Concentrations of $^{214}$Pb, $^{214}$Bi in $^{238}$U series and $^{208}$Tl, $^{228}$Ac in $^{232}$Th series in granite rock in (Kadugli) Sudan

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A survey study on natural radioactivity of $^{214}$Pb, $^{214}$Bi in $^{238}$U series and $^{208}$Tl, $^{228}$Ac in $^{232}$Th series in granite rock collected from the state of southern Kordofan (Kadugli), Sudan has been carried out by using γ-ray spectroscopy technique. The gamma-ray spectra have been measured for daughter decaying of $^{238}$U and $^{232}$Th series. Elemental and activity concentrations were found to be 1.41±0.02 ppm and 17.41±0.19 Bqkg$^{-1}$ for $^{214}$Pb, 1.66±0.02 ppm and 25.60±0.21 Bqkg$^{-1}$ for $^{214}$Bi and 9.56±0.11 ppm and 38.81±0.43 Bqkg$^{-1}$ for $^{208}$Tl, 8.55±0.06 ppm and 34.71±0.23 Bqkg$^{-1}$ for $^{228}$Ac, respectively. Moreover, average activity concentrations of these radio-nuclides for granite samples have been compared with the typical values of different countries.

Keywords: Radio-nuclides, Concentrations, Uranium, Thorium series, Granite rock, Kadugli

1 Introduction

The radioactive decay process of $^{238}$U series is complex, passing through fourteen steps, each with characteristic disintegration and daughter products before reaching the final stable end product $^{206}$Pb. It is most important to realize that the principle gamma emission is associated with $^{214}$Pb and $^{214}$Bi and not directly with $^{238}$U. It should be noted that one of the steps in the decay chain preceding the formation of $^{214}$Pb and $^{214}$Bi is radon gas ($^{222}$Rn), a noble gas which represents a major source of $^{238}$U disequilibrium because it has a half-life of 3.8 days and can migrate substantial distances from its source. It is very important to seal samples in air-tight containers for a period of 3–4 weeks to allow time to reach equilibrium in the $^{238}$U series$^{2,3}$. The radioactive nucleus $^{232}$Th, like $^{238}$U, undergoes a complex decay process before reaching $^{206}$Pb. Most of the gamma rays originate from decaying $^{228}$Ac, $^{208}$Tl and $^{212}$Pb. Uranium occurs as a trace element in the earth’s crust and is typically present in concentrations of 1 to 10 ppm in granite and in elatic sediments of granitic origin, while thorium is typically present in concentrations ranging between 3 and 30 ppm in crustal minerals. Most thorium is found in sediments$^{4,5}$. $^{238}$U and $^{232}$Th and their decay products represent the main external source of irradiation to the human body$^{4,6}$. Specific levels are due to naturally occurring radionuclides are related to the types of the rock from which the soils originate. Therefore, natural environmental radiation depends mainly on the geological and geographical composition of each lithological separated area$^{5,7}$. Granite is widely distributed throughout the continental crust and is the most abundant plutonic rock in mountain belts. These rocks occur in great batholiths that may occupy thousands of square kilometres and are usually closely associated with quartz monzonite, granodiorite, diorite and gabbro. They consist mainly of coarse grains of quartz, potassium and sodium feldspars. Other common minerals that contain granite include mica and hornblende. Typical granite is chemically composed of 75% silica, 12% aluminium, < 5% potassium oxide, < 5% soda, as well as lime, iron, magnesia and small quantities of tatania. Originally, it was widely believed that granites were formed mainly from magmatic differentiation of basaltic magma, evidence that was considered to indicate a metamorphic origin. However, because of the large quantities of granites that occur in nature, geologists believe now that most granites have been formed either by melting, partial melting or metamorphism of deeply buried shale and sandstone. Granites, therefore, are the result of rapidly injected coalescing sheets of magma, each of which cooled independently of the other sheets. Evidence of intrusion or great mobility indicates an igneous origin.
that stems from the melting of sediments and, consequently, granite dykes are clearly igneous\(^7\).

In terms of natural radioactivity, granites exhibit an enhanced elemental concentration of uranium (U) and thorium (Th) compared with the very low abundance of these elements observed in the mantle and the oceanic crust of the Earth. Geologists provide an explanation of this behaviour in the course of partial melting and fractional crystallization of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into more silica-rich products\(^7\). For that reason, igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5 ppm of U and 15 ppm of Th), compared with the Earth’s crust (average 1.8 ppm for U and 7.2 ppm for Th), the upper continental crust (average 2.7 ppm for U and 10.5 ppm for Th) and rocks of basaltic or ultramafic composition\(^8\) (0.1 ppm of U and 0.2 ppm of Th). Higher radiation levels are usually associated with igneous rocks such as granite and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have a relatively high content of radionuclides\(^3,4,9\).

The assessment of gamma radiation from natural sources is particularly important as natural radiation is the largest contributor to the external dose absorbed by the world’s population. To determine the concentration of radionuclide’s in granite rock samples, a study was carried out using gamma ray spectroscopy, which is very important as a component of environmental health surveillance program projects to develop a subject of interest in the environmental sciences.

2 Experimental Details

2.1 Sample collection and preparation

Four granite samples, each of 1.65 kg mass were collected from Kadugli, a city in the southern state of Kordofan, Sudan. Granite stones were crushed to fine powder form and sieved to a particle size less than 120 μm. To remove moisture, the sample\(^10,11\) was dried at 110°C for 24 h, and then carefully weighed using an electronic balance with a sensitivity of 0.01 mg. The powdered samples were sealed in standard Marinelli beakers. After properly tightening the lid, the containers were set aside for 28 days in order to ensure radioactive equilibrium\(^11-14\). After 28 days, the samples were analyzed using \(\gamma\)-ray spectrometer.

2.2 Standard preparation

Standard reference material used as a basis to calculate the concentration of \(^{238}\)U and \(^{232}\)Th and their daughters decaying in the granite rock was provided by the International Atomic Energy Agency (IAEA). Samples of pure quartz (silicon dioxide, \(\text{SiO}_2\)) were used for background measurements\(^3\). Standard samples were prepared by mixing 15 g of IAEA-313 with 400 g of \(\text{SiO}_2\) and sealing the mixture in Marinelli beakers for 28 days in order to reach secular equilibrium.

2.3 Experimental method for \(\gamma\)-spectroscopy

Experiments were carried out using the gamma ray spectroscopy facilities at the Nuclear Laboratory, Physics Department, Universiti Teknologi, Malaysia. A gamma ray spectroscopy system consists of a high purity germanium (HPGe) detector with a counting efficiency of 20% which is connected to an 8192 channel computerized multi-channel analyzer (MCA). The detector used in these measurements was a Canberra GC2018 with Genie 2000 software, \(p\)-type HPGe detectors of 2”\(\times\)2” size and an energy resolution of (FWHM) 1.8 keV at 1.3 MeV \(\gamma\)-energy of Co-60. The detector and pre-amplifier were placed inside a lead shield of 12 cm thickness to reduce background scattering and the detector was cooled by liquid nitrogen. The integrated signal processor consists of a pulse height analysis system to transform pulses, which are finally collected by a computer based MCA. Energy calibration was performed using six standard gamma point sources \(^{214}\)Am (59.5 keV), \(^{57}\)Co (122.1 and 136.5 keV), \(^{133}\)Ba (302.9 and 356.0 keV), \(^{152}\)Eu (344.3 keV), \(^{137}\)Cs (661.7 keV), and \(^{60}\)Co (1173 and 1333 keV).

The concentrations of \(^{238}\)U and \(^{232}\)Th have been calculated from their progeny photopeaks. The gamma background level at the counting room was determined with an empty Marinelli beaker. The background was measured under the same conditions as the measurement of the samples. Finally, the background was subtracted from the area of each spectrum of sample. Similar measurements were done for standard samples, which have certified values of U and Th measured in parts per million (ppm). However, based on their percentage of gamma disintegration, only four clear intense energy peaks were selected for analysis in this work which are: \(^{214}\)Pb (352 keV and gamma yield of \(~35\%)\), \(^{214}\)Bi (609.4 keV, \(~43\%)\), \(^{208}\)Tl (583.1 keV, \(~30\%)\) and \(^{228}\)Ac (911.1 keV, \(~29\%)\). Each sample was measured thrice before an average value was taken.
2.4 Calculation of elemental concentrations

The current concentration of uranium and thorium is given by the expression:

\[ C_{\text{samp}} = \frac{W_{\text{std}}}{W_{\text{samp}}} \times \frac{N_{\text{samp}}}{N_{\text{std}}} \times C_{\text{std}} \]  

where \( C_{\text{samp}} \) is the concentration of the sample collected (ppm), \( W_{\text{std}} \) the weight of the standard sample (g), \( W_{\text{samp}} \) the weight of the sample collected (g), \( N_{\text{samp}} \) the net counts of the photopeak area for the sample collected, \( N_{\text{std}} \) the net counts of the photopeak area for the standard sample and \( C_{\text{std}} \) is the concentration of the standard (ppm).

The uncertainty of the sample concentration could be calculated by using the following formula:

\[ \Delta C_{\text{samp}} \text{(ppm)} = \left( \frac{\Delta W_{\text{std}}}{W_{\text{std}}} + \frac{\Delta W_{\text{samp}}}{W_{\text{samp}}} + \frac{\Delta N_{\text{samp}}}{N_{\text{samp}}} + \frac{\Delta N_{\text{std}}}{N_{\text{std}}} \right) \times C_{\text{samp}} \]  

\( \Delta C_{\text{samp}} \) is the uncertainty of the sample concentration (ppm), \( \Delta W_{\text{std}} \) the uncertainty of the weight of the standard sample (g), \( \Delta W_{\text{samp}} \) the uncertainty of the weight of the sample collected (g), \( \Delta N_{\text{samp}} \) the uncertainty of the net counts of the photopeak area for the sample collected, \( \Delta N_{\text{std}} \) is the uncertainty of the net counts of the photopeak area for the standard sample.

The results are obtained in units of ppm for elemental concentrations, which are converted into corresponding activity concentrations according to the convention factors given by the International Atomic Energy Agency: The activity concentration of a sample containing 1 ppm by weight of \( ^{238}\text{U} \) is 12.35 (Bqkg\(^{-1}\)) and 1 ppm of \( ^{232}\text{Th} \) is 4.06 (Bqkg\(^{-1}\)).

3 Results and Discussion

Figure 1 shows the single gamma-ray spectra of granite sample measured by Ge detector. The gamma energies of 351.9 keV from \( ^{214}\text{Pb} \) and 609.4 keV from \( ^{214}\text{Bi} \) firmly identified the uranium daughter series. The gamma-ray peaks 583.1 keV from \( ^{208}\text{Tl} \) and 911.4 keV from \( ^{228}\text{Ac} \) have been identified as the daughter of thorium series. The population yields of \( \gamma \)-rays for selected nuclei have been estimated from single gamma ray data by correcting the efficiency of the detector. The yields of \( ^{214}\text{Pb}, ^{214}\text{Bi}, ^{208}\text{Tl} \) and \( ^{228}\text{Ac} \) were found using the \( \gamma \)-yield by setting the area of interest in the energy spectrum.

The elemental and activity concentration for granite stone under this study are presented in Table 1. The average value of elemental and activity concentration for selected nuclei are indicated in brackets: \( ^{214}\text{Pb} \) (1.41±0.02 ppm and 17.41±0.19 Bqkg\(^{-1}\)), \( ^{214}\text{Bi} \) (1.66±0.02 ppm and 25.60±0.21 Bqkg\(^{-1}\)), \( ^{208}\text{Tl} \) (9.56±0.11 ppm and 38.81±0.43 Bqkg\(^{-1}\)), and \( ^{228}\text{Ac} \) (8.55±0.06 ppm and 34.71±0.23 Bqkg\(^{-1}\)). Consequently, we have estimated the average concentration and activity of uranium and thorium as 1.54±0.02 ppm, 21.51±0.20 Bqkg\(^{-1}\) and 9.06±0.09 ppm, 36.76±0.33 Bqkg\(^{-1}\), respectively.

Moreover, we carried out a comparison of the activity concentrations of uranium and thorium in the present study of granite stone with previous studies and the results are presented in Table 2. It was found that the maximum activity value 78.78±2.34 Bqkg\(^{-1}\) of \( ^{238}\text{U} \) was in Egypt which is larger than the present work by a factor of 2.3. The maximum value 83.19±1.1 Bqkg\(^{-1}\) of \( ^{232}\text{Th} \) was found in Turkey which is greater than the present work by a factor of 2.3. The results obtained in the present work are consistent with previous results. The values of \( ^{238}\text{U} \) and \( ^{232}\text{Th} \) activities are distinctly lower than the corresponding
ones obtained from other countries and, in general, all results existed within the range given in UNSCEAR report\(^\text{15}\). According to the geological composition of the southern Kordofan, more systematic environmental monitoring studies are required, as preliminary studies in the area show radioactivity levels 10 times higher than that of more normal areas\(^\text{16}\). The present study of the concentration of \(^{238}\text{U}\) and \(^{232}\text{Th}\) in granite suggests an investigation of the concentrations of natural radio-nuclides \(^{40}\text{K}\) in these regions which would provide valuable and detailed information concerning the external hazard value.

### 4 Conclusions

We have carried out this experiment to provide information about the concentration of radionuclides in granite rock in Kadugli, Sudan by using gamma ray spectrometer. The activity of thorium is 36.76±0.33 Bq kg\(^{-1}\), and the activity of uranium in granites is 21.51±0.20 Bq kg\(^{-1}\). These values are found to be within the range of the countries in the world reported in the literature\(^\text{7,11,14,15,17–20}\). In general, all reported results are within the range as given in the UNSCEAR report of 1993.

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### References


### Table 2 — Average activity concentration (Bq kg\(^{-1}\)) for granite sample from work conducted worldwide

<table>
<thead>
<tr>
<th>Country</th>
<th>Area of study</th>
<th>(^{238}\text{U})</th>
<th>(^{232}\text{Th})</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Egypt</td>
<td>East desert</td>
<td>78.78±2.34</td>
<td>44.48±1.43</td>
<td>17</td>
</tr>
<tr>
<td>Egypt</td>
<td>Bir El-sid</td>
<td>57.4±4.5</td>
<td>53.4±5.4</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>Wadi El-Gemal</td>
<td>39±3.2</td>
<td>47.9±5.1</td>
<td>14</td>
</tr>
<tr>
<td>Egypt</td>
<td>Aswan to Wadi El-Alaqi</td>
<td>14.94±0.22</td>
<td>15.50±0.36</td>
<td>7</td>
</tr>
<tr>
<td>Saudi Arabi</td>
<td>Riyadh</td>
<td>18.7±0.08</td>
<td>11.5±0.13</td>
<td>18</td>
</tr>
<tr>
<td>Turkey</td>
<td>—</td>
<td>69.74±0.83</td>
<td>83.19±1.1</td>
<td>19</td>
</tr>
<tr>
<td>Iran</td>
<td>Tehran</td>
<td>74.08±3.5*</td>
<td>69.31±3.7</td>
<td>11</td>
</tr>
<tr>
<td>Sudan</td>
<td>Kadugli</td>
<td>21.51±0.20</td>
<td>36.78±0.33</td>
<td>Present Study</td>
</tr>
<tr>
<td>Worldwide</td>
<td>—</td>
<td>20-120*</td>
<td>20-80</td>
<td>15</td>
</tr>
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</table>

*Taken from \(^{226}\text{Ra}\)