Measurement of natural radioactivity in brick samples used in the construction in Iraq

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ABSTRACT
The activity concentrations of the natural radionuclides, namely, ²²⁶Ra, ²³²Th and ⁴⁰K are measured for eleven brick samples collected from some locations of Iraq. An HPGGe detector, based on high-resolution gamma spectrometry system is used for the measurement of activity concentrations. The average activity concentration values of ²²⁶Ra, ²³²Th and ⁴⁰K from the studied are 26.34 Bq/kg, 26.97 Bq/kg and 530.24 Bq/kg respectively. In order to evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity, absorbed dose rate, annual effective dose rate, internal and external hazard indices, gamma index, alpha index, excess lifetime cancer risk and annual gonadal dose equivalent have been calculated.

Keywords: Natural radioactivity, Brick samples, HPGe detector, Radiological hazards, IRAQ

INTRODUCTION
An established fact that all the construction material contains trace amount of the natural radioactivity. This activity is a major source of external and internal radiation exposure to the occupant of the dwelling. The most commonly encountered radionuclides in the construction materials are ²³⁸U, ²³²Th their decay products and ⁴⁰K. Radon isotopes are amongst the members of radioactive series of Uranium and Thorium. The internal exposure due to radon and its radioactive daughters present in the environment, result in the largest contribution to the average effective dose received by human beings [1]. All building materials such as concrete, brick, sand, aggregate, marble, granite, limestone, gypsum, etc., contain mainly natural radionuclides including ²³⁸U, ²³²Th their decay products and ⁴⁰K [2,3]. Bricks are used as one of the main building materials, so the knowledge of the basic radiological parameters and radioactive contents in the bricks and other construction materials is important since it allows us to calculate the exposure of the population of the radiation from natural sources [4]. The aim of this investigation is to estimate the radiological effect to the human being which result from some type of bricks used in Iraq by determine the activity concentration of the ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides', and some of the radiological parameters as the radium equivalent activity, gamma absorbed dose rate, external and internal hazard indices, and other parameters.

MATERIALS AND METHODS
2.1 Sampling and Sample preparation:
Eleven type of brick samples are collected from different districts of Iraq: (Baghdad factory)(S1), (Al-Taji district)(S2), (Diayla province) (S3), (Erbil factory)(S4), (Kirkuk province)(S5), (Dhululya district )(S6), (Yellow Iranian 1)(S7), (Suylamina province)(S8), (Yellow Iranian 2) (S9), (Karbala province)(S10) and (Red Iranian)(S11). The samples dried by placing it in oven of 110°C about 24 h, then crushed to pass through 2 mm sieve to be homogenized in size. The homogenized brick samples were sealed in plastic containers and left for at least one
month, before gamma spectrometric analysis, to attain secular equilibrium between radon and its decay products[3].

2.2 Radioactivity measurement:
The concentration of the natural radioactivity ($^{226}$Ra, $^{232}$Th and $^{40}$K) in the brick samples were determined using a high-resolution high purity germanium (HPGe) - spectroscopy system with 40% counting efficiency, and resolution (1.75 keV), normally based on the measurement of 1.332 MeV gamma ray photo peak of Co-60 source and Cooled with liquid nitrogen. A Multichannel analyzer (MCA) with 4096 channel was used. A detector shield had a cavity adequate to accommodate large samples. The Shield had a lead walls with 10 cm thickness lined inside with graded absorber of Cd ~ 1.6 mm Cu ~ 0.4 mm. Calibration and efficiency of the system were carried out using multi-gamma ray standard source (MGS-5, Canberra) of Marinelli beaker geometry. A library of radionuclide's which contained the energy of the characteristic gamma emissions of each nuclide was analyzed and their corresponding emission probabilities were built from the data supplied in the software.

In order to determine the background distribution due to naturally occurring radionuclide's in the environment around the detector, an empty Marinelli beaker container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentrations were calculated.

The activity concentrations for the natural radionuclide's in the samples were computed using the following relation [5,6].

$$A = \frac{(cps)_{net}}{I \times \epsilon \times M} \quad (1)$$

where $A$ is the activity concentration in Bq/kg, (cps) net is the (count per second) and equal { (cps)sample - (cps) background}, $I$ is the intensity of the $\gamma$-line in a radionuclide, $\epsilon$ is the measured efficiency for each $\gamma$-line observed and $M$ is the mass of the sample in gram.

### Estimation of $^{226}$Ra and $^{232}$Th and $^{40}$K:

$^{226}$Ra concentration was determined from $\gamma$-peak of 1765keV from $^{214}$Bi, $^{232}$Th concentration was determined from $\gamma$-peak of 911keV from $^{228}$Ac and $^{40}$K concentration was measured from 1460 keV $\gamma$–line.

### RESULTS AND DISCUSSION

3.1 Activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K:
The activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K measured in brick samples are shown in table 1. As seen from table 1, the activity concentration of $^{226}$Ra ranged from 15.02Bq/kg (S4) to 41.29 Bq/kg (S5), with an average value of 26.34Bq/kg, while the activity concentration of $^{232}$Th ranged from 15.63Bq/kg (S4) to 37.62Bq/kg (S11), with an average value of 26.97Bq/kg and finally the activity concentration of $^{40}$K ranged from 293.03Bq/kg (S2) to 786.71Bq/kg (S11), with an average value of 530.24 Bq/kg. The activity concentration of $^{226}$Ra, $^{232}$Th and $^{40}$K are illustrated in figs.(1) and (2).

![Fig.1: The activity concentration of $^{226}$Ra and $^{232}$Th in brick samples in different locations of Iraq](image-url)
Table(1): The activity concentration of $^{226}$Ra, $^{232}$Th, and $^{40}$K in Brick samples in different locations of Iraq

<table>
<thead>
<tr>
<th>Samples codes</th>
<th>Activity concentration (Bq/kg)</th>
<th>Samples weight (gm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{226}$Ra</td>
<td>$^{232}$Th</td>
</tr>
<tr>
<td>S1</td>
<td>21.14</td>
<td>23.28</td>
</tr>
<tr>
<td>S2</td>
<td>21.95</td>
<td>26.14</td>
</tr>
<tr>
<td>S3</td>
<td>22.72</td>
<td>23.66</td>
</tr>
<tr>
<td>S4</td>
<td>15.02</td>
<td>15.63</td>
</tr>
<tr>
<td>S5</td>
<td>41.29</td>
<td>35.13</td>
</tr>
<tr>
<td>S6</td>
<td>25.94</td>
<td>17.41</td>
</tr>
<tr>
<td>S7</td>
<td>33.21</td>
<td>34.61</td>
</tr>
<tr>
<td>S8</td>
<td>38.46</td>
<td>36.56</td>
</tr>
<tr>
<td>S9</td>
<td>24.28</td>
<td>26.78</td>
</tr>
<tr>
<td>S10</td>
<td>22.53</td>
<td>19.94</td>
</tr>
<tr>
<td>S11</td>
<td>23.20</td>
<td>37.62</td>
</tr>
<tr>
<td>Average±S.D.</td>
<td>26.34±7.96</td>
<td>26.97±7.96</td>
</tr>
</tbody>
</table>

3.2 Radiological parameters:

3.2.1 Radium equivalent activities (Ra$_{eq}$)

To represent the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K by a single quantity, which takes into account the radiation hazards associated with them, a common radiological index has been introduced. The index is called radium equivalent activity (Ra$_{eq}$) which is used to ensure the uniformity in the distribution of natural radionuclides $^{226}$Ra, $^{232}$Th, and $^{40}$K and is given by the expression [7]:

$$ Ra_{eq} (\text{Bq/kg}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (2) $$

Where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the specific activities concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K in (Bq/kg) respectively.

As we seen from table(2), the calculated value of (Ra$_{eq}$) varied from 81.89 Bq/kg (S2) to 143.78 Bq/kg (S5), with an average value of 105.74 Bq/kg. These values are less than 370 Bq/kg, which are acceptable for safe use [8].

3.2.2 External hazard index (H$_{ex}$)

To limit the external gamma-radiation dose from building materials, an extensively used hazard index, the external hazard index (H$_{ex}$) was calculated from the equation [9].

$$ H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_{K}/4810 \leq 1 \quad (3) $$

The calculated values of H$_{ex}$ are ranged from 0.221 (S2) to 0.388 (S5) with an average of 0.285 as in table 2, where all values of H$_{ex}$ are less than the critical values of unity [8].

3.2.3 Internal hazard index (H$_{in}$)

Radon and its short-lived products are also hazardous to the respiratory organs. So internal exposure to radon and its short-lived products is quantified by internal hazard index and is expressed mathematically [10].
H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \leq 1 \tag{4}

The calculated values of \( H_{in} \) are ranged from 0.261 (S4) to 0.499 (S5), with an average of 0.356 as in table 2, which is less than the critical values of unity \([8]\).

### 3.2.4 Gamma Index (I_{\gamma})

The gamma index (I_{\gamma}) for soil samples was calculated by using the following equation \([11]\).

\[
I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500} \leq 1 \tag{5}
\]

I_{\gamma} varies from 0.603 (S2) to 1.079 (S5) with an average of 0.798. The types of bricks which the I_{\gamma} values was higher than the critical values of unity are (S5, S8 and S11), as listed in table 2.

### 3.2.5 Alpha Index (I_{\alpha})

Also several indexes dealing with the assessment of the excess alpha radiation due to the radon inhalation originating from building materials (brick) "called alpha indexes or internal indexes" have been developed \([5]\). In the present work, alpha indexes were determined through the following formula \([12]\):

\[
I_{\alpha} = \frac{A_{Ra}}{200 \text{Bq/kg}} \leq 1 \tag{6}
\]

I_{\alpha} varies from 0.075 (S4) to 0.206 (S5) with an average of 0.131 (table 2), which is less than the critical values of unity \([8]\).

The values of \( H_{ex}, H_{in}, I_{\gamma} \) and I_{\alpha} are shown in fig.3.

### 3.2.6 Absorbed Gamma Dose Rate (D)

Outdoor air gamma absorbed dose rate (D) in (nGy/h) due to terrestrial gamma rays at (1m) above the ground surface which can be computed from specific activities \( A_{Ra}, A_{Th} \) and \( A_{K} \) of \(^{238}\text{U}\), \(^{232}\text{Th} \) and \(^{40}\text{K} \) in (Bq/kg) respectively using the following relation \([13]\):

\[
D (\text{nGy/h}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \tag{7}
\]

The absorbed dose in the brick samples ranges from 38.14 nGy/h (S2) to 68.59 nGy/h (S5) with an average of 50.75 nGy/h, as presented in table 3.

### Table (2): Radium equivalent activity, external and internal hazard indices, gamma index and alpha index for brick samples in different locations of Iraq

<table>
<thead>
<tr>
<th>Samples codes</th>
<th>( R_{eq} ) (Bq/kg)</th>
<th>( H_{ex} )</th>
<th>( H_{in} )</th>
<th>I_{\gamma}</th>
<th>I_{\alpha}</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>82.61</td>
<td>0.223</td>
<td>0.280</td>
<td>0.617</td>
<td>0.105</td>
</tr>
<tr>
<td>S2</td>
<td>81.89</td>
<td>0.221</td>
<td>0.280</td>
<td>0.603</td>
<td>0.109</td>
</tr>
<tr>
<td>S3</td>
<td>91.63</td>
<td>0.247</td>
<td>0.308</td>
<td>0.691</td>
<td>0.113</td>
</tr>
<tr>
<td>S4</td>
<td>81.95</td>
<td>0.221</td>
<td>0.261</td>
<td>0.642</td>
<td>0.075</td>
</tr>
<tr>
<td>S5</td>
<td>143.78</td>
<td>0.388</td>
<td>0.499</td>
<td>1.079</td>
<td>0.206</td>
</tr>
<tr>
<td>S6</td>
<td>97.40</td>
<td>0.263</td>
<td>0.333</td>
<td>0.750</td>
<td>0.129</td>
</tr>
<tr>
<td>S7</td>
<td>130.48</td>
<td>0.352</td>
<td>0.442</td>
<td>0.981</td>
<td>0.166</td>
</tr>
<tr>
<td>S8</td>
<td>136.72</td>
<td>0.369</td>
<td>0.473</td>
<td>1.020</td>
<td>0.192</td>
</tr>
<tr>
<td>S9</td>
<td>96.99</td>
<td>0.261</td>
<td>0.327</td>
<td>0.727</td>
<td>0.121</td>
</tr>
<tr>
<td>S10</td>
<td>82.17</td>
<td>0.221</td>
<td>0.282</td>
<td>0.619</td>
<td>0.112</td>
</tr>
<tr>
<td>S11</td>
<td>137.57</td>
<td>0.371</td>
<td>0.434</td>
<td>1.055</td>
<td>0.116</td>
</tr>
</tbody>
</table>
| Average± S.D  | 105.74                 | 0.285       | 0.356±      | 0.798±    | 0.131±    | 0.039±
3.2.7 Annual Effective Dose Equivalent (AEDE)

The estimated annual effective dose equivalent received by a member was calculated by using a conversion factor of (0.7 SvyGy), which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [14]:

\[
(AEDE)_{out} = D_{y}(nGy/h) \times 10^{-3} \times 8760 \text{ h/y} \times 0.20 \times 0.7 \text{Sv/Gy} \tag{8}
\]

\[
(AEDE)_{in} = D_{y}(nGy/h) \times 10^{-3} \times 8760 \text{ h/y} \times 0.80 \times 0.7 \text{Sv/Gy} \tag{9}
\]

(AEDE)_{out} varies from 46.78 \mu Sv/y (S2) to 84.12 \mu Sv/y (S5), with an average of 62.02 \mu Sv/y, the present results are given in table 3 the values of (AEDE)_{out} were lower than the value of the outdoor annual effective dose equivalent global limit which is equal to 460 \mu Sv/y [8].

(AEDE)_{in} varies from 187.14 \mu Sv/y (S2) to 324.97 \mu Sv/y (S11) with an average value of 248.10 \mu Sv/y, as mentioned in table 3, which is lower than the value of the indoor annual effective dose equivalent global limit which is equal to 460 \mu Sv/y [8].

3.2.8 Excess Lifetime Cancer Risk (ELCR)

This gives the probability of developing cancer over a lifetime at a given exposure level. It is presented as a value representing the number of extra cancers expected in a given number of people on exposure to a carcinogen at a given dose, and we can calculate (ELCR) by eq. (10) if considering 70 years as the average duration of life for human being [15].

\[
ELCR = AEDE \times DL \times RF \tag{10}
\]

Where AEDE is the Annual Effective Dose Equivalent, DL is the average Duration of Life (estimated to be 70 years) and RF is the risk factor (Sv^{-1}), fatal cancer risk per Sievert. For low dose background radiations which are considered to produce stochastic effects, ICRP 60 uses values of 0.05 for the public exposure [15]. This value-free units because it represents the probability of cancer incidence through this we can deduce the equation above.

From table 3, the (ELCR) values varied from 133.52 x 10^{-6} (S2) to 240.08 x 10^{-6} (S5), with an average of 177.01 x 10^{-6}, for all samples is less than the world average of 290 x 10^{-6} [15].

3.2.9 Annual Gonadal Dose Equivalent (AGDE)

The annual gonadal dose equivalent (AGDE) in mSv /y due to the activity concentrations of ^{226}\text{Ra}, ^{232}\text{Th}, and ^{40}\text{K} was calculated using the following formula [16]:

\[
AGDE(\text{mSv}/\text{y}) = (3.09A_{Ra} + 4.19A_{Th} + 0.314A_{K})/1000 \tag{11}
\]

As seen in table 3, the AGDE values of brick samples varied between 0.269 mSv/y (S2) and 0.487 mSv/y with an average value of 0.360 mSv/y. This result is slightly higher than the permissible safety limit of 0.3 mSv/y [17].
The values of \( I_\alpha, \text{AGDE} \), \{\text{AEDE}_{\text{out}} & \text{AEDE}_{\text{in}} \} \) and \( \text{Ra}_{eq}, D \text{ ELCR} \) are shown in figs.4,5 and 6 respectively.

Table (3): Absorbed Gamma Dose Rate, Annual Effective dose equivalent (outdoor & indoor), Excess Lifetime Cancer Risk and Annual Gonadal Dose Equivalent for brick samples in different regions of Iraq

<table>
<thead>
<tr>
<th>Samples codes</th>
<th>( D_{\text{nGy/h}} )</th>
<th>\text{AEDE}_{\text{out}}\mu\text{Sv/y}</th>
<th>\text{AEDE}_{\text{in}}\mu\text{Sv/y}</th>
<th>\text{ELC}\times10^{-6}</th>
<th>\text{AGDE}\text{mSv/y}</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>39.09</td>
<td>47.94</td>
<td>191.76</td>
<td>136.81</td>
<td>0.277</td>
</tr>
<tr>
<td>S2</td>
<td>38.14</td>
<td>46.78</td>
<td>187.14</td>
<td>133.52</td>
<td>0.269</td>
</tr>
<tr>
<td>S3</td>
<td>43.78</td>
<td>53.70</td>
<td>214.80</td>
<td>153.25</td>
<td>0.312</td>
</tr>
<tr>
<td>S4</td>
<td>40.52</td>
<td>49.69</td>
<td>198.78</td>
<td>141.82</td>
<td>0.293</td>
</tr>
<tr>
<td>S5</td>
<td>68.59</td>
<td>84.12</td>
<td>236.50</td>
<td>240.08</td>
<td>0.487</td>
</tr>
<tr>
<td>S6</td>
<td>47.71</td>
<td>58.52</td>
<td>234.08</td>
<td>167.01</td>
<td>0.343</td>
</tr>
<tr>
<td>S7</td>
<td>62.12</td>
<td>76.18</td>
<td>304.74</td>
<td>217.42</td>
<td>0.442</td>
</tr>
<tr>
<td>S8</td>
<td>64.75</td>
<td>79.41</td>
<td>317.65</td>
<td>226.63</td>
<td>0.459</td>
</tr>
<tr>
<td>S9</td>
<td>46.03</td>
<td>56.45</td>
<td>225.81</td>
<td>161.10</td>
<td>0.327</td>
</tr>
<tr>
<td>S10</td>
<td>39.30</td>
<td>48.20</td>
<td>192.83</td>
<td>137.58</td>
<td>0.280</td>
</tr>
<tr>
<td>S11</td>
<td>66.24</td>
<td>81.24</td>
<td>324.97</td>
<td>231.86</td>
<td>0.476</td>
</tr>
</tbody>
</table>

Average\(\pm\) S.D

\(\text{AEDE}_{\text{out}}\)\(\text{AEDE}_{\text{in}}\)

Fig. 4: Annual Gonadal Dose Equivalent and alpha index for brick samples in different locations of Iraq

Fig. 5: Annual Effective dose equivalent (outdoor & indoor) for brick samples in different locations of Iraq
CONCLUSION

The activity concentration of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ measured in commonly used building materials (brick samples) used for construction purpose in Iraq have been determined by high purity germanium detector. For each sample in this study, the activity concentration, radiation equivalent activity, external and internal hazard indices, gamma index, alpha index, Annual effective dose equivalent (outdoor and indoor), Excess lifetime cancer risk and Annual gonadal dose equivalent have been determined to assess the radiological hazards from brick samples.

The values obtained in the study are within the recommended safety limit, showing that the brick samples do not pose any significant radiation hazard and hence the use of these brick samples in the construction of dwelling is considered to be safe for the inhabitants. This study can be used as a reference for more extensive studies of the same subject in future.

REFERENCES


