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Environmental safety of natural and manufactured building materials

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Abstract

Natural radioactivity was estimated in building materials using γ-spectroscopic method. Samples of granite, bricks, concrete and ceramic were collected from different places in Egypt. Samples were prepared for physical and mechanical properties measurements as well as the radioactive content. Gamma spectrometer composed of NaI crystal connected to ORTEC analyser was used for radioactive measurements. Standard sample was prepared with the same geometry factor in NIS using a standard source traceable to NIST. Data of 238U, 232Th and 40K activities were collected, where the effective dose was calculated by the aid of UNSCEAR. Diffusion equation was used to estimate Radon emissions rate from building materials used in proposed model rooms.

It was found that the average concentrations of 238U, 232Th and 40K in the studied materials were for granite 63.4, 2.42, 1010.91 Bq/kg, for bricks 20.12, 3.75, 27.25 Bq/kg and for concrete 34.23, 2.36, 506.36Bq/kg. In spite of using materials with permissible activity concentration, the radon emission in model rooms was beyond the safe limits for inhabitants. The maximum dose from Rn concentration was 1.23 mSv/y. This concentration was affected by the space dimension, passing elapsed time and building material radioactivity as well as ventilation. It was also found that the most powerful factor affecting radon concentration is the ventilation.

Keywords: environment, safety, building materials, radioactivity.

1. Introduction

Construction materials and interior finish products should be chosen with zero or low emissions to improve indoor air quality. Many building materials and cleaning/maintenance products emit toxic gases, such as formaldehyde and Radon. These gases can have a detrimental

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impact on occupant's health and productivity. Another source of Radon is the radiation in the environment, which is either natural or artificial. All exposure from natural background radiations, except for direct cosmic radiation, is produced by radiation coming from natural radionuclide in the environment. Thorium and Uranium in their natural occurrence undergo radioactive decay in three different series, headed by 238U, 235U (uranium) and 232Th (thorium). Without chemical or physical separation, each of the three series attains a state of secular radioactive equilibrium [1]. Each one of these series produces Radon, which is a chemically inert radioactive element at normal temperatures. This gas does not chemically interact with other elements. It is difficult to be trapped. It is an extremely toxic gas [2]. Airborne radon in the mines may probably be the most essential factor in the production of lung cancer. Prolonged breathing of an atmosphere containing radon may be responsible for the increase in the incidence of lung cancer [2].

Many researches were conducted in Egypt to estimate the radioactivity of building materials. El Afifi et al [3] investigated the activity concentration of granite, ceramic, cement and phosphor-gypsum. Using these building materials need some careful regulations to reduce the hazardous radiation released into surroundings. The radiation hazards can be reduced using natural wood or minimizing the areas of granite or ceramic. Sharaf et al. [4] investigated radioactivity of bricks, tiles, ceramic and marble. All investigated samples were safe to be used except for blast furnace slag cement. The activity of clay bricks is higher than that of cement bricks. Concentrations of natural radionuclide in Egyptian bricks were compared with those of imported bricks. The activity of Egyptian bricks was found to be significantly lower in all cases [5]. Clay bricks have the highest brick radioactivity. On the contrary, local samples of marble and granite contain higher levels of radioactivity than those of imported samples [6, 7].

Many investigations on building materials were conducted in different countries. All building materials showed Radium-equivalent activities lower than the limit set in OECD report (370 Bq/kg/y), equivalent to external γ-dose ranging from 1mSv/y to 2mSv/y [8, 9, 10]. Another research calculated the annual effective dose of γ that was found to be in safe limits [11]. A few number of the investigated building materials proved to be unsafe such as Portland cement (Cem II) with high addition of fly ash [12]. Samples of Macedonian gypsum contribute to an annual effective dose of more than 1mSv so these samples were not safe [13].

The results indicated that magmatic rocks are generally characterized by higher natural radioactivity than those of other natural building materials [14]. The results showed that the radioactivity level is low in marble but high in some granite samples.

Radon concentrations in dwellings up to 100kBq/m3 were found in some regions. This is a relatively high concentration. The soil, outdoor environment, shows high uranium content. Additionally, a fast radon transport in the soil is possible. To reduce the radon exposure of the inhabitants in these radon prone areas, it is necessary to look for building and insulating materials with low radon permeability [15, 16]. The presence of cracks and holes in concrete may aggravate the high radon indoors concentrations [17]. It should be emphasized that the dosage of radiation received by individuals depends on many factors such as rate of ventilations, pattern of air flow and time [18].

The aim of this work is to determine the ratio of radioactive content in the building materials to be taken in consideration during the selection of such materials in different construction applications.

2. Experimental Program

Common used building materials were selected such as bricks, concrete, granite and ceramic. A person living in a room is surrounded by two floors and four walls. The two floors are made of concrete. The four walls are built using bricks. The ordinary domestic room covered by ceramic. Granite is commonly used in Egyptian kitchens, entries, basements, etc.

The most used types of granite in the building market were selected. Most of the granite samples are locally produced. The samples used and their sources are shown in Table 1. One sample of ceramic was investigated. It is produced by Prima factory called Geranitio. The main component of Geranitio is granite. Its dimensions were 50*50*0.5 cm. Four types of bricks were used. Cement and clay bricks are the most popular types used in Egyptian market. Sand bricks and tiles 2.5cm thick with trade name Sornaga are out covering items used widely in Delta and North of Egypt. Five samples of clay bricks were collected from Nile-Delta. Table 2 shows the source of the bricks. Three samples of cement bricks were collected. One sample of Sornaga and another of sand bricks were investigated. Since the coarse aggregate of silica gravel becomes very rare in Egypt and expensive, crushed stones is a proper alternative in the construction field. Referring to the previous studies, granite shows a high radioactivity. The concrete aggregate used was crushed granite to get the worst case of radioactive concrete .Three different mixes of plain concrete were cast. Table 3 shows the design of these concrete mixes.

A full scale investigation was conducted on the different samples of granite and bricks used in this investigation according to the Egyptian Code of Practice of masonry works (ECP 204-2005). The concrete samples were tested according to Egyptian Code of Practice of Reinforced concrete (ECP 203-2007). Tables 1, 2 and 3 illustrate the mechanical and physical properties of materials used in this investigation. The properties of samples were within the specifications limits.

TABLE1: PHYSICAL, MECHANICAL PROPERTIES AND RADIOACTIVE CONCENTRATION OF GRANITE SAMPLES USED IN THIS INVESTIGATION

No.	Type	Source	% Absorption	Density gm/cm^3	Comp. Strength (kg/cm^2)	Fracture modulus (kg/cm^2)	Activity concentration (Bq/kg)		
							A_U	A_{Th}	A_K
G 1	Rosa Elnaser	Aswan	0.4%	3.17	1072	30	70.727	8.91	1101.07
G2	Gandola	Aswan	0.2%	3.53	396	49	84.997	3.26	1025.58
G3	Verdy	Aswan	0.3%	3.3	316	27	38.120	1.40	1135.54
G4	Red Aswan	Aswan	0.1%	3.37	464	30	74.847	0.91	1153.27
G5	Rozeta	Aswan	0.2%	3.32	304	54	46.543	3.92	995.55
G6	Hody	Hurgada	0.3%	3.56	540	27	120.760	1.07	1189.28
G7	Ghardka	Hurgada	0.3%	3.54	1008	42	78.470	3.63	915.18
G8	Gray	St. katreen	1%	3.4	1648	39	36.473	0.73	553.869
G9	Royal	Red sea	0.1%	3.57	764	35	61.380	1.07	1143.09
G10	Halayab	Halayab	0.5%	3.17	364	56	10.470	0.64	590.88

TABLE 2: PHYSICAL, MECHANICAL PROPERTIES AND RADIOACTIVE CONCENTRATION OF THE BRICKS SAMPLES USED IN THIS INVESTIGATION

No.	Type	Dimension (mm)	Density gm/cm^3	% Absorption	% voids	Strength kg/cm^2	Activity concentration (Bq/kg)		
							A_U	A_{Th}	A_K
B 1	Clay	59*102*225	1.65	9.2	32.1	95	33.35	3.24	13.86
B 2	Clay	62*108*224	1.5	9.2	28	111	35.04	0.89	2.07
B 3	Clay	61*103*220	1.49	8.8	32.23	106	31.27	10.65	34.13
B 4	Clay	60*106*228	1.6	9.5	33.7	124	35.5	1.96	9.28
B 5	Clay	60*103*227	1.68	8.9	32.4	84	27.64	0.89	2.07
B 6	Cement	55*114*237	2.07	4.4	-	79	3.84	5.13	89.17
B 7	Cement	70*100*190	2.13	4.7	-	71	4.07	4.91	86.90
B 8	Cement	54*117*237	2	4.7	-	78	8.41	1.63	4.53
B 9	Sandy	250*120*60	1.8	12	-	135	6.91	2.28	16.17

B 10	Sornaga	24*40*200	-	-	-	-	15.25	5.87	14.34

TABLE 3: CONCRETE MIX DESIGN, COMPRESSIVE STRENGTH AND RADIOACTIVE CONCENTRATION

No.	Constituent of 1 m^3				Strength (kg/cm^2)		Activity concentration (Bq/kg)		
	Weight of cement kg	Weight of water (kg)	Vol. coarse agg.	Vol. fine agg.	7days	28days	A_U	A_{Th}	A_K
C 1	250	125	0.7	0.4	256	334	31.04	1.72	451.65
C 2	250	125	0.8	0.4	326	422	33.31	2.07	531.48
C 3	250	125	0.9	0.4	270	407	38.34	3.28	535.91

2.1. Activity Measurements

The collected samples were grinded to reduce the particle size to get some form of homogeneity. Then, they were sieved to remove any undesirable particle size. The samples were weighed and sealed in a suitable standard container similar to the source to be used with the gamma ray detector. Each container was carefully sealed and stored for four weeks to achieve the secular equilibrium between 226Ra and its daughter 222Rn [1]. As for concrete, it was mixed and cast in the standard container.

Twenty containers were manufactured with the same dimension of the standard container to accommodate the large number of investigated samples. The containers were manufactured of metal sheets to be used in the gamma-ray spectrometer.

The measurements were conducted in the Radiation Physics Department in the Faculty of Science, Menoufiya University. The calibration was conducted using a sample measured in the National Institute of Standards (NIS).

The detection system is a low-level gamma-ray spectrometer including 3"x3"NaI(Tl) detector and the associated electronics, PCA, 1024MCA data acquisition card mounted in a PC. The detector is surrounded by cylindrical lead shield with a moving cover in order to suppress the soft component of cosmic rays and the background due to the surrounding building materials and air. The shield contains an inner concentric cylinder of copper. The Cu-liner is used to attenuate the X-rays stimulated in the lead shield itself. The counting geometry was selected in order to minimize the backscattering radiation. The spectrometer was adjusted and calibrated. The background was carefully measured and subtracted for each sample [1].

The calibrated γ-spectrometer was used to estimate U, Th and K activity. Efficiency-Energy Calibration Curve for NaI(Tl)3"x3"Spectrometer Detector is shown in Fig. 1. It shows that the efficiency decreases as the gamma energy increases. Fig. 2 shows a spectrum of one of the studied samples. The line at 230,350keV of 214Pb and line at 609keV of 214Bi were used to determine 238U activity, the line at 510keV of 208Tl for 232Th and 1460 keV for 40K concentration where the estimated activity values are represented in Tables 1, 2 and 3.

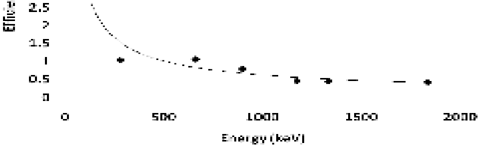

Figure 1: Efficiency-Energy Calibration Curve for NaI(Tl)3"x3"Spectrometer detector.

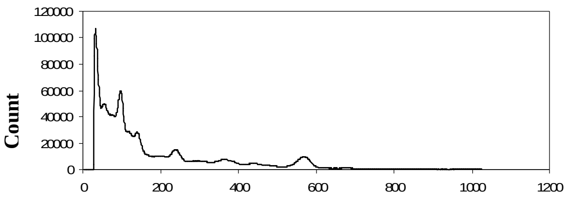

Figure 2: Spe ranite Sample (G1) Rosa El-Naser.

The absorbed dose and effective dose rate were determined from the following relations:

$$\boldsymbol{D = 0.472A_U + 0.662\,A_{Th} + 0.043\,A_K \quad (nGy/h)} \qquad (1)$$

Where A_U, A_{Th} and A_K are the concentration of uranium, thorium and potassium respectively in (Bq/kg) [19].

$$\boldsymbol{D_{eff} = D \times 24h \times 365.25d \times 0.8 \times 0.7 \times 10^{-3} \; \mu Sv/y} \qquad (2)$$

Where $\boldsymbol{D_{eff}}$ is the effective dose rate in µSv per year and D is the dose rate in nGy/h. Average absorbed dose, average effective dose D_{eff}, are listed in Table 4.

TABLE 4: AVERAGE ABSORBED DOSE AND AVERAGE EFFECTIVE DOSE

Material	Average values.		Standard deviation	
	Dose rate (nGy/h)	D_{eff} (mSv/y)	Dose rate	D_{eff}
Granite	73.241	0.374	21.69563	0.121033
Concrete	39.489	0.202	5.857169	0.016978
Brick	13.358	0.068	3.323683	0.029777

3. Determination of Rn emanation

An extensive theoretical study was performed for Rn emanation. This extensive study is an approach to calculate the indoor radon concentration. An ordinary domestic room built using the studied materials was virtualized. Measuring the radioactivity of building materials of the virtual room was used to determine radon emission.

The mass balance equation was used for indoor radon concentration. Basic form of the mass balance equation is as follows:

$$Accumulation = Input - Output - Reaction \qquad (3)$$

It is assumed that:

1. Radon gas is homogeneously mixed with the room air.
2. 2.Radon gas does not react with any substance or disappear by any process except for ventilation and natural decay.
3. Minor sources of radon such as water, natural gas and Liquid Petroleum Gas (LPG) in the house are negligible.

The time dependency of the gas concentration C_i(t) inside a single room with volume V can be given by the differential equation as follows [20]:

$$\frac{dC_i(t)}{dt} = J\frac{S}{V} + C_o l_v - C_i(l + l_v) \qquad (4)$$

where:
C_i(t) radon concentration (Bq/m^3) in room at time t,
J radon exhalation rate of concrete (Bq/m^2/s),
S exhalation surface area (m^2),
V volume of room (m^3),
C_o radon level (Bq/m^3) of outside air,
l_v ventilation rate (s^{-1}),
l decay constant of radon ($2.1\times10^{-6}\,s^{-1}$).

From Eq. (4) it can be seen that indoor radon levels depend on the source of radon such as emanation from building materials and outdoor radon levels, radon flux (area radon exhalation rate, J), ventilation rate (l_v) and surface to volume ratio of room besides other removal processes. The differential equation was solved by Matlab program. Fig. 3 shows the sequence of solving the problem.

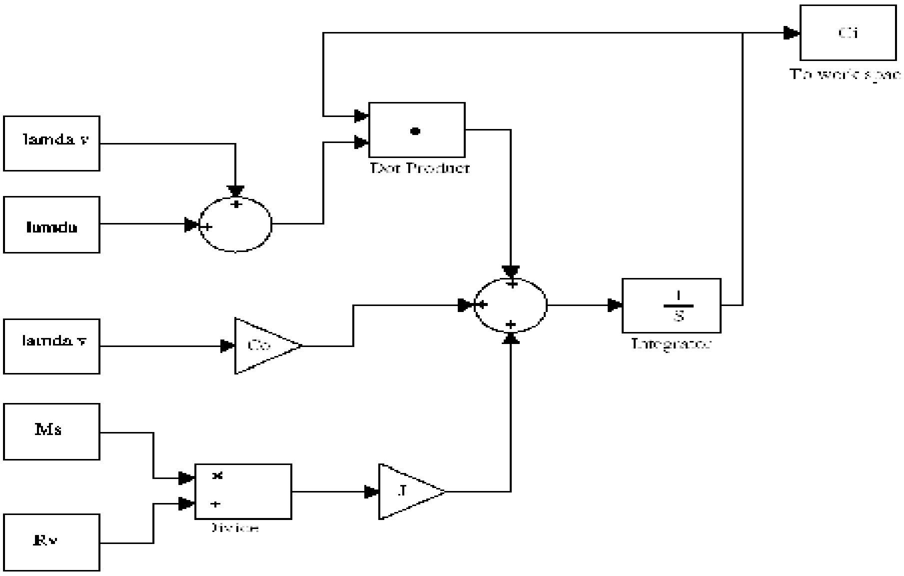

Figure 3: Matlab Program Flow Chart.

The effect of parameters in equation (4) is time, dimensions, mass exhalation rate and ventilation. The estimated values of ventilation rates were in the range of 0.5 h^{-1} [21]. Radon decay constant is $2.1\text{x}10^{-6}s^{-1}$ [20]. Radon emanation was studied versus different cases using the parameters of material variation, dimensions of rooms and ventilations. The highest values of mass exhalation rate of the materials investigated were chosen. Those values are illustrated in Table 5 to study Radon emanation in a virtual room for four cases shown in Table 6.

TABLE 5: MASS EXHALATION RATE OF THE MATERIAL USED

No.	Sample type	D_{eff} (mSv/y)	Mass exhalation rate (Bq/kg/s)
G6	Hody granite	0.555986	0.00147
C2	Concrete	0.221246	0.00130
B3	Clay brick	0.118901	0.00068
B6	Cement brick	0.046190	0.00017
G11	ceramic	0.472808	0.00130

TABLE 6: THE BUILDING MATERIALS OF EACH CASE

case	**floor**	**wall**	**Ceiling**
1	Concrete (C2)	Clay brick(B3)	Concrete(C2)
2	Ceramic (G11)	Ceramic(G11)	Concrete(C2)
3	Ceramic(G11)	Granite + Clay brick (G6+B3)	Concrete(C2)
4	Concrete(C2)	Cement brick (B6)	Concrete(C2)

4. Analysis and discussion of test results

4.1. Activity Measurements

The activity concentrations of natural radionuclides in the measured samples were calculated for ^{238}U, ^{232}Th and ^{40}K as shown in Tables 1, 2 and 3. The total air absorbed dose rates, D, in $nGy.h^{-1}$ due to the mean specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K

(Bq/kg) were calculated from equation (1). The total air absorbed dose rate of samples is illustrated in Fig. 4. Effective dose rate indoors in units of µSv per year is calculated by equation (2). Figure 5 illustrates the values of effective dose rate of samples.

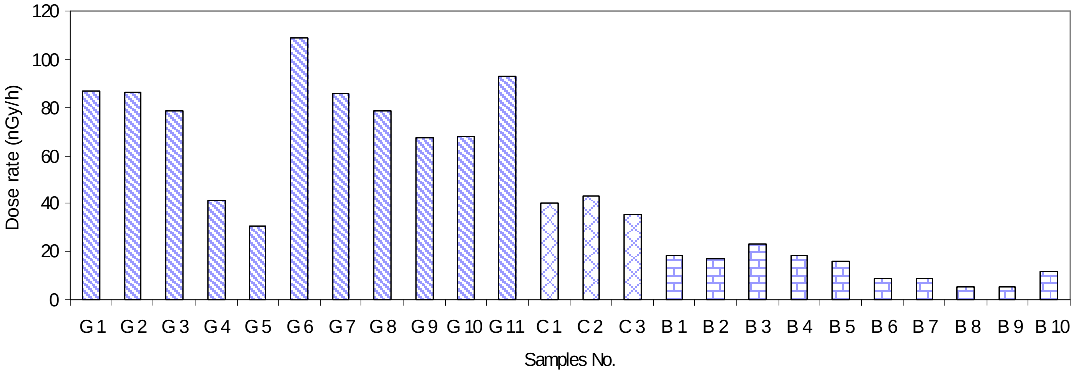

Figure 4: Total Air Absorbed Dose Rate of Investigated Samples.

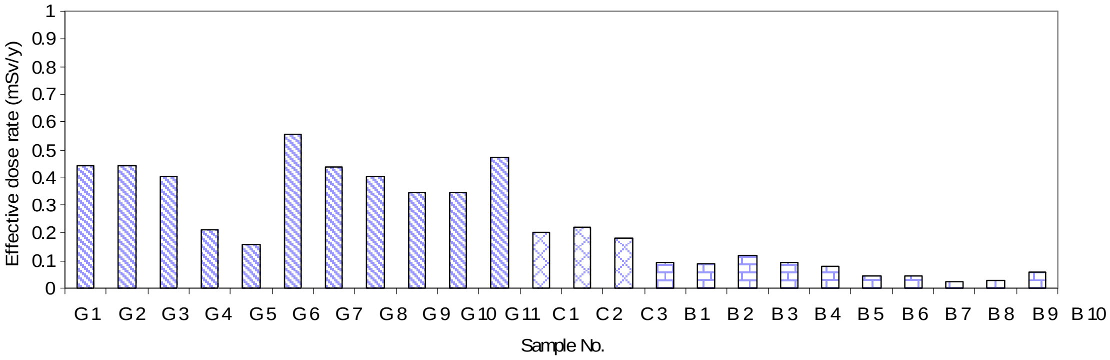

Figure 5: Effective Dose Rate of Investigated Samples.

From the data listed in Tables 1, 2 and 3, it appears that the dose rate and the effective dose rate are found to be from 30 to 180.8 nGy/h and 0.157 to 0.556 mSy/y for granite samples , from 35.21 to 3.3 nGy/h and 0.18 to 0.22 mSv/y for concrete samples, from 5.5 to 23.277 nGy/h and 0.028 to 0.119 mSv/y for bricks samples and 92.56 nGy/h and 0.47 mSv/y for ceramics sample.

Granite samples have the highest absorbed dose and effective dose. The igneous rocks tend to have high uranium, thorium and potassium. Concrete mixes containing granite aggregate have a relative high absorbed dose.

Table 1 illustrate that Hody granite has the highest activity of^{238}U. It has the highest dose and effective dose. Halayab granite has the lowest activity concentration of^{238}U, ^{232}Th, and ^{40}K.

4.2. Emanation from the Solution of the Differential Equation

The theoretical study in hand is an extension to the experimental work in this investigation. The idea of the theoretical study is based on using more than one building material with permissible activity in a virtual room would lead to impermissible dose of radon emission. The radioactive properties of the building material investigated were taken as reference to compute radon concentration of an ordinary domestic room built with the materials investigated in this research.

The effective parameters were as follows:

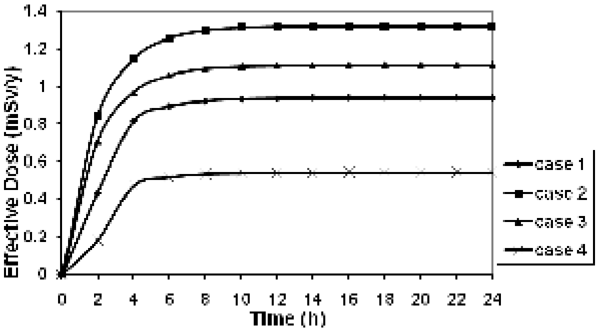

Figure 6: The relationship between Effective dose and time

4.2.1. Time

The parent radioactive materials such as ^{238}U, ^{232}Th produce radon with time passing. The concentration of radon with time is illustrated in figure (6). It shows that The Rn concentration increases with time until the full build up after 10 hrs. This behavior is the same as obtained from the buildup equation of any radioactive material that reflects the accuracy of the method used in this work. The Rn emission is high for cases 2 and 3. This high dose could be decreased by raisingλ_v .

4.2.2. Material

Using a material with high radioactivity leads to high radon concentration. This is obvious in case (2), where the materials used, C2 and G11 have the highest values of effective dose rates as shown in table (6). Case (3) shows better behavior, since the material effective doses is less than that of case (2), and so for other cases.

4.2.3. Dimensions

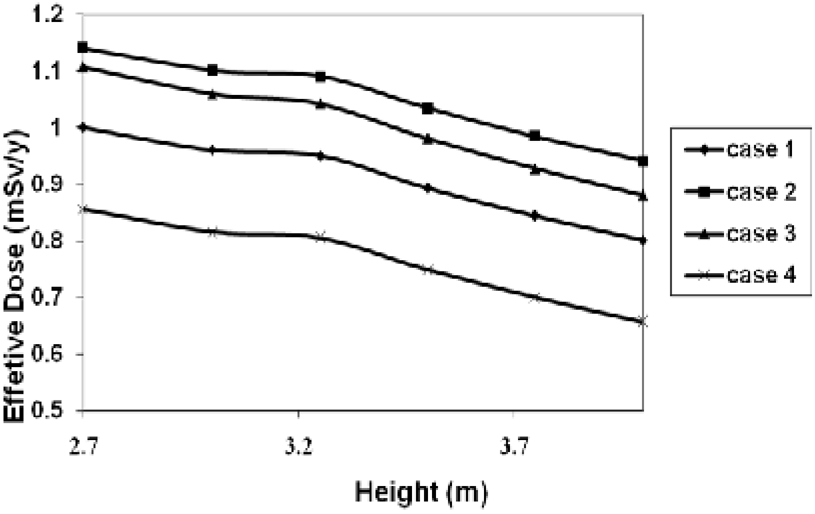

Figure 7: The Relation between Effective Dose and Height.

a. Height: A case was studied representing a basement with bad ventilationλ_v=0.05 h^{-1} and dimensions 10*20 m and height 2.7m. The maximum height is 4m. The relation between height and radon concentration is shown in figure (7). It results unsafe dose for cases 1, 2, 3 at height 2.7 m. Raising height makes the effective dose safe for case (2) at height 3.75m. It becomes safe for case (3) at height 3.5 m. In case (1) increasing height with less than 10% of total height changes the case into safe mode. The second case (the worst case) raising height with 38% changes the case into safe mode.

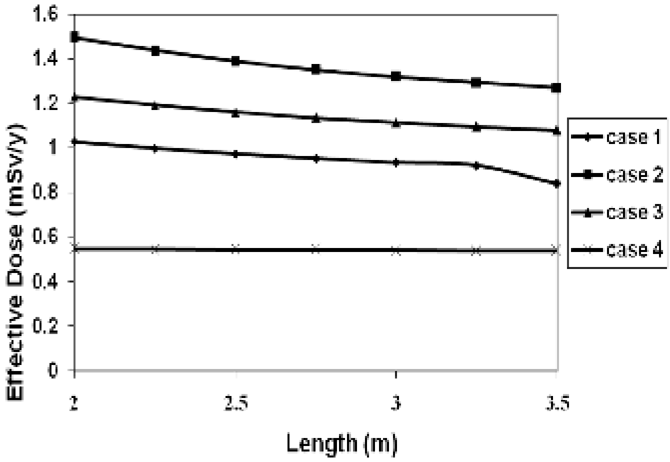

Figure 8: The Relation between Effective Dose and Length of the Room

b. Length: The dimensions of room were taken 2*3*2.7m. The length varied from 2m to 3.5m with an interval 0.25m and λ_v=0.5h^{-1}. The relation between room length and radon concentration is shown in figure (8). Case (2) was unsafe for the original dimension till 20.00m. The safe length in case (3) is 6.00 m. Changing the dimension of plan is very difficult either in existing building or planned to be constructed. Increasing the dimensions does not affect the dose seriously.

c. Volume: The dimensions of room were taken 2*3*2.7m. The plane dimensions varied from 2*3 to 3.5*3m. The relation between the volume and the radon concentration was drawn in figure (9). It has the same trend as figure (8). The plan dimensions are very difficult to be changed, where the construction area in Egypt is very limited. So, changing the height with very small values not exceeding 20% of total height makes a positive effect on the radon emission safety.

4.2.4. Ventilation

Since the ventilation is a superior factor of Radon emission in buildings, it was taken as a parameter. Bad ventilation is an existing case as in a garage basement. That was taken as the first case, which the $\lambda\upsilon$ the ventilation rate equals 0.05h^{-1}.Good ventilation was assumed 3h^{-1}. Intermediate cases in between were calculated.

Four cases of an ordinary domestic room are studied. The room dimensions were 3*3*2.7m. The relation between the ventilation and the radon concentration was drawn in figure (10) and it shows that in this case in natural ventilation the dose is more than 1mSv/y but when the ventilation rate increases it become safe for all cases. Increasing the ventilation is very effective on the results.

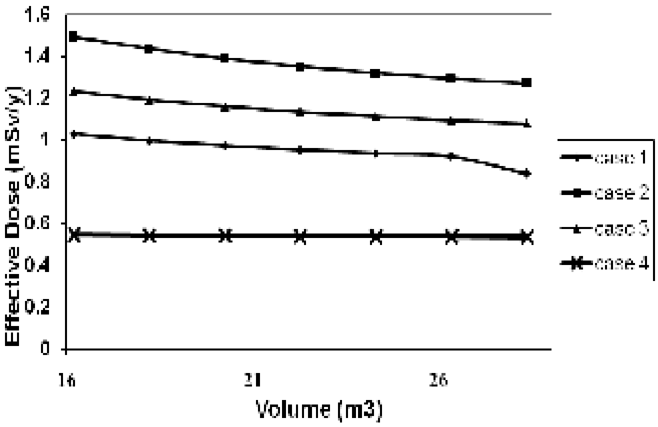

Figure 9: The Relation Between The Effective Dose (mSv/y) and The Volume (m^3)

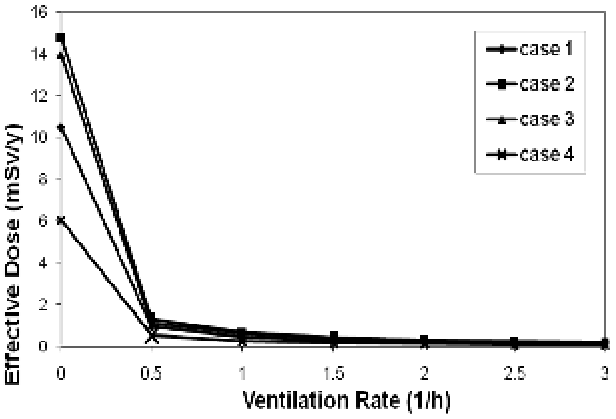

Figure 10: The Relation Between the Effective Dose (mSv/y) and the Ventilation

5. Conclusions

1. All the investigated materials have an activity within the permissible dose limit. Different samples of granite show wide range of radioactivity as well as the effective doses values. Hody granite from Hurghada gives the highest dose and effective dose (0.56 mSv/y). Halayb granite from Halayb gives the lowest dose and effective dose (0.16mSv/y).
2. Using natural materials such as igneous rocks tend to contain high radium, uranium, thorium and potassium concentrations in ceramic industry. This affects ceramic tiles radioactivity. Large areas of ceramic tiles should be minimized. Using natural aggregate with high radioactivity produces concrete with high radioactivity. Increasing aggregate content leads to the increase of concrete radioactivity.
3. Different brick samples of same type show different spectrometric data, because their raw materials are of different origin. Brick samples show wide range of radioactivity. Still, clay bricks have the highest effective dose. The raw material of the clay bricks contains a relatively high amount of Uranium and Thorium. Clay brick has the highest radioactivity of the investigated bricks (0.12mSv/y). Cement brick has the lowest radioactivity of the investigated bricks (0. 0.03mSv/y).
4. Although all the investigated materials have safe effective doses, they cause unsafe radon concentration, when they are collected in a room. Using natural materials of high radioactivity causes high radon concentrations compared with others. Radon concentration becomes saturated in a room atmosphere in about ten hours with moderate ventilation rate.

5. The height is the most effective parameter in room dimension; it should be concerned in building design. The plan dimensions do not affect the dose seriously. The ventilation rate is a very powerful factor securing air emission. The outdoor radon concentration affects the indoor radon concentration.

6. Recommendations

1. Using cement brick instead of clay brick is recommended
2. Calculating the radioactivity, effective dose and radon concentration of building materials planned to be used in the designing stage is very important.
3. Using ventilators reduces the radon concentration in unsafe cases.
4. Ventilating areas should be increased to reduce radon emission.
5. Good maintenance of the structure prevents a lot of health hazard.

Acknowledgment

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