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Radiological Assessment of Naturally Occurring Radioactive Material (NORMs) in Selected Building Materials

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ABSTRACT

The use of building materials containing high concentration of naturally occurring radioactive materials (NORMs) may result in the increase of radiation dose rate inside the building. The assessment of radiological hazard in building materials is necessary due to the concern of radiological impact to the people in the building. NORMs present naturally in the environment, but the concentration of radionuclide such as uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) may increase due to anthropogenic activities. NORM at certain limit becomes hazardous to environment and human. Thus, the objectives of this study were to determine the level of natural radioactivity in Malaysia's building materials and to assess the associated radiological risk of selected building materials. The selected material used were sand, soil, cement (white and black), bricks (sand and clay), marble and gravel. The assessment of NORMs in building materials were carried out and the concentrations of were determined using Energy Dispersive X-ray Fluorescence (EDXRF). Activity concentrations of ^{238}U, ^{232}Th and ^{40}K were found to be in the range of 13.93-60.09 Bq/kg, 3.44-128.14 Bq/kg and 2.11-1294.93 % respectively. The external hazard index for all building material samples is under the permissible limits (<1.0) except gravel (1.10). The external hazard index must be less than unity in order to keep the radiation hazard to be negligible. It is suggested that the radiological risk of raw material is predetermined before being used as building material.

1. Introduction

The ionizing radiation of naturally occurring radioactive material (NORM) may cause continuously exposed to humans. The main radionuclide in NORM is ^{238}U, ^{232}Th, and ^{40}K can be originated from primordial era. NORMs are known to be present permanently in rocks and soils since their half-live are very long (up to 1010 years) [1,2]. All building materials contain small amounts of radioactive substances. The most important naturally occurring radionuclides present in building materials including soil are ^{226}Ra, ^{232}Th and ^{40}K. The radionuclides are not uniformly distributed; they can vary considerably according to the geological origin of the raw materials [3]. The main sources of all

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product and raw building material in peninsular Malaysia are made from soil and rock based such as cement, bricks, tiles, and sand. It increases the exposure of NORM to people based on the rapid corporeal development of the country. UNSCEAR 2000 report shows that every person on the earth annually receives, on average, an effective dose of about 0.48 mSv due to external exposure to radiation due to the presence of terrestrial sources [4]. Therefore, the concentration of some pollutants such as NORMs, may increase 2 to 5 times higher than typical outdoor concentration. So, measuring the activity concentrations of radionuclides in building materials is important for the radiological assessment of population exposures. Therefore, the awareness of the natural radioactivity in building materials is essential for the determination of population exposure to radiations [5-7].

The assessment of NORM in building material of peninsular Malaysia to provide baseline data since at present is very limited. So, the current data of NORM in building materials need to be established. In this work, the concentrations of natural radionuclides were measured in 27 samples of nine types of building materials that are commonly used in the peninsular Malaysia, by means of Energy Dispersive X-ray Fluorescence (EDXRF), with the aim of assessing the radiological hazards due to external radiation exposure in residence. The potential radiological hazards associated with these materials were assessed by calculating the external hazard (Hex) indices [8-10]

Natural radioactivity of building materials originating from natural sources (soil, rock) is connected mainly with the radium (^{226}Ra), thorium (^{232}Th), and potassium (^{40}K) radionuclides. The built-in materials can cause both external and internal exposures indoors. The external exposure is caused by gamma radiation resulting from the decay of the radionuclides present in material; the internal exposure is caused by inhalation of the decay products of radionuclides present in materials, e.g., radon gas. Knowledge of hazards is required to take protective precautions to decrease the exposure of the population to ionizing radiation [1,11-13].

The main effects of ionizing radiation on living organisms are cell death, loss of reproductive capacity, or mutation. Exposure to gamma radiation in the indoor environment would result in low doses of radiation [14]. Since the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in building materials is not uniform, the real radioactivity levels in building materials can be assessed in the form of a single quantity by using several radiological indices [15]. The activity concentrations of all three radionuclides, was proposed by the European Commission and it is most often used to assess the dose level of external gamma radiation from building materials. However, based on EC RP 112 (119), the radiation protection principles in the EU, controls are recommended for building materials contributing to overall dose by the value of 0.3–1 mSv/y.

2. Method

2.1 Materials

All building material samples were purchased from three different hardware shops in peninsular Malaysia. The sampling locations were chosen according to their population and the amount of building material used in the study area. Three locations in the district were chosen for this investigation, i.e., Malacca, Kedah and Selangor. The selected material used were sand, soil, cement (white and black), bricks (sand and clay), marble and gravel.

2.2 Sample Preparation and Sample Measurement

Each sample was properly catalogued, marked and coded according to its origin and the location of the sampling site. After crushing and grinding, the representative samples with a maximum grain size of 250 µm to get more homogeneity. Samples were dried in an oven at approximately 110 °C until the sample weight became constant. Approximately 5.0 g of powdered samples were pressed into pallet form before analysed by Energy Dispersive X-ray Floresence spectrometer [3,16,17]

2.3 Radiological Assessment

The concentration of all three elements which is uranium, thorium and potassium determine by using the conversion factor. Concentration of uranium (^{238}U) is calculated from Eq. (1), thorium (^{232}Th) from Eq. (2), potassium (^{40}K) from Eq. (3).

$$1 \text{ Bq } ^{238}U/kg = 0.081 \text{ ppm} \tag{1}$$

$$1 \text{ Bq } ^{232}Th/kg = 0.246 \text{ ppm} \tag{2}$$

$$1 \text{ Bq } ^{40}K/kg = 32.3 \text{ ppm} \tag{3}$$

It is calculated through the following relation proposed by (Ademola, 2009), if 370 Bq/kg ^{232}Th or 4810 Bq/kg ^{40}K produces the same gamma dose rate. Next, all activity concentration of radionuclides has been calculated. Absorbed dose (D), annual effective dose (E), and external hazard index (H_{ex}) can be determined. The radium equivalent (Ra_{eq}) in Bq/kg is calculated using the formula proposed by UNSCEAR 2000.

The conversion factors used to calculate the absorbed dose rates are given as using Eq. (5). Then, from the absorbed dose (D), To estimate the annual effective dose rates, the conversion coefficient from the absorbed dose in air to the effective dose (0.7 Sv/Gy) and the outdoor occupancy factor (0.2) proposed by UNSCEAR, 2000 is used. Hence the annual effective dose rate can be calculated by using the Eq. (6). Finally, to evaluate the hazard of the natural gamma radiation, the external hazard index (H_{ex}) is calculated from the following formula in Eq. (7).

$$D \text{ (nGy/h)} = 0.427\text{Conc. } ^{238}U + 0.662\text{Conc. } ^{232}Th + 0.0432\text{Conc. } ^{40}K \tag{5}$$

$$E \text{ (Sv/y)} = D*8760*0.2*0.7*10\text{^-}6 \tag{6}$$

$$H_{ex} = (\text{Conc. Ra-226}/370) + (\text{conc. T} + (\text{conc.K-40}/4810) \tag{7}$$

3. Results and Discussion

3.1 Concentration of Radionuclide

The mean concentration of ^{238}U, ^{232}Th and ^{40}K present in building materials samples are given in Table 1. ^{238}U and ^{232}Th measured in mg/kg, while potassium in percent (%). The concentrations of ^{238}U, ^{232}Th and ^{40}K in all samples are in the range sand range 1.13- 4.87 Bq/kg, 0.85-36.28 Bq/kg and 0.53-4.42 Bq/kg respectively. From the Table 1 it shown that the highest means value of ^{238}U concentration is 4.87±0.09 mg/kg in sand, while the lowest ^{238}U concentration is 1.13±0.07 mg/kg in cement (grey). Then, the highest means value ^{232}Th concentration is 36.28±0.52 mg/kg that was found in gravel while the lowest ^{232}Th concentration is 0.85± 0.16 mg/kg in cement (grey). Furthermore, the highest ^{40}K concentration is 7.01±0.01 mg/kg in cement (white) while the lowest ^{40}K concentration is 0.53±0.02 mg/kg in soil.

Table 1
Mean value of concentrations of NORM in building material samples in mg/kg

Samples	^{238}U	^{232}Th	^{40}K (%)
Sand	4.87±0.09	10.96±0.19	1.69±0.18
Cement (white)	1.21±0.05	0.89±0.07	7.01±0.01
Cement (black)	2.37±0.05	4.63±0.19	0.68±0.01
Cement (grey)	1.13±0.07	0.85±0.16	4.17±0.06
Gravel	4.57±0.11	36.28±0.48	4.42±0.01
Brick (Sand)	2.68±0.07	2.95±0.17	0.89±0.01
Brick (Clay)	3.91±0.07	24.79±0.36	2.71±0.02
Soil	2.28±0.03	26.32±0.24	0.53±0.02
Tiles	3.17±0.09	14.22±0.10	1.13±0.01
Range	1.13 – 4.87	0.85 – 36.28	0.53 – 7.01

3.2 Activity Concentrations of Radionuclide

The results obtained are calculated using a conversion factor as mentioned in radiological assessments. As seen in Table 2 the range for activity concentrations of ^{238}U, ^{232}Th and ^{40}K are 13.93 - 60.09 Bq/kg, 3.57 – 147.47 Bq/kg and 211.75 – 1373.71 Bq/kg respectively. Commonly, the mean concentration of ^{238}U, ^{232}Th and ^{40}K in the samples are relatively less than the world average (50 Bq/kg for ^{238}U and ^{232}Th, and 500 Bq/kg for ^{40}K) present in building materials except for ^{238}U in sand (60.09 ± 1.11 Bq/kg) and gravel (56.41 ± 1.36). For ^{232}Th the samples that are relatively higher than the world average is gravel (147.47 ± 1.95 Bq/kg), brick (clay) (101.55 ± 1.46 Bq/kg) and soil (128.14 ± 0.98 Bq/kg). While in ^{40}K the samples that are higher than world average is sand (524.88 ± 55.90 Bq/kg), cement (white) (898.65 ± 3.11 Bq/kg), cement (grey) (1294.93 ± 18.63 Bq/kg) and brick (clay) (845.83 ± 6.21) Bq/kg. Conversely, the mean values of all radionuclide in most of the building material samples are close or slightly above the world average value of soils (45 Bq/kg for ^{238}U, 33Bq/kg for ^{232}Th, and 370 Bq/kg for ^{40}K) (UNSCEAR). Studies conducted on natural radionuclides in Malaysian building material in 2006 found the mean concentration of most of building material was lower than world average different from present studies the increase of mean concentration especially for ^{232}Th in brick (clay), soil and tiles [2].

Table 2
Mean Activity concentration of radionuclide in Bq/kg

Samples	^{238}U	^{232}Th	^{40}K
Sand	60.09 ± 1.11	44.56 ± 0.77	524.88 ± 55.90
Cement (white)	14.72 ± 0.62	3.57 ± 0.28	898.65 ± 3.11
Cement (black)	29.25 ± 0.62	18.82 ± 0.77	211.75 ± 3.11
Cement (grey)	13.93 ± 0.86	3.44 ± 0.65	1294.93 ± 18.63
Gravel	56.41 ± 1.36	147.47 ± 1.95	1373.71 ± 3.11
Brick (Sand)	33.23 ± 0.86	11.93 ± 0.69	274.59 ± 3.11
Brick (Clay)	49.16 ± 0.86	101.55 ± 1.46	845.83 ± 6.21
Soil	30.67 ± 0.37	128.14 ± 0.98	213.25 ± 6.21
Tiles	39.19 ± 1.11	57.80 ± 0.41	349.28 ± 3.11
Range	13.93 – 60.09	3.57 – 147.47	211.75 – 1373.71
UNSCEAR 1993	50	50	500

3.3 Radiological Assessment

From the Table 3, represents the radium equivalent values range was found from 91.37 to 406.91 Bq/kg. The lowest value was found in this study is cement (black) while the maximum value was found in gravel. The radium equivalent permissible values for all analysed samples are less than the world average value 370 Bq/kg (UNSCEAR, 2000) except the sample of gravel, while the value is 406.91 Bq/kg. Thus, the building materials used in the present study are quite safe to be used as building materials except gravel. This is because according to UNSCEAR 1982 where a radium equivalent of 370 Bq/kg in building materials yields an exposure of about 1.5 mSv/y to inhabitants that should be consider taking for radiation protection. Absorbed dose rate ranged from 33.95 to 181.05 nGy/h. According to UNSCEAR 2000, the world average absorbed dose rate is 55 nGy/h. The average absorbed rate for certain samples for this study was above the world average value which is more than 55nGy/h.

Table 3
Radium equivalent activity, absorbed dose rate, annual effective dose, and external hazard index

Samples	Radium equivalent act. (Bq/kg)	Absorbed dose rate (nGy/h)	Annual effective dose (mSv/y)	External Hazard index (Hex)
Sand	200.27	77.83	0.10	0.54
Cement (white)	97.85	47.47	0.06	0.26
Cement (black)	90.02	34.10	0.04	0.24
Cement (grey)	126.91	64.16	0.08	0.34
Gravel	406.91	181.05	0.22	1.10
Brick (Sand)	91.37	33.95	0.04	0.25
Brick (Clay)	289.00	124.76	0.15	0.78
Soil	248.73	107.14	0.13	0.67
Tiles	172.26	70.09	0.09	0.47
Range	90.02 – 406.91	34.10 – 181.05	0.04 – 0.15	0.24 -1.10
UNSCEAR 2000	370	55	< 1	< 1

The highest value (0.22 mSv/y) of annual effective dose in this study was observed in gravel while the lowest value (0.06 mSv/y) was found in cement (black). However, the annual effective dose for all samples is less than 1.0 mSv/y as prescribed by UNSCEAR 2000, as the maximum tolerable dose from building material. According to El-Taher, the external hazard index must be less than unity to keep the radiation hazard to be negligible. For external hazard index the maximum suggested value is 1. However, the results show highest value (1.10) was found in gravel. Any value more than 1 show that the external hazard index is more than the maximum suggested dose by UNSCEAR 2000. So, the external hazard index for all building material samples is under the permissible limits except gravel.

4. Conclusions

The results indicate that the activity concentration of naturally occurring radioactive material of ^{238}U,^{232}Th and ^{40}K in most of the building materials studied are approximately lower than worldwide average except in ^{238}U (Sand and gravel), ^{232}Th (gravel, brick, soil, and tile) and ^{40}K (sand, cement (black), cement (grey) and brick). The determined of radiological assessment are also not exceeding the maximum suggested values except gravel. Therefore, these could be explained the use of these building material samples in the construction of resident is safe for dweller, except the gravel samples which are critical points for safety in construction. According to the results, the use of some restricted building materials in Peninsular Malaysia is discouraged, and the replacement of them by building materials with low radioactivity contents is suggested.

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