Determination of factors affecting indoor doses from Malaysia’s ceramic tiles containing natural radionuclides

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Abstract. The level of natural radioactivity and radiological risks attributed to forty (40) different ceramic tiles using gamma-ray spectroscopy employing high-purity germanium detector were studied. The average activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K range from 37.5 ± 0.3 to 215.9 ± 5.8 Bq kg$^{-1}$, 42.2 ± 0.1 to 181.8 ± 3.8 Bq kg$^{-1}$ and 349.5 ± 25 to 1589.2 ± 21.1 Bq kg$^{-1}$, respectively. The radium equivalent activity and activity concentration index were calculated to estimate the potential radiological hazards to the dweller. Furthermore, the factors affecting the total effective dose equivalent (TEDE) such as ventilation rate, room size and dweller position were investigated from the measured activity concentrations using RESRAD-BUILD computer code. The simulation results of TEDE from the variations of ventilation rate in a room range from 0.26±0.01 to 0.61±0.01 mSv y$^{-1}$, on the other hand, the percentage variation of TEDE due to dweller position and room size are 35% and 33%, respectively. The calculated radiological risks parameters were all below the recommended maximum limit. Therefore, the radiological impact attributed from the ceramic tiles under study is negligible.

1. Introduction

Natural background radiations are responsible for over 80% of the radiation exposure received by members of the public [1]. The critical ones’ present in the environment are Uranium ($^{238}$U) and Thorium ($^{232}$Th) decay progeny and isotope of Potassium ($^{40}$K). These radionuclides are naturally found in all earth materials including soil, rock and building materials. However, the radiation exposure from natural radionuclides are unavoidable since they can be almost found everywhere. Building materials such as tiles can significantly increase exposure due to natural radionuclides. Moreover, the level of contributions of the exposure from building materials to the general public depends on the selection of raw materials, quantity of each material in the building, geological origin, manufacturing process and added opacifier during industrial processes. The emphasis here is ceramic tiles which are mostly used for decorative purposes in both indoor and outdoor environment. Ceramic tiles are produced from earth materials by pressing into shape and firing at high temperature [2]. The surface of the tile may be glazed or unglazed depending on the desired needs [3]. The important properties of ceramic tiles are less water absorption level (< 0.5 to 10%), high chemical and mechanical properties [2–4]. Therefore, the aim of
this study is to determine the factors affecting the indoor doses attributed to Malaysia’s ceramic tiles containing natural radionuclides.

2. Material and Method

The materials under study are ceramic tiles sourced from various locations in Malaysia. The number of the samples considered in the study are forty (40). The samples were cleaned and dried in an oven at 105 °C for 48 hours until constant weight is achieved. The choice of the temperature and time were in accordance with International Atomic Energy Agency (IAEA) [1,5,6]. The samples were ground, sieved using 500 µm mesh, weight, packed and sealed in an air tight merinelli beaker. The samples were then kept in a laboratory for 30 days to achieve secular equilibrium; the steps mentioned are important in the present study, since the activity concentration of $^{226}$Ra and $^{232}$Th were determined from their daughter nuclides.

The prepared samples were studied using High-Purity Germanium (HPGe) detector located at Nuclear Science Program, Universiti Kebangsaan Malaysia. Each sample was prepared in three replicates and counted for 12 hours. Prior to counting of the samples, energy calibration of the HPGe detector was performed using a standard mixture of $^{228}$Na, $^{60}$Co and $^{137}$Cs radionuclides. Furthermore, the background measurements were also performed by routine counting of empty merinelli beaker for 12 hours and the background contributions were deducted during spectrum analysis of the samples. The activity concentrations of $^{226}$Ra and $^{232}$Th were determined through gamma-ray energy peaks of 1765 keV ($^{210}$Bi) and 2615 keV ($^{208}$Tl), respectively. While, the activity concentration of $^{40}$K was determined directly from 1460 keV gamma-ray energy peak.

The present study adopted a relative method for determination of $^{226}$Ra, $^{232}$Th and $^{40}$K natural radionuclides. The certified IAEA-375 soil of known activity concentrations was prepared in two replicates counted several times using the preset counting time of 12 hours. The activity concentrations of IAEA-375 soil are 20, 20.5 and 424 Bq kg$^{-1}$ for $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively. The obtained gamma-ray energy peaks of the IAEA-375 soil and its activity concentrations ($C_{std}$) in Bq kg$^{-1}$ were compared with the obtained gamma-ray energy peaks of the samples with a view to determine the individual activity concentrations of the sample ($C_{s}$) in Bq kg$^{-1}$ as shown in Eq. (1) [1][7][8].

$$C_s = \left( \frac{M_{std} \times A_s \times C_{std}}{M_s \times A_{std}} \right)$$  (1)

where $M_{std}$ and $M_s$ are Mass of the standard (IAEA-375 soil) and sample in kg, respectively. $A_{std}$ and $A_s$ are activity of the standard and sample in count per second (cps), respectively.

3. Result and discussion

The measured activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K are presented in Fig. 1(a and b) with values ranging from 37.5 ± 0.3 to 215.9 ± 5.8 Bq kg$^{-1}$, 42.2 ± 0.1 to 181.8 ± 3.8 Bq kg$^{-1}$ and 349.5 ± 25 to 1589.2 ± 21.1 Bq kg$^{-1}$, respectively. It can be observed that the variations of the natural radionuclides are not uniform across the studied tiles. The observed non-uniform behavior may be associated with different raw materials selection and manufacturing processes. Furthermore, most of the measured activity concentrations in the studied tiles are above the world average values of $^{226}$Ra, $^{232}$Th and $^{40}$K in building materials as reported by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) of 50, 50 and 500 Bq kg$^{-1}$ for $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively [9]. On the contrary, the measured activity concentrations are comparable with reported Malaysia’s soil activity concentrations of 67, 82 and 310 Bq kg$^{-1}$ for $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively [10].

3.1 Radiological Hazard Parameters

The potential radiological hazard associated with ceramic tiles were studied through radium equivalent activity ($Ra_{eq}$) and activity concentration index (ACI) as shown in Eq. (2) and Eq. (3), respectively [4][7].
\[ Ra_{eq}(Bq\ kg^{-1}) = \left( \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810} \right) \times 370 \]  

\[ ACI = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_{K}}{3000} \]  

where \( C_{Ra}, C_{Th} \) and \( C_K \) are the activity concentrations of \(^{226}\text{Ra}, \, ^{232}\text{Th} \) and \(^{40}\text{K} \) in Bq kg\(^{-1} \), respectively. The two radiological hazard parameters are the main parameters used to determine the external radiological hazard due to gamma radiation from building materials. The contributions of building materials to external radiological hazard is considered negligible if the \( Ra_{eq} \) in the materials is less than 370 Bq kg\(^{-1} \) [7]. On the other hand, the ACI of building materials should be less than the exemption level of 2 for the materials used in limited quantities such as tiles and board, while the action level is 6. However, the building materials used in bulk quantities such as cement, bricks and sand have the same exemption and action level of 1 [4,11]. The variations of \( Ra_{eq} \) and ACI from the ceramic tiles are presented in Fig. 1(c) and Fig. 1(d), respectively with values ranging from 146 to 565 Bq kg\(^{-1} \) and 0.5 to 2.0 for \( Ra_{eq} \) and ACI, respectively. The highest and the least value of \( Ra_{eq} \) and ACI are found in tile 5 and tile 2, respectively. And, over 90 % of both \( Ra_{eq} \) and ACI reported herein are below the recommended maximum limits. Therefore, the external radiological hazards due to gamma radiation from the studied ceramic tiles are negligible.

Fig. 1 (a and b) Variations of activity concentrations of \(^{226}\text{Ra}, \, ^{232}\text{Th} \) and \(^{40}\text{K} \) in various ceramic tiles; (c) variations of Radium equivalent activity from various ceramic tiles; (d) variations of activity concentration index from various ceramic tiles.
3.2 Evaluation of Factors Influencing Total Effective Dose Equivalent using RESRAD-BUILD Computer Code

RESRAD-BUILD Computer code is a model designed to determine the indoor doses from Residual Radioactivity in Buildings (RESRAD-BUILD) [12]. The RESRAD code is developed by Argonne National Laboratory United States (US). The design of the code was sponsored by the department of energy and federal agencies in the US [12]. The code is flexible and permit user to calculate indoor doses over the desired integrated time.

The measured activity concentrations were projected to RESRAD-BUILD computer code from initial year to 70 years to evaluate the effects of room size, dweller position and air exchange rate (ventilation rate) on the total effective dose equivalent (TEDE). TEDE refers to the combined indoor external and internal doses related with the measured activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in the ceramic tiles as discussed in the previous section. The code determined the indoor doses by assuming a uniform distribution of the radiation doses in the buildings. However, the code may over-estimate the indoor doses from Thoron ($^{220}$Rn) due to its known variability nature, the variability nature of $^{220}$Rn is associated with its short half-life of 56 s, which would make it difficult to attain a uniform distribution in the buildings.

The variations of room size and air exchange rate considered in this study are 6 to 42 m$^2$ and 0.1 to 2.0 h$^{-1}$, respectively. The role of room size, dweller positions and air exchange rate on the variations of

![Fig. 2 (a) Effects of room surface area on total effective dose equivalent over time; (b) effects of dweller position relative to the wall on total effective dose equivalent over time; (c) effects of air exchange rate on total effective dose equivalent over time.](image-url)
TEDE over time are presented in Fig. 2(a), Fig. 2(b) and Fig. 2(c), respectively with values ranging from 0.2 to 0.4 mSv y$^{-1}$, 0.2 to 0.3 mSv y$^{-1}$ and 0.3 to 0.6 mSv y$^{-1}$ for TEDE due to room size, dweller position and air exchange rate, respectively. The variations of TEDE with room size is presented in Fig. 2(a), it can be observed that the TEDE decrease as the room size increase from 6 m$^2$ up to 36 m$^2$ however from 36 m$^2$ upwards the TEDE seem to be relatively stable. In addition, the variations of TEDE over time indicate nearly identical behavior. The impact of dweller position from the wall on TEDE is presented in Fig. 2(b) with TED$E$ increase as the dweller is moving away from one extreme end of the room toward to the other end. The initial position of the dweller was assumed to be the location of the air exchange systems of the room. Therefore, the air exchange systems play a significant role in the variations of TEDE as show in Fig. 2(c). It can be observed that the TEDE decrease with increasing air exchange rate. Even though, the highest TEDE recorded from the studied factors was below the recommended maximum limit of 1 mSv y$^{-1}$ as reported by European Commission (EC) [13], there are still significant variations observed from initial year to 70 years as shown in Fig. 2(a), Fig. 2(b) and Fig. 2(c), respectively. The behavior of the TEDE with the studied factors were also in agreement with reported previous studies [14][15].

4. Conclusion

The activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in the studied tiles materials were mostly above the world average values of 50, 50 and 500 Bq kg$^{-1}$, respectively. On the contrary, the calculated radium equivalent activity and activity concentration index were generally below the recommended maximum values except for tile 1 and 5. The factors influencing the indoor doses were also investigated using RESRAD-BUILD computer code. It was observed that increase in room size and ventilation decrease the indoor doses receive by the dweller. Similarly, the closer the dweller position is to the wall the higher the indoor doses. Therefore, the radiological hazards associated with ceramic tiles under study is negligible.

5. References

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