STUDY ON RADON EMANATION FROM SELECTED BUILDING MATERIALS IN MALAYSIA

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ABSTRACT

Radon-222 emanation from selected locally produced samples of building materials, used in Malaysia were measured using the Professional Continuous Radon Monitor Model 1027, which is a patented electronic detecting-junction photodiode sensor to measure the concentration of radon gas. Each sample was placed for 72 hours inside a 3.11 x 10^{-2} m^3 sealed container. It was found that the average radon concentration A_t Bqm^{-3} of air for concrete bricks, concrete brick with cemented coatings, concrete brick with cemented coatings and paint samples were, 303.7 Bq/m^3, 436.6 Bqm^{-3}, and 410.7 Bqm^{-3}, respectively. A_t (Bqm^{-3}) for brown clay brick, brown clay brick with cemented coatings, brown clay brick with cemented coatings and paint were 166.5 Bqm^{-3}, 166.5 Bqm^{-3}, and 148 Bqm^{-3}, respectively. A_t (Bqm^{-3}) for sample of compact ceramic tile was 0 Bqm^{-3}. The findings show that concrete brick samples are important source of radon emanation, while brown clay brick have been accepted as the recommendation of the U.S. Environmental Protection Agency (EPA), and ceramic tiles had no emanation of radon gas due to their compact surface, or the glazed layer created on the tile surface during the manufacturing process, which blocks radon emanation. A positive correlation between radon emanation and radium content has been observed for both brown clay brick and concrete brick samples whereas a negative correlation for ceramic tile has been observed. Consequently from the findings, in order to reduce radon emanation and radon exposure in house dwellings and in addition to EPA recommendation of sealed cracks and established good ventilation, we recommend concrete walls to be painted and concrete floors to be paved with ceramic tiles.

ABSTRAK

Pembebasan Radon-222 dari bahan binaan tempatan yang digunakan di Malaysia telah ditentukan dengan menggunakan Professional Continuous Radon Monitor Model 1027, iaitu alat pengesan elektronik-sensor fotodiod yang digunakan untuk mengukur gas radon. Setiap sampel diletakkan selama 72 jam di dalam bekas kedap udara bersaiz 3.11 x 10^{-2} m^3. Didapati bahawa purata kepekatan radon A_t Bqm^{-3} dalam udara bagi bata konkrit, bata konkrit disaluti simen, bata konkrit disaluti simen dan cat adalah masing-masing 303.7 Bq/m^3, 436.6 Bqm^{-3}, dan 410.7 Bqm^{-3}. Nilai A_t (Bqm^{-3}) bagi bata tanah liat, bata tanah liat disaluti simen, bata tanah liat disaluti simen dan cat adalah masing-masing 166.5 Bqm^{-3}, 166.5 Bqm^{-3}, dan 148 Bqm^{-3}. Nilai A_t (Bqm^{-3}) bagi jubin seramik padat adalah 0 Bqm^{-3}. Hasil kajian mendapati bahawa sampel bata konkrit adalah sumber utama pembebasan radon, manakala bata tanah liat, dan jubin seramik tidak membebaskan gas radon disebabkan permukaannya yang padat, atau lapisan kilat di atas permukaan jubin yang sewaktu ia dihasilkan, di mana ia menghalang pembebasan radon. Kedua-dua sampel bata tanah liat dan bata konkrit didapati mempunyai korelasi positif antara pembebasan radon dan kandungan radium manakala jubin seramik didapati mempunyai korelasi negatif. Berdasarkan hasil kajian ini, *bagi memngurangkan pembebasan radon dan dedahan terhadap radon di dalam bangunan dan berdasarkan cadangan EPA, kami mencadangkan dinding konkrit dicat dan lantai konkrit dilitupi dengan jubin seramik.*

Keywords: Radon emanation, Building materials, Radon reduction.

INTRODUCTION

Radon is tasteless, odorless, and colorless, radioactive gas that is a decay product of radium. An emitter of ionizing radiation, it is the second leading cause of lung cancer after smoking (Laboratory, 1988). Moreover, the U.S. EPA has estimated that the average radon concentration in outside air is 14.8 Bqm^{-3} (0.4 $pCiL^{-1}$) and indoor is 148 Bqm^{-3} (4 $pCiL^{-1}$). If the average of the first and the second test is 4 $pCiL^{-1}$or higher the average year-round residential indoor radon level is estimated to be about 48.1 Bqm^{-3} (1.3 $pCiL^{-1}$), and about 14.8 Bqm^{-3} (0.4 $pCiL^{-1}$) of radon is normally found in outside air (EPA, 2005).

Radon comes from a decay of radium, present in all soils and rocks, water, and building materials. Rn-222 spontaneously breaks down into its four relatively short-lived progenies (Po-218, Pd-214, Bi-214, and Po-214) which mix with room air, then are inhaled by human lung, they emit alpha-particle, and may strike sensitive cells in the bronchial tubes and increase the risk of lung cancer. However, long lived radon progeny (Pd-210, Bi-210, and Po-210) contribute little to the dose because they are eventually removed by the mucous and cilia in GI tract before they can decay (Keller *et al*., 2001; Wang *et al*., 2002; Tung *et al*., 2005).

Since there are many factors control the emanation of radon from building materials: material structure, meteorology conditions, and radium, the later is the most important factor of radon emanation comparing other factors, which will allow us to measure and estimate radon (Chambaudet *et al.,* 1997).

Our main hypothesis is that the content of radium will regulate the radon emanation from construction and covering building materials in dwellings. If selected building materials are significant radon source in dwellings, curative medicine should be taken to addition to provide a good ventilation to reduce radon concentration in dwellings in Malaysia. Considering above-mentioned hypothesis and also null hypothesis there is no observable difference in radon emanation from construction building materials as compared to covering building materials.

In the current study, we measure radon emanation from selected locally building materials, which placed inside tightly radon container with Professional Continuous Radon Monitor Model 1027 for 72 h. Result shows that selected concrete bricks are notable source of radon emanation, while brown clay bricks have been accepted with the U.S. EPA's recommendation, and ceramic tiles had no emanation of radon gas due to their compact surface.

MATERIALS AND METHODS

Some chosen locally produced samples of construction and covering materials (2 brown clay bricks, 2 concrete bricks, one ceramic tile, cement, sand, and paint) commonly used in Penang were purchased from the supplier of building materials.

The Professional Continuous Radon Monitor Model 1027, which detected the alpha particles emitted by radon and its two daughters, polonium-218 and polonium-214 in the detection volume of the tight sealed container (3.11×10^{-2} m^3) was attached to the ceiling of the tight sealed container, and connected with a PC-based data acquisition and analysis system as shown in Figure 1. Background radon emanation for each sealed sample in the sealed container was measured for 24 h. Next, each selected sample (2 brown clay bricks, 2 concrete bricks) was placed inside the tightly radon container and radon concentration was allowed to build up with time and measured by radon monitor for 72 h. The experiment was repeated with 2 cement coated clay bricks and 2 cement coated concrete bricks. Next, each cemented sample was painted, dried, and measurements again taken. Finally, two equal parts of ceramic tile were placed separately inside the tightly radon container, the radon concentration inside the tightly radon container was allowed to build up with time and measured by radon monitor for 72 h.

RESULTS AND DISCUSSION

Results of measurements as presented in Table 1 show the overall average radon concentration At in concrete bricks was 303.4 Bqm^{-3}; in concrete bricks with cemented coatings, 421.8 Bqm^{-3}; in concrete bricks with cemented coatings and paint, 395.9 Bqm^{-3}; in brown clay bricks was 166.5 Bqm^{-3}; in brown clay bricks with cemented coatings, 166 Bqm^{-3}; in brown clay bricks with cemented coatings and paint, 148 Bqm^{-3}; and in ceramic tile was 0 Bqm^{-3}. Figure 2 shows a good correlation (R^2 = 0.5814, R^2 = 0.7211, and R^2 = 0.5436) between radium content and radon concentration, which has been observed in brown clay brick, cemented coating brown clay brick, and cemented coating concrete brick and painting respectively. But in case of concrete brick samples, cemented coating concrete brick, and cemented coating concrete brick and painting respectively, samples quite a good correlation (R^2 = 0.7635, R^2 = 0.8419), and R^2 = 0.9008)) between radium content and radon emanation (Figure 3) indicates that concrete brick samples are rich in uranium and radium.

The Professional Continuous Radon Monitor Model 1027 measures radon concentration of ambient air of the sealed container by unit $pCiL^{-1}$, which is unit for American System. The conversion factor is 1 $pCiL^{-1}$ equals 37 Bq/m^3. Thus, the radon concentration in air is expressed as $pCiL^{-1}$ or Bqm^{-3} (Lockey and Ross, 1994). The concentration, exhalation rate, and the maximum activity concentration of radon at infinite time detected in the building material samples collected from Penang, Malaysia are shown in Table 1.

Samples of concrete bricks, cemented concrete bricks, brown clay bricks, cemented brown clay bricks, show detectable radon exhalation, but painted ones and ceramics did not show detectable radon exhalation, which indicates that they did not contain detectable traces of uranium. The total area of the concrete brick samples varied from 4.35×10^{-2} to 4.887×10^{-2} m^2 with different thicknesses f 6.6 cm to 7.8 cm. Total area of the brown clay bricks varied from 4.444×10^{-2} m^2 to 5.945×10^{-2} m^2 with different thicknesses from 7.5 cm to 8 cm, and the total area of the ceramic tile is 4.47 x 10 m^{-2} with different thicknesses is 0.6 cm. Radon mass exhalation rate Es was significantly influenced by all factors; radium, porosity; water content, grain size, and sample mass (Dinh Chau *et al.*, 2005).

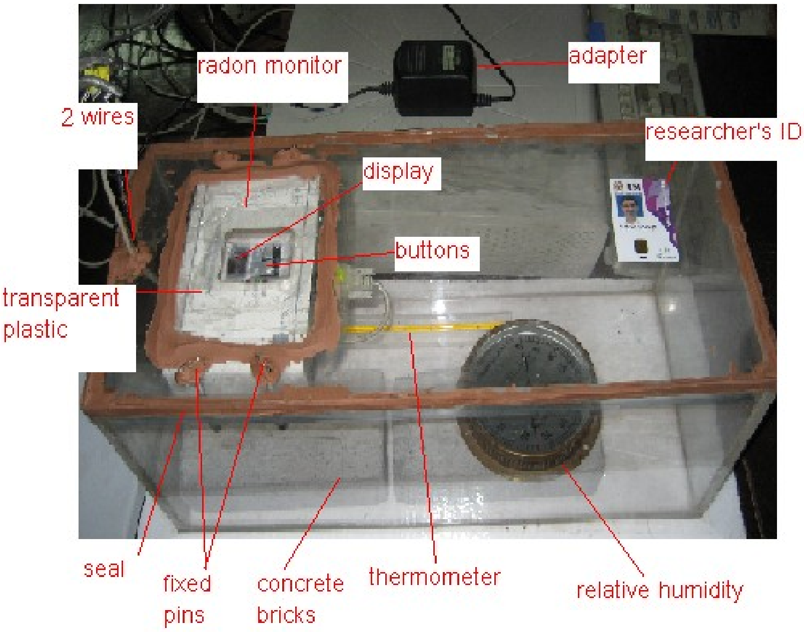

Figure 1: Radon detector attached to the ceiling of container. The selected sample placed at the bottom of the tight sealed container, while wires passed through small two openings. Also, relative humidistat and a thermometer placed at the bottom of the container.

Table 1: Mean radon concentration A_t, mean radon activity of the sample A_m, and E_s, and the maximum activity concentration of radon at infinite time A_0 from samples for 72h.

Samples	Mass (kg)	A_t, (Bq/m^3)	E_m (Bq/kg)	E_s Bq/m^{2}h	A_0 (Bq/m^3)
Two concrete bricks	5.71	303.7	3.938	3.905	723
Two concrete bricks with cemented coatings	7.16	436.6	4.515	5.01	1039.4
Two concrete bricks with cemented coatings and paint	7.25	410.7	4.194	4.71	977.73
Two brown clay bricks	4.75	166.5	2.595	2.099	396.4
Two brown clay bricks with cemented coatings	7.48	166.5	1.648	1.569	396.4
Two brown clay bricks with cemented coatings and paint	7.55	148	1.451	1.395	352.3
Part no. 1 of ceramic tile	1	0	0	0	0
Part no. 2 of ceramic tile	1	0	0	0	0

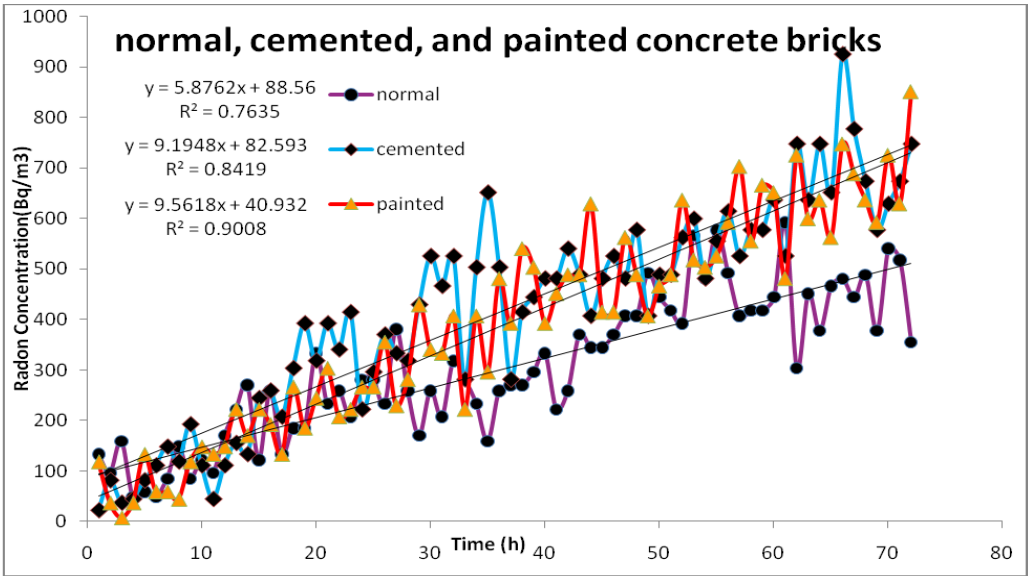

Figure 2: The typical observed curve of radon concentration growth in the tightly sealed exhalation chamber containing concrete brick samples collected from Penang, Malaysia.

In this study radon concentration (A_t) from each sample inside the tightly radon container was allowed to build up with time and was measured in 1 h cycle for 72 h radon activity given by equation:

$$A_t = A_o (1 - e^{-\lambda t}) \tag{1}$$

where A_t radon concentration (Bqm^{-3}) at time t (h); A_0 (Bqm^{-3}) is the complete saturation radon concentration when $t \approx 7\ T_{1/2}$; decay factor $e^{-\lambda t}$ is the fraction of radioactive atoms remaining after time (t); λ is the decay constant radon ($7.567 \times 10^{-3}\ h^{-1}$); t is the time of the buildup of radon activity inside the tight exhalation container (72 h). The half-life ($T_{1/2}$) of any nuclide is acquired by equation:

$$T_{1/2} = \ln/\lambda = 0.693/\lambda \tag{2}$$

where λ, $T_{1/2}$ are the decay constant and the half life of the nuclide concerned (interest); the number of elapsed half-lives in any time (n) = $t/T_{1/2}$.

In order to contrast exhaled radon from the measured samples, the radon mass exhalation rates A_m (radon activity of the sample A_m) ($Bqkg^{-1}$) for the samples is related to the units of mass of the selected sample in (kg) was calculated as follows:

$$A_m = (A_o V)/m \tag{3}$$

where V is the volume of the emanation container ($3.11 \times 10^{-2}\ m^3$), m is the mass of the selected sample in (kg) and A_0 (Bqm^{-3}) is the complete saturation radon concentration when $t \approx 7\ T_{1/2}$. The radon mass exhalation rates Es ($Bqm^{-2}h$) for the samples is related to the units of surface area of the selected samples in m^2 was calculated as follows:

$$E_s = A_o \lambda (V/S) \quad (4)$$

where λ, is the decay constant of radon (7.567x $10^{-3}h^{-1}$), V is the volume of the emanation container (3.11x10^{-2} m^3) and S is the total surface area of the selected samples. Complete details of the measurement method are given elsewhere (Al-Jarallah *et al.*, 2001), *(Aksoy et al., 2002)*, (Simon R. Cherry, 2003), (Al-Jarallah *et al.*, 2005), (Kovler *et al.*, 2005), and (Kovler, 2006).

The radon concentration of concrete bricks with cemented coating was higher due to the function of the superficial, inner pores, tough-grains, and lack of cement of concrete bricks. Furthermore, they had a little water content that allowed the diffusion rate of radon would be faster in the pore system. However, when they were coated with cement, the value of At increased, this mean that cement itself would contained uranium or radium. In addition, it was found that the radon exhalation is reduced inside the area of the sealed container due to paint, the age of the 4 concrete blocks and the progressive dehydration of concrete as it ages also reduced the water content in the pores of the concrete, so reducing the probability of trapping radon within the pores and hence the probability of radon emanation from these pores.

In the case of brown clay bricks, radon emanated only from the superficial pores of them, despite they had pores, cracks, and tough-grained surface. This is due to the fact that clay bricks were heated during their manufacturing process, had more tight space, and obtained poor source of radon.

When the bricks were coated with cement, the At increased, this means that cement itself may contained traces of uranium or radium. In addition, it found that the paint reduced radon exhalation from selected brown clay bricks. According to (Bossew, 2003), and (Righi and Bruzzi, 2006) ceramic tile did not exhibit detectable radon concentration even if they contain of the source of radon due to the difficulty for radon to escape from condensed solid matrixes where a reduction of the micro-porosity of the grain or by the glazed layer created on the tile surface during the manufacturing process, which blocks radon emanation.

CONCLUSION

In the study, selected building materials were left inside the tight sealed container for 72 hours in order to reach equilibrium radon concentrations, and its daughters. It was found that the average radon concentrations $\mathbf{A_t}$ t (Bqm^{-3}) for concrete bricks, concrete brick with cemented coatings, concrete brick with cemented coatings and paint samples were 303.7 Bqm^{-3}, 436.6 Bqm^{-3}, and 410.7 Bqm^{-3}, respectively; for brown clay brick, brown clay brick with cemented coatings, brown clay brick with cemented coatings and paint were 166.5 Bqm^{-3}, 166.5 Bqm^{-3}, and 148 Bqm^{-3} respectively; for sample of compact ceramic tile was 0 Bqm^{-3}. We may deduce that a negative correlation has been observed between the radon emanation and the radium presence of ceramic tile samples due to their a compact surface, or by the glazed layer created on the tile surface during the manufacturing process, which blocks radon emanate, and a positive correlation of brown clay brick to acceptable levels with the U.S. EPA's recommendation whereas comparatively significant correlation (R^2 = 0.9008) for concrete brick samples has been observed. Points to the conclusion that a reduction in radon emanation from building materials is to recommend residents for painting walls, paving the floors with ceramic tiles, and providing a good ventilation of dwellings.

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