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Original Article

A preliminary study on real-time Rn/Tn discriminative detection using air-flow delay in two ion chambers in series

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ABSTRACT

Due to its short half-life, thoron gas has been assumed to have negligible health hazards on humans compared to radon. But, one of the decay products with a long half-life can make it to be transported to a long distance and to cause a severe internal dose through respiration. Since most commercial radon detectors can not discriminate thoron signals from radon signals, it is very common to overestimate radon doses which in turn result in biased estimation of lung cancer risk in epidemiological studies. Though some methods had been suggested to measure thoron and radon separately, they could not be used for real-time measurement because of CR-39 or LR-115. In this study, an effective method was suggested to measure radon and thoron separately from the free air. It was observed that the activity of thoron decreases exponentially due to delay time caused by a long pipe between two chambers. Therefore from two ion chambers apart in time, it was demonstrated that thoron and radon could be measured separately and simultaneously. We also developed a collimated alpha source and with this source and an SBD, we could convert the ion chamber reading to count rate in cps.

1. Introduction

Radon (radon-222, Rn) is a decay product of ^{238}U presents in soil and rocks of granite specially. When radon gas is inhaled, high LET(linear energy transfer) alpha particles emitted by its short-lived decay products can interact with the biological tissue of a lung leading to DNA damage. Some degree of DNA damage can greatly increase the pool of cells available for the development of cancer [1]. According to the recent review of the International Agency for Research on Cancer (IARC), radon is one of the groups – 1 carcinogenic agents [2]. Epidemiological studies confirmed that radon in homes increased the risk of lung cancer in the general population. The proportion of all lung cancers linked to radon was estimated to lie between 3% and 14% depending on the average radon concentration in the country [1].

Collaborative analysis of individual data from 13 case-control studies of residential radon and lung cancer in 9 European countries where 7,148 cases of lung cancer and 14,208 controls, it was found that for the case of lung cancer the mean concentration was 104 Bq/m^{3}and the risk of lung cancer increased by 8.4% per 100 Bq/m^3 [3]. EPA (US Environmental Protection Agency) estimated that out of a total of 157,400 lung cancer deaths nationally in 1995, 21,100 (13.4%) were radon-related [4].

Since radon half-life is 3.82 days and thoron (radon-220, Tn) half-life is 55.6, radon can move a longer distance than thoron before their decay. For this reason, it was believed that thoron could not travel from its production site to the close environment of human beings. Hence, thoron was treated less significantly for radiation exposure evaluation and sometimes its contribution to dose calculation was totally omitted. But, the relatively long half-life of one of its decay products, ^{212}Pb (10.6 h), allows it to migrate away from its source before converting into an important alpha-emitter ^{212}Bi (60.6 min) [5,6] which plays an important role in human health like radon. Several studies have shown thoron emission can be higher than radon emission depending on many conditions. Syarbaini et al. [7]showed radon exhalation rates were in the range

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3.73 ~ 326 mBq.m^{-2}s^{-1} whereas thoron exhalation rate varies from 144 ~ 9,470 mBq.m^{-2}s^{-1}.M. Hosoda et al. [8] showed thatthe average exhalation rate of radon and thoron at seven measuring points in Kobe city were 12 ± 6 mBq.m^{-2}s^{-1} and 1,210 ± 1,110 mBq.m^{-2}s^{-1} respectively. M. Chege et al. [9] showed that in Mrima Hill radon concentration varies from 16 to 56 Bqm^{-3} and thoron concentration varies from 132 to 1,295 Bqm^{-3}.S. Gierl et al. [10] showed that thoron concentration varies from 10 to 90 Bqm^{-3} at 20 cm distance from the wall which contributes significantly to the inhalation dose of the dwellers 0.6 ~ 4 mSva^{-1}.

As radon and thoron decay products are positively charged, they can attach to tiny atmospheric aerosol and form radioactive aerosol. Depending on the diffusion properties of the aerosol, the decay products present in the air deposit in the nasal cavities, on the walls of bronchial tubes, and in the deep lung which then emit alpha particles. The energy deposited by these alpha particles then leads to severe health effects [11]. For this reason, both radon and thoron have the same importance in lung cancer risk estimation. As thoron is everywhere with radon and it is possible to get a higher concentration of thoron than radon in different places, thoron concentration should consider during risk assessment of radon.

Though lung cancer risk from radon is higher than that of thoron, we cannot skip the risk from thoron due to its alpha emitting daughter nucleus. If the concentration of radon and thoron are not differentiated well, the detector can detect both radon and thoron at the same time and can give overestimated data. In that case, the lung cancer risk will be given as biased estimate during epidemiological study.

A few years ago in Korea, there was a nationwide commotion of the so called ‘radon bed’. Somel bed companies in Korea had often used packs of monazite powder as special components of their mattresses. Monazite had been known to produce negative ions which were believed to give a healthy effect when inhaled. One day, a customer coincidentally detected radon around his bed and it has been known to the public through newspapers and broadcastings. Then KINS (Korea Institute of Nuclear Safety) had been ordered to investigate radon activities of huge number of beds in use and reported that there could be hazardous effect when one sleeps on such a bed for a long time. Since this investigation, nearly all the bed with monazite were retrieved and a new regulation on the use of monazite was made. Now it is recommended by law in Korea, new radon detectors should be able to measure thoron concentration separately from radon concentration.

There are many types of radon detectors to measure radon concentration in air depending on the measurement time scale. Some of them can be operated in real-time measurement and some other measure the radon activity on a long-term accumulated filter. Among them, AirThings Corentium Home, FT Lab RadonEye, Rad7 etc. are real time measuring system. Most of radon detectors do not have a capability to detect radon and thoron separately. For these reasons, they cannot distinguish thoron from radon so the radon activity measured may include thoron activity or purely thoron itself. Also most of such detectors depend on the natural diffusion process. But diffusion process varies depending on atmospheric temperature, pressure, humidity, and turbulence, etc. Hence, it is very much possible to get inaccurate measurement from most of those detectors. So, it is demanded to develop a detector in which both radon and thoron can be measured separately and in real-time so that the long-term environmental dependency can be minimized.

To discriminate Radon and Thoron measurements, there has been several ideas developed in the past. New York University developed a detector system that had 4 separate ion chambers for duplicate measurement of radon and a total gas (radon plus thoron). Each chamber had a 9 mm square CR-39 film for alpha detection. Then the thoron concentrationwas calculated by the signal difference between the two sets of measurement [12].G. Sciochheti et al. [13] demonstrated another thoron and radon detector based on 25 mm square CR-39 films where a piston was used to balance the radon gas inside and outside of the diffusion chamber. The gas sampling active mode based on syringe functionality was used for sampling radon in various areas. A small air inlet hole acted as a barrier of thoron due to its short half-life. B.K. Sahoo et al. [14] described an LR-115 track detector which could give a time-integrated measurement of radon and thoron and has a single entry for gas. The device consisted of two identical chambers of which the first chamber is the reference chamber and the second one is the pin-hole chamber. Gas enters into the first chamber through a filter paper and then into the second chamber through a pin-hole. Alphas from radon and thoron were measured by using 3 cm square LR-115 films. S. Tokonami et al. [15] developed a CR-39 film-based detector that has two different diffusion chambers, where radon gas in the air diffuses into one chamber through an invisible air gap between its lid and the bottom which create a high barrier for thoron to enter into the chamber. To detect thoron, 6 holes of 6 mm diameter are opened at the side of the other chamber.

All the above detection systems use CR-39 or LR-115 films, tracking detectors, so they need a chemical etching process. Hence the process is time consuming and expensive. Those systems are not good for real time detection system.

In this study, a new real-time radon and thoron discriminative measurement system(RT-Rn/Tn-DMS) was proposed which uses the difference of half-life of radon and thoron using an air-flow delay pipe and two ion chambers before and after the pipe. The first chamber measures both radon and thoron activity in air sample but the second chamber measures only the activity of radon in the same air sample. Since radon has a very long half-life, its activity does not change through a delay pipe but thoron has a very short half-life so it decays almost completely before reaching the second ion chamber. The difference between the two signals reflects the thoron activity in the air. The proposed system is real time and we don’t need to use any secondary detection system hence the system is accurate and reliable.

There are many advantage of this new detection method over conventional system. 1). the system is real time. 2). it is independent of natural diffusion process. 3). we don’t need high voltage to operate it. 4). we don’t need to use any secondary film or chemical. 5). It is cheap and user friendly.

Like other detector, we have some limitations e.g. sensitivity of the system should be high.

2. Materials & method

2.1. The proposed Rn/Tn discriminative measurement system

The proposed system is basically composed of an air pump, two ion chambers, and 3 pipes, and its schematic is shown in Fig. 1. The air pump is a mechanically vibrating pump, Grovita GAP-603, with a very low pumping speed, ~4 L/min. To make 2 L per minutes flow, the knob was set at the middle of its range. It samples air and send to the first ion chamber and the second ion chamber sequentially through 3 PVC pipes of the 6.6 mm inner diameter (the inner air volume of the pipe is 0.0342 L/m); pipe #1 or the inlet pipe of the first chamber (~1 m long), pipe #2 or the air-flow delay pipe between two chambers (the length is a variable), and finally pipe #3 or the exit pipe of the second chamber. All pipes are equipped with a manual valve each to open and close them.

The ion chamber is a cylinder made of a thin steel foil with the length of 118 mm and the diameter of 104 mm. Hence the physical

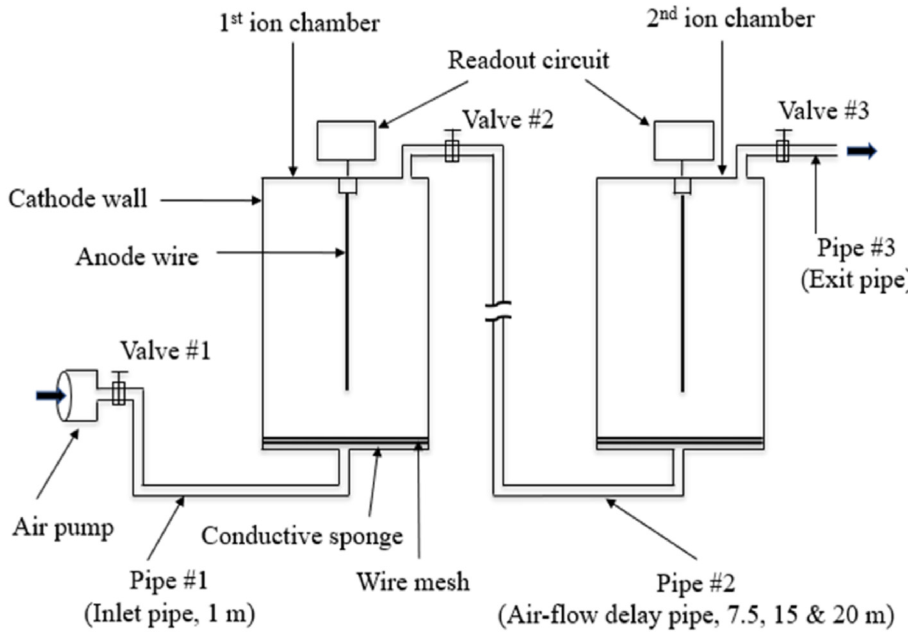

Fig. 1. A schematic diagram of the proposed Radon/Thoron discriminative measurement system showing an air pump, two ion chambers and three pipes.

volume of the chamber,

$$\begin{aligned} V &= \pi r^2 h \\ &= 3.14 \times 5.2^2 \times 11.8 \\ &= 1001.88 cm^3 \\ &= 1.00188\ liter \end{aligned}$$

The length of the anode wire is 77 mm length shorter than the whole length of the cylinder so the sensitive volume of the ion chamber,

$$\begin{aligned} V &= \pi r^2 h \\ &= 3.14 \times 5.2^2 \times 7.7 \\ &= 654\ cm^3 \\ &= 0.654\ liter \end{aligned}$$

During the operation of the system both chambers are placed uprightly so the air flows in from a hole in the bottom surface and leaves through a hole in the top surface. A conductive sponge with a metal mesh was connected to the cathode wall electrically and was installed on the bottom surface where the air inlet hole is located. The function of conductive sponge and the metal mesh is to remove positively charged aerosols in the air produced by daughters of radon and thoron. Otherwise these daughters (radioactive Po, Pb, Bi etc.) may undergo alpha decay during the operation of the system then add the extra alpha signals to the ion chamber in addition to radon or thoron signals. Therefore the removal of these charged aerosols is necessary from the air before entering the sensitive volume of the ion chambers. Table 1 shows the physical dimensions of the components of the proposed system in the order of air flow.

The anode was biased to +12 V and the cathode wall, the top and bottom surfaces and conductive sponge and the metal mesh were grounded. The radiation detector signal is basically an induced current pulses made by the ionization of air by alpha particles in the sampled air but because the proposed system works in continuous sampling mode so the anode signal can be read as a slowly varying saturation voltage by a current-sensitive amplifier circuit with a 10 kOhm load resistance at the output stage. Then the signals from both ion chambers were continuously sampled and recorded into a PC through a BNC adaptor, BNC-2110 and a Multi-function I/O device, PCI-6221 (both made by National Instrument) by a custom-made LabView program. Finally the voltage output of both ion chambers are calibrated by a certified Am-241 check source and a silicon surface barrier detector of Ortec Alpha Spectrometer Alpha Aria. The Fig. 2. shows the air pump and two chambers with a long yellow helical (air-flow delay) PVC pipe and a part of two transparent (inlet and exit) pipes.

2.2. Test sources of thoron and Radon and a calibration source

To demonstrate the proposed Rn/Tn discriminative measurement system, a thoron source and a radon source were prepared as shown in Fig. 3 (a) and (b). Korean land is mainly possessing granite rocks and the average concentration of U and Th is 2.3 and 11.7 ppm respectively [16]. The prepared monazite powder has 5.47 wt% ThO_2 and 0.34 wt% U_3O_8 [17,18] suitable as a thoron gas emitter for the test of the system. The thoron concentration in the air was controlled by the amount of the monizite power put in the tray. Also a Pylon RN-1025 calibrated gas source was used as a radon

Table 1
Physical parameters of pipes and chambers in the order of air flow.

Pipe #1		First chamber	Pipe #2		Second chamber
Length (m)	Volume (liter)	Volume (liter)	Length (m)	Volume (liter)	Volume (liter)
1.0	0.034	1.002	7.5	0.255	1.002
			15	0.51	
			20	0.68	

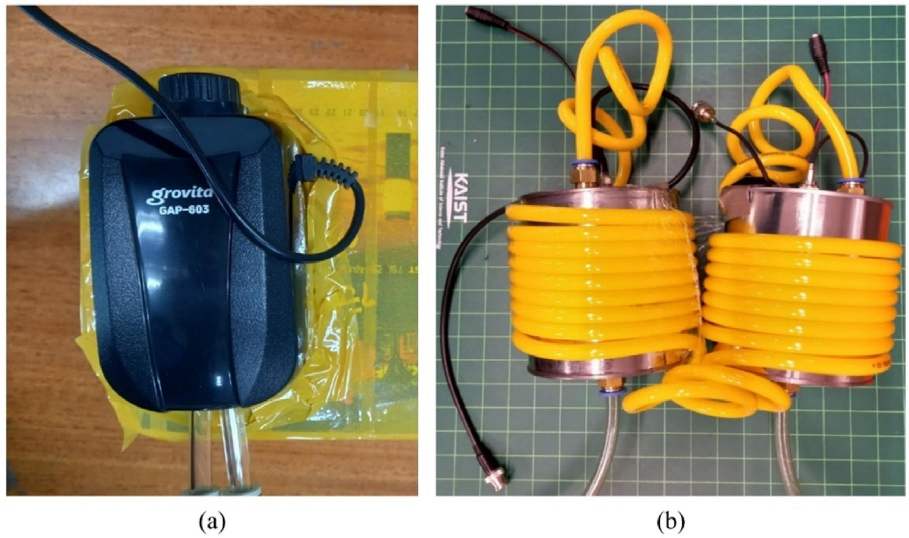

Fig. 2. (a) The air pump and (b) two ion chambers and a long PVC pipe (yellow) and an inlet and an exit pipe (transparent) of the proposed Radon/Thoron discriminative measurement system. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

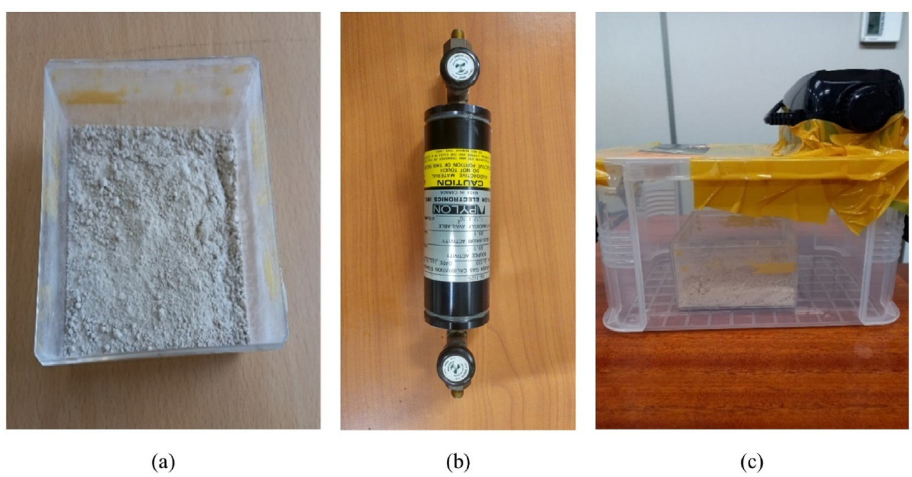

Fig. 3. (a) A tray containing monazite power as thoron source, (b) a radon source of Pylon RN-1025 containing Ra-226, (c) a big box for mixing radon and thoron and a pump placed on the box.

source. The parent nuclide of this radon source is Ra-226 which emits Rn-222 [19].

To control radon concentration, the gas valve of the RN-1025 open partially. It is because the activity of the RN-1025 is 89.1 kBq. As this activity is very high compared to our collimated source and used monazite powder, it is required to use almost same activity radon source so that, we can clearly observe the signal change for both radon and thoron. To control the activity, the inlet pipe of the RN-1025 was open slightly so that small amount of radon gas can enter into the big chamber and mix up with fresh air. Another big box was used as a buffer container for mixing gases when both thoron and radon gas are needed then the mixed gas enters into the inlet pipe through the air pump of the system as shown in Fig. 3 (c).

2.3. Calibration of the proposed system using a collimated alpha source and an SBD

Since the ion chamber is operated in a continuous current mode, to estimate the activity concentration (Bq/liter) in the ion chamber, a kind of calibration process to convert the output signal (its unit is voltage) of the ion chamber into the count rate (CR, its unit is count per second or cps) of alpha particles is necessary.

For this purpose, a collimated alpha source with a fixed intensity was prepared. An aluminum collimator cap with a thickness of 6 mm and with a hole of 3 mm diameter was fabricated to cover an Americium-241 check source as shown in Fig. 4.

The alpha particles from this source was measured by the ion chamber with the alpha source inside and also by a counting-mode alpha detector, a silicon surface barrier detector (SBD), in a vacuum system as illustrated in Fig. 5 (a) and (b). The SBD has an area of ~706.86 mm^2 and the vacuum spectrometer used was Ortec Alpha Aria. To guarantee all alpha particles emitted from the collimated source for a given time to be detected by the SBD, the solid angle of the collimated beam should be smaller than that of the SBD. To verify this the distance from the top surface of the collated source to the entrance surface of SBD was varied from 3 to 15 mm by changing the Teflon block with different heights. Similarly, the distance from the top surface of the collimated source to the anode wire in the ion chamber was also varied.

Radon and thoron can emit alpha particle at any position of the chamber. As the range of the alpha is within the limit of the radius

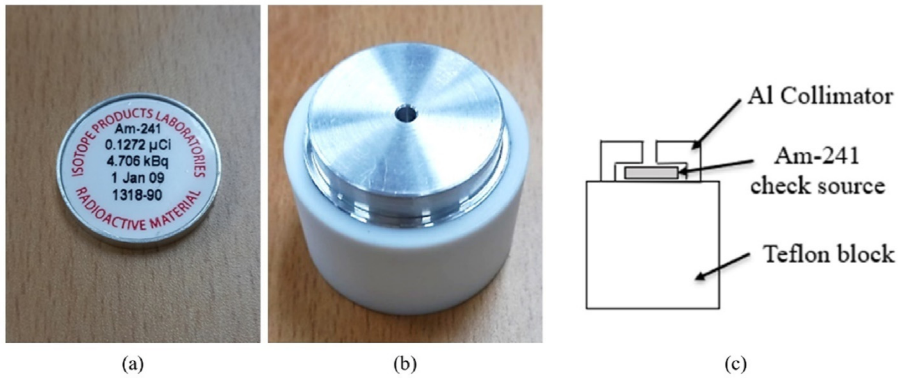

Fig. 4. (a) An Am-241 check source, (b) a photo of the collimated alpha source showing an Al collimator cap with a 3 mm hole (top) and a Teflon block supporter (bottom) and (c) a schematic of the collimated alpha source.

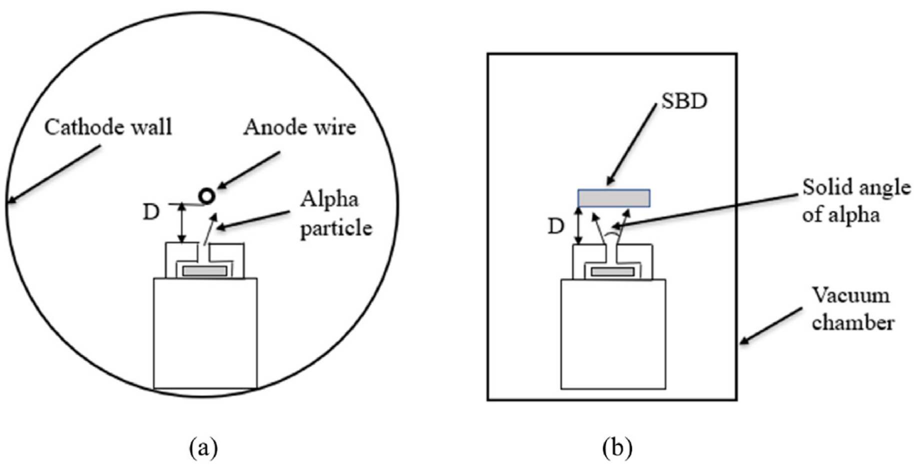

Fig. 5. (a) A collimated alpha source placed inside an ion chamber, (b) a collimated alpha source placed inside the Ortec alpha spectrometry vacuum system with an SBD.

of the ion chamber, we need to find out the average count regardless of the position of alpha emitting point. To find the average count for alpha, emitting from Am-241 source placed at different position of the chamber, the distance was varied. Ideally for both cases, the distance does not affect the measured values.

The result for both measurements is shown in Fig. 6 (a) and (b). For both cases, there seem to be dependencies on the distances defined in the above. This might be due to a scattering effect in the collimator channel wall by alpha particles for both cases. The dependency in the ion chamber is bigger probably due to the scattering with air molecules may get serious because of low bias, 12 V, operation of the ion chamber. If we apply higher bias to operate it in a proportional counting mode its dependency may decrease. Nevertheless the existence of the distance dependencies, both signal seems to saturate at longer distance. Using the extrapolated saturation values of both signals, it was found that 1 V signal in the ion chamber of the proposed system is equivalent to5,746 ± 260 cps in SBD. This value can be converted into activity concentration in the gas, 8,786 Bq/liter for each Volt in ion chamber output and 1 cps is equivalent to 1.529 Bq/liter for this ion chamber.

The conversion process of volts to cps and cps to Bq/liter is shown below –

As alpha emitter is a gas and fully enclosed inside the ion chamber which radius is within the limit of alpha particle range in the air, we assumed that the detection efficiency is ~100%. The average value for ion chamber = 0.0595918 ± 0.002 V.

Average value for SBD = 342.39 ± 0.46 count.

Hence for 0.0595918 V, there is a 342.39 count.

Therefore, for 1 V in the ion chamber, there are 5746 ± 260 counts. As the sensitive volume of the chamber is 0.654 L, for 5746 cps it will be 5746 ÷ 0.654 = 8786 Bq/liter.

This indicates 1 cps equivalent to 1.5 Bq/liter.

3. Result and discussion

3.1. Characterization of the proposed system by measuring thoron

After the calibration, at first, we performed a thoron measurement in the first and second chambers to demonstrate the proper performance of the ion chamber and the effect of the air-flow delay on the 2nd chamber data by using different lengths of pipe #2. To do this experiment, we used monazite powder only in a big box then turned on the air pump and opened the all valves in Fig. 1 (at 60 s in the CR curve of Fig. 7). Hence the air containing radioactive gas from monazite started to flow to the proposed system.

We observed (1) the times for the CR curves to start to rise and (2) the times to reach saturation levels as well as (3) the saturated values of count rates (SVCR) for both chambers. The initial delay pipe (pipe #2) had a length of 7.5 m. Then we repeated the measurements with other delay pipes with lengths of 15 and 20 m. All measurement results are shown in Fig. 7 and Table 2 is the numerical data for Fig. 7.

We observed that the CR of the first chamber starts to rise in about 3 s after opening the valves. This delay time is certainly due

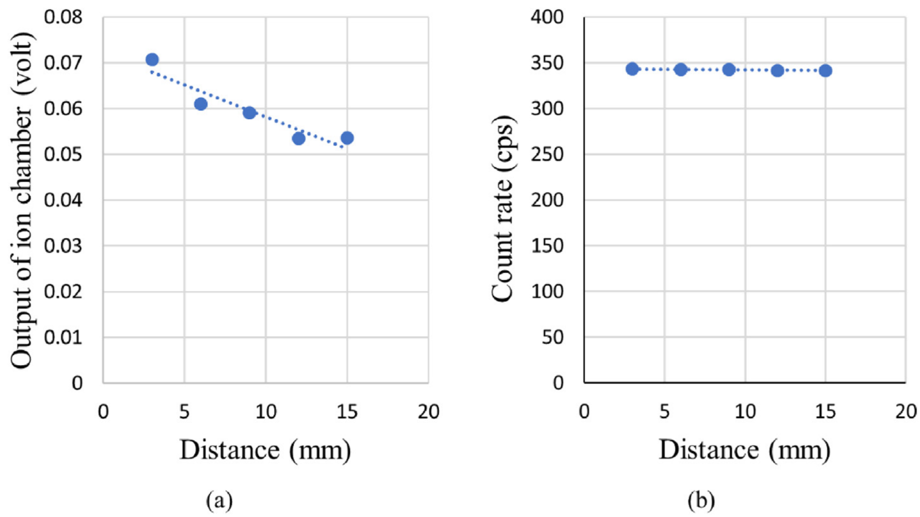

Fig. 6. Measured signals of the collimated alpha source showing a slight dependency on the distance, D, defined Fig. 5 (a) and (b) for (a) the ion chamber in the proposed system and (b) the surface barrier detector (SBD) of OrtecAlpha Aria respectively.

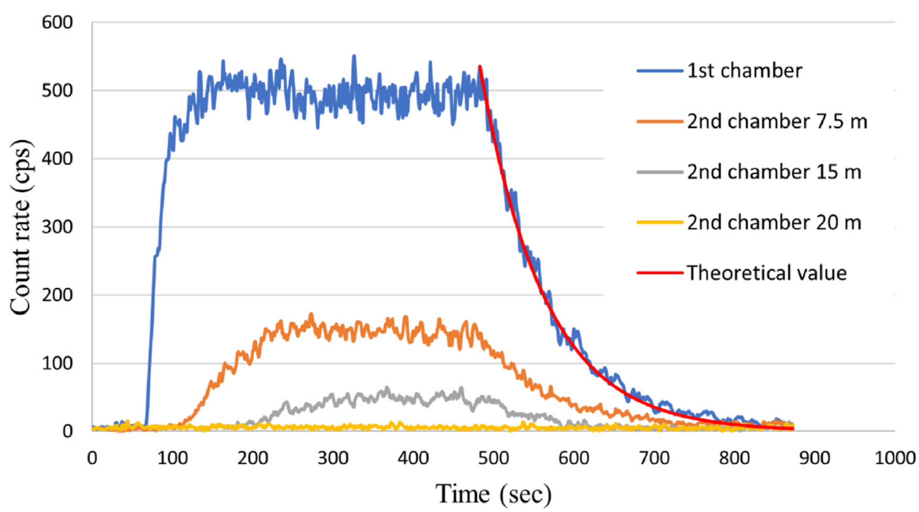

Fig. 7. Measured count rates of thoron in the first and the second chamber with different lengths of pipe #2(Opened all valves at 60 s and closed all valves at 500 s in the graph, the x-axis is the time passed since the LabView starts to record CR data).

to the time required by the air pass through the 1 m long inlet pipe (pipe #1). Similarly, the delay time for the second chamber with a longer pipe case takes a longer time. These times could also be interpreted as the time delay for filling the first chamber fully and passing through pipe #2. The rise times were given in the table which is defined as the time required for the magnitude of CR increase from 10% to 90% of SVCR. It is also observed that once the CR of both chambers reaches a saturation level, it remains at a constant value steadily as long as the air flows continuously and steadily. The SVCR of the second chamber is certainly lower than that of the first chamber and decreases as the #2 pipe length increases. This was an expected result because thoron from monazite will continuously decay as the time passes i.e. the air flows to a longer distance so that thoron of the lower activity concentration will be supplied to the second chamber steadily after its saturation time. From the background fluctuation of the first ion chamber, the RMS (root-mean-square) noise level of ~2 cps (equivalent to ~3 Bq/liter in the air inside of the ion chamber) was observed. This seems to be rather high but it can be improved with a high-class amplifier and the data acquisition system with careful grounding.

In addition to the above measurements, we performed another measurement in a closed chamber condition. At around 480 s since

Table 2
Measurement results of thoron in the first and the second chamber with different lengths of pipe #2(Delay means the time passed since the valves open).

Pipe	Length of pipe (m)	Chamber	Delay time until CRstarts to rise (sec)	The rise time of CR up to saturation level (sec)	SVCR (cps)
#1	1.0	First	3	45	486 ± 18
#2	7.5	Second	51	94	142 ± 11
	15		120	112	44 ± 6
	20		Not observed	Not observed	0

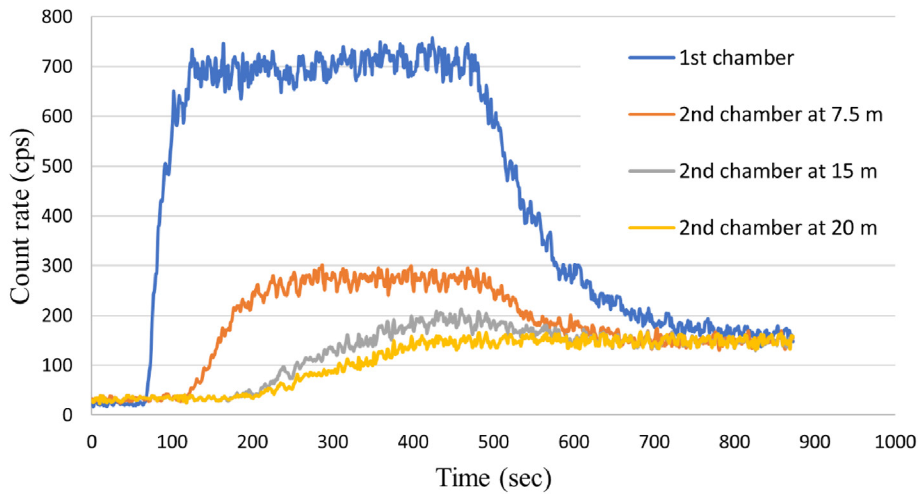

Fig. 8. Measured CR of both radon and thoron in the first and second chamber with pipe #2 of different lengths (Opened all valves at ~60 s and closed all valves at ~480 s in the graph).

Table 3
Measurement results of radon and thoron in the first and the second chamber with different lengths of pipe #2 (Delay means the time passed since the valves opened).

Pipe	Length of pipe (m)	Chamber	Delay time until CRstarts to rise (sec)	Rise time of CR up to saturation level (sec)	SVCR (cps)
#1	1.0	First	3	45	675 ± 21
#2	7.5	Second	59	94	242 ± 12
	15		109	159	162 ± 9
	20		118	186	115 ± 7

the LabView start to record as seen in the Fig. 7, all valves were closed simultaneously so that there would be no more supply of thoron into the chambers. Then as one could expect, the CR dropped exponentially as shown in most curves of Fig. 7 according to the half-life of thoron. As also shown in the figure, the measured count rate values in the first chamber during this time matches well with a theoretically calculated values of due to the decay of thoron. Hence, it was clear that the emitted gas from monazite was certainly thoron and the proposed ion chamber was demonstrated to work well to detect alpha particles from thoron.

3.2. Discriminative measurement of radon and thoron by the proposed system

In order to verify the capability of discriminative measurement of radon and thoron by the proposed system, the radon gas from RN-1025 was supplied the big box where the monazite powder tray was placed. Similarly to the previous measurement with thoron only, the valves opened and closed at 60 s and 480 s of the LabView recording time and the CR of both chambers was observed as a function of time as shown in Fig. 8 and Table 3.

The timing response of the system such as delay times and rise times of CR for both ion chambers are very much similar to the previous measurement of thoron only case. However, the saturated values of CRs for both chambers were increased compared to thoron source only case obviously due to radon is added to thoron except the case of pipe #2 is 20 m long. In this case, as we had already seen, all thoron decays out when the air arrived at the second ion chamber.

We also can notice that after all valves closed, the SVCRs approach to the same level of 115 cps (this is the net magnitude after subtraction of the background) which certainly is the SVCR due to radon only because its half-life of 3.8 days is a relatively long in comparison with the measurement time of this experiment. Therefore if we subtract this value from the SVCR of the first ion chamber, the remaining is the CR due to thoron which is 560 cps in this case. If we convert these values into the activity concentrations then they will be 173 Bq/liter and 840 Bq/liter for radon and thoron respectively.

But these values are only the activity concentrations of the air inside the ion chambers and not an activity concentration of the outside air. For measuring the environmental radon and thoron activity concentration in free air, the pump speed must be considered to find the corresponding conversion coefficient. When the pump speed changes, the conversion ratio of cps into Bq/liter in the environment will be changed accordingly.

When the proposed system is utilized in a continuous mode in a practical situation, radon and thoron activity concentrations should be estimated from output values of the first and second ion chambers with a 20 m long pipe between them. From this study, another way of measuring radon and thoron separately using a single ion chamber can be suggested in such a way that inlet and exit valves open and close periodically with a minimum period of 5 min of a waiting time so that thoron decays out enough.

4. Conclusions

We proposed a simple method using two ion chambers and a long air pipe as a time delay device between two chambers. While the first chamber measures both radon and thoron, the second chamber only measures radon because of the short half-life of thoron which shall decay out during it passes through the long air pipe. We also successfully demonstrated that our system could measure radon and thoron separately by using natural monazite powder and a commercial Ra-226 source simultaneously. We also developed a collimated alpha source using an aluminum collimator and an Am-241 check source and were able to convert the output signal of ion chambers in volt to the count rate in cps by using an SBD in a vacuum chamber.

To remove daughter nucleus, the conductive sponge are connected to the cathode of the chambers. As the daughter nucleus are positively charged and the conductive sponge are at the entrance of each of the chamber, during air flow the daughter nucleus can easily captured by conductive sponge. Hence, before entering into the active volume of the chamber, it is possible to remove all the daughter product.

There are a few issues to be studied further for improvement; The first one is to improve sensitivity and reduce the noise level. This can be improved by replacing the current readout circuit by a high-class electronics and a low noise preamplifier. Secondly, the conversion of the count rate of ion chambers into the activity concentration in free air also needs to be addressed.

Nevertheless the key idea of the proposed system can be a practical solution to measure radon and thoron separately and in real-time mode.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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