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Review

An Overview of Passive-Type Detectors for Radon and Its Progeny Measurement

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Radon is well known as a radioactive gas, and the inhalation of radon and its progeny could lead to health risks, especially inducing carcinogenesis that potentially generates lung cancer. Therefore, it is essential to monitor concentrations of radon and its decay products to estimate the radiological hazards and risks to human health. In this paper, numerous studies on passive techniques for radon and its progeny have been reviewed and summarized. The mainstream of passive devices can be classified into four groups: (i) alpha track detectors, (ii) activated charcoal detectors, (iii) electret ion chambers, and (iv) thermo-luminescent dosimeters. The principle of passive devices, materials, designs, and the factors affecting their performance are discussed. This review aims to provide options and understanding of the passive techniques for radon and its progeny measurement, which have been developed for radiation protection and surveillance of radon and its progeny in various environments.

Key words: passive radon detector, radon, radon and its progeny, radon measurement

1. Introduction

Radon (Rn-222) is a naturally occurring radioactive gas and a transient product of the uranium (U-238) decay series. As a noble gas, radon can easily diffuse through soil and rocks from the earth's crust into the air and tends to concentrate in the atmosphere, enclosed spaces, and environments[1)]. Radon has a half-life of 3.82 days, and its decay produces and emits other short-lived radionuclides with alpha particles and beta rays[2)]. The decay products of radon or radon progenies can adhere to dust and aerosol and deposit in the lungs and airways when inhaled. Radon and its progeny continue to emit intensely ionizing alpha particles and beta rays, affecting biological cells in the lungs, leading to DNA damage and inducing carcinogenesis that potentially generates lung cancer[3)]. The World Health Organization (WHO) has identified radon and its progeny as the second largest risk factor for the cause of lung cancer only after smoking in the general population[4)]. Approximately 50% of the world's average annual effective dose to the human by natural radioactivity has been recognized due to internal exposure to radon progeny[5)]. Therefore, it is vital to monitor concentrations of radon and its products in the environment to estimate the radiological hazards and risks to human health.

Most techniques of measuring radon and its progeny are based on the measurement of the alpha particles emitted from the radionuclides during their radioactive decay. Techniques for measuring radon and its progeny can be classified as active or passive method. The active

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methods are usually used for short-term measurements of radon, and require power to function under inspection for investigations of individual sites. On the other hand, the passive methods generally do not require power to function, and are more suitable for long-term measurements and large-scale surveys6). The radon and its decay product in air typically enter those passive detectors via diffusion, and the radiation detector medium itself normally does not require any power supply.

This paper aimed to propose an overview of passive-type monitoring techniques for measuring radon and its progeny, namely alpha track detectors, activated charcoal canisters, electret ion chambers, and thermoluminescent dosimeters. Special attention needs to be paid to the application and conceptual design description of mainstream technologies, such as types of detectors, sample collection strategies and measurement techniques.

2. Properties of Radon and Its Progeny

Radon is a naturally radioactive inert gas, and it has 36 known isotopes, ranging from Rn-193 to Rn-228. The most mentioned three radioisotopes are Rn-222 (radon), Rn-220 (thoron), and Rn-219 (actinon) and are decay products of U-238, Th-232, and U-235, respectively. Among these three isotopes, Rn-222 is most present in the environment due to the abundance of U-238 in nature and its longest half-life (3.82 days). Therefore, if radon was mentioned without isotopes specification, it usually means Rn-222.

In the radioactive decay series of radon, mainly three groups of emitting alpha particles that alpha detectors may detect are considered, namely, those that emit alpha particles with energies of 5.49 MeV from Rn-222, those with most energies of 6 MeV from Po-218, and others with energies of about 7.69 MeV from Po-214. When radon and its progeny are inhaled, the primary result of radiation dose to the lung is not due to radon itself but due to some of radon's short-lived decay products (mainly Po-218 and Po-214) are retained in the lungs and irradiate cells in the respiratory tract with alpha-particles before they have been expelled from the lung.

The short-lived radon progeny concentrations are collectively expressed as Potential Alpha Energy Concentration (PAEC) or Equilibrium Equivalent Radon Concentration (EERC). These two quantities combine the radon progeny concentrations into a single value using weighting factors that consider the progenies' half-lives and alpha energies emitted. The historical unit called the Working Level (WL) was proposed to PAEC of radon progeny in air. Originally, one WL refers to the concentration of the short-lived decay products of radon in equilibrium with 3700 Bq m^{-3} (100 pCi L^{-1}) of EERC that will result in the ultimate emission of 1.3 x 10^5 MeV of potential alpha energy7). The International System (SI) unit for PAEC is J m^{-3}, one WL is equivalent to 20.8 μJ m^{-3}. The SI unit for EERC is Bq m^{-3}. A PAEC in the unit of 1 WL corresponds to EERC about 3700 Bq m^{-3}.

3. Passive Measurement Techniques

3.1. Alpha track detectors

Alpha Track Detectors (ATDs) are also called Nuclear Track Detectors (NTDs) or Solid-State Nuclear Track Detectors (SSNTDs). These detectors work on a principle of alpha particles emitted from radon and some of its progeny, creating microscopic damages to the detector materials and resulting in latent tracks. The tracks can be visualized under an optical microscope through a chemical or electrochemical etching process8). The track density is proportional to the radon concentration accumulated over the exposure period. The most popular materials employed for ATDs are polymeric plastics, such as poly-allyl diglycol carbonate (also known under the trademark as CR-39), cellulose nitrate (commercially well-known as LR-115), and polycarbonate (e.g., Makrofol)9). Generally, a minimum detectable concentration of ATDs was found about 30 Bq m^{-3} or lower, for a month of exposure. And there is typical uncertainty of 10-25%4).

The possibility of using ATDs for nuclear applications was first introduced by Fleischer *et al.* in 196510). After that, many studies proposed using track detectors for long-term monitoring of radon and its progeny11-13). In general, track detectors used for radon monitoring in the atmosphere can be categorized into two main modes: open/bare detector mode and close detector mode.

In the case of the open/bare detector mode, ATDs are open-faced without a filter or barrier and directly exposed to alpha particles present in the air. Alpha particles emitted by radon and all radionuclides in the ambient atmosphere can damage the track detectors and produce latent tracks in them. Therefore, the resulting measurement from the open detector is related to radon concentration, radon progeny concentrations, as well as to other alpha-emitting radionuclide concentrations14). Thus, this technique is not very suitable for measuring radon only. The information from the open detectors does reveal the concentrations of radon and its progeny in the air. Unfortunately, this measurement mode has significant uncertainties because the calibration factor (k) depends on the equilibrium factor (F) between radon and its progeny15). In the past, the radiation dose quantities due to exposure to radon and its progeny have been commonly estimated using the typical assumed equilibrium factor value of 0.4 for the indoor environment, as recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)5). However, in reality, the value of F varies widely with meteorological and environmental conditions, such as ventilation, humidity, time, place, etc.[16-18]. Therefore, using an assumed constant value for F does not always reflect the actual conditions. Moreover, thoron (Rn-220) and its decay products can interfere with open/bare detectors, which may cause an overestimation of derived radon concentrations. At the same time, radon progeny can be deposited onto the surface of ATD materials, especially in the case of CR-39. This radon progeny deposition process is called "plate-out," and alpha tracks from the plate-out radon progeny are usually larger than those produced by alpha particles in the air. So, the accuracy of radon measurements will be affected significantly by the overlap of alpha tracks.

The closed detector mode is proposed to avoid the entry of radon decay products present in the ambient atmosphere. The ATD materials are positioned at different places inside the container, with the open end covered by a filter or barrier to prevent radon and thoron decay products from entering the sensitive volume of the devices. Consequently, the number of tracks on the detector is related only to alpha particles originating from radon entering the sensitive volume of the container as well as its progeny formed in the container. Therefore, the closed detector mode has solved some major problems related to the open detector mode and can provide measurements related to the actual radon concentration in the air. However, many factors affect the stability of ATD detection response, including humidity, temperature, presence of oxygen, thoron activity concentration, deposited progenies, diffusion rate, and electrostatic charges on the detector surface, especially ultraviolet radiation can hardly affect the measurement in closed detector mode[19-21].

Several designs of containers or chambers have been developed and are used to improve the efficiency and accuracy of radon measurement. Studies dealing with the filters to protect radon detectors measuring in extreme environmental conditions have also been published[20, 22, 23]. For example, Bochicchio *et al.*[24] used a sealed polyethylene bag to enclose the radon concentration measurement device (LR-115 detectors), which prevented the entrance of dust, humidity, and radon decay products. It also reduced thoron from entering the device to reduce the uncertainty of radon concentration measurement. Regarding shape, the containers can be cylindrical, conical, hemispherical, or rectangular. Some containers are equipped with more than one piece of ATD. Furthermore, a closed detector, which is covered with the polyethylene membrane to permit only radon to enter the detector and prevents thoron, radon progeny and dust, and an open detector, which was exposed freely to radon as well as radon progeny are occasionally used together for the determination of F. In this method, F can be found as a function of the track density ratio (D/D_0) between open detector (D) and closed detector (D_0), respectively[25].

However, the bare mode or open detectors are not recommended to use in the high thoron areas or high concentrations of thorium. Some researchers reported the higher values of indoor radon concentrations in the bare mode study were mainly due to interference of thoron and its decay products in the environment[26]. Many of the nationwide surveys were carried out with radon concentration measuring devices in the single closed chamber. Examples of such devices used in nationwide surveys are the NRPB/SSI type (first developed by the National Radiation Protection Board and was subsequently improved by the Swedish Radiation Protection), which had been used in Albania[27] and Sweden[28], and the Karlsruhe type detectors were used in Austria[29], Germany[30] and Luxembourg[31].

As ATDs cannot separate radon and thoron signals, a two-chamber design measurement system is widely used to separate various radon and thoron measurements. Sahoo *et al.*[32] developed a type of two-chamber device based on a pinhole design (Fig. 1(A)) for radon/thoron

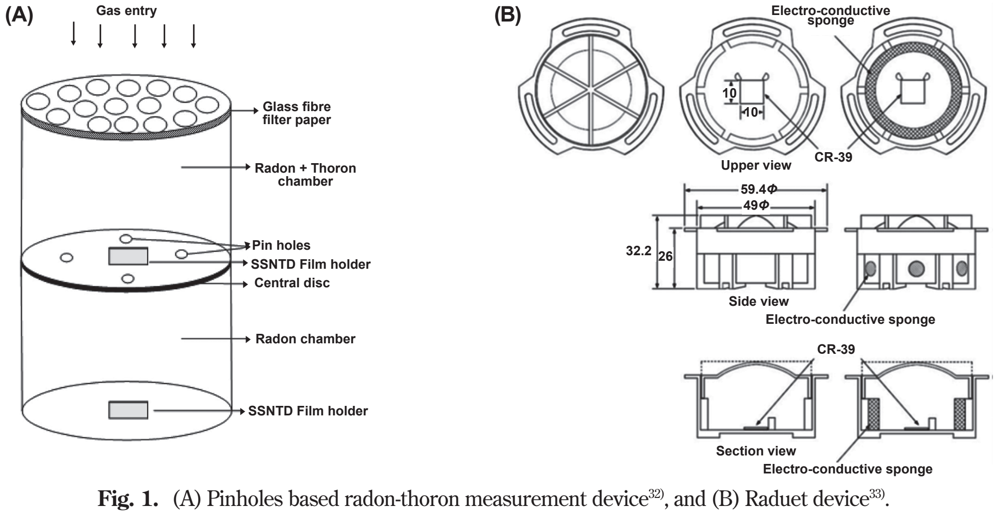

Fig. 1. (A) Pinholes based radon-thoron measurement device[32], and (B) Raduet device[33].

discriminative measurement. By using a diffusion barrier, separate measurements can be made of radon and thoron because of the different entry rates of radon/thoron through two chambers. So, when air diffuses through a filter, cutting off the aerosol component into the upper chamber, alpha particles of radon, thoron, and their progenies are detected with ATD. Air enters the lower chamber through the pinholes as a diffusion barrier, and nearly only radon permeates.

Another example of a two-chamber design, called Raduet, proposed by Tokonami *et al.*[33], is shown in Figure 1(B). Using CR-39 as detector material, the primary measurement principle remains almost the same as the pinhole-based device with the different air diffusion rates into two-chamber. However, the design was somewhat changed as chambers were separated. Specialty plastic and sponge were respectively used as the wall chambers and hole filters to eliminate electrostatic effects, which influenced the distribution of charged progenies.

Recently, the decay products of radon and thoron were widely measured using the direct radon progeny sensors (DRPS) and direct thoron progeny sensors (DTPS) method of passing alpha particles through appropriate absorber materials[34-36]. This method uses absorber-mounted ATD, which selectively records the tracks due to alpha emissions at different energies from radon and thoron progeny. For example, Po-212 with alpha energy 8.78 MeV and Po-214 with alpha energy 7.69 MeV from the deposited atoms of thoron and radon progeny, respectively.

After exposure period, latent tracks in the ATDs can be made visible for easily counting by a chemical etching with alkaline solutions of sodium hydroxide (NaOH) or potassium hydroxide (KOH). The main etching parameters, that can affect etching results are temperature, etchant concentration, and etching time. Daci *et al.*[37] examined several etching conditions of CR-39 to determine optimal etching condition. For the track density counting, ATDs can be measured with different systems. For example, detectors can be counted with an optical microscope and are manually or automatically counted, or they can be counted with a non-optical system, such as a spark-counter[38].

3.2. Activated charcoal detectors

The Activated Charcoal Detector (ACD) technique is widely used to measure radon concentrations for a short-term survey, typically 2-7 days, to provide rapid and cost-effective results. At this survey period, a minimum detectable concentration was calculated and shown around 20 Bq m^{-3}, and typical uncertainty 10-30%[4]. Regarding the measurement principle, activated charcoal is used to adsorb and desorb radon by the active sites of the carbon particles. During the exposure period, radon is being adsorbed to carbon particles and it is undergoing radioactive decay from radon decay. After ACDs are closed more than three hours, radioactive equilibrium of radon and its progeny is reached. Radon activity is measured via its progeny activity at the equilibrium point. Bi-214 and Pb-214 are typically detected by gamma spectrometers due to their gamma lines[39]. Commonly, NaI(Tl) detectors and High Purity Germanium (HPGe) detectors are used. For this process, three gamma energy peaks are typically analyzed at 295 keV and 352 keV for Pb-214 photons and 609 keV for Bi-214 photons. Furthermore, radon absorbed on the ACD can be measured by the liquid scintillation technique. This technique observed the count rate of radon and its progeny from the eluting the radon from the charcoal is accomplished by liquid scintillation cocktail and measured using a liquid scintillation counter[40].

In 1987, the United States Environmental Protection Agency (US EPA) published the protocols for radon measurement using charcoal detectors[39]. In the following decades, radon measurement systems with charcoal detectors were applied worldwide due to their advantages of high sensitivity, low-cost operation, and operation simplicity. Furthermore, they are often recyclable by driving off radon and other organic gases after gamma radiation analysis. In 2019, Pantelic and his team[41] provided the overview of indoor radon surveys in Europe and included around 7% of radon measurements were performed by charcoal detectors. On the other hand, the disadvantages of this technique are that they require quick analysis, generally within seven days after the end of exposure periods. Moisture also affects the adsorption efficiency of activated charcoal. Moreover, the costs of measuring instruments and analysis skills of reading operators should be considered, because this technique require gamma spectrometers for reading results.

The ACD technique does not require electricity or special operating skills. After exposure, detectors can be sent by mail to measure and analyze at other locations. However, ACDs cannot be respected as suitable integrating devices, mainly when the radon concentration changes rapidly during the measurement period[42]. Furthermore, as they allow continual radioactive decay of radon, their response gives a high weight to the last day of exposure and is affected by humidity and temperature[43]. Therefore, radon measurement with ACDs is generally used for screening purposes in the indoor environment.

As activated charcoal is an excellent water vapor absorber, this property reduces absorption capability for radon. Therefore, humidity correction is required for this method. Generally, the activated charcoal is heated at over 100 °C for several hours to remove the moisture from charcoal before measuring radon. And after exposure, the device is tightly sealed and quickly returned to the laboratory to avoid the effect of humidity and maintain the maximum sensitivity44). In some research, a diffusion barrier or desiccant, such as silica gel, has been incorporated in containers to separate the activated charcoal from the air to reduce the effect of moisture45, 46). Sometimes, the ACD is weighted before and after deployment to evaluate the amount of adsorbed humidity. Because the change of detector mass due to water adsorption is used to calculate calibration factor and to select adjustment factor curve for correcting radon concentration47).

3.3. *Electret ion chambers*

An electret detector or Electret Ion Chamber (EIC) is an integrated ionization chamber wherein the electret functions both as a sensor and a source of the electrostatic field. Generally, the electret is an electrostatically charged plastic disc mounted inside a small container made from conducting plastic, where radon can diffuse through a filter-covered opening of the chamber. The filter precludes radon progeny into the chamber. EIC provides the information on a time-integrated radon activity concentration. The popular commercial version of EIC is often known under the trade name E-PERM (Electret-Passive Environmental Radon Monitor). It is assumed that E-PERMs measure radon gas concentrations as low as 370 Bq m^{-3} in air with lower 10% error when measured over 1 day48).

During the exposure period, radon gas can enter the EIC device, where radon and its progeny undergo radioactive decay and ionize the air within the chamber. The positively charged electret collects negative ions that have been produced from the ionization of air, resulting in a discharge of the electret, which is related proportionally to the integrated concentration of radon inside the chamber. The electric charge on the electret is determined before and after the exposure period by using a voltage reader (an electret surface potential voltmeter)49). In general, the electrets should be read and analyzed as soon as possible after the end of exposure to avoid continual discharge due to background radiation. Furthermore, unlike alpha track detectors that are only sensitive to alpha and particles radiation, electrets are also sensitive to gamma and other background radiation, thus necessitating a separate determination of gamma background radiation. The following equation (Eq. 1) calculates radon concentration from the EIC device50).

$$C_{Rn} = \frac{(V_i - V_f)}{(T)(CF)} - BG \qquad \text{(Eq. 1)}$$

C_{Rn} is the actual radon concentration in Bq m^{-3}, V_i and V_f are the measured initial and final electret voltages, respectively, T is the exposure time in days, CF is the conversion factor in the unit of V per Bq m^{-3} d and BG is the radon concentration equivalent of the gamma background. Note that BG of 0.1 μGy h^{-1} was measured to be equivalent to 32 Bq m^{-3}. And a linear correction can be made to accommodate other BG values.

The EIC can be applied to different exposure periods, from a few days to a year50, 51), depending on the EIC design, which is determined by the thickness of the electret and the volume of the ion chamber. Short-term devices are designed to expose radon for about two days to two weeks; thick electret and large volume ion chamber are used to enhance the high sensitivity of devices. Alternatively, thin electret and small volume ion chambers are used for less sensitive electrets in long-term EIC devices46), which can provide an average of integrated radon concentration for the duration of exposure periods spanning several weeks to a year.

The first advantage of EIC devices is that conducting measurements do not require many skills, and results can be obtained in the field, if necessary. Moreover, the devices are reusable. However, its sensitivity to gamma radiation can interfere with radon measurement, and humidity can affect EICs. For example, Sorimachi *et al.*52) indicated a slight influence of humidity, ambient aerosols, and thoron on the responsibility of EICs for radon measurements. Thus, an area with high moisture and extremely high thoron concentrations should be avoided, and attention must be paid to these factors when using EICs. Furthermore, EIC is also sensitive to altitude change, which relates to pressure and air density change. At higher altitudes (or at low pressures), fewer air molecules are available for producing ion pairs in a traveling path length of radiation compared to the ions produced at lower elevations (or at higher pressures). The corrections of elevation effect should be applied for obtaining more accurate results whenever EIC is used for making radon measurements at very high altitudes53), due to differences in air density that can affect the distance alpha particles can travel54).

3.4. *Thermo-luminescent dosimeters*

Thermo-luminescent dosimeters (TLDs) are based on using crystalline materials in which ionizing radiation transfers energy to an electron and creates electron-hole pairs. When TL materials are irradiated, they store energy inside their structure, causing electrons and holes to move independently throughout the media. The amount of energy required to create electron-hole pairs is dependent on the energy difference between the valence band and conduction band of materials. After the exposure period, the TL materials are generally heated, causes the rapid recombination of electrons and holes, resulting in the release of light (photons). The intensity of emitted light can be measured using a photomultiplier tube (PMT) in the TLD reader device, and the amount of emitted light is proportional to the radiation dose accumulated over the exposure period55). For radon measurement using the TLD technique, the information stored in TLDs is not the number of alpha particles but the total energy of alpha particles. Therefore, the readout information of TLDs shows directly potential alpha energy concentration (PAEC), with no dependence on the equilibrium factor.

TLDs were found to function as a radon detector. In 1981, Ho and Weng56) published the newly developed TLDs technique using TLD ($CaSO_4$: Dy) as a thin foil as part of a specially designed device for radon measurement near-surface soil. CaF_2: Dy is a type of TLD that Brits and Van57) calibrated to respond to alpha and beta radiation from radon progeny. In addition, TLDs are also used to measure background gamma radiation together with another detector type. Additionally, Stranden *et al.*58) showed that TLDs could be applied with the charcoal detector to record the continual gamma emission from radon progeny captured in charcoal. The average radon concentration can subsequently be analyzed using a TLD reader, but this approach is not in general use.

However, TLDs have a limited utilization for radon dosimetry because most TL materials are sensitive to whole radiation, including gamma, beta, and alpha radiation. Therefore, one of the problems of radon measurement using TLDs is separating the TL signal associated with alpha particles emitted from radon from the signal caused by accompanying beta and gamma radiation. Generally, this problem can be solved using at least two TLDs, one sensitive to all ionizing radiation for determining all radiation emitted from radon and its progeny, and another to indicate background gamma and beta radiation59).

4. Recent Advances in Passive Radon Detector

Recently, several research have been conducted in the field of the development of the passive devices for monitoring and modeling radon and its progeny.

MetroRADON Project, which running in Europe is one of the radon research projects aim to develop reliable techniques and methodologies to enable in the international system of units (SI) traceable radon activity concentration measurements and calibrations at low radon concentrations. The calibration methods and measurement techniques for some passive devices are developed in the project. As part of this project, compact discs (CDs) are used for radon measurements, novel techniques for radon exhalation measurement from soil based on polymers or track-etching of CDs and liquid scintillation detector have been developed and evaluated. It is based on radon absorption in the polycarbonate material of CDs. Pressyanov and his team60) proposed the application of CDs method for radon measurements at workplaces by testing under highly variable exposure conditions. Good correspondence was observed, small systematic bias of 3.7% at a depth 80 μm and 13.5% at a depth 120 μm beneath the front surface of CDs are explained by the significant variability of the temperature and radon concentrations during exposure. Furthermore, some topics related to thoron interference in radon by passive detectors are investigated in the project61). The single-chamber detectors with no discriminative function are used in some countries to measure radon concentrations, which may be overestimated due to thoron interference. In order to calibrate and reduce the influence of thoron, the influence of thoron on the radon activity concentration measurements has been observed and investigated with some radon detectors in this project. Related to the case, Sorimachi *et al.*62) examined the performance of passive detectors (radon-thoron discriminative RADUET detector) at exposure to low-level radon concentration, and they reported good results compared to active radon detectors. The agreements between the active detectors were within 15%, while the passive detectors were consistent with those by the monitors within approximately 20%.

Another advanced research project in field of radon metrology was performed at IFIN-HH, in Romania63). One of their work objectives which related to passive devices for radon measurement, is the controlled radon exposure of several nuclear track detectors placed in the radon chamber. The TASTRAK solid-state nuclear track detectors, Polyallyl Diglycol Carbonate (PADC; CR-39) were used in this work together with the optical reading system (TASLIMAGE system) as a new high-performance radon dosimetry. Their system showed good result of the main technical characteristics, such as fast reading per detector between 40 s and 100 s, lower detection limit about 10 Bq/m^3, upper detection limit about 10 MBq/m^3 and linear response within 5% accuracy for a radon exposure of 500 $kBq \cdot h \cdot m^{-3}$.

5. Conclusions

In the present paper, an overview of the passive measurement techniques of radon and its progeny has been proposed. The mainstream passive devices for measuring radon and its progeny are classified into alpha track detectors, activated charcoal detectors, electret ion chambers, and thermo-luminescent dosimeters. The principles of these devices, their main designs, influencing factors, the application status of detection devices, sampling collections, and data analysis have been described and summarized.

From the comparison of the passive devices: ATDs and TLDs are suitable for long-term measurement, but ACDs are potentially for short-term measurement. While EICs are able to serve for both long-term and short-term measurement. After the sampling or exposure period, ATDs, ACDs and TLDs must be sent to a laboratory for analysis, but EICs are possible to read the results on-site. However, the cost of EIC is high than other devices, and it require correction for background radiation. As the same problems of radon measurement using TLDs is separating the TL signal associated with alpha particles emitted from radon from the signal caused by accompanying beta and gamma radiation.

In general, passive devices do not require any power supply for sampling collections. Therefore, they are usually simple to use, cost-effective, and some of them are repeatable. For these reasons, passive techniques are suited for radon measurement in long-term and large-scale measurement. However, the efficiency, accuracy, and stability of the passive radon measurement devices may be affected by humidity, temperature, thoron activity concentration, and deposited progenies of radon and thoron. Furthermore, several passive devices detect not only radon but also thoron and its progeny, resulting in a possibility for overestimating the level of radon concentration. This could lead to a biased estimate of the health and lung cancer risk in epidemiological studies. Therefore, there are still many limitations that must be subject to an important future development of passive-type radon detectors. Although, the future trend of passive-type radon detectors development can be changed in many directions, but the demands for cheaper prices, longer lifetime, less maintenance (cost-effectiveness), more accuracy and easy to read-out results are to be expected. And also, consecutive demands for automatically read-out electronics, interconnection technologies, data acquisition, and quality assurance.

Author Contributions

Conceptualization, S.T.; Data acquisition and interpretation, W.P.; Supervision, C.K. and S.T.; Validation, W.P., C.K. and S.T.; Writing—original draft, W.P.; Writing—review and editing, W.P., C.K. and S.T. All authors have read and agreed to the published version of the manuscript.

Conflict of Interest

The authors declare that they have no conflict of interest.

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