# Environmental Risk Assessment of Radon from Ceramic Tiles

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Abstract

Radon-222 exhalation from different ceramics tiles depends upon the radium ($^{226}Ra$) concentration and porosity. Raw zirconium sand is one of the substances widely used in the ceramic industry and it is naturally radioactive. This can produce unjustified concern and subsequently perturb the market of these products. The radon exhalation rates for all ceramic tiles companies in the vicinity of ceramic surface were in the range 28-44 mBq.m$^{-2}$.h$^{-1}$ or 2.0 to 4.8 mBq.kg$^{-1}$ .h$^{-1}$. The porosity of ceramic tiles is found in the range 0.19-0.28. The assessment of radium activity of ceramic tiles was found in the range 16-38 Bq.kg$^{-1}$ for glaze surface and 23-64 Bq.kg$^{-1}$ for clay surface, respectively. The average equivalent dose in contact to the ceramic surface was found 22 mSv.y$^{-1}$. The exposure in working level in the vicinity of ceramic tile was found in the range 2.4-3.8 WL, respectively. This give risk indication to people whom spend long time in closed ceramic tiles stores to avoid staying for long time in such places.

Key words: Radon-222, tracks density, exhalation rate, ceramic tiles, porosity.

## 1. Introduction

Radon is a colorless, odorless, naturally occurring gas, continuously generated from $^{226}Ra$ by the radioactive decay of $^{238}U$ together with ionizing radiation. Radon emanates from the earth and on a smaller scale from certain construction materials (EC, 1999; Tufail et al., 2007; Cortina et al., 2008; Maged, 2009). It is present everywhere and in the open air it rapidly disperses and rarely reaches hazardous concentrations. Daily and seasonal cycles are common features of indoor radon time series, which would cause higher radon levels to be observed in general at night and in the winter (Miles, 2001; Karpinska et al., 2004; Crockett et al., 2006; Denman et al., 2007). The gas is much more harmful when present indoors, where it tends to accumulate, reaching levels likely to represent a risk to health. Most radon inhaled with indoor air is exhaled and remains in the lungs for only a short time. The first radon daughter Po-218 is very reactive, heavy metal and electrostatically attracted to tiny particulates in air. These particulates are inhaled and deposited (as solid state materials) in the lung. Radon's daughter then decays sequentially, releasing damaging alpha and beta particles. Therefore, it is radon's progeny, not radon, that actually cause damage to the bronchial epithelium, because only the progeny remain in the lungs long enough to decay significantly (Cothern, 1987). Ceramics can be particularly radioactive if some compound of uranium (e.g., uranium oxide, sodium urinate) has been used to impart color (e.g., orange-red, green, yellow, black) to the glaze. The glaze can serve two functions: it provides color, and it seals the ceramic. There remain cases of elevated radioactivity in glaze and sometimes the higher readings are due to uranium in the glaze; sometimes they are due to the radionuclides in the clay that was used to produce the ceramic. It is true that the clay used to produce the body of the ceramic (rather than the glaze) can be a source of radon, but this is true for all ceramics, with or without a uranium glaze. Raw zirconium sand is one of the substances (naturally occurring radioactive material, NORM) which is widely used in the ceramic industry (Verita et al., 2009; ICRP, 2007; Luisa et. al., 2008). This sand contains varying concentrations of natural radionuclides: mostly $^{238}U$. Particular attention from a radiological point of view is given to ceramic tiles, which are eventually considered responsible for undue radioactivity (EC, 1996). Consequently, this study was undertaken with the purpose of determining radon concentration in the vicinity of ceramic tiles, and to assess the annual effective dose rate. The radon exhalation rate for different ceramics tiles companies used in Egyptian market and assessment of radium activity were measured.

## 2. Materials and Methods

A total of 50 samples of 13 different ceramic tiles manufacturers were collected from the Egyptian market for the measurements of radon exhalation rate. The ceramic tiles (floor and wall tiles) were obtained from suppliers or buildings under construction. The period of the survey was about 15 days for ceramic tile. The radon exhalation rate of ceramic tiles for both the clay and glaze surface was measured by tracks technique

The radon concentration per unit volume was calculated using the relation given (Maged et al., 1993),

$$\mathbf{C=K^{-1}\rho} \qquad \textbf{(1)}$$

where ρ is the number of tracks per $cm^2$, K is the calibration factor, 1.37 track $cm^{-2}$ $kBqh^{-1}m^3$ inside the hollow holder. The setup to measure the radon exhalation rate of ceramic tiles using CR-39 track detectors and the hollow holder is shown in Fig. 1. The CR-39 plastic detector were removed from the hollow holder and etched chemically in a 6 M NaOH solution at 70 °C for 18 h to display and enlarge the latent alpha tracks due to radon decay. The etched tracks on the detectors were counted, using an optical microscope with objective lens 4× magnification which attached to image analyzer. The average area of one hundred fields of view was calculated by using image analyses system and the track density was calculated in terms of tracks per $cm^{-2}$. The background track density was determined by processing a virgin detector under the same etching conditions. The background was subtracted from the measured track density. In order to obtain realistic statistics of the tracks, 100 fields of view were scanned continually to cover the detector surface.

## 3. Results and discussion

### 3.1. Radon exhalation rate

Expressions for the concentration of radon from building materials were derived several authors (Krisiuk, 1980; Quindos, et al., 1987). The radon concentration inside the hollow holder in equilibrium is:

$$\mathbf{C} = \frac{\mathbf{E}.S}{\mathbf{V}.\ (\lambda+\lambda_v)} + \frac{C_0\,.\,\lambda_v}{\lambda+\lambda_v} \qquad \textbf{(2)}$$

where E is the exhalation rate, S is the surface area of the hollow holder, V is the volume of the hollow holder, λ is the radon decay constant (7.6 x $10^{-3}$ $h^{-1}$), $C_o$ is the outdoor radon concentration, $\lambda_v$ is the ventilation rate. For calculating the exhalation rate of ceramic tile when it is considered that $\lambda_v \cong 0$, so the second term in equation (2) equal zero. The amount of radon transferred from the ceramic into a structure is affected by many factors, including radium content, porosity, construction type, and meteorological conditions. The radon exhalation rates for different ceramic tiles manufacturers ranged from 2.0 to 4.8 $mBqkg^{-1}.h^{-1}$. A comparison study for geometric mean for both of the mass exhalation rate (3.4 $mBq.kg^{-1}.h^{-1}$) and the area exhalation rate (38 $mBq.m^{-2}.h^{-1}$) for the average of all ceramic tiles with the other authors (Rashmi, et al., 2009) was found less than soil sample (38 $mBq.kg^{-1}.h^{-1}$ and 185 $mBq.m^{-2}.h^{-1}$, respectively).

### 3.2. Assessment of effective radium concentration

The radon emanation power or emanation coefficient, denoted by ε, is defined as the fraction of $^{222}Rn$ produced by the disintegration of $^{226}Ra$ in the grains of the material that can escape from it. The emanation power is dimensionless and ranges from 0 (no radon escapes from the material) to 1 (all radon escapes). The rate of radon exhalation is proportional to the gradient of the radon concentration in the internal pores (Culot et al., 1976). The principle factors affecting the radon exhalation rate from a building material per unit activity concentration of Ra-226 are the porosity and the density of the material, the diffusion coefficient, the water, the age and the composition of the material as seen in equation (3).

$$E = \boldsymbol{\varepsilon}\,.\,\mathbf{C_{Ra}}\,.\,\boldsymbol{\rho}\,.\sqrt{\frac{\lambda \mathbf{D}}{\mathbf{P}}}\,.\mathbf{tanh}[\sqrt{\frac{\lambda P}{D}}\,.\,\boldsymbol{l}] \qquad \textbf{(3)}$$

Where $\boldsymbol{\varepsilon}$ is the emanation power, $C_{Ra}$ is the effective radium concentration, $\boldsymbol{\rho}$ is the density, D is the effective diffusion coefficient, $\lambda$ is the decay constant of $^{222}$Rn, P is the porosity of the material, and $\boldsymbol{l}$ is the half thickness of the material.

The activity concentrations of NORM in ceramic tiles vary according to the type and origin of the ceramic tiles. The activity concentration ($Bq.kg^{-1}$) in the most common building materials in Europe, e.g. concrete and sand-lime bricks is 40 and 10 for $^{226}$Ra, respectively (EC, 1999). The radium-226 activity values of NORM for the present study and the other authors are shown in Table 1. The $^{226}$Ra activity for all ceramic tiles either the floor or wall tile was in the range 16-64 $Bq.kg^{-1}$ as shown in Table 2. It is clear that the average of radium-226 activity in ceramic tiles in Egypt was less than the others authors (Verita et al., 2009; EC, 1999) as shown Table 3. The average radium concentrations (79 $Bq.kg^{-1}$) in clays which used in ceramic industry in Serbia (Todorovic et al., 1999) was found more the present work (41±12 $Bq.kg^{-1}$). The dependence of the radon-222 exhalation rate on the porosity of ceramic tiles shows no significant difference. The use of industrial by-products and residues containing elevated concentrations of radioactive material in building materials is increasing due to economic and environmental reasons. The national regulatory authorities should ensure that “annual doses are restricted to a few mSv for the worst-case scenarios (ICRP, 2007). The dependence of the $^{222}$Rn exhalation rate on the radium concentration of ceramic tiles either floor or wall for all manufacturers (glaze and clay) show a little increase as in Figs. 2-5.

3.3 Effective dose equivalent due $^{222}$Rn

The effective dose equivalent due to $^{222}$Rn and its progenies was estimated using the following equation (UNSCEAR, 2000),

$$\mathbf{H(mSv.y^{-1}) = ((0.17 + 9F)C_{Rn}) \times 8760(h) \times 0.8 \times 10^{-6}} \qquad \mathbf{(4)}$$

Where, F is the equilibrium factor between radon and its progeny, $C_{Rn}$ is the concentration of $^{222}$Rn and 0.8 is the indoor occupancy factor.

The equivalent dose due to radon concentration inside the hollow holder which is in contact to the ceramic tiles surface is found 22±2 $mSv.y^{-1}$. It is found higher twice more than the recommended action level (10 $mSv.y^{-1}$). The track density, radon concentration, equilibrium equivalent concentration and exposure were found in the range 288-444 $No.cm^{-2}$, 486-751 $Bq.m^{-3}$, 243-375 $Bq.m^{-3}$ and 2.4-3.8W.L, respectively as in Table (3.4). The exposure in working level in the vicinity of ceramic tile was found in the range 2.4-3.8 WL, respectively.

Conclusion

The recycling of industrial waste or by-products is extensively used in the construction industry. The use of fly ash, coal slag, phosphogypsum, and red mud in ceramic and the increment in radiation exposure from these materials it has been of concern for several years. The radon exhalation rates for all ceramic tiles companies in the vicinity of ceramic surface were in the range 28-44 $mBq.m^{-2}.h^{-1}$ and 2.0 to 4.8 $mBq.kg^{-1}.h^{-1}$. Mean value of radium concentrations of ceramic tiles were in the range 16-38 $Bq.kg^{-1}$ for glaze and 23-64 $Bq.kg^{-1}$ for clay, respectively. The equivalent dose in contact to the ceramic tiles surface is found 22±2 $mSv.y^{-1}$. The equivalent dose due to radon concentration in the vicinity of ceramic tile surface is found higher twice more than the recommended action level (10 $mSv.y^{-1}$). These data must be regarded, as preliminary and further more extensive studies should be done on large scale for all building materials.

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**Table 1. Average values and ranges for activity concentrations of naturally occurring $^{226}Ra$, (EC, 1999).**

| Building material | $^{226}Ra$ ($Bqkg^{-1}$) |
|---|---|
| Ceramic | 25-193 |
| Granite | ND[a)]-160 |
| Tiles | 33-61 |
| Marble | 1-63 |
| Ceramic [b)] | 16-64 |

a) ND= No data

b) Present work

**Table 2. Average $^{226}Ra$ concentration ($Bq.kg^{-1}$) from all ceramic tiles of different manufacturers.**

| Code No. | Average radium concentration ($Bq.kg^{-1}$) Floor tile | | Average radium concentration ($Bq.kg^{-1}$) Wall tile | |
|---|---|---|---|---|
| | Glaze | Clay | Glaze | Clay |
| 1 | 33 | 54 | 17 | 24 |
| 2 | 36 | 52 | 23 | 31 |
| 3 | 30 | 59 | 22 | 34 |
| 4 | 24 | 48 | 21 | 32 |
| 5 | 23 | 50 | 16 | 33 |
| 6 | 34 | 56 | 27 | 37 |
| 7 | 31 | 64 | 21 | 34 |
| 8 | 30 | 54 | 26 | 39 |
| 9 | 38 | 56 | 24 | 34 |
| 10 | 26 | 48 | 16 | 29 |
| 11 | -- | -- | 22 | 30 |
| 12 | -- | -- | 17 | 23 |
| 13 | -- | -- | 18 | 27 |
| Mean | 31 | 54 | 21 | 31 |
| Max | 38 | 64 | 27 | 39 |
| Min | 23 | 48 | 16 | 23 |
| S.D | 5 | 5 | 4 | 5 |

**Table 3. Activity concentrations of $^{226}$Ra in zircon materials and ceramic tiles (Verita et al., 2009).**

| Material | Samples | Average $^{226}$Ra radioactivity (Bq.kg$^{-1}$) |
|---|---|---|
| **Zircon materials** | **27** | **2640±650** |
| **Ceramic tiles** | **12** | **90±60** |
| **Ceramic tiles[a)]** | **50** | **33±13** |

**a) Present work**

**Table 4**. **The average value of ceramic tiles for all companies.**

| Code | Track density No.$cm^{-2}$ | Radon concentration $Bq.m^{-3}$ | Exhalation rate $mBq.m^{-2}.h^{-1}$ | Equivalent dose $mSv.y^{-1}$ | E.E.C[a)] $Bq.m^{-3}$ | Exposure W.L |
|---|---|---|---|---|---|---|
| 1 | 377 | 637 | 37 | 21 | 319 | 3.2 |
| 2 | 431 | 728 | 43 | 24 | 364 | 3.6 |
| 3 | 430 | 727 | 43 | 24 | 363 | 3.6 |
| 4 | 387 | 654 | 37 | 21 | 327 | 3.3 |
| 5 | 381 | 644 | 43 | 21 | 321 | 3.2 |
| 6 | 430 | 728 | 42 | 24 | 364 | 3.6 |
| 7 | 416 | 703 | 41 | 23 | 351 | 3.5 |
| 8 | 434 | 733 | 43 | 24 | 367 | 3.7 |
| 9 | 444 | 751 | 64 | 25 | 375 | 3.8 |
| 10 | 394 | 673 | 38 | 22 | 337 | 3.4 |
| 11 | 331 | 559 | 33 | 18 | 279 | 2.8 |
| 12 | 288 | 486 | 22 | 16 | 243 | 2.4 |
| 13 | 337 | 569 | 33 | 18 | 285 | 2.8 |
| Mean | 390 | 661 | 41 | 22 | 331 | 3.3 |
| Max | 444 | 751 | 64 | 25 | 375 | 3.8 |
| Min | 288 | 486 | 22 | 16 | 243 | 2.4 |
| SD | 46 | 68 | 5 | 2 | 55 | 0.2 |

[a)]**E.E.C= Equilibrium Equivalent concentration**

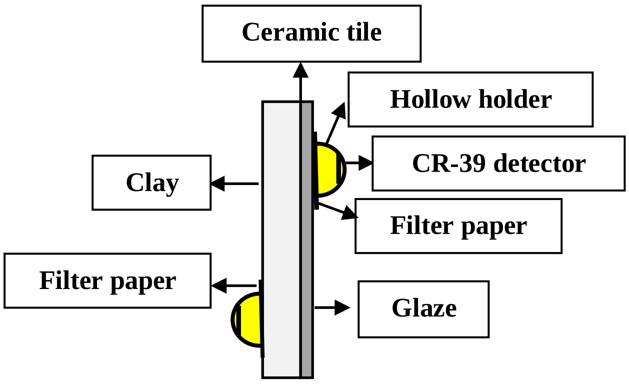


**Fig. 1. The setup diagram for measuring radon exhalation rate of ceramic tile.**

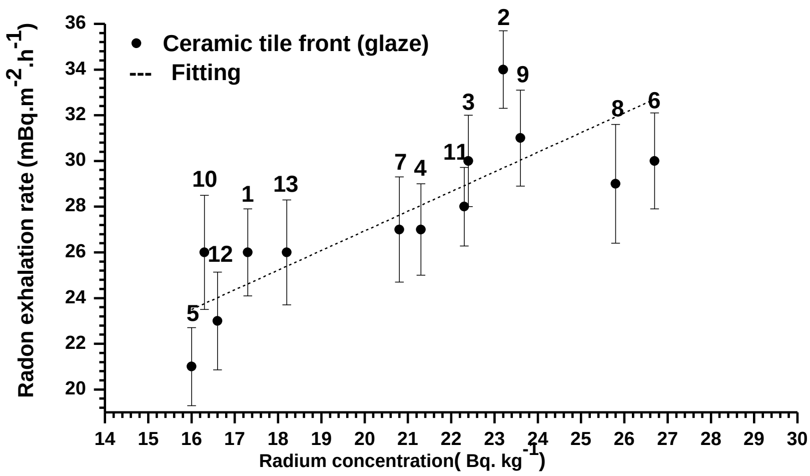


**Fig.2.The dependence of the radon exhalation rate from the glazed side on the $^{226}$Ra concentrations of wall tiles. Numbers with each point indicate the source manufacturers.**

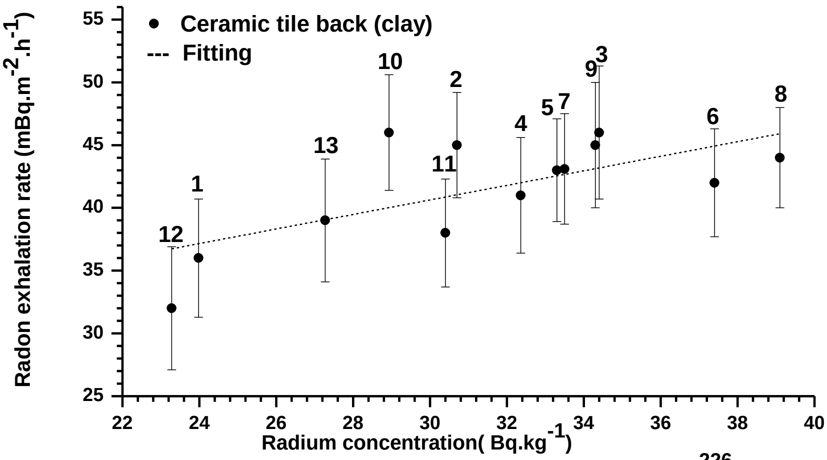


**Fig.3.The dependence of the radon exhalation rate from the clay side on the $^{226}$Ra concentrations of wall tiles.Numbers with each point indicate the source manufacturers.**

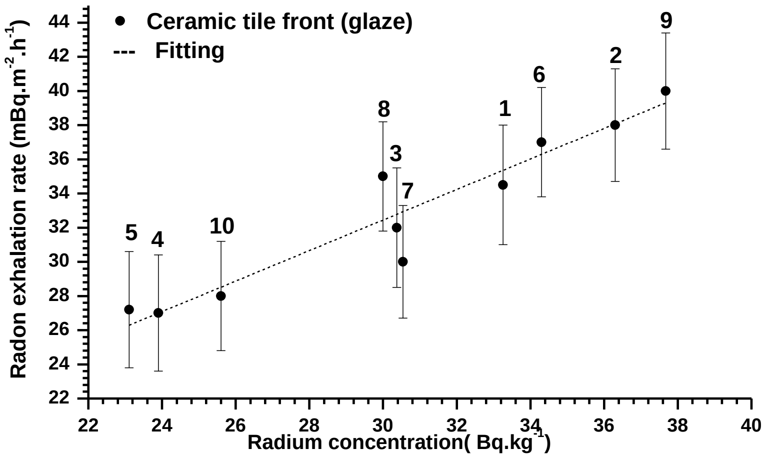


**Fig.4.The dependence of the radon exhalation rate from the glazed side on the $^{226}$Ra concentrations of floor tiles.Numbers with each point indicate the source manufacturers.**

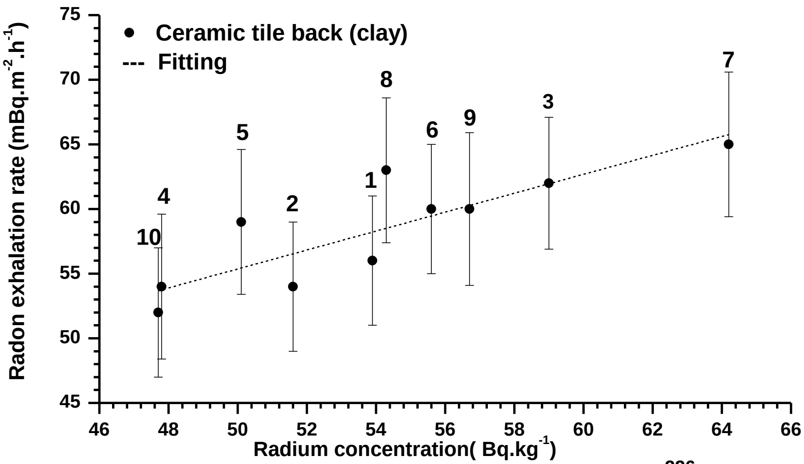


**Fig.5.The dependence of the radon exhalation rate from the clay side on the $^{226}$Ra concentrations of floor tiles.Numbers with each point indicate the source manufacturers.**