NATURAL RADIOACTIVITY CONTENT AND RADON EXHALATION FROM MATERIALS USED FOR CONSTRUCTION AND DECORATION

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Abstract

The present work deals with the measurement of radioactivity and radon exhalation rate from geological samples manufactured in Douala city and used as building materials. Nine types of building materials were surveyed for their natural radioactivity content using a hyper purity germanium (HPGe) detector. The absorbed dose rate in the samples investigated ranged from 28.5 to 66.6 nGy h$^{-1}$ for brick samples, from 32.4 to 63.1 nGy h$^{-1}$ for roofing tiles and was 30.3 nGy h$^{-1}$ for concrete. External and internal hazard indexes were also estimated as defined by the European Commission.

The study of radon exhalation rate from building materials is important for well understanding the individual contribution of each material to the total indoor radon exposure. Solid state nuclear track detectors, CR-39 were used for measuring the radon concentration from different materials. Samples were hermetically closed in glass vessels and the radon growth was followed as a function of time. Exploring the one-dimension radon transport equation, we derived the radon exhalation rate from the experimental measurement of α-track densities. The radon exhalation varied from $(5.77\pm0.06) \times 10^{-5}$ to $(7.61\pm0.07) \times 10^{-5}$ Bq cm$^{-2}$ h$^{-1}$ in bricks, from $(5.79\pm0.05) \times 10^{-5}$ to $(11.6\pm0.12) \times 10^{-5}$ in tiles and was $(6.95\pm0.03) \times 10^{-5}$ Bq cm$^{-2}$ h$^{-1}$ in concrete. A positive correlation was found between uranium concentration measured with HPGe detector radon exhalation rate and radium content obtained using nuclear track detectors.
1 Introduction

Knowledge on geological materials contaminated with naturally occurring radioactive materials and used as building materials has become a focus of great attention in assessing the population exposure. Indeed, most of the dose rate of natural radioactivity is due to the members of the radioactive decay chains of \(^{238}\text{U}\) (55.8\%), \(^{232}\text{Th}\) (14\%), along with \(^{40}\text{K}\) (13.8\%) (UNSCEAR, 1993). The contribution of \(^{235}\text{U}\) to radiation exposure is negligible because the ratio \(^{235}\text{U} / ^{238}\text{U}\) is less than 1\%. Human being is exposed to natural radiation through external component due to \(\gamma\)-rays, and internal irradiation due to inhalation of radon and its short-lived decay products.

Gamma-ray spectrometry is a non destructive method suitable to quantify radionuclides in the environment. It has been shown that dose rate indoors increases when building materials with elevated natural radioactivity are used for constructions (Ahmad et al., 1997; Stoulos et al., 2003).

The use of nuclear track detector CR-39 is convenient since it has high registration sensitivity and the possibility to be used for long period without any fading. This method has been applied worldwide by many authors to determine the radon exhalation from building materials and soils (Al-Jarallah et al., 2001; Hafez et al., 2001; Sroor et al., 2001; Walley El-Dine et al., 2001; Shafiur-Rehman et al., 2006). Exhalation of radon \(^{222}\text{Rn}\) (\(T_{1/2} = 3.825\) d) from building materials is associated with the presence of radium \((^{226}\text{Ra})\) and its ultimate precursor uranium \((^{238}\text{U})\). In fact people spend most of their time indoor (80\%) and inhalation of radon and its decay products (especially \(^{218}\text{Po}\) and \(^{214}\text{Po}\) attached to aerosols) constitute a significant radiation hazard to human lung. In poorly ventilated spaces the radon concentration may reach levels of great concern so that precautions must be taken. The United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1994) estimated that the inhalation of radon and its short-lived decays products contributes on average for about one half of the effective dose from all natural sources of radiation. As a noble gas, radon can move through pores of building materials and rocks in large distances. This motion within building materials is governed by a radon transport equation and dominated by molecular diffusion (Nazaroff and Nero, 1988).

This study deals with the determination of the natural radioactivity content and the radon exhalation rate from bricks, tiles and concrete in order to give guidelines and legislative protection against natural radiation for people living in Cameroonian dwellings. In fact assessment of specific activity and radiation hazards in some materials of local origin have just being initiated (Ngachin et al., 2007). Data on radon exhalation rates, emanation and diffusion coefficients in building materials used in Cameroon are unavailable. Measured values of \(^{238}\text{U}, ^{232}\text{Th}\) and \(^{40}\text{K}\) concentrations, external and internal indexes, surface exhalation rate are reported and compared with the exempt levels proposed by the United Nation Scientific Committee on the Effects of

\begin{equation}
\end{equation}
Atomic Radiation (UNSCEAR, 1988). The total radiation dose is also evaluated and could be of great interest to manufactories, national regulatory board and construction companies.

2 Theoretical approach of radon transport in porous media

Radon is being produced in soil as a result of the presence of trace amounts of $^{238}\text{U}$. Depending on the properties of the soil such as porosity, permeability, presence of cracks and the conditions such as moisture content, pressure gradient and among others, radon can be transported through building materials pores and reach the dwelling indoor. $^{226}\text{Ra}$ decays to $^{222}\text{Rn}$ through $\alpha$-disintegration. Once radon is generated into the porous material through emanation ($^{222}\text{Rn}$ escape from a grain), it is transported to the indoor environment. Two mechanisms play an important role in the transport of radon: diffusive transport due to the difference in radon concentration and convective transport due to the pressure gradient induced by air flow. The transport of radon in building material is described by a general equation of continuity which includes processes such as molecular diffusion, radon decay, convective process and radon generation. This time dependent radon transport in air filled pores is given by the following equation assuming that the gas transport in the soil water phase can be neglected. This assumption remains true for $^{222}\text{Rn}$ since its molecular diffusion coefficient is $10^4$ times higher in air than in water (Dueñas et al., 1997).

\begin{equation}
\beta \frac{\partial C_a}{\partial t} = \nabla \cdot (D \nabla C_a) + \frac{K \nabla P(x, y, z) \nabla C_a}{\mu} - \beta \lambda C_a + G_o
\end{equation}

with

\begin{align*}
\beta &= \text{partition-corrected porosity} = (1 - m + Lm)\epsilon + \rho_b k_a. \text{ It depends on the moisture content in the material.} \\
\epsilon &= \text{total porosity (air phase and water phase)} \\
m &= \text{volume fraction of pores occupied by water.} \\
L &= \text{Ostwald coefficient or solubility coefficient, } L = \frac{C_w}{C_a} = 0.26 \text{ at 293K} \\
\rho_b &= \text{bulk density of dried material (kg m}^{-3}) \\
k_a &= \text{radon surface adsorption coefficient ( m}^3 \text{ kg}^{-1}). \ k_a = \frac{C_s}{C_a} \\
C_a &= \text{radon concentration in air-filled pores (Bq m}^{-3}) \\
C_w &= \text{radon concentration in water-filled pores (Bq m}^{-3}) \\
C_s &= \text{radon concentration in solid phase, ie the adsorbed radon activity (Bq kg}^{-1})
\end{align*}
\(D\) = bulk diffusion coefficient. It relates the interstitial concentration of radon to the flux density across the geometric area (m\(^2\) s\(^{-1}\))

\(K\) = intrinsic permeability (m\(^2\))

\(\mu\) = dynamic viscosity for air (\(\mu = 1.83 \times 10^{-5}\) Pa s)

\(P\) = pressure disturbance field at a given point in the sample (Pa)

\(\lambda\) = decay constant of radon (\(\lambda = 2.1 \times 10^{-6}\) s\(^{-1}\))

\(G_o\) = radon generation rate per unit bulk volume (Bq m\(^{-3}\) s\(^{-1}\))

In this study, the following simplifications have been considered

1. Pressure disturbance field remains constant because of low pressure gradient in building materials sealed in the chamber
2. Samples are dried (\(m = 0\))
3. Radon diffuses only in z-direction
4. Surface adsorption is negligible (\(k_a = 0\))

Under these four considerations, \(\beta = \epsilon\) and Eq. (1) becomes

\[
\epsilon \frac{\partial C_a}{\partial t} = \frac{\partial}{\partial z}(D \frac{\partial C_a}{\partial z}) - \epsilon \lambda C_a + G_o
\]

As measurements take place when equilibrium is established, the steady state equation derived from Eq. (2) is reduced to the following form

\[
D \frac{\partial^2}{\partial z^2} - \epsilon \lambda C_a + G_o = 0 , \quad G_o = \lambda C_{Ra} \rho_b \eta
\]

where \(C_{Ra}\) is the radium concentration per unit mass of the material (Bq kg\(^{-1}\)) and \(\eta\) the radon emanation coefficient. The general solution of Eq. (3) is given by

\[
C_a(z) = k_1 \exp(\frac{\sqrt{\lambda \epsilon}}{D}z) + k_2 \exp(-\frac{\sqrt{\lambda \epsilon}}{D}z) + \frac{G_o}{\lambda \epsilon}
\]

Constants \(k_2\) and \(k_2\) will be defined under boundary conditions. The diffusion length \(l\) (m) is often used in the literature. It is related to the diffusion coefficient according to the following relation

\[
l = \sqrt{\frac{D \epsilon}{\lambda}} = \sqrt{\frac{D e}{\lambda}}
\]
$C_a(z) \to C_0$ as $z \to z_0$ and $\frac{\partial C_a(z)}{\partial z} \to 0$ as $z \to 0$.

For these boundary conditions, constants $k_1$ and $k_2$ in Eq. (4) are determined.

$$k_1 = k_2 = \frac{C_0 - \frac{G_o}{\lambda \epsilon}}{\exp(\frac{z}{l}) + \exp(-\frac{z}{l})} \tag{6}$$

Substituting Eq. (6) in Eq. (4), we obtain

$$C_a(z) = \left( C_0 - \frac{G_o}{\lambda \epsilon} \right) \cosh\left(\frac{z}{l}\right) + \frac{G_o}{\lambda \epsilon} \tag{7}$$

As the thickness of the sample $z_o$ (few centimeters) is small compared to the $^{222}$Rn diffusion length $l(\sim 1 \text{ m})$, then $\cosh\left(\frac{z}{l}\right) \approx 1$ and Eq. (7) becomes

$$C_a(z) = \left( C_0 - \frac{G_o}{\lambda \epsilon} \right) \cosh\left(\frac{z}{l}\right) + \frac{G_o}{\lambda \epsilon} \tag{8}$$

The radon flux at the surface $S$ of the sample, usually called exhalation rate is given by Fick’s law of diffusion, and determined as follows

$$J = -D \frac{\partial C_a(z)}{\partial z} \bigg|_{z=z_o} = D \left( C_0 - \frac{G_o}{\lambda \epsilon} \right) \frac{\sinh\left(\frac{z}{l}\right)}{l} \tag{9}$$

In this case of thick samples $(z_o < l)$, $\sinh\left(\frac{z}{l}\right) \to \frac{z}{l}$, and Eq. (9) becomes

$$J = \lambda C_{Ra} \rho_b \eta z_o - \epsilon \frac{D_c z_o C_0}{l^2} \tag{10}$$

The second term in the right-hand side of Eq. (10) represents the back diffusion characterized by the back diffusion parameter $\beta = \frac{D_c z_o}{l^2}$. This back diffusion could be avoided with short growth time (Nazaroff and Nero, 1988) or by using diffusion chamber with effective volume that is 10 times higher than the volume of the sample. That was the case in this study since the effective volume of the chamber $V_e$ defined as below was about 2 liters compared to about 0.195 liter occupied by sample. So no “back diffusion” correction was necessary.

$$V_e = V_c - V_s$$

where $V_c$ is the volume of the chamber without sample and $V_s$ the volume of the sample.

The rate of change in radon activity concentration in the volume of the chamber can therefore be written as follows

$$\frac{dC(t)}{dt} = -\lambda C(t) + \eta \lambda \rho_b C_{Ra} \tag{11}$$

The first term on the right-hand side describes the loss of radon through decay and the second term describes the generation of radon through radium decay.
where $S$ is the surface area of the sample ($\text{m}^2$) and $V$ the effective volume of the chamber ($\text{m}^3$).

Solving Eq. (12) by assuming that $C(t) = 0$ at the beginning of the measurement, the radon concentration $C(t)$ in the air volume of the chamber is given by the following equation

$$C(t) = \frac{J_o S}{\lambda V} \left(1 - \exp(-\lambda t)\right), \quad J_o = \eta \lambda \rho_b C_{Ra} z_o$$  \quad (13)

The etched track detector CR-39 provides a measure that indicates the integrated exposure of a few weeks. Then with a given detector efficiency $\varepsilon$ (in tracks $\text{m}^{-2} \text{h}^{-1} \text{per Bq m}^{-3}$), the track densities observed during the exposure time $T$ is given by

$$\varrho = \varepsilon \int_0^T C(t) dt = \varepsilon \frac{J_o S}{\lambda V} \left(T - \frac{1}{\lambda} (1 - \exp(-\lambda T))\right)$$  \quad (14)

Rearranging Eq. (14), the exhalation rate is deduced and given by

$$J_o = \frac{\varrho V \lambda}{\varepsilon S T_e}$$  \quad (15)

where $T_e = T - \frac{1}{\lambda} (1 - \exp(-\lambda T))$ is the effective exposure time.

Comparing Eq. (15) and the expression of $J_o$ in Eq. (13), the effective radium content is derived as

$$\eta C_{Ra} = \frac{\varrho V}{\varepsilon T_e M}$$  \quad (16)

where $M$ (kg) is the mass of the sample.

3 Experimental procedures

3.1 Method for $\gamma$-ray spectrometry

Nine types of building materials (Three types of bricks, Five types of roofing tiles and concrete) were obtained from manufacturers in the locality of Douala, known to be the biggest city in Cameroon. Three samples were obtained from each type of material for more accuracy. Therefore 27 samples were prepared for experiments. Samples were weighed (0.600-0.680 kg), sealed and stored for one month to allow radioactive equilibrium in the $^{238}\text{U}$ and $^{232}\text{Th}$ decay series. The experimental method for $\gamma$-ray spectrometry and the determination of concentration of primordial radionuclides in samples were previously described (Ngachin et al, 2007). The
detector efficiency and energy calibration have been carried out using a multi-gamma standard in Marinelli beaker shape provided by Cerca Framatome in France. The range of energy in the standard was 59.54-1836 keV, including radionuclides such as $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{203}$Hg, $^{113}$Sn, $^{85}$Sr, $^{137}$Cs, $^{60}$Co, $^{88}$Y. The counting time was 80000 s to obtain the spectrum with good statistics, such that the overall uncertainty was less than 10% at the 95% confidence level. Since the standard and the samples were different in density, a self absorption correction (Sima, 1992) was performed to obtain the correct activities.

3.2 $^{222}$Rn exhalation with solid state nuclear track detectors

A large number of methods for determining radon exhalation have been developed (Stranden, 1988; Stoulos et al., 2003). In this work a tight closed cylindrical jar was used for radon exhalation measurement. The samples were dried at room temperature and crushed to fine grain size (100 $\mu$m). Long term time-integrated measurements were performing by placing samples in glass containers of 10 cm diameter and 25 cm height. The exhaled radon was measured using polycarbonate CR-39 detectors supplied by Radosys Co., Ltd., Hungary. The CR-39 detectors used were 1cm $\times$ 1cm. Detection system were built by installing detectors in the bottom of the chamber covers at a distance of 22 cm from the surface of the sample. Before installing CR-39, samples were stored for 1 month so that radon has achieved equilibrium with radium. The detectors were carefully installed and the containers were hermetically sealed for a known period of time. During the exposure, $\alpha$ particles from radon and its decay products strike the CR-39 and cause damage tracks. At the end of the exposure detectors were collected and chemically etched. The etching conditions adopted in our laboratory were 6.25% NaOH at 70°C for 4.5 h etching time, immediately followed by a 15 min neutralization with diluted acidic solution (4 $\times$ 10$^{-5}$ M at 60°C). Finally detectors were washed in distilled water during 15 min in order to remove any chemicals excess and air-dried for 1 day. The number of tracks were scanned in 144 fields using an optical microscope of 40× magnification objective lens. The tracks density left on track films allowed to evaluate the radon exhalation rate using Eq. (15). An amount of about 300 g of each sample with 78.5 cm$^2$ surface area was considered for radon exhalation rate measurement.

4 Results and discussion

4.1 Natural radioactivity concentration

A total of 27 samples were investigated for their natural radioactivity content. The mean radionuclide activity in tiles, brick and concrete are reported in Tables 1 and 2, respectively.

The average activities of $^{238}$U, $^{232}$Th, and $^{40}$K in samples investigated are reported in Table 3. The maximum value of $^{238}$U activity is found in sample (4) (49±1 Bq kg$^{-1}$) whereas the
minimum is found in sample (1) (11.2±0.3 Bq kg⁻¹). For ²³²Th, the maximum activity was found in sample (7) (37±4 Bq kg⁻¹) and the minimum in sample (1) (16±1 Bq kg⁻¹). The values of ⁴⁰K concentrations ranged from 306±8 Bq kg⁻¹ in sample (5) to 774±21 Bq kg⁻¹ in samples (7). Results are comparable to those measured worldwide (Savidou et al., 1995; Safdar et al., 1996; Giuseppe et al., 1997; Petropoulos et al., 2002; Righi et al, 2006; Ngachin et al., 2007). Figures 2a and 2b show the spectra of brown tile and red brick, respectively with more than 16 resolved photo peaks. From these results, it can be seen that the mean concentrations of ²³⁸U and ²³²Th in the samples investigated are less than their corresponding world typical average of 50 and 50 Bq kg⁻¹, respectively, whereas the mean activities of ⁴⁰K in black tile and red brick exceed the world average value of 500 kg⁻¹ (UNSCEAR, 1993).

The γ-dose rate $\dot{D}$ given in column 7 of Table 3 was found to range from 28.5 nGy h⁻¹ in black brick to 66.6 nGy h⁻¹ in red brick. Except samples (2) and (7) which exceed the world average of 55 nGy h⁻¹ all the other materials lie within the range 18-93 nGy h⁻¹ reported in the UNSCEAR (1988) report.

The radium equivalent activity: Its is defined to compared the activity concentration of materials containing different amount of ²³⁸U, ²³²Th and ⁴⁰K since their distribution in building material is not uniform. It is calculated through the following expression

$$Ra_{eq} = 0.077 \times C_K + 1.43 \times C_{Th} + C_U$$

where $C_K$, $C_{Th}$ and $C_U$ are the concentrations of ⁴⁰K, ²³²Th and ²³⁸U, respectively. Table 3 indicates that the range of radium equivalent was 65.2-127 Bq kg⁻¹ in tiles and was 28.5-133
Figure 2: A part of Spectra showing: a) high concentration in brown tile and b) low concentration in red brick
Table 1: Activity in investigated roofing tiles (in Bq kg\(^{-1}\))

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Black tile</th>
<th>Brown tile</th>
<th>Gray tile</th>
<th>Red tile I</th>
<th>Red tile II</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{238})U decay series</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{214})Bi</td>
<td>27±1</td>
<td>51±1</td>
<td>51±1</td>
<td>19±1</td>
<td>12.0±0.4</td>
</tr>
<tr>
<td>(^{214})Pb</td>
<td>23±1</td>
<td>46±1</td>
<td>39±1</td>
<td>21±1</td>
<td>13±1</td>
</tr>
<tr>
<td>(^{232})Th decay series</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{228})Ac</td>
<td>35±1</td>
<td>23±1</td>
<td>21±1</td>
<td>28±2</td>
<td>17±1</td>
</tr>
<tr>
<td>(^{212})Bi</td>
<td>41±3</td>
<td>29±4</td>
<td>29±3</td>
<td>29±4</td>
<td>21±1</td>
</tr>
<tr>
<td>(^{212})Pb</td>
<td>29±1</td>
<td>24±1</td>
<td>25±1</td>
<td>32±3</td>
<td>17±1</td>
</tr>
<tr>
<td>(^{40})K</td>
<td>683±15</td>
<td>351±12</td>
<td>337±11</td>
<td>404±13</td>
<td>350±12</td>
</tr>
</tbody>
</table>

Table 2: Activity in investigated bricks and concrete (in Bq kg\(^{-1}\))

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Black brick</th>
<th>Brown brick</th>
<th>Red brick</th>
<th>Concrete</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{238})U decay series</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{214})Bi</td>
<td>11±0.3</td>
<td>11±1</td>
<td>21±2</td>
<td>12.0±0.3</td>
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<tr>
<td>(^{214})Pb</td>
<td>12±0.4</td>
<td>13±1</td>
<td>20±1</td>
<td>13.3±0.3</td>
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<tr>
<td>(^{232})Th decay series</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{228})Ac</td>
<td>15.0±0.3</td>
<td>14±1</td>
<td>35±2</td>
<td>17.4±0.3</td>
</tr>
<tr>
<td>(^{212})Bi</td>
<td>17±1</td>
<td>17±3</td>
<td>47±7</td>
<td>18±1</td>
</tr>
<tr>
<td>(^{212})Pb</td>
<td>16±1</td>
<td>17±2</td>
<td>30±2</td>
<td>18±1</td>
</tr>
<tr>
<td>(^{40})K</td>
<td>307±10</td>
<td>320±12</td>
<td>774±21</td>
<td>306±8</td>
</tr>
</tbody>
</table>

Bq kg\(^{-1}\) in bricks. The value of \(Ra_{eq}\) in concrete was 61.5 Bq kg\(^{-1}\). It can be seen that the calculated \(Ra_{eq}\) does not exceed the recommended maximum value of 370 Bq kg\(^{-1}\) (OECD, 1979).

The external index: The gamma-index (\(I_\gamma\)) is defined in order to examine the applicability of using building materials in construction. For a typical material it is given by the following expression

\[
I_\gamma = \sum_x \frac{C_x}{A_x} \leq 1 \tag{18}
\]

where \(C_x\) (Bq kg\(^{-1}\)) is the measured activity of each nuclide in the building material. \(A_x\) (Bq kg\(^{-1}\)) is the activity concentration of each nuclide in the material and assumed to produce the same gamma dose rate, i.e 300, 200 and 3000 Bq kg\(^{-1}\) for \(^{238}\)U, \(^{232}\)Th and \(^{40}\)K, respectively (European Commission, 1999). Column 8 of Table 3 indicates that \(I_\gamma\) is less than unity in all
materials investigated.

The internal index: Several α-indexes have been proposed to assess the exposure level due to radon inhalation originating from building materials (EC, 1999; Stoulos et al., 2003). It is defined as follows

\[ I_\alpha = \frac{C_U}{200 \text{ Bq kg}^{-1}} \]  

(19)

where \( C_U \) (Bq kg\(^{-1}\)) is the activity concentration of \(^{238}\)U assumed in equilibrium with \(^{226}\)Ra. The recommended exemption level and the recommended upper level for \(^{226}\)Ra suggested by the International Commission on Radiological Protection (ICRP, 1994) are 100 Bq kg\(^{-1}\) and 200 Bq kg\(^{-1}\), respectively. As indicated in Table 3, \(^{238}\)U activities in all samples investigated were lower than 200 Bq kg\(^{-1}\) so that the alpha-indexes in building material did not exceed the recommended upper level.

Table 3: Mean activities, radium equivalent Ra\(_{eq}\), absorbed dose rate \( \dot{D} \), gamma index \( I_\gamma \), and alpha index \( I_\alpha \) in building materials investigated

<table>
<thead>
<tr>
<th>Sample</th>
<th>Code</th>
<th>( ^{238})U Activity (Bq kg(^{-1}))</th>
<th>( ^{226})Th Activity (Bq kg(^{-1}))</th>
<th>( ^{40})K Activity (Bq kg(^{-1}))</th>
<th>Ra(_{eq}) (Bq kg(^{-1}))</th>
<th>( \dot{D} ) (nGy h(^{-1}))</th>
<th>( I_\gamma )</th>
<th>( I_\alpha )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Black brick</td>
<td>1</td>
<td>11.2±0.3</td>
<td>16±1</td>
<td>307±10</td>
<td>57.5</td>
<td>28.5</td>
<td>0.22</td>
<td>0.056</td>
</tr>
<tr>
<td>Black roofing tile</td>
<td>2</td>
<td>25±1</td>
<td>35±2</td>
<td>683±15</td>
<td>127</td>
<td>63.1</td>
<td>0.48</td>
<td>0.12</td>
</tr>
<tr>
<td>Brown brick</td>
<td>3</td>
<td>12±1</td>
<td>16±2</td>
<td>320±12</td>
<td>59.5</td>
<td>29.5</td>
<td>0.23</td>
<td>0.059</td>
</tr>
<tr>
<td>Brown roofing tile</td>
<td>4</td>
<td>49±1</td>
<td>25±2</td>
<td>351±12</td>
<td>111</td>
<td>52.5</td>
<td>0.4</td>
<td>0.24</td>
</tr>
<tr>
<td>Concrete</td>
<td>5</td>
<td>12.6±0.3</td>
<td>18±1</td>
<td>306±8</td>
<td>61.5</td>
<td>30.3</td>
<td>0.23</td>
<td>0.063</td>
</tr>
<tr>
<td>Gray roofing tile</td>
<td>6</td>
<td>45±1</td>
<td>25±2</td>
<td>337±11</td>
<td>107</td>
<td>50.5</td>
<td>0.39</td>
<td>0.22</td>
</tr>
<tr>
<td>Red brick</td>
<td>7</td>
<td>20±2</td>
<td>37±4</td>
<td>774±21</td>
<td>133</td>
<td>66.6</td>
<td>0.51</td>
<td>0.10</td>
</tr>
<tr>
<td>Red roofing tile type I</td>
<td>8</td>
<td>20±1</td>
<td>29±3</td>
<td>404±13</td>
<td>92.9</td>
<td>45.3</td>
<td>0.35</td>
<td>0.098</td>
</tr>
<tr>
<td>Red roofing tile type II</td>
<td>9</td>
<td>12±1</td>
<td>18±1</td>
<td>350±12</td>
<td>65.2</td>
<td>32.4</td>
<td>0.25</td>
<td>0.061</td>
</tr>
</tbody>
</table>

4.2 Radon exhalation rate and effective radium content

Table 4 summarizes the track density, the radon exhalation rate and the effective radium content in materials investigated. The maximum of radon exhalation rate was observed in brown tile and the minimum in black brick. For brick samples the radon exhalation rate ranged from \((5.77±0.06)\times 10^{-5}\) in black brick to \((7.61±0.07)\times 10^{-5}\) Bq cm\(^{-2}\) h\(^{-1}\) in red brick with an average of \((6.81±0.06)\times 10^{-5}\) Bq cm\(^{-2}\) h\(^{-1}\), while the effective radium content ranged from \((2.98±0.12)\times 10^{-3}\) to \((3.93±0.16)\times 10^{-3}\) Bq kg\(^{-1}\) with an average of \((3.52±0.12)\times 10^{-3}\) Bq kg\(^{-1}\). In roofing tiles the radon exhalation rate was found to vary from \((5.79±0.05)\times 10^{-5}\) in red tile II to \((11.60±0.12)\times 10^{-5}\) Bq cm\(^{-2}\) h\(^{-1}\) in brown tile with an average of \((8.28±0.06)\times 10^{-5}\), whereas the radium content ranged from \((2.99±0.17)\times 10^{-3}\) to \((5.98±0.07)\times 10^{-3}\) Bq kg\(^{-1}\) with
an average of $(4.28 \pm 0.09) \times 10^{-3}$. In concrete sample the radon exhalation and the radium content was $(6.95 \pm 0.03) \times 10^{-5}$ Bq cm$^{-2}$ h$^{-1}$ and $(3.59 \pm 0.21) \times 10^{-3}$ Bq kg$^{-1}$, respectively.

The effective radium content in brick samples are close to the mean value (between 3%-15%). We observed almost the same behavior in tile samples excepted brown tile with the highest uranium activity. This obviousness could be due to the chemical composition which is almost similar for the same type of building material. They can be therefore differentiated just by the mixture used for their color. One can finally notice that for all samples investigated, radon exhalation rate was found to be lower than the world average value of $1.25 \times 10^{-4}$ Bq cm$^{-2}$ h$^{-1}$ (30 Bq m$^{-2}$ d$^{-1}$) set in the UNSCEAR report (1993).

Table 4: The measured track density $\rho$ (tracks cm$^{-2}$), the calculated effective radium content $\eta C_{Ra}$ (Bq kg$^{-1}$) and radon exhalation rate $J_o$ (Bq cm$^{-2}$ h$^{-1}$) in materials investigated

<table>
<thead>
<tr>
<th>Code</th>
<th>$\rho$</th>
<th>$\eta C_{Ra} \times 10^{-3}$</th>
<th>$J_o \times 10^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1960±13</td>
<td>2.98±0.08</td>
<td>5.77±0.04</td>
</tr>
<tr>
<td>2</td>
<td>2789±32</td>
<td>4.24±0.02</td>
<td>8.81±0.05</td>
</tr>
<tr>
<td>3</td>
<td>2391±35</td>
<td>3.63±0.12</td>
<td>7.05±0.06</td>
</tr>
<tr>
<td>4</td>
<td>3934±71</td>
<td>5.98±0.07</td>
<td>11.6±0.12</td>
</tr>
<tr>
<td>5</td>
<td>2359±12</td>
<td>3.59±0.21</td>
<td>6.95±0.03</td>
</tr>
<tr>
<td>6</td>
<td>2919±12</td>
<td>4.44±0.11</td>
<td>8.60±0.06</td>
</tr>
<tr>
<td>7</td>
<td>2582±32</td>
<td>3.93±0.16</td>
<td>7.61±0.07</td>
</tr>
<tr>
<td>8</td>
<td>2453±10</td>
<td>3.73±0.09</td>
<td>7.23±0.04</td>
</tr>
<tr>
<td>9</td>
<td>1966±38</td>
<td>2.99±0.17</td>
<td>5.79±0.05</td>
</tr>
</tbody>
</table>

The risk of radon in a given environment was evaluated by checking the relationship between concentrations of uranium and exhalation of radon in samples under investigations. A positive correlation was found as depicted on Figure 3.

5 Conclusion

The natural radioactivity and the radon exhalation rate in building materials of local origin were determined using a combination of gamma-ray spectroscopy and polymeric nuclear track detector (NTD). A mathematical model was explored and applied for radon exhalation rate. The study of radon release from building materials proved to be necessary to better understand the individual contribution of each material to the total indoor radon exposure. The results of this study show that the materials investigated can be safely used in buildings and decorations according to the international recommendations (UNSCEAR, 1993 and 1988). A linear relationship between uranium and radon concentration was found so that the rate of radon exhalation in building materials can be easily estimated in a given atmosphere from the knowledge of the
Figure 3: Radon exhalation rate versus uranium concentration in building materials investigated uranium concentration.

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