample air flow to the desired humidity conditions. We investigated the optimal operation of the air humidity conditioner, including such items as response time to humidity, ability to control the humidity in the air and the losses of ^{220}Rn gas in passing through the air humidity conditioner. Our first experiment showed that the performance of the air humidity conditioner depended on the flow rate conditions of sample and purge air flows. Only small losses due to decay of ^{220}Rn and adsorption of ^{220}Rn gas onto the tube surfaces were also evident. In the second experiment with the ^{220}Rn chamber, we were able to adjust and control the humidity in the chamber using the humidity control system with the air humidity conditioner.

Key words: humidity, Nafion®; sulfonic acid, thoron, time constant

1. Introduction

Thoron (^{220}Rn) is a natural radon (^{222}Rn) isotope with a relatively short half-life (55.6 s), comparing with ^{222}Rn (3.82 days). Although United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) reports estimated that the contribution of ^{220}Rn and its progeny to the total annual effective dose from ^{222}Rn was about 8%[1, 2], in some cases the dose contribution due to the inhalation of ^{220}Rn and its progeny could equal to or even exceeded that of ^{222}Rn and its progeny[3]. In order to determine the radiation hazard from ^{220}Rn and its progeny, it is necessary to make accurate measurements in indoor surveys using proper detectors. Determining the radiation hazard from ^{220}Rn and its progeny has become imperative from the viewpoint of radiation protection. Additionally, developing an accurate measurement procedure for ^{220}Rn and its progeny would be beneficial, because such measurements are overestimated due to interference from ^{220}Rn and its progeny[4].

Passive time-integrating ^{222}Rn-^{220}Rn discriminative

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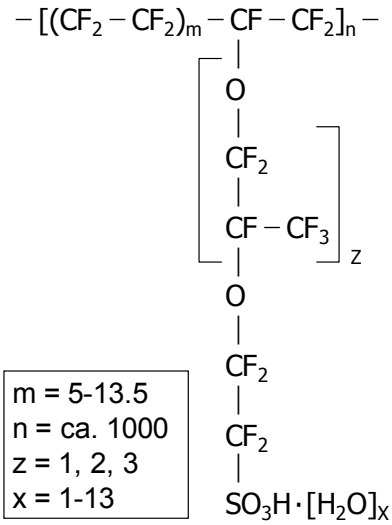

Fig. 1. Chemical structural formula of Nafion®. Obtained from http://www.permapure.com[28].

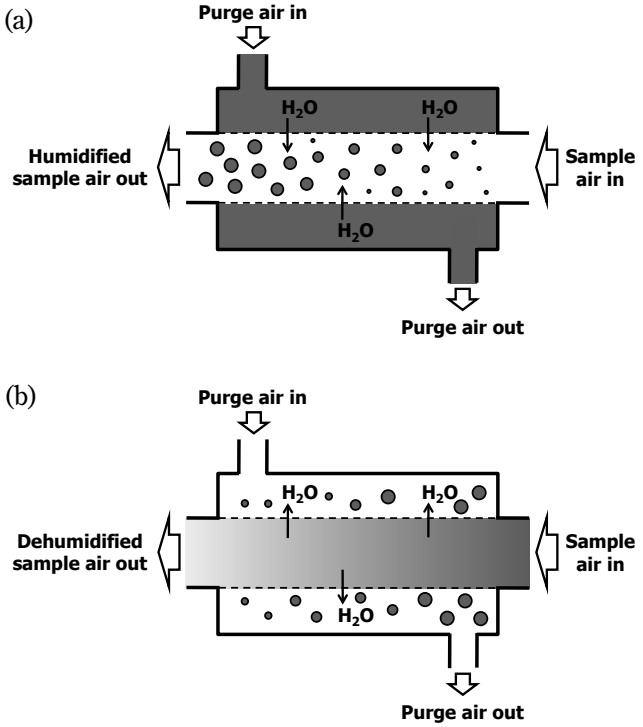

Fig. 2. Schematic diagrams of the air humidity conditioner at (a) humidification and (b) dehumidification experiments.

detectors have been developed[5, 6], which have been used in many worldwide indoor surveys[7-17]. We have concurrently established a ^{220}Rn chamber to calibrate such detectors[18-22] and have checked detector performance[23]. This ^{220}Rn chamber system is able to generate a nearly constant ^{220}Rn concentration, as well as get a certain temperature and humidity in the chamber, depending on humidity in the air passed through the source and the total weight of the source[21]. However, it is difficult to adjust the humidity in the ^{220}Rn chamber while keeping the ^{220}Rn concentration constant, because the humidity condition in the ^{220}Rn chamber depends on the humidity in the air passed through the source.

In recent years, various exposure experiments have been carried out to elucidate the influence of humidity, ambient aerosols and ^{220}Rn on the detection responses of some passive detectors[23-26]. It has also been reported that the response of the passive detectors showed a significant deviation from the true value for the different environmental conditions in laboratory experiments[27]. Accordingly, it is important to establish a chamber in which the environmental conditions can be controlled, because it is necessary to determine how those conditions influence the detection responses of passive detectors. In this study we establish the humidity control system to adjust the environmental conditions while keeping the ^{220}Rn concentration constant and we describe the characteristics of the humidity control system. As well, we investigate the optimal operation and the losses of ^{220}Rn gas in the humidity control system.

2. Experimental method

In order to achieve our purpose, we used a perfluorosulfonic membrane (commercially available as Nafion®) tube (Model MH-050-12F-2, Perma Pure LLC, USA; with 1.1 mm i.d. × 1.3 mm o.d. × 292 mm length) as the air humidity conditioner. The membrane is a copolymer of tetrafluoroethylene and perfluoro-3,6-dioxa-4-methyl-7-octene-sulfonic acid and has the chemical formula shown in Figure 1[28]. The tube consists of tube moisture exchangers that allow the transfer of water vapor between a flowing sample gas stream and a flowing purge gas stream over the exterior surface of the Nafion® tube (Figs. 2a and 2b). Thus the driving force is the difference in water vapor concentration between the inside and outside of the tube by absorption, as water-of-hydration, of the sulfonic acid group in the polymer. The tube has a wide range of applications, such as medical devices, gas analyzers and analytical devices, gas sample conditioning systems for emission and process monitoring, and so on[28].

The optimal operation of the humidity control system depends on the tube size (diameter and length) and the combination of flow rates of the sample and purge air flows. Residence time in one tube used in this study was less than 0.1 s at the flow rate of 1 L min^{-1} for the sample air flow. The performance experiments on the humidity response of the air humidity conditioner were carried out using the experimental setup shown in Figure 3a.

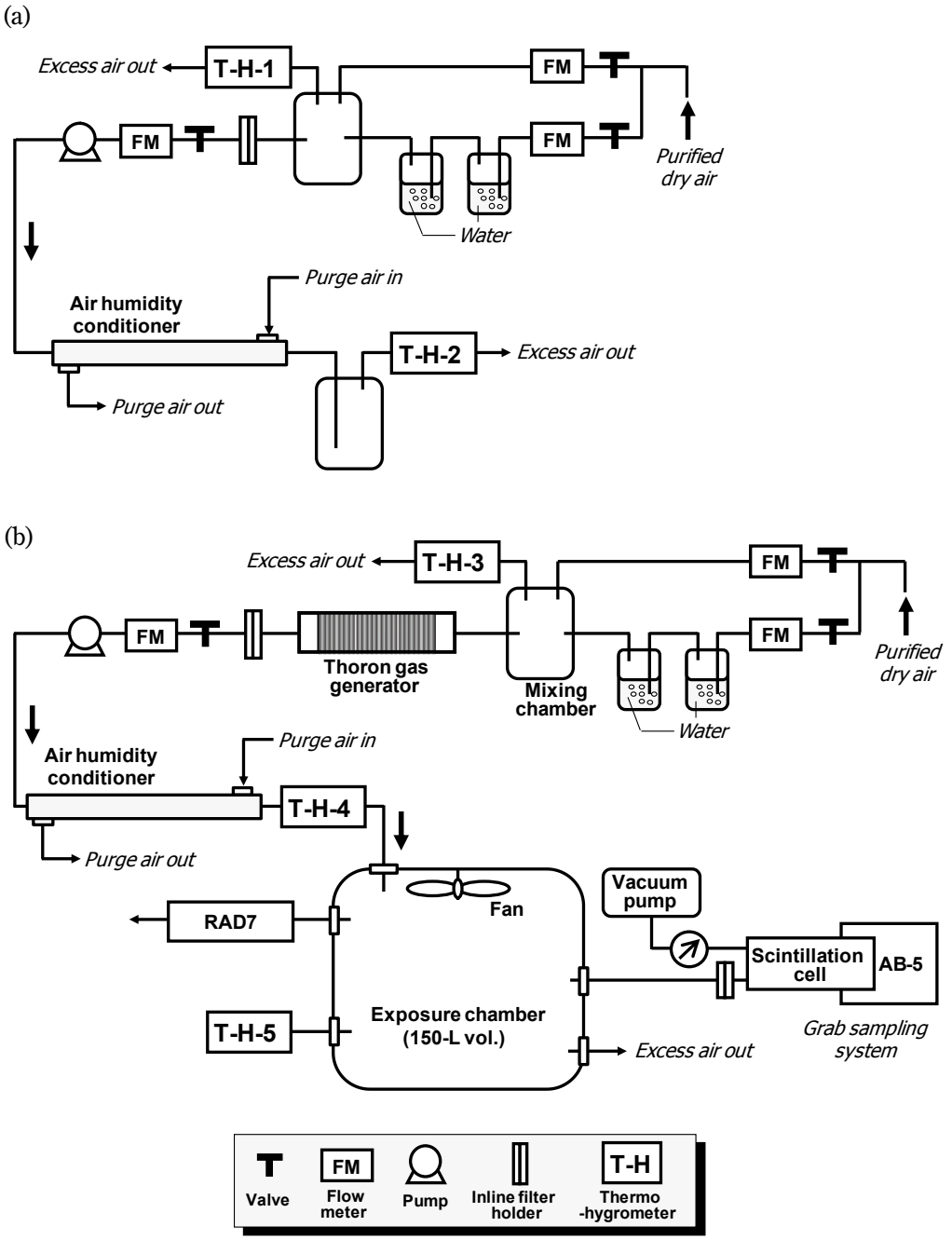

Fig. 3. Schematic diagrams of the experimental setup in (a) the performance experiments for the humidity control system and (b) ^{220}Rn gas generation with the humidity control system.

The chamber air temperature and relative humidity (RH) were monitored at 1-min intervals using a hygrometer Model TR-73U (T & D Corporation, Japan).

In order to evaluate the charactersitics of humidity in the air in the ^{220}Rn chamber using the air humidity conditioner, the ^{220}Rn chamber system of National Institute of Radiological Sciences (NIRS)[21, 22] was used (Fig. 3b). The ^{220}Rn concentrations were measured once every hour using an electrostatic collection (EC) monitor, the RAD7 (Durridge Co. Inc., U.S.A.). In addition to this continuous measurement, intermittent measurements by grab sampling were also made during the experiment using a ZnS scintillation cell (SC) Model 300A (inner volume: 270 mL) with a portable radiation monitor Model AB-5 (Pylon Electronics Inc., Canada) so as to obtain the quality assurances of the measured ^{220}Rn concentration[19]. The chamber air temperature and RH were monitored at 1-h intervals using a hygrometer Model HygroPalm-2

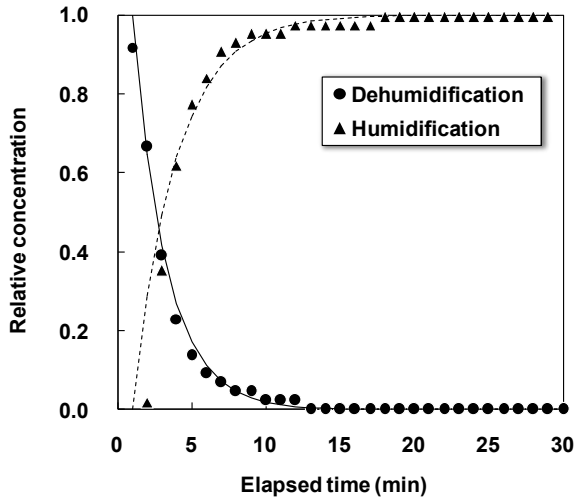

Fig. 4. Responses of the air humidity conditioner to humidity step changes at dehumidification and humidification experiments. The solid and dashed lines are fits of Eqs. 1 and 2 to measurements, respectively.

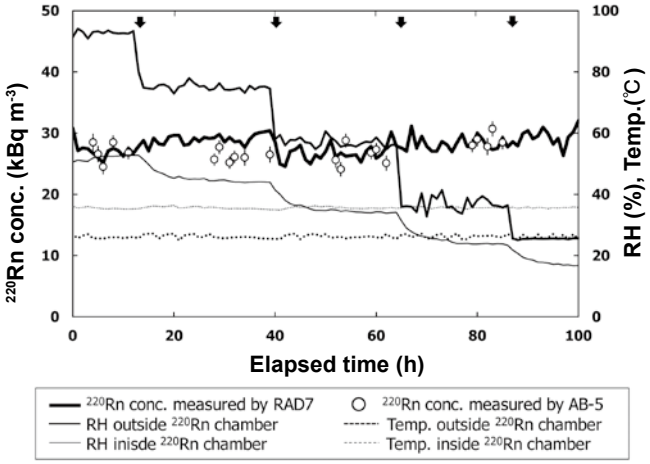

Fig. 5. Time variation of ^{220}Rn concentrations, temperature and relative humidity using the air humidity conditioner. Two sets of air humidity conditioners were used. The flow rates of sample and purge air flows of the air humidity conditioner were set to 1 and 2 L min^{-1}, respectively. The arrows show the time when the humidity in the purge air flow was changed.

(ROTRONIC AG, Switzerland) for the ^{220}Rn chamber.

3. Results and discussion

3.1. Performance experiments of air humidity conditioner

In the ^{220}Rn chamber the flow rate through the ^{220}Rn source was set to about 2 L min^{-1} during generation of ^{220}Rn gas to take into account configuration of the ^{220}Rn chamber[21]. Accordingly, in this study performance experiments of the air humidity conditioner were carried out at the flow rate from 0.5 to 2 L min^{-1}.

In order to derive the response time of the air humidity conditioner, humidified and dried air flows were introduced to the air humidity conditioner for the sample and purge air flows (dehumidification experiments), respectively, and vice versa (humidification experiments).

For the first-order response the formulas for the dehumidification and humidification experiments are Eqs. 1 and 2, respectively,

$$f(t) = \frac{C_{\text{Outlet}}}{C_{\text{Inlet}}} = \exp(-\frac{t - t_0}{\tau}) \quad (1)$$

$$f(t) = \frac{C_{\text{Outlet}}}{C_{\text{Inlet}}} = 1 - \exp(-\frac{t - t_0}{\tau}) \quad (2)$$

where C_{Inlet} and C_{Outlet} are the RHs at the inlet and outlet of the air humidity conditioner in %, respectively, t is time in min, t_0 is time of the humidity step in min and τ is system time constant in min. τ is fitted to dehumidification experiments at time, t_0, and the analogue exponential growth model to humidification experiments at time, t_0, in order to determine τ of the system. With the applied exponential mode, a 95% signal response is achieved after 3τ.

Figure 4 exemplifies the response of the air humidity conditioner to the humidity step change at dehumidification and humidification experimnets. The mean (± standard deviation) temperature was 25.4 ± 0.5 ℃ within 2% relative standard deviation (RSD) for all experiments. As shown in the graph, the measured points at both dehumidification and humidification experiments were fitted to the Eqs. 1 and 2, respectively, in order to determine τ of the air humidity conditioner.

For the flow rate of sample air flow at a constant flow rate of purge air flow (runs 1 and 4, 3 and 5), the equilibrium RH and τ at dehumidification experiments became higher with increasing the flow rate of sample air flow. The inverse tendency at humidification experiments was observed for the equilibrium RH. This indicates that the higher flow rate ratio of purge to sample air flows exchanged more water vapor over the exterior surface of the tube, and consequently, lower (higher) equilibrium RH at dehumidification (humidification) experiments was achieved and faster response was found.

The variability in equilibrium RH and τ associated with the flow rate ratio of purge to sample air flows was examined. By increasing the flow rate of purge air flow at a constant sample flow rate of 1 L min^{-1} (runs 1, 2 and 3), which means the flow rate ratio of purge to sample air flows, a decrease in equilibrium RH at dehumidification experiments and the increase at humidification experiments was observed, while the correspomding τ values seemed to become short at both experiments. Thus equilibrium RH and τ of the sample air flow depended on the flow rate ratio of purge to sample air

Table 1. Equilibrium relative humidity (RH) and time constant (τ) at given flow rates of sample and purge air flows of the air humidity conditioner ($F_{Sam.}$ and $F_{Pur.}$) at dehumidification and humidification experiments[a]

Run	$F_{Sam.}$	$F_{Pur.}$	N[b]	Dehumidification experiment			Humidification experiment		
				C_{Inlet}[c, d]	$C_{Eq.}$[c]	τ	C_{Inlet}[c, d]	$C_{Eq.}$[c]	τ
	(L min^{-1})			(%)	(%)	(min)	(%)	(%)	(min)
1	1	0.5	1	95	61 (1)	5.4 (0.0)	10	26 (1)	7.1 (2.7)
2	1	1	1	95	55 (2)	2.8 (0.4)	10	40 (2)	3.3 (0.3)
3	1	2	1	95	54 (2)	3.1 (1.1)	10	52 (1)	3.0 (0.1)
4	0.5	0.5	1	95	43 (1)	4.1 (0.7)	10	53 (1)	3.5 (0.2)
5	2	2	1	95	65 (4)	3.6 (0.6)	10	34 (0)	2.1 (0.2)
6	1	2	2[e]	95	36 (1)	2.6 (0.2)	10	76 (3)	2.7 (0.0)

[a] Values in parentheses are standard deviations (n = 2 except run 2 (n = 4)).
[b] Number of air humidity conditioners.
[c] C_{Inlet} and $C_{Eq.}$ represent the RH at the inlet of the air humidity conditioner and the equilibrium RH measured at the outlet of the air humidity conditioner, respectively. The RHs at the inlet and outlet of the air humidity conditioner were measured using the hygrometers "T-H-1" and "T-H-2" shown in Fig. 3a, respectively.
[d] Detectable range of the Model TR-73U hygrometer ranged from 10 to 95% RH. When the readings from the hygrometers were out of the detectable range, the values of C_{Inlet} were represented as upper and lower detection limits.
[e] Two sets of air humidity conditioners were connected in series.

Table 2. Mean (standard deviation) ^{220}Rn concentration and relative humidity (RH) in the ^{220}Rn chamber using two sets of air humidity conditioners[a]

RH[b]	^{220}Rn concentration[c]			^{220}Rn ratio of EC to Cal.
	EC	SC	Cal.	
(%)		(kBq m^{-3})		(−)
52.5 (0.2)	27.2 (1.3)	25.8 (0.5)	25.8 (0.5)	1.05 (0.05)
44.7 (0.7)	29.0 (0.8)	26.2 (0.9)	26.5 (0.9)	1.09 (0.05)
34.6 (0.7)	26.8 (1.3)	26.5 (0.8)	26.5 (0.8)	1.01 (0.06)
24.3 (0.8)	28.2 (1.1)	27.0 (1.0)	27.0 (1.0)	1.04 (0.06)
17.2 (0.4)	29.4 (1.4)	NM[d]	26.0 (0.9)	1.13 (0.07)
RSD (%)[e]	4.0	1.9	1.8	

[a] All data are derived from Fig. 5.
[b] The values were the equilibrium RH after changing RH outside the chamber. The RH in the ^{220}Rn chamber was measured using the hygrometer "T-H-5" shown in Fig. 3b.
[c] "EC", "SC" and "Cal." respectively represent the ^{220}Rn concentrations measured by the RAD7 electrostatic collection monitor, the 300A scintillation cell with the AB-5 portable radiation monitor, and calculated from the equation reported by Sorimachi et al.[21] [C_{Tn} = 0.592exp(0.234AH), where C_{Tn} and AH are the ^{220}Rn concentration and absolute humidity in the chamber, respectively].
[d] Not measured.
[e] Relative standard deviation.

flows of the air humidity conditioner.

The effect of the number of air humidity conditioner on the variability in equilibrium RH and τ was investigated (runs 3 and 6). From Table 1, the efficiency of both dehumidification and humidification experiments by the air humidity conditioner increased by approximately 30% with an increase of the number of air humidity conditioners.

3.2. Time variation of ^{220}Rn and RH in the ^{220}Rn chamber using the air humidity conditioner

The variations of the temperature and RH are plotted as a function of time in Figure 5. Using Figure 3b, we gradated the RH outside the chamber from 93% to 26% by passing through two sets of air humidity conditioners in series at sample and purge flow rates of 1 and 2 L min^{-1}, respectively. As a result, a systematic temporal pattern was observed that the RH inside the chamber decreased with time until achieving equilibrium within the first 6 h after changing the humidity outside the chamber. Using Eq. 1, we estimated τ for the chamber to be approximately 4 h. These may correspond to the residence time in the ^{220}Rn chamber (2.5 h) and the time to reach equilibrium of adsorption and desorption of water vapor on the chamber surfaces. Accordingly, the air humidity conditioner was able to make adjustments from 17 to 53% RH inside the ^{220}Rn chamber by changing outside RH from 26 to 93%.

The variations of ^{220}Rn concentrations are also plotted as a function of time in Figure 5. The ^{220}Rn concentration remained nearly constant for about 100 h, with RSD of 4%. This stability of the ^{220}Rn concentration was in agreement with results reported by Sorimachi *et al.*[21]

The adsorption of ^{220}Rn gas in the sample air on the

interior surfaces of the tube was evaluated. From Table 2, the ^{220}Rn concentration with the air humidity conditioner at each RH level was 28.1 (±1.1) kBq m^{-3}, which was measured by the RAD7 monitor. On the other hand, the estimated ^{220}Rn concentration without the air humidity conditioner was 26.4 (±0.5) kBq m^{-3}, which was calculated from the expression reported by Sorimachi *et al.*[21] [C_{Tn} = 0.592exp (0.234AH), where C_{Tn} and AH were the ^{220}Rn concentration and absolute humidity in the chamber, respectively]. From Table 2, the ^{220}Rn concentration ratio of the measured value to the estimated value was constant with a value of approximately 1 at 17-53% RH. At a sample flow rate of 1 L min^{-1} with one air humidity conditioner, the losses of ^{220}Rn gas due to decay were less than 0.1%. Thus, the losses of ^{220}Rn gas can be ignored in passing the air through the air humidity conditioner.

4. Conclusions

We have established and performed the humidity control system using the Nafion® tube as the air humidity conditioner in the ^{220}Rn chamber. The conditioner has an advantage of being able to adjust the sample air flow to the desired humidity conditions. Consequently, there were only small losses due to decay of ^{220}Rn gas and adsorption of ^{220}Rn gas in the sample gas on the tube surfaces. The humidity control system was also able to adjust and control the humidity in the ^{220}Rn chamber. The feature of the ^{220}Rn chamber with the humidity control system satisfies the experimental conditions carried out in the calibration experiments[29], intercomparison experiments[30, 31] and performance experiments of the passive radon and thoron detectors[24-26] and then is available to similar experiments to them. Thus the technique established through the present study may be able to supply useful information about the generation technique of ^{220}Rn gas in the ^{220}Rn chamber for calibration and performance checks of passive time-integrating detectors.

Acknowledgments

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