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Evaluation of radionuclide concentrations and associated radiological hazard in marble indices and granite used as building materials in Al-Madinah Al-Munawarah

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Abstract

Sixty samples of different marble and granite tiles used as building materials, from local and foreign origins, were collected from different factories in the Al-Madinah Al-Munawarah region of Saudi Arabia. Natural radioactivity of the U–Th series and ^{40}K in the samples were measured using an HPGE detector to conduct gamma-ray spectrometry. From the measured γ-ray spectra, the activity concentrations for different marble samples of Turkish, Egyptian, and Omani origin were ^{232}Th (0.10 ± 0.07, 0.9 ± 0.09, 0.31 ± 0.06 Bq/kg), ^{226}Ra (5.7 ± 0.3, 7.3 ± 0.40, 2.0 ± 0.1 Bq/kg), and ^{40}K (0.9 ± 0.2, 0.96 ± 0.1, 0.8 ± 0.09 Bq/kg), respectively, whereas for Saudi, Chinese, and Indian granite, the concentrations were ^{232}Th (48.6 ± 2.5, 22.9 ± 1.20, 80 ± 4 Bq/kg), ^{226}Ra (56.4 ± 3.0, 32 ± 1.7, 10.1 ± 1.1 Bq/kg), and ^{40}K (1492.8 ± 79.7, 762.5 ± 40.7, 1906.0 ± 43.6 Bq/kg), respectively. The calculated values of the radium equivalent activity Raeq for the studied marble samples ranged from 2.504 to 8.66 Bq/kg, and for granite, the values ranged from 127.8 to 244.19 Bq/kg. Moreover, a comparison study of the measured activity concentrations was presented. The values of external Hex and internal Hin hazard indices for the studied marble and granite samples are less than unity. The values of gamma-radiation hazard indices $I_{\gamma r}$ for the marble and granite samples are below 0.058 and 1.905, respectively. The values of the annual effective dose for the studied marble samples ranged from 0.011 mSv/y to 0.038 mSv/y, and that of granite ranged from 0.577 mSv/y to 1.169 mSv/y. Therefore, the use of these marble and granite samples under investigation in the construction of dwellings is considered to be safe for inhabitants, and the order of preference for marble is first Omani marble, then Turkish, and finally Egyptian marble. For granite, the preference is first Chinese granite, then Saudi granite, and finally Indian granite because the annual effective doses are comparatively lower.

Keywords: Radioactivity concentration; Radium equivalent; Marble; Granite; Hazard; Annual dose rate; Natural radioactivity

1. Introduction

Thousands of varieties of igneous rocks, marketed generally as "marble" and "granite," are widely used in the Al-Madinah Al-Munawarah province in Saudi Arabia as home faces and large-area countertops in kitchens and other rooms.

These stones can contain admixtures of Potassium ^{40}K and the decay series of natural Uranium (U_{-nat})

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and Thorium ^{232}Th. These include a number of progeny radioisotopes that emit gamma rays with characteristic intensities and energies ranging from some tens of "keV to 2.6 MeV". The presence of these radionuclides in the materials can cause external and internal exposure radiation for occupants who live in these buildings.

The external exposure is caused by direct gamma irradiation, and internal exposure is caused by the inhalation of the radioactive inert gas radon and radon decay products, which emanate from the building materials [1]. The use of such marble and granite as building materials in a home can thus result in long-term whole-body exposure of the occupants to this radiation.

Marble is a metamorphic rock, composed mainly of calcite (a crystalline form of calcium carbonate ($CaCO_3$)) and other minerals.

Any calcareous and/or dolomite rock with a polishable surface is called marble.

Marble rock is essentially formed from sediments and may contain large amounts of radioactive materials. The presence of other minerals in marble gives it a variety of attractive colours [2]. It is mostly used as a lining on walls and floors in dwellings.

However, granite is usually suitable as a building and ornamental material for interior and exterior use. It is a hard, natural stone that requires harder tools to cut, shape, and polish it, compared to marble. Granite is an igneous rock and usually medium- to coarse-grained, occasionally with some individual crystals. Granites are pink to dark grey or even black, depending on their chemistry and mineralogy [3].

Usually, marble and granite have commercial names, petrographic and technological characterizations, and their producer countries identified. Numerous works [4–13] have quantified the ^{40}K, U-$_{nat}$, and ^{232}Th activity concentrations, in units of Bq/kg, in specific stones and other building materials.

To estimate the health risks, these studies have commonly provided the values of various hazard indices [14–16] and/or dose rates at a distance above a large area of the material using conversion factors from UNSCEAR [17–21] in units of (nGy/h)/(Bq/kg). The objective of the present study is to determine the specific radioactivity concentrations of natural radionuclides of ^{232}Th, ^{226}Ra, and ^{40}K in selected marble and granite rocks used as building materials in the Al-Madinah Al-Munawarah region in Saudi Arabia with the aim of contributing and establishing a baseline data of radioactivity levels in building materials.

The average radium equivalent activity (Ra_{eq}), total absorbed dose rate (D), external (H_{ex}), internal (H_{in}), and gamma radiation ($I_{\gamma r}$) hazard indices have been estimated and compared with the recommended limits from the UNSCEAR report. Additionally, a comparison with the published results of activity concentrations of thorium, radium, and potassium in marble and granite samples from different countries is reported. The results are of great interest in the environmental radiological protection study because marble and granite are widely used as building and ornamental materials.

Local authorities can limit the use of building materials that cause a significant increase in radiation exposure due to higher levels of indoor radon and external gamma exposure. According to international recommendations quoted in the Basic Safety Standards No. 115 from the IAEA, the use of building materials containing enhanced concentrations of NORM should be controlled and restricted via the application of the radiation safety standards. Several publications address measuring low levels of naturally occurring radioactive isotopes in marble and granite rocks [22–27,8].

2. Experimental

2.1. Sampling and sample preparation

Sixty samples of different marble and granite tiles used as building materials were collected from different countries of origin. Three different types of marble, Type 1 (Turkish), Type 2 (Egyptian), and Type 3 (Omani), and three different types of granite, Type 1 (Saudi), Type 2 (Chinese), and Type 3 (Indian), were collected from factories.

The samples were crushed using a laboratory jaw crusher. Each sample was homogenized and sieved through approx. 100 mesh and then dried in an oven at 110 °C.

The samples were placed in (500 g) Marinelli beakers and stored for at least one month to allow the short-lived daughter nuclides to attain equilibrium with the long-lived parents.

2.2. Analysis of samples

Radioactivity measurements were carried out on the samples using a gamma-ray spectrometer, model Gem 80 p4, made up of a coaxial hyper pure germanium detector connected to a multichannel analyser manufactured by Canberra. The detector crystal has a diameter of 74.7 mm and height of 92.9 mm.

The detector has a relative efficiency of 80% and energy resolution of 2 keV (FWHM) for the 1.33 MeV gamma ray line of the ^{60}Co gamma source. Both energy