Research Article

The Accumulation of Radon Gas in Shophouses: Case Study of Using Concrete with Fly Ash and Phosphogypsum Additives as Building Materials

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Abstract

Radon is a ubiquitous radioactive noble gas which cannot be detected by any human senses. Moreover, it is known to be the leading cause of lung cancer among non-smokers. Soil is the main source of radon, although another major source of indoor radon is derived from building materials, particularly building materials which are by-products from industrial processes containing relatively high concentrations of radium (such as fly ash and phosphogypsum). These building materials produce more radon gas than the others. This research is divided into two parts. The first part focuses on radon-exhalation rates from cementitious materials which contain variations of fly ash (FA) and phosphogypsum (PG). The second part focuses on the study of the concentrations and distributions of radon in shophouses, one of the most popular residential building types in Thailand, which use fly ash and phosphogypsum as an additive in cement. From the first part, the results from measuring radon concentration in closed test chambers by using a pulse-counting ionization chamber radon gas monitor (Atmos 12 dpx) show that the radon exhalation rate is significantly increased when more fly ash is added (from 40 to 60% in concrete); whereas, a clearly exponential correlation is found between radon exhalation rate and phosphogypsum content. From the second part, the CFD simulations show that, in the case of using 50% PG additive, the interior partitions in shophouses affect the radon concentration level and this noble gas accumulates in areas of poor ventilation, which could reach 648.10 Bq/m3 (higher than the average world value of 39 Bq/m^3), resulting in the exceedance of the annual effective dose of 16.33 mSv/y.

Keywords: Radon, Phosphogypsum, Fly ash, building material, indoor air quality

1. Introduction

Radon (^{222}Rn) is a colorless radioactive gas which cannot be detected by any of the human senses but it becomes yellow when it is cooled below its freezing point. Radon arises from the decay of radium (^{226}Ra), which originates from the ^{238}U (Uranium) decay series[1] . Its half-life of 3.82 days allows time for substantial diffusion to the atmosphere, compared to the half-life of thoron (^{220}Rn) and actinon (^{219}Rn) (55 s and 3.96 s, respectively), which contributes less significantly to the airborne inventory of radon. When radon decays, it produces a series of radioactive isotopes (called radon daughters) of polonium (^{218}Po), bismuth (^{214}Bi), and lead (^{214}Pb) which can attach to particulate matter in the air and, upon inhalation, deposit in the respiratory tract.

These radon daughters emit alpha-particles, equivalent to the nucleus of a helium atom, where the unstable nucleus releases an electron, damaging the DNA of epithelial cells in the respiratory tract; this may result in lung cancer (World Health Organization [WHO], 2009). This risk is exacerbated by the deposition of these particles in the lung tissue, where they deliver a higher radiation dose compared to when they are suspended in the air. Moreover, the absorbed dose from deposited radon progeny is significantly higher than from suspended particles, with alpha decay contributing most to the lung absorbed dose (Danaei et al., 2020). While the primary concern is the carcinogenic potential of radon decay products, it is also important to consider the broader context of radon exposure. Seasonal variations in indoor radon levels, influenced by environmental and structural factors, can affect exposure risk.

Although the principal source of indoor radon is from soil and rock adjacent to buildings' substructures, another potential source that can elevate indoor radon concentrations is building materials themselves (United Nations Scientific Committee on the Effects of Atomic Radiation [UNSCEAR], 2000; 2008). Since radium is widely distributed in the earth's crust, most building materials contain radium-226 (^{226}Ra). The study of ^{226}Ra concentration values in construction materials in different countries shows that the highest values derive from gypsum (1480 Bq/kg) and granite (111 Bq/kg) in the USA and Russia, respectively; whereas, the fine aggregate (23.3 Bq/kg), cement (44.4 Bq/kg), and concrete (26 Bq/kg) in Russia, Sweden, and Norway give the lowest respective values (Bulut & Şahin, 2024). These building materials cause radon gas emissions and contribute approximately 25% of indoor radon concentrations (Turkish Atomic Energy Authority [TAEA Guide], 2017). The Organization for Economic Co-operation and Development (OECD) Surveys from 30 countries revealed that the highest arithmetic mean of indoor radon concentration levels was found in Mexico (140 Bq/m^3) and the lowest levels were found in Iceland (10 Bq/m^3); the worldwide average indoor radon concentration has been estimated at 39 Bq/m^3 (WHO, 2009).

Indoor radon concentrations have also been investigated in Thailand. In Phu Wiang (Khon Kaen Province), Saraphi District (Chiang Mai Province), and the Na Mhom District (Songkhla Province), the indoor levels were 21±70, 21±6 and 52±17 Bq/m3, respectively (Wanabongse et al., 2005).

The measurement of indoor radon is important because people spend 80-90% of their time indoors, according to UNSCEAR's report in 1992, and doses of inhaled radon in dwellings is more than from all other natural radiation sources (UNSCEAR, 1992). Furthermore, the by-products from industrial processes, such as fly ash and phosphogypsum, are currently used in cement production as pozzolanic additives, due to its economic price, even though some studies have found that the radon levels in dwellings using fly ash and phosphogypsum as building material additives are higher than in normally cemented dwellings (Chauchun et al., 2003). The use of these by-products as building material additives may affect doses from external radiation and the inhalation of radon decay products to occupants, especially in residencies with poor ventilation, like shophouses.

This research investigates the radon exhalation rates from concrete specimens with fly ash (FA) and phosphogypsum (PG) additives at different contents using hermetically closed chambers equipped with ATMOS 12 dpx radon gas monitors. In the following section, the radon concentrations were estimated using CFD simulation. The annual dose contributions were assessed using these results.

[1] The three radioactive isotopes are (1) ^{222}Rn – called radon, belongs to the ^{238}U decay series, (2) ^{220}Rn – called thoron, belongs to the ^{232}Th decay series, and (3) ^{219}Rn – called actinon, belongs to the ^{235}U decay series.

2. Basic equations and effective radon dose

The effective radon dose measures the potential health risk from radon exposure, calculated using various factors such as radon concentration, equilibrium factor, occupancy factor, and dose conversion factor. This calculations are crucial for assessing the risk of radon exposure in different environments, such as workplaces, homes, and schools. The effective dose is typically expressed in millisieverts (mSv) per year and is used to estimate the potential detriment to human health from radon exposure. Below are the key aspects of calculating and understanding effective radon dose.

The radon gas concentration ($C_i(t)$) inside a room can be described by equation 1 (Man & Yeung, 1999; Sahota et al., 2005; UNSCEAR, 1982):

$$\frac{dC(t)}{dt} = \frac{E \cdot S}{V} + C_0\lambda_v - C(\lambda + \lambda_v) \quad (1)$$

where C(t) is the time dependency of radon concentration inside a room (Bq/m3), E is the radon exhalation rate from the material ($Bq/m^2{\cdot}s$), S is the exhaling surface area of material (m^2), V is the volume of the room (m^3), C_0 is the radon concentration in the outside air (Bq/m^3), λ_v is the ventilation rate (s^{-1}) and λ is the decay constant of radon (2.1×10^{-6} s^{-1}).

At the steady state; $\frac{dC(t)}{dt} = 0$, and hence:

$$0 = \frac{E \cdot S}{V} + C_0\lambda_v - C(\lambda + \lambda_v) \quad (2)$$

$$C = \frac{\frac{E \cdot S}{V} + C_0\lambda_v}{\lambda + \lambda_v} \quad (3)$$

Since the hermetically closed chambers were very tight, radon from the outside air was not taken into account, and so:

$$C = \frac{E \cdot S}{V(\lambda + \lambda_v)} \quad (4)$$

According to the UNSCEAR 2000 Report, the annual absorbed dose due to indoor exposure to radon can be estimated by using the radon concentration (C_{Rn}), balance factor (F), occupancy factor (O), number of hours per year (T) and the dose conversion factor (D), as follows:

$$E_{indoor} = C_{Rn} \times F \times O \times T \times D \quad \text{mSv} \quad (5)$$

where E_{indoor} is the annual effective dose (mSv),C_{Rn} is the indoor radon concentration (Bq/m3), F is the equilibrium factor which depends on the level of ventilation (0.4 for natural ventilation and 0.2 for forced ventilation), O is the occupancy factor (0.8; estimated from the fact that people spend 80% of their time indoors), T = 24 hours x 365 days = 8760 hours, and D converts the radon concentration into a dose (9 nSv/ ($Bq{\cdot}hr/m^3$)).

The effective dose from radon exposure is calculated using a dose conversion factor, which the International Commission on Radiological Protection (ICRP) revised from 8 nSv / ($Bq{\cdot}hr/m^3$) to 33 nSv / ($Bq{\cdot}hr/m^3$) in 2017, reflecting an updated understanding of radon's health impacts (Chung et al., 2018). A radiation weighting factor of about 10 for alpha particles is suggested for consistency between epidemiological and dosimetric approaches, differing from the ICRP's previous value 20 (Beck, 2024). The equilibrium factor (F) is crucial in dose assessment, as it varies significantly across environments, from 0.15 to 0.94, with an average of 0.55. If a single coefficient is used, this variability can lead to underestimation or overestimation of the effective dose (Grygier & Skubacz, 2024). In industrial settings, such as gypsum and cement manufacturing, radon concentrations and effective doses vary, with annual doses ranging from 0.11 to 0.63 mSv depending on the process and location (Chung et al., 2018) and in educational settings, such as primary schools, radon concentrations in tap water result in annual effective doses well within global safety limits, indicating minimal risk from radon ingestion and inhalation (Jabar et al., 2023).

The ICRP has evaluated the risk of inducing lung cancer at 1.65×10^{-2} per Sv (Nazaroff and Nero, 1988); whereas, UNSCEAR reported that the worldwide average value of effective annual dose is 1.15 mSv/y (UNSCEAR, 2000). Whilst there is no 'safe' radon level, the EPA (Environmental Protection Agency) and the WHO (World Health Organization) have conducted studies and proposed a recommended radon action level at 2.0–4.0 pCi/L. If the radon level is higher than 4.0 pCi/L, radon mitigation should be considered immediately.

3. Materials and methods

In order to measure radon emissions, 11 concrete block samples (20 x 20 x 5 cm) with different FA and PG contents (from 0-60% by mass) were manufactured. The cement used throughout this experiment was ordinary Portland cement, type I. Fly ash was produced by the Mae

Moh power station in Lampang province and phosphogypsum was obtained from the NFC Public Company Limited, Thailand.

All of the samples were dried at 105 °C for 24 hours and placed in hermetically sealed chambers made of 8 mm thick, clear acrylic sheets (40 x 40 x 40 cm) (Figure 1), which was over 10 times larger than the pore volume of the sample, in order to minimize the back diffusion effect (Samuelsson, 1990). Nevertheless, the air inside each test chamber was purified by circulating it through activated charcoal and a radon-thoron filter for at least 20 minutes, in order to minimize radon and its progeny. The radionuclides content was measured after 27 days, to achieve secular equilibrium of the ^{226}Ra progeny (Figure 2).

After 27 days of exposure, the radioactivity of radium and the radon concentration in the enclosed test chambers were measured by using a pulse-counting ionization chamber radon gas monitor (Atmos 12 dpx). The device, connected via a closed circuit with a pump system (Figure 3), facilitates the continuous monitoring of radon levels by pumping filtered air into a pulsed ion chamber at a rate of 1 l/min. The radon concentration is then displayed in Bq/m^3 on a computer screen, providing real-time data on the radon levels within the test chamber. Using ionization chambers in pulse mode allows for precise measurement of radon concentrations, which is crucial for environments like mining where radon levels can fluctuate (Calin & Calin, 2011).

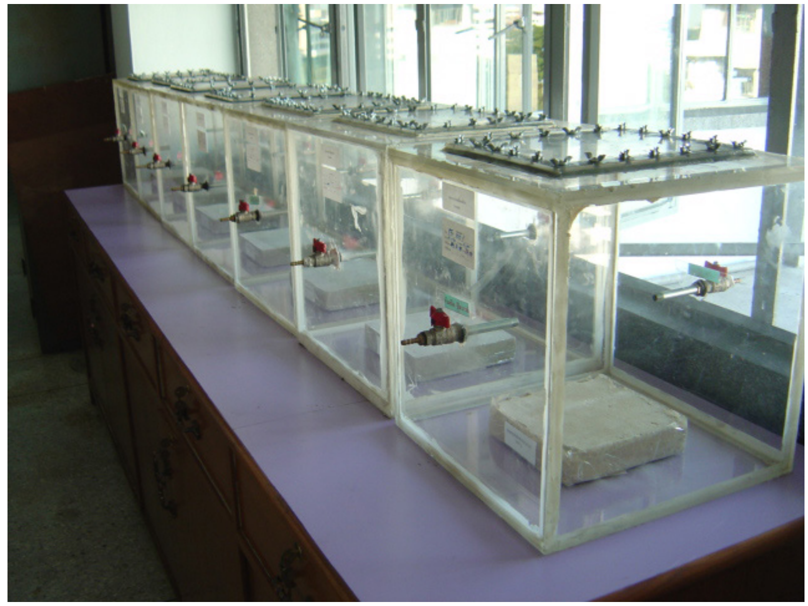

Figure 1. Samples in the radon test chambers

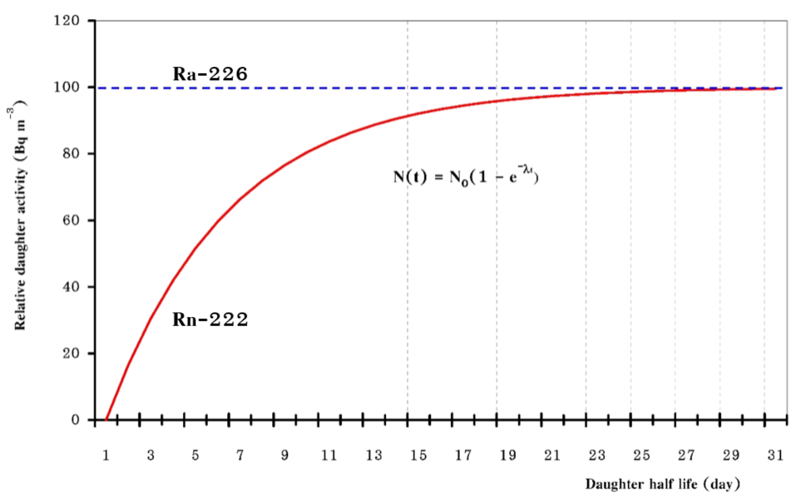

Figure 2. Secular equilibrium of ^{226}Ra progeny in the test chamber

Figure 3. Test chamber connected to an Atmos 12 dpx for radon measurement

Table 1. Radon exhalation rate from concrete with fly ash (FA) additives.

Material	Time (t) (hour)	λt	^{222}Rn Concentration (Bq m^{-3})	^{222}Rn Exhalation rate (Bq m^{-2} h^{-1})
Ce 100*	748	5.654	122 ± 6	0.395
FA 20	818	6.184	134 ± 0	0.429
FA 30	814	6.153	114 ± 1	0.312
FA 40	795	6.010	124 ± 3	0.380
FA 50	743	5.617	144 ± 1	0.556
FA 60	839	6.342	156 ± 1	0.564

Radon-decay constant (λ) = 7.56 x 10^{-3} h^{-1}

**Ordinary concrete without any by-product additive.*

Table 2. Radon exhalation rate from concrete with phosphogypsum (PG) additives.

Material	Time (t) (hour)	λt	^{222}Rn Concentration (Bq m^{-3})	^{222}Rn Exhalation rate (Bq m^{-2} h^{-1})
PG 20	749	5.662	321 ± 12	2.736
PG 30	795	6.010	335 ± 11	2.774
PG 40	745	5.632	395 ± 13	4.170
PG 50	846	6.395	604 ± 18	8.655
PG 60	821	6.206	670 ± 19	10.679

Radon-decay constant (λ) = 7.56 x 10^{-3} h^{-1}

4. Results

The results of radon concentration growth inside the chamber were measured by the Atmos 12 dpx for about 27 days and are presented in Tables 1-3.

As can be seen from the results in Tables 1 and 2, the radon exhalation rates from the sample materials significantly increased, in relation to increases in the amount of additives, both fly ash and phosphogypsum. In the case of concrete additives with phosphogypsum, the results reveal an obvious exponential correlation between radon exhalation rate and phosphogypsum content. The case of PG 20 only contained 20% phosphogypsum but exhaled 7 times more radon gas than ordinary concrete (Ce 100); whereas, in the case of fly ash additives, it was found that the radon exhalation rate of concrete additives with fly ash only increased slightly or even decreased, despite the higher radon exhalation rate, by mass, in fly ash compared with Portland cement (Table 3). This paradoxical behavior is consistent with the study conducted by Roelofs and Scholten, which concluded that, under certain conditions, the addition of fly ash to concrete could reduce the radon exhalation (Roelofs and Scholten, 1994).

The incorporation of fly ash and phosphogypsum into concrete materials significantly impacts the radon exhalation rate, with both materials offering potential benefits and challenges. When used in moderation, fly ash can reduce radon exhalation rates, while phosphogypsum's influence is more complex due to its inherent radioactivity. The influence of adding fly ash and phosphogypsum into concrete material on the radon exhalation rate is shown in Figure 4.

Material	Mass	λV	^{222}Rn Concentration	^{222}Rn Exhalation rate
	(kg)		(Bq m^{-3})	(Bq kg^{-1} h^{-1})
Cement	0.50	4.45 x 10^{-4}	12 ± 9	0.0106
Fly ash	0.50	4.45 x 10^{-4}	14 ± 7	0.0124
Phosphogypsum	0.50	4.45 x 10^{-4}	397 ± 24	0.3535

Radon-decay constant (λ) = 7.56 x 10^{-3} h^{-1}
Volume of test chamber (V) = 0.0589 m^3

Table 3. Radon exhalation rate from cement, fly ash and phosphogypsum.

5. CFD analysis: case study of a shophouse

Shophouses are one of the most popular building types in Thailand and have been used for both commercial and residential purposes for a long time. However, since they are situated on a limited amount of land and comprise more than two units, this causes problems in terms of a lack of daylight and poor ventilation, which leads to the accumulation of indoor radon.

To understand the distribution of radon in a poorly ventilated closed space, Computational Fluid Dynamics (CFD) software was used for simulation analysis.

The shophouse model studied was based on the general size of a shophouse: 3.5-4.0 m in width, 8.0-12.0 m in depth and 2.7-3.0 m height, with some internal partitions. The ventilation in each floor was provided by two windows in the front and the back of building (Figure 5).

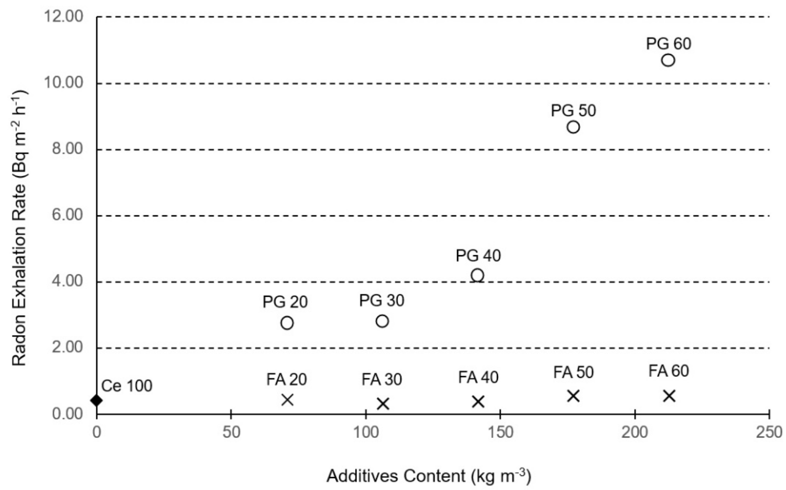

Figure 4. Radon exhalation rate from concrete with fly ash and phosphogypsum additives

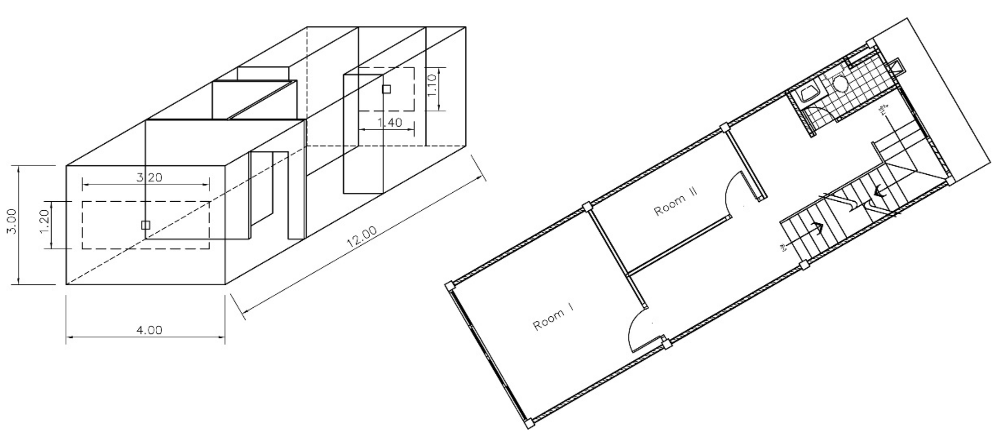

Figure 5. Shophouse model used for CFD simulation

In order to investigate the radon concentration inside the shophouse, two conditions were taken into account: the building materials (Ce 100, FA 50 and PG 50) and the ventilation, i.e., the indoor ventilation rate 0.35 ACH but not less than 15 cfm (the minimum ventilation rate in residential buildings recommended by ASHRAE, Standard 62.2-2016). The probe's position was set at a level 1.50 m above the floor, which is the approximate height of the inhalation region around a human head (Zhu et al., 2005). Radon gas emanated from the floor and walls but radon from the outside environment was neglected;
a low flow rate from the opening was set at 0.14 m^3/s. The boundary conditions for the simulation are given in Table 4.

Table 4. The boundary conditions for CFD simulation.

Parameter		Area (m^2)	Exhalation rate (Bq/m^2s)	Flow rate (m^3/s)
Space volume	144 m^3	48.00	-	-
Ventilation rate	0.35 ACH	-	-	0.14
Source material	Ce 100	145.72	1.12×10^{-4}	-
	FA 50	145.72	1.52×10^{-4}	-
	PG 50	145.72	2.40×10^{-3}	-

Velocity inlet boundary condition for the air inlet was considered uniform and the flow was normal to the inlet section.

Table 5. Indoor radon concentrations of CFD results.

Source Material	Rn Exhalation rate (Bq/m^2s)	Rn Concentration (Bq/m^3)		Annual Effective Dose (mSv/y^1)
		Ave. value	Probe value	
Ce 100	1.12×10^{-4}	5.82	30.24	0.147
FA 50	1.52×10^{-4}	7.90	41.04	0.199

Ventilation rate = 0.35 ACH

In the simulation, the shophouse model tested was kept close in order to imitate the poorly ventilated condition. The simulation time was set long enough to achieve a steady state radon concentration. The results from the CFD simulations are presented in the contour plots in Figure 6 (A – C) and Table 5.

Figure 6 shows the horizontal distribution of radon concentrations in a typical shophouse at the air change rate of 0.35 by CFD simulation. In general, most accumulated radon is found in Room II due to a lack of ventilation windows. The simulation shows that the average indoor radon concentrations, in the case of Ce 100 and FA 50, are 5.82 and 7.90 Bq/m^3, respectively. The case of using PG 50 as a building material presents the higher value of 124.90 Bq/m^3. Moreover, it is also found that the radon concentration in Room II shows the highest value of 398.10 Bq/m^3, which is 2.68 times higher than the limit of indoor radon concentration (148 Bq/m^3) recommended by USEPA (the U.S. Environmental Protection Agency).

Finally, according to the simulation results, the effective annual dose from the inhalation of radon was calculated as shown in Table 5. The effective annual dose, in the case of using PG 50 as a building material, reached up to 3.147 mSv y^{-1}, which is higher than the safety limit of 1 mSv recommended by UNSCEAR (UNSCEAR, 1993). This elevated dose is concerning, especially in poorly ventilated areas where radon can accumulate (e.g. Room III), increasing the risk of exposure and potential health hazards such as lung cancer.

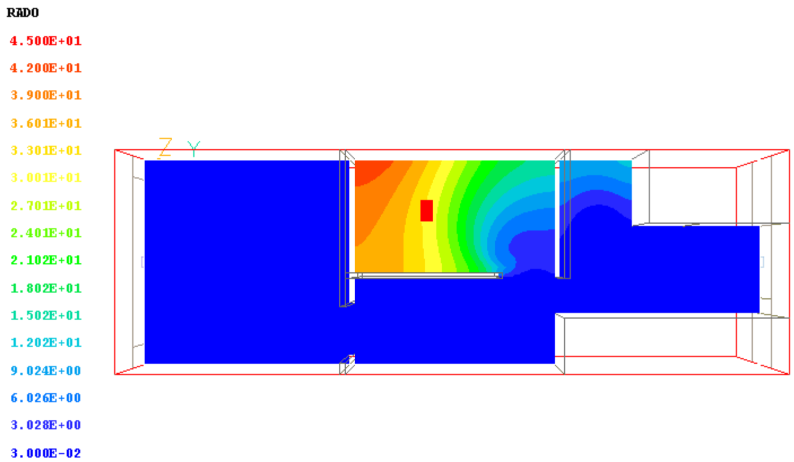

Figure 6A. Horizontal indoor radon concentrations: Ce 100

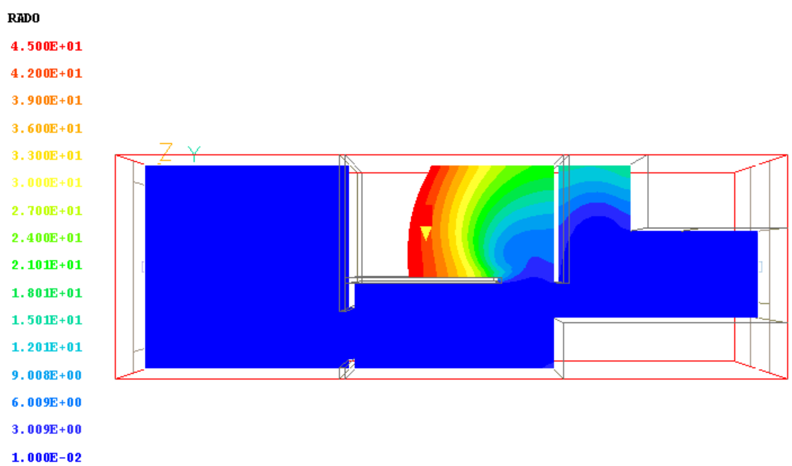

Figure 6B. Horizontal indoor radon concentrations: FA 50

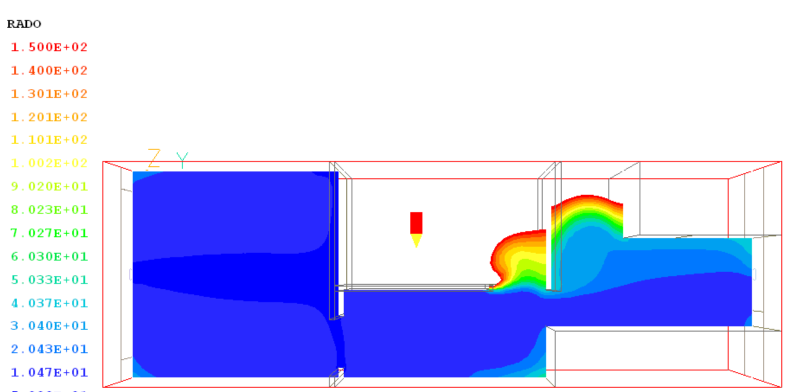

Figure 6C. Horizontal indoor radon concentrations: PG 50

6. Conclusion

This research investigated the exhalation of radon gas from concrete additives with industrial by-products, such as fly ash and phosphogypsum, which contain high amounts of radium. Samples of 11 concrete blocks with diverse of fly ash and phosphogypsum proportions were evaluated in hermetically sealed chambers utilizing an Atmos 12 dpx radon gas monitor. The findings demonstrated that while concrete containing fly ash manifested a gradual augmentation in radon emanation rates, the increase was relatively negligible compared to pure concrete (Ce100). In certain instances, such as FA 30 and FA 40, the radon emanation rates were even inferior to those of Ce 100. Conversely, concrete with phosphogypsum substantially escalated radon concentrations and emanation rates across all scenarios.

CFD simulations were executed with an assumed air change rate of 0.35 ACH to examine radon accumulation in a poorly ventilated shophouse. The results revealed mean radon concentrations of 5.82 Bq/m^3 (Ce 100), 7.90 Bq/m^3 (FA 50), and 124.90 Bq/m3 (PG 50) in Room II. Consequently, the annual effective dose for inhabitants utilizing PG 50 as a construction material attained 3.147 mSv/y, significantly surpassing the UNSCEAR safety threshold of 1.0 mS/y for indoor radon exposure. While fly ash presents minimal risk, phosphogypsum can amplify radon exposure, underscoring the necessity for meticulous assessment prior to its application in construction materials, particularly in inadequately ventilated environments.

CRediT Authorship Contribution Statement

Touchaphong Srisuwan: conceptualization, formal analysis, writing - original draft, writing – review and editing, conceptualization, writing – review and editing

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