Gamma Radiation Measurements in Tunisian Marbles

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ABSTRACT

The radioactivity of 15 kinds of different granites collected in Tunisia was determined by gamma-ray spectrometry using hyper-pure germanium (HPGe) detector. The average activity concentrations for primordial radionuclides ²³⁸U, ²³²Th and ⁴⁰K were respectively 33.24, 8.01 and 116.98 Bq/kg. The activity concentrations ranged from 3.59 to 87.37 Bq/kg for ²³⁸U, from 0.45 to 25.34 Bq/kg for ²³²Th and from 24.06 to 380.23 Bq/kg for ⁴⁰K. The measured activity concentrations were used to assess of the radium equivalent activity ranged from 22.2 to 995.8 Bq/kg, the absorbed dose rate in air from 7 to 1209 nGy/h and the internal (0.1 to 2.8) and external (0.1 to 2.7) h azard indices. The data obtained in this study may be useful for natural radioactivity mapping.

Keywords: Radioactivity; Marble; Radium Equivalent; External and Internal Hazard Indices

1. Introduction

Natural radioactive materials (rocks, soil, sediment, water and aliments) contain low-level radioactivity. Marble used in building and decoration are also possible sources of radioactivity. Radioactivity in the marbles consists of the ²³⁸U series [1-3] with a relative abundance of 99.284% and a half-life of 4.468 × 10⁹ years, the thorium series ²²²Th with a relative abundance of 100% and a half-life of 14.05 × 10⁶ years and the actinium series ²³⁵U with a relative abundance of 0.716% and a half-life of 9.47 × 10¹⁰ years. There are also several singly occurring radionuclides, the most important one is ⁴⁰K with a relative abundance of 0.0117% and a half-life of 1.248 × 10⁹ years. The emanation of radon (²²²Rn) is associated with the presence of ²²⁶Ra and its ultimate precursor ²³⁸U in the ground. The inhalation of its short-lived daughter products is a major contributor to the total radiation dose.

The objective of the work is to estimate the radioactivity concentrations of primordial radionuclides; ²³⁸U, ²³²Th and ⁴⁰K in marble samples collected in Tunisia. The absorbed and effective dose rate, the external and internal hazard indices in marble samples have been determined. The measurements have been carried out using a HPGe spectrometer.

2. Experimental Setup

A total of 15 different kinds of marble in Tunisia have been collected. Figure 1 shows the Tunisian states where the commercial marble samples were collected. The samples were pulverised, dry-weighed [4,5], sealed in 1 l plastic Marinelli beakers and stored for 4 weeks in order to allow the reaching of equilibrium between ²²⁶Ra and ²²²Rn and its decay products [6,7].

Spectra for different samples were measured with a hyper-pure germanium (HPGe) detector of high-resolution gamma-ray spectrometer. Figure 2 show a spectrum of Tunisian marble sample 1. The detector has a photopeak efficiency of 30% and an energy resolution of 1.85 keV (FWHM) for the 1.332 MeV reference transition of ⁶⁰Co. The measurements have been carried out at the Center for Nuclear Sciences and Technology, Sidi Thabet, Tunisia (CNSTN).

To obtain the gamma spectrum with good statistics, the accumulation time for each sample is set at 20 hours. Marble samples container were placed inside a shield, in order to minimize the background radiation. The background distribution in the environment around the detector was counted in the same manner and in the same geometry as the samples. It was determined using an empty sealed beaker. The background spectra were used to correct the net peak area of gamma rays of measured isotopes.

3. Results and Discussion

3.1. Calculation of Activity Concentrations

The specific activity $A_{E_i}$ (Bq/kg) of a nuclide $i$ and for
Figure 1. Localization of the Tunisian states where the commercial marble samples were collected.

A peak at energy $E_i$ is given by:

$$A_{E_i} = \frac{N_{E_i}}{n \times \epsilon_g \times \gamma_D \times t \times M_s}$$

where $N_{E_i}$ is the net peak area, $\epsilon_g$ the detection efficiency at energy $E_i$, $\gamma_D$ the gamma ray yield per disintegration of the specific nuclide for the transition at energy $E_i$, $M_s$ the mass of the measured sample (kg) and $t$ the counting live-time (s). If there is more than one peak in the energy range of analysis, then the result is the weighted average nuclide activity.

The gamma-ray transitions of $^{214}$Pb, $^{214}$Bi were used to determine the concentration of the $^{238}$U series. The gamma-ray transitions $^{228}$Ac, $^{209}$Tl $^{228}$Ac were used to calculate the concentration of the $^{232}$Th series. The gamma-ray
transition of $^{40}$K was used to determine the concentration of $^{40}$K in different samples.

The activity concentrations of the natural radionuclides of the 15 kinds of marbles are shown in Figure 3. The activity concentrations ranged from 3.59 to 87.37 Bq/kg for $^{238}$U, from 0.45 to 25.34 Bq/kg for $^{232}$Th and from 24.06 to 380.23 Bq/kg for $^{40}$K. The average activity concentrations are 33.24 Bq/kg for $^{238}$U, 8.01 Bq/kg for $^{232}$Th, and 116.98 Bq/kg for $^{40}$K.

From the 15 samples measured in this study, Sample (2) appears to present the highest concentrations for $^{238}$U, Sample (11) presents the highest concentration for $^{232}$Th and Sample (7) presents the highest concentration for $^{40}$K.

3.2. Absorbed and Effective Dose Rate

Absorbed dose rate $D$ (in nGy/h) can be calculated using the following formula [8,9]:

$$D = A_{E_i} \times C_F$$

(2)

where $A_{E_i}$ is the activity concentration (in Bq/kg), and $C_F$ is the dose conversion factor (absorbed dose rate in air per unit activity per unit of soil mass in nGy/h per Bq/kg).

To estimate the annual effective dose, one has to take into account the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor [10]. A value of 0.7 Sv/year was used for the conversion coefficient from absorbed dose in air to effective dose received by adults, and 0.8 for the indoor occupancy factor, implying that 20% of time is spent outdoors. The effective dose rate ($D_{eff}$) indoors (in mSv/year), is calculated by the following formula:

$$D_{eff} = D \times 8760(h/year) \times 0.2 \times 0.7 \times 10^{-6}$$

(3)

where $D$ (nGy/h) is the calculated dose rate, 0.7 (Sv/Gy) is the conversion coefficient from the absorbed dose rate in air to the effective dose rate and 0.2 is the outdoor occupancy factor [11].

Table 1 shows the absorbed and effective dose rate. The average absorbed and effective dose rates are 25.7 nGy/h and 0.23 mSv/year respectively. The highest value of the dose rate is for Sample (15), 51.20 nGy/h and the minimum value is for Sample (1), 4.31 nGy/h. The maximum effective dose rate is 0.45 mSv/year and the minimum is 0.04 mSv/year.

3.3. Radium Equivalent Activity

The distribution of $^{238}$U, $^{232}$Th and $^{40}$K in marbles is not uniform. Uniformity in respect of exposure to radiation
has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq/kg to compare the specific activity of marbles containing different amounts of 238U, 232Th and 40K. It is assumed that 370 Bq/kg of 232Th and 4810 Bq/kg of 40K produce the same gamma dose rate [12]. A radium equivalent of 370 Bq/kg in marbles will produce an exposure of about 1.5 mSv/year to the inhabitants [11]. The radium equivalent activity which can be calculated by the following formulae [13,14]:

\[
Ra_{\text{eq}} = A_U + A_{\text{Th}} \times 1.43 + A_K \times 0.077
\]  

(4)

where \(A_U\) is the 235U activity concentration (Bq/kg), \(A_{\text{Th}}\) is the 232Th activity concentration (Bq/kg) and \(A_K\) is the 40K activity concentration (Bq/kg).

The average calculated (Ra_{eq}) values for marble samples were ranging from 9.04 to 110.9 Bq/kg with an average value of 54.88 Bq/kg.

3.4. External and Internal Hazard Indices

To estimate the radiation dose expected to be delivered externally if a building is constructed using marbles, the external hazard index (H_{ex}), due to the emitted γ-rays, can be calculated using the following equation [8,15]:

\[
H_{\text{ex}} = \frac{A_U}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_K}{4810}
\]

(5)

In addition to the external irradiation, radon and its daughter products are also hazardous to the respiratory organs. The internal exposure to radon and its short-lived products is proscribed by the internal hazard index (H_{in}). For the safe use of marble in the construction of dwellings H_{in} should be less than unity. The Internal hazard index is calculated by the following formula [15-17]:

\[
H_{\text{in}} = \frac{A_U}{185} + \frac{A_{\text{Th}}}{259} + \frac{A_K}{4810}
\]

(6)

The calculated external and internal hazard indices values (Table 1) are ranged from 0.05 to 0.3 and from 0.04 to 0.51 respectively with an average value of 0.15 and 0.24 respectively.

4. Conclusions

Natural radioactivity of 15 samples of marbles collected in Tunisia was determined by gamma-ray spectrometry using hyper-pure germanium (HPGe) detector. In the case of uranium the highest level was found in the “Thala royal” marble sample and the lowest level in the “Gris Marmis” marble sample. For thorium the highest level was found in “Courteau” marble sample and the lowest level in “Thala imperial” marble sample, while for potassium the highest level was found in “Chamtou” marble and the lowest level in the “Kadhel rose” marble sample.

Based on the results obtained in this study for absorbed and effective dose rate, radium equivalent activity, external and internal hazard indices, we conclude that the radioactivity levels of marbles are within the international recommended values from world-wide areas due to terrestrial gamma radiation [11]. According to the recommended values and the calculated external and internal hazard indices values, the marble samples are acceptable for use as building materials and decoration.

REFERENCES


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