X-ray diffraction and gamma-ray analysis of rock samples from Haradh Region in Saudi Arabia

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ABSTRACT
Rock samples collected from Haradh region, Saudi Arabia were analyzed by using X-ray diffraction (XRD) spectroscopy to determine mineral composition. The XRD spectroscopy results indicate that the major, minor, and trace constituents varied from one sample to another. Also, high purity germanium (HPGe) detector was used to determine the activity concentration of U-238, Th-232 series, and K-40. The average concentration levels were 16.71, 4.41, and 55.51 Bq/kg for ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. For assessing the potential radiological risks to human health, absorbed dose rate, radium equivalent activity, annual effective dose, and internal and external hazards were determined and compared with limits recommended by UNSCEAR. Results were within recommended safe ranges, meaning that the area under study is radiologically safe for habitation and can be used as construction materials.

1. Introduction
Many natural rocks contain radioactive elements such as ²³⁵U, ²³⁴Th, and ⁴⁰K (Abbady et al., 2006). Although these radionuclides are widely distributed, and their concentrations depend on geological and geographical conditions, they vary from place to place (Iqbal, Tufail, & Mirza, 2000). Some igneous rocks such as granite and basalt have other purposes in industries and commercials due to their high clay mineral element (El-Taher, Uosif, & Orabi, 2007). Studies of natural radioactivity are necessary not only because of their radiological impact but also because they act as an excellent biochemical and geochemical tracer in the environment (Sroor, El-Bahi, Ahmed, & Abdel-Haleem, 2001). Therefore, the assessment of the radioactive contents of various radionuclides in rocks may play an essential role in preventing health hazards and environmental pollutions by helping us to control and minimize such radiation exposure. Several studies have been made in many countries to determine the concentration of the natural radionuclides in rock samples, e.g. Masok, Masiteng, Mavunda, Maleka, and Winkler (2018), Prakash, Kaliprasad, and Narayana (2017), and Uosif, Hashim, Issa, Tamam, and Zakaly (2016). The objectives of the present study were to determine metal composition and measure the natural radioactivity levels of ²²⁶Ra, ²³²Th, and ⁴⁰K in rock samples collected from an important site in Saudi Arabia (Haradh region) also to assess the radiological hazard indices in the air and to compare the results with international levels. The results are significant since these types of rocks are usually found in buildings and ornamental materials. The data generated in this work are of great interest in the environmental radiation protection study and may be helpful as a baseline in making estimations of population’s exposure in Saudi Arabia, as well as to plan and conduct further studies on this issue. Radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K were measured with a well-type gamma-ray detector. Metal concentrations were determined by X-ray diffraction (XRD), and patterns were recorded using X’Pert PRO Powder X-Ray Diffraction with CuKα radiation (λ = 1.5418 Å), Ni-filter, and the general area detector.

1.1. Description of the study area
This study was carried out in a part of the eastern region in Saudi Arabia (Haradh), which lies on the longitude 49°04’ east and latitude 24°08’ north, 273 km northeast of Riyadh city, the capital of Saudi Arabia. There are two main reasons behind choosing this specific region: first, because it represents the foundation of vast agricultural and commercial industries, and second, because of its exposure to many companies that are engaged in oil and gas exploration. The concentration of the natural radionuclides in rock samples collected from Haradh region, which are used in various constructions as building materials, has not been verified and no available data of the rock activities was done in this area. Figure 1 is a map that shows the area of the study.

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2. Material and methods

2.1. Samples

Ten granite rock samples were collected randomly from different locations in Haradh region, Saudi Arabia. In the laboratory the rocks were crushed and pulverized to a uniform mixture and were sieved to a particulate size of about 2 mm. The ground samples were dried at 110°C for 12 h to remove all moisture and then weighed. For radiometric analysis, about 500 g of powder samples were filled with polyethylene containers and kept for 4 weeks to attain a secular equilibrium between $^{226}$Ra and $^{232}$Th and their decay products (El-Taher, 2010). Each sample was analyzed using an advanced powder X-ray diffraction system (Bruker, USA) to determine the composition of minerals.

2.2. Measurements

Ten grams of the dried samples were analyzed by XRD model X’Pert PRO powder diffract meter equipped with Cu anode, for the chemical and mineral composition. The X-ray source used in this research has a wavelength of $\lambda = 1.5418$ Å. The diffract grams were recorded in the $2\Theta$ range of 0.5–70° with the step size of 0.02 Å and a step time of 0.6 s. Also, the gamma rays emitted by the rock samples were measured by using high purity germanium (HPGe) detector gamma-ray spectrometry system of 1.85 keV resolution and 25% relative efficiency with coaxial type vertical dipstick cryostat. It is surrounded by lead and copper, which provides an efficient suppression of background gamma radiation present at the laboratory. The system has a resolution (Full Width at Half Maximum [FWHM]) of 3.0–3.5 keV for 1332.5 keV gamma-ray peak of $^{60}$Co and a peak to a Compton ratio of 41:1. The spectra were analyzed by commercially available software GENIE-2000 obtained from Canberra, USA. The system was calibrated for energy and absolute efficiency (IAEA, 1989). Counting of samples and background was done for 36000s duration. The activity concentrations of $^{232}$Th, $^{226}$Ra, and $^{40}$K were specified by using the following obvious and explicit peaks: $^{232}$Th (238.63 keV of $^{212}$Pb, 911.21 of $^{228}$Ac, and 583.02 keV of $^{208}$Tl), $^{226}$Ra (351.9 keV of $^{214}$Pb; 609.3, 1120.3, and 1764 keV of $^{214}$Bi), and $^{40}$K (1460.83 keV).

2.3. Calculations

The radioactivity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the environmental samples were calculated using the following relation (El-Taher & Al-Zahrani, 2014):

$$A = N_{C}/\varepsilon \gamma m$$

(1)

where $A$ is the activity concentration in Bq/kg, $N_{C}$ is the net gamma counting rate (counts per second), $\varepsilon$ the detector efficiency of the definite gamma ray, $\gamma$ is the absolute transition probability of gamma-decay, and $m$ the mass of the sample (kg).

The radium equivalent activity ($Ra_{eq}$) was calculated by using the following formula (NEA-OECD, 1979):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$

(2)

This formula is based on the assumption that 370 Bq/kg of $^{226}$Ra, 259 Bq/kg of $^{232}$Th, and 4810 Bq/kg of $^{40}$K produce the same gamma-ray dose rate (Stranden, 1976). The external hazard ($H_{ex}$) and internal hazard ($H_{in}$) indices for the samples can be used to estimate the radiation hazards. They can be calculated using the following equations (Beretka & Matthew, 1985):

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_{K}/4810$$

(3)

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_{K}/4810$$

(4)

where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively. For the safe use of a material in the construction of dwellings, it is proposed that both $H_{in}$ and $H_{ex}$ should be less than

![Figure 1. The rock samples location (Haradh).](image-url)
unity to keep the radiation hazard insignificant (Beretka & Matthew, 1985).

The total air-absorbed dose rate (nGy/h) due to the activity concentrations of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$ (Bq/kg) can be calculated using the activity concentration (Bq/kg) of $^{40}\text{K} (A_k)$, $^{226}\text{Ra} (A_{Ra})$, and $^{232}\text{Th} (A_{Th})$ substituted into the formula (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000).

$$D = 0.0417A_k + 0.462A_{Ra} + 0.604A_{Th} \quad (5)$$

The annual effective dose equivalent (AEDE) was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv/Gy with an outdoor occupancy factor of 0.2 (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000).

$$\text{(AEDE)outdoor} = D(\text{nGy/h}) \times 8766(\text{h/year}) \times 0.7 \times (103\text{mSv/nGy} \times 109) \times 0.2 \quad (6)$$

3. Results and discussion

3.1. Natural radioactivity determination

The activity concentrations of natural radionuclides ($^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$) in the granite rocks of Haradh region in Saudi Arabia were presented in Table 1 and Figure 2. As shown in this table, the activity concentration of $^{226}\text{Ra}$ ranged from 2.07 to 53.79 Bq/kg, $^{232}\text{Th}$ ranged from 0.21 to 15.70 Bq/kg, and $^{40}\text{K}$ ranged from 4.11 to 2.06 Bq/kg. The corresponding average values were 16.71, 4.41, and 55.51 Bq/kg for $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$, respectively. These average values were lower than the international radioactivity levels of 50, 50, and 500 Bq/kg for $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$, respectively (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). It was found that the activity contributions of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ of the analyzed samples were 33.42%, 8.82%, and 11.10%, respectively, when comparing with the average granite rock concentrations as reported by UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation (2000). So, the activity concentrations of the radionuclide in the rock samples are quite uniform and do not show any significant variation.

The radiological hazard indices of radium equivalent activity ($\text{Ra}_{eq}$), absorbed dose rate ($D$), effective dose (AEDE) outdoor, internal hazard index ($H_i$), and external hazard index ($H_e$) were presented in Table 1. The radium equivalent activity ($\text{Ra}_{eq}$) calculated for granite rock in the area of study ranged from 0.3 to 54.98 Bq/kg with an average 25.38 Bq/kg which is lower the maximum value 370 Bq/kg of $\text{Ra}_{eq}$ for the material to be considered safe for use (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). The calculated $H_e$ in the rock samples ranged from 0.01 to 0.15 with an average value of 0.07. Similarly, for the $H_i$, the values ranged from 0.01 to 0.29 with an average of 0.11. These values of the indices hazard were below the recommended limit one, which does not cause any harm to workers in the studied area. The calculated absorbed dose rates of the samples were shown in Table 1 and Figure 3. Absorbed dose in rock samples ranged from 1.33 to 24.01 with an average value of 11.28 nGy/h. It is clear that absorbed dose rate values for all rock samples are lower than the international recommended limit of 60 nGy/h (UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). The corresponding average annual effective dose was calculated to be 0.014 mSv in a range of 0.002 to 0.029 mSv. The annual effective doses calculated from the various samples are considered insignificant in respect to the recommended limit of 1 mSv for the public. So, the results included that all the rock samples do not pose any significant source of radiation hazard and the use of the rock samples in the construction of dwellings is considered to be safe for inhabitants.

![Figure 2](image-url)  
**Figure 2.** Activity concentrations of radionuclides $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in granite rock samples from Harada region, Saudi Arabia.
A comparison between the average activity concentration results of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the present study of granite rocks and activity concentrations in different countries of the world were presented in Table 2. It was found that the current results are consistent with previous findings. The variation in the natural radioactivity levels is due to variations in concentrations of the radionuclides in the geological formations (Xinwei, Lingqing, & Xiaodan, 2006). In general, all results are existed within the range given in the UNSCEAR 2000.

### 3.2. XRD analytical results

XRD is a nondestructive analytical technique, which provides detailed information about the atomic
Table 3. The compound name of mineral constituents of granite rock samples analyzed by XRD spectrometer T.

| Sa. no. | Major | Minor | Trace |
|--------|-------|-------|-------|
| 1      | Labradorite | Diopside, Calcite | –     |
| 2      | XRD peaks are not distinct enough to determine the mineral constituents of the sample |
| 3      | XRD peaks are not distinct enough to determine the mineral constituents of the sample |
| 4      | XRD peaks are not distinct enough to determine the mineral constituents of the sample |
| 5      | XRD peaks are not distinct enough to determine the mineral constituents of the sample |
| 6      | XRD peaks are not distinct enough to determine the mineral constituents of the sample |
| 7      | Anorthite, Albite | Diopside | –     |
| 8      | XRD peaks are not distinct enough to determine the mineral components of the sample |
| 9      | Richterite, Albite | – | Dufrenite |
| 10     | Anorthite | Fluoropargasite | Biotite, Magnetite, Hurlbutite |

Table 4. The mineral composition of samples analyzed by XRD spectrometer.

| Sa. No. | MINERAL | CHEMICAL COMPOSITION       |
|---------|---------|----------------------------|
| 1       | Albite  | NaAlSi3O8                  |
| 2       | Anorthite | CaAl2Si2O6                |
| 3       | Biotite | K(MgFe3+2)4AlSi3O10(OH)2F2 |
| 4       | Calcite | CaCO3                      |
| 5       | Diopside | CaMgSi2O6                 |
| 6       | Dufrenite | Fe3+Fe5+2[PO4]3(OH)22H2O |
| 7       | Fluoropargasite | NaCa2(MgFe3+2AlSiAlO22)F2 |
| 8       | Hurlbutite | CaBe2[PO4]2   |
| 9       | Labradorite | NaCa2Si2AlO22O8 |
| 10      | Magnetite | Fe3+Fe5+O4               |
| 11      | Richterite | Na2CaMg2Fe3+2Si2O5(OH)2 |

structure of crystalline substances, chemical composition, and physical properties of materials. In the present study, the XRD results indicate that the major, minor, and trace constituents varied from one sample to another. Table 3 details the mineral content and description of 10 rock samples obtained by XRD. Table 3 details the mineral content and description of 10 rock samples obtained by XRD. In this table, the major elements are Labradorite in samp. 1, Anorthite, Albite in samp. 7, Richterite, Albite in samp. 9, and Anorthite in samp. 10; the minors are Diopside, Calcite in samp. 1, Diopside in 7, and Fluoropargasite in 10; and the trace elements are Dufrenite in samp. 9 and Biotite, Magnetite, and Hurlbutite in samp. 10.

XRD peaks in samples 2, 3, 4, 5, 6, and 8 are not distinct enough to determine the mineral constituents. Table 4 is a review of the chemical composition of each mineral, which shows the formation of the samples (Leet, Judson, & Kauffman, 1982).

4. Conclusion

The paper deals with the study of natural radioactivity of 238U, 232Th, and 40K and their possible radiological effects in the granite rock of an important region in Saudi Arabia (Haradh). The samples were analyzed by HPGe-based gamma-ray spectrometer. The results indicate that all the concentration values of 226Ra, 232Th, and 40K are below the world average values and are comparable to similar studies carried out previously. The average value of radium equivalent (Ra eq) for rock samples in the studied area is lower than the internationally accepted value 370 Bq/kg. The present average absorbed dose rate in the air outdoors is consistent with the results reported in UNSCEAR 2000. The internal and external radiation hazard index for all the samples is lower than unity. Therefore, the radiological impact in granite rocks of the selected area is found to be within safety limits; the rock samples do not cause any hazard to workers as well as members of the public and can be safely used as construction material. Generally, the level risk is considered insignificant. Results of the analysis by XRD indicate that the main major, minor, and trace constituents varied from one sample to another.

Disclosure statement

No potential conflict of interest was reported by the author.

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