Radon Mass and Surface Exhalation Rates in Ceramic Tiles available in Ibadan and associated Radiological Hazards

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Abstract

Radon mass and surface exhalation rates in selected ceramic tiles commonly used in Ibadan Southwest Nigeria were determined and the associated radiological parameters estimated. Forty-five samples of different types of floor and wall tiles were obtained from local markets. Each prepared sample was sealed an airtight radon sample container and kept for 28 days before analyses were carried out using a radon monitor (SARAD Radon Scout Plus). The mean values obtained for radon concentration, radon mass and surface exhalation rates were; 9.71 Bqm^{-3}, 4.78 $mBqkg^{-1}hr^{-1}$ and 282.44 $mBqm^{-2}hr^{-1}$ respectively. These values are lower than the corresponding world population-weighted average values. Again the estimated average values obtained for the annual effective dose rates due to radon inhalation, exposure to radon (radon effective dose equivalent) and excess lifetime cancer risk values were; 0.24 $mSvyr^{-1}$, 2.27 x 10^{-2} $WLMyr^{-1}$ and 7.95 x 10^{-4} (0.0795%) which are also below the world average values. This indicates that the use of these tiles in dwellings does not pose any significant radiological risks.

Keywords: ceramic tiles, radon exhalation rates, hazard parameters, radon monitor, sarad radon scout plus

1.0 Introduction

Human beings are exposed to radiation arising from sources in the lower and upper atmosphere some of these sources includes cosmic rays, naturally occurring radioisotopes in water, air, soil and plants and artificial radioisotopes due to fallouts from nuclear experimentation and medical uses. (Ademola *et al*, 2014).

The main naturally occurring radioisotope of concern in terrestrial environments are ^{226}Ra, ^{232}Th, ^{40}K and radon, which is the radioactive gas produced when radium radioisotopes decay. At normal levels these radioisotopes are usually of no radiological concern. However, some building materials may contain elevated concentrations of these radionuclides due to their origin, thereby posing radiological hazards when used for building purposes. The gamma radiation from terrestrial radionuclides and rays from the upper atmosphere such as cosmic rays constitute external exposure while radioisotopes inhaled and taken in by ingestion through air, foods particles and drinking water accounts for the internal exposure to the general public. Radon emanates from the ground as a result of the disintegration of naturally occurring radium (uranium) and it is a major source of radiation exposure to people (EPA, 2007). The exhalation of radon gas from building into the atmosphere is a function of the concentration of radium content in the soil and the type of building materials used for the construction of house which can in turn be inhaled by dwellers. Radon gas leak out of rocks and soil into the atmosphere or from ground water into buildings (Akerblom *et al*., 1984). The rate at which radon emanates from the soils and rocks depends on many factors, some of the factors includes temperature, moisture content and activity concentrations of radioisotopes (^{238}U and ^{226}Ra) present in the soils and rocks (Mayya *et al*., 1998). Radon and thoron exposure can be heightened or decreased by human activities, one of which is noteworthy is house construction. It may step up the concentration of airborne indoor radioactivity to a level that makes it unacceptable especially in places having low air-exchange rates (Mehta *et al*., 2015; Verma *et al.*, 2014). The concentrations of ^{222}Rn and ^{220}Rn indoors, is a function of the geological plenteousness of their parent elements and on their access to interiors of building. Hence, types of soil and rocks around dwellings are the main source of ^{222}Rn and ^{220}Rn to which general population is exposed (UNSCEAR, 2000).

Floor and wall tiles are one of the commonly used decorative building materials. In recent times, the use of these tiles in indoor decoration has increased in Nigeria and due to advancement in technology many building materials are now being produced from different combination of raw materials from the earth crust and industrial wastes such as fly ash (Kent *et al.*, 2010). The building materials of interest in this study are ceramic wall and floor tiles commonly used in Ibadan, Southwest Nigeria. The primary objective is to determine the radon exhalation rates and potential radiological hazards associated with the use of tiles in buildings.

2.0 Materials and Methods

A total of forty-five samples comprising five replicates of nine different types of wall and floor tiles were obtained from five building materials markets in Ibadan town. The different types were given codes as illustrated in Table 1. The samples were packed in polythene bags, marked appropriately and then taken to the laboratory where they were washed and sun-dried to remove any form of moisture.

Afterwards they were pulverized into powder from which 200 g of each was kept sealed in plastic cylindrical sample containers of the same dimensions as that of the reference sample (height 90 mm and diameter 70 mm) for over 30 days which is reasonable to attain which is reasonable to attain an equilibrium which is unperturbed over time between ^{226}Ra and its decay products of short half-lives. (Ravisankar *et al*., 2012). The activity concentrations of ^{222}Rn in these samples were determined using a radon monitor (Sarad Radon Scout Plus) and an accumulation chamber made of Perspex. The Sarad Radon Scout Plus equipment employs a solid state silicon detector to obtain the alpha particles emitted by radon gas and its decay products due to passive diffusion of radon gas from the indoor air into the device's diffusion chamber (Dawodu *et al*., 2015). The radon concentration for each sample from the accumulation chamber was detected and recorded by the radon monitor at every 3600 seconds. The monitor was later connected to a computer installed with the radon vision software which handles all setups and data transfer functions.

3.0 Results and Discussion

3.1 Radon Mass and Surface Exhalation Rates

The mass and surface exhalation rates due to radon gas from the samples using appropriate radiometric parameters were calculated using the following expression (Al-Saadi *et al*, 2015; Zubair *et al*, 2012):

$$E_m = \frac{CV\lambda}{M[T+\lambda^{-1}(e^{-\lambda T}-1)]} \quad 1$$

$$E_s = \frac{CTV\lambda}{S[T-\lambda^{-1}(1-e^{-\lambda T})]} \quad 2$$

Where E_m is the radon mass exhalation rate ($mBqkg^{-1}hr^{-1}$); E_s is radon surface exhalation rate ($mBqm^{-2}hr^{-1}$); M is mass of the sample (kg); S is the surface area exposed (m^2); T is the exposure time (hours); C is the average radon concentration (Bqm^{-3}); V is volume of the chamber (m^3) and λ is the decay constant of ^{222}Rn.

The average radon concentrations, radon mass and surface exhalation rates in the tiles are presented in Table 1. The average radon concentration varies from 11.85 Bqm^{-3} (Glazed floor tiles) to 8.48 Bqm^{-3} (Vitrified floor tile) with an arithmetic mean value of 9.71 Bqm^{-3} which is below the world average value of 200 Bqm^{-3} (UNSCEAR, 2006). Also the values of the mass exhalation rate for radon ranges from 4.18 $mBqkg^{-1}hr^{-1}$ (Vitrified floor tiles) to 5.84 $mBqkg^{-1}hr^{-1}$ (Glazed floor tiles) with an average of 4.78 $mBqkg^{-1}hr^{-1}$ and that of surface exhalation rates for radon ranges from 247 $mBqm^{-2}hr^{-1}$ (Ceramic floor tiles) to 345 $mBqm^{-2}hr^{-1}$ (Glazed floor tiles) with an average of 282.44 $mBqm^{-2}hr^{-1}$. From the result, radon exhalation rate is less than the world population average of 0.016 $Bqm^{-2}s^{-1}$ or 57,600 $mBqm^{-2}hr^{-1}$ (UNSCEAR 2000; Al-Saadi *et al*, 2015). Figure 2 illustrates a strong positive correlation (R^2 = 0.9999) between radon mass and surface exhalation rates. This implies that the radon mass and surface exhalation rates have corresponding characteristics.

3.2 Assessment of Radiological Hazards

The exposure to radon daughters in the tiles coupled with appropriate dose conversion factors form the basis for the evaluation of the radiological health hazard parameters (UNSCEAR 2000, EPA 2003; Asere and Ajayi, 2017; Omeje *et al*, 2018; Farhood *et al*, 2017)

3.2.1 Radon Annual Effective Dose Rate (indoor)

The mean radon concentration obtained for each sample is used to calculate the annual effective dose equivalent in dwellings. In doing this calculation, an average occupancy time of 7000 $hryr^{-1}$ and equilibrium factor of 0.4 and dose conversion factor of 9 x 10^{-6} $mSvhr^{-1}$ $(Bqm^{-3})^{-1}$ are assumed which is then used to convert radon concentration to population effective dose using the expression (UNSCEAR 2000, Asere and Ajayi, 2017):

$$\boldsymbol{H_e = C\ x\ F\ x\ T\ x\ D} \qquad 3$$

Where H_e is radon annual effective dose rate ($mSvy^{-1}$); C is the mean radon concentration (Bqm^{-3}); F is the equilibrium factor; T is the indoor occupancy time (hyr^{-1}) and D ($mSvhr^{-1}/Bqm^{-3}$) is the dose conversion factor. The calculated annual effective doses due to radon inhalation are presented in Table 1. The estimated value of radon annual effective dose (indoor) ranges from 0.21 $mSvyr^{-1}$ (Ceramic and Vitrified floor tiles) to 0.30 $mSvyr^{-1}$ (Glazed floor tiles) with an average of 0.24 $mSvyr^{-1}$ which is less than the world average value of 1.1 $mSvyr^{-1}$ (UNSCEAR, 2000).

3.2.2 Exposure to Radon Daughters and Excess Lifetime Cancer Risk

The exposure to radon daughters (radon effective dose equivalent) in the tile samples was calculated on the basis of the measured radon concentration using the following equation (Farhood *et al*, 2017; EPA 2003; UNSCEAR 2000):

$$\boldsymbol{E_R = R_C\ x\ (2.7\ x\ 10^{-4})\ x\ F\ x\ n\ x\ \frac{8766}{170}} \qquad 4$$

Where E_R = exposure to radon daughters in $WLMyr^{-1}$; R_c = radon concentration in Bqm^{-3}; (2.7 x 10^{-4}) is the factor for the conversion of radon concentration to the WL per Bqm^{-3}; F = the indoor equilibrium factor (= 0.4), n is the occupancy factor (= 0.42). Again, the excess lifetime cancer risk deals with the estimation of the probability of developing cancer over a lifetime at a given exposure level (Avwiri *et al*., 2013). It is presented as a value representing the number of cancers expected in a given number of people on exposure to a carcinogen at a given dose. It is important to note that an increase in the excess lifetime cancer risk (ELCR) causes a proportionate increase in the rate at which an individual can get cancer of the breast, prostate or even blood (Avwiri *et al.*, 2013). The excess lifetime cancer risk (ELCR) due to radon exposure in the tiles was determined using the following equation (Farhood *et al*, 2017; EPA 2003):

$$\boldsymbol{ELCR = E_R\ x\ T\ x\ F_R} \qquad 5$$

Where E_R is exposure to radon daughter (radon effective dose equivalent) in WLMyr^{-1}; T is average lifetime expectancy (estimated to be 70 years) and F_R is the risk coefficient factor for exposure to radon in equilibrium with its daughters. Based on the recommendations of ICRP 2009, the F_R is taken as 5 x 10^{-4} per WLM (Working Level Month). The calculated values for E_R and ELCR are presented in Table 1. The E_R values varied from 1.98 x 10^{-2} to 2.77 x 10^{-2} WLMyr^{-1} with an average value of 2.27 x 10^{-2} WLMyr^{-1} which is much lower than the average values 4 WLMyr^{-1} (UNSCEAR 2000, Farhood *et al*, 2017). Again the ELCR values ranged from 9.70 x 10^{-4} (0.097%) to 6.94 x 10^{-4} (0.0694%) with an average of 7.95 x 10^{-4} (0.0795%) which is less than the estimated lifetime cancer risk of 1.3% due to a radon exposure of 148 Bqm^{-3} (EPA 2003)

Table 1: Average radon concentration, Radon Mass and Surface exhalation rates, Annual, effective dose, Exposure to radon daughter and Excess lifetime cancer risk

Sample Type	**^{222}Rn (Bqm^{-3})**	**E_M (mBqkg^{-1}hr^{-1})**	**E_S (mBqm^{-2}hr^{-1})**	**H_E (indoor) (mSvyr^{-1})**	**E_R x 10^{-2} (WLMyr^{-1})**	**ELCR x 10^{-4}**
1	8.48	4.18	247	0.21	1.98	6.94
2	9.10	4.48	264	0.23	2.13	7.45
3	11.05	5.44	322	0.28	2.58	9.05
4	9.62	4.74	280	0.24	2.25	7.88
5	11.85	5.84	345	0.30	2.77	9.70
6	10.23	5.04	298	0.26	2.39	8.37
7	8.71	4.29	253	0.22	2.03	7.13
8	9.83	4.84	286	0.25	2.30	8.05
9	8.48	4.18	247	0.21	1.98	6.94
Mean	**9.71**	**4.78**	**282.44**	**0.24**	**2.27**	**7.95**
Minimum	**8.48**	**4.18**	**247**	**0.21**	**1.98**	**6.94**
Maximum	**11.85**	**5.84**	**345**	**0.30**	**2.77**	**9.70**

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Figure 1. Radon Scout Plus and the Sample in the accumulation chamber

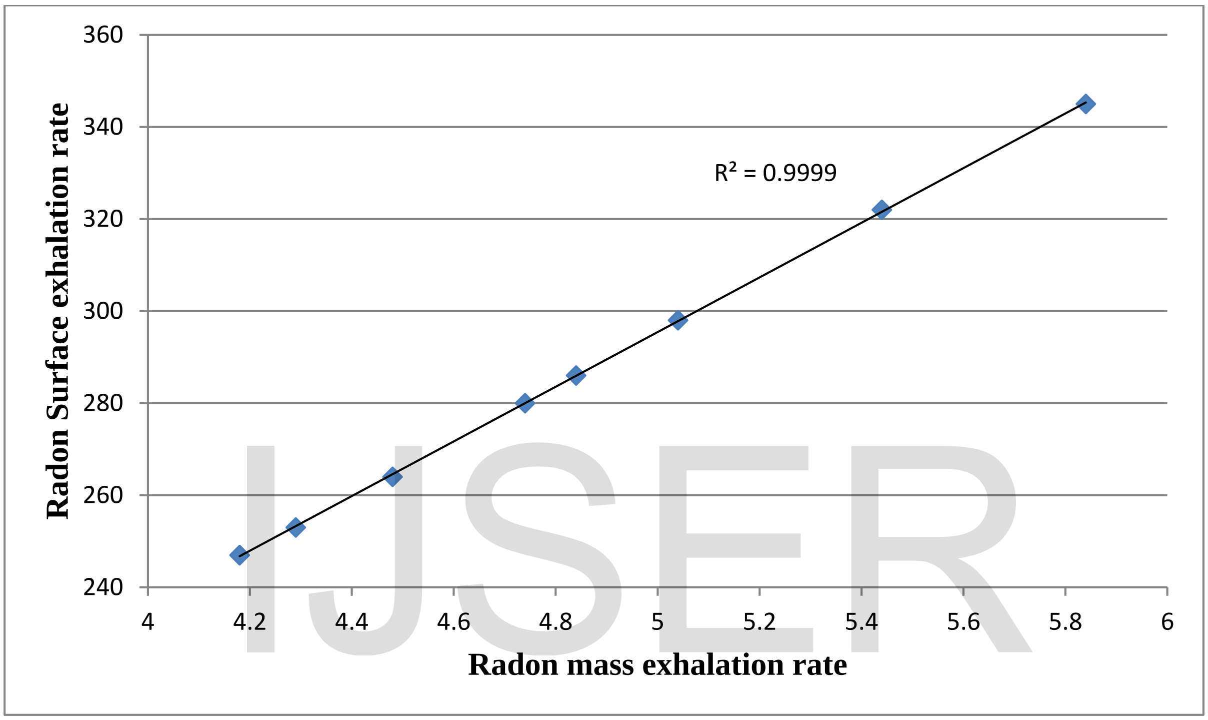

Figure 2. Radon mass exhalation rate against surface exhalation rate

Conclusion

The average radon concentration for the analyzed samples is less than the world population-weighted average value of 200 Bqm^{-3}. This may be due to the fact that the tiles samples may have different radium content which accounts for variation in radon emanation. Again the average radon exhalation rate is also below 0.016 $Bqm^{-2}s^{-1}$ (57,600 $mBqm^{-2}hr^{-1}$) for radon surface exhalation rate (UNSCEAR 2000; Al-Saadi *et al*, 2015) and the mean annual effective dose rate due to radon inhalation, exposure to radon daughter and excess lifetime cancer risk due to radon inhalation are also below the world population weighted average of 1.1 $mSvyr^{-1}$ for annual effective dose due to radon inhalation (UNSCEAR, 2000), 4 $WLMyr^{-1}$ for exposure to radon daughter (UNSCEAR

2000; Farhood *et al*, 2017) and 1.3% (EPA 2003). This shows that the samples analyzed do not have high radon exhalation rates and hence will not present radiological risk if used in dwellings.

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