Evaluation of Natural Radioactivity and its Radiation Hazards in Some Building and Decorative Materials in Iraq

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ABSTRACT

Twenty-nine samples of different types of building materials such as bricks, cement, ceramic, granite, gravel, marble, paint, plaster, sand and soil were analyzed by a gamma spectrometer based on HPGe detector. The total average contents of $^{226}$Ra, $^{232}$Th, and $^{40}$K for all the building samples are 35.26 Bq kg$^{-1}$, 26.99 Bq kg$^{-1}$, and 321.19 Bq kg$^{-1}$ respectively. The estimated radium equivalent activities ($Ra_{eq}$), representative index ($I_{γ}$), absorbed $γ$-dose rate, external and internal hazard indices and the annual effective dose equivalent (AEDE) are lower than the recommended safe limit 1 mSv$^{-1}$. The results indicated that all the building materials in Iraq are safe except the materials that used as decorative materials that have to be regulated.

Keywords: Water/Spectrometry/Gamma/Building/Materials/IRAQ

INTRODUCTION

The world is naturally radioactive, and around 90% of human radiation exposure arises from natural sources such as cosmic radiation, exposure to radon gas and terrestrial radiation. Reviewing the published literature shows that some studies have determined the levels of exposure due to natural radiation in Iraq¹,². Recently, effects from NORM in building materials and its relation with the building construction were studied by Eltahe et al., 2010³, Kamal; 2012⁴, Gharbi; 2012⁵, Amran et al., 2013⁶, Sharaf & Hamideen; 2013⁷ and Xinwei L et al., 2014⁸.

Hussain et al., 2010⁹ evaluated the natural radioactivity of some local building materials and their associated absorbed doses from some building materials used in the middle Euphrates of Iraq such as cement, bricks, blocks, and sand which contain low concentrations of NORM. The existence of radioactivity in building materials increases the internal and external absorbed dose to the residence. The analysis of building materials for the concentrations of either natural or man-made radioactivity give good information about the internal as well as the external dose to the inhabitance. The source of the increase of the absorbed dose was found to be mostly of $^{222}$Rn or $^{220}$Rn which are members of the radioactive $^{238}$U, $^{226}$Ra and $^{232}$Th series respectively. About 45% of the inhabitant areas of the world are recorded by UNSCEAR, 2000¹⁰. The average dose per person range is from 84 nGy h$^{-1}$ to 200 nGy h$^{-1}$. The lowest value in Newzeland, Iceland and USA 40 nGy h$^{-1}$ because the construction of buildings mostly of wood. The highest value (95-115 nGy h$^{-1}$) in Hungary, Malezia and some other countries which use stones in construction ¹⁰. The aim of the present work is to quantify the presence of natural radionuclides in local building materials available in Iraq as well as to assess the radiological impact to the dweller of the building made from their materials.

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EXPERIMENTAL AND METHODS

Sample preparation

Twenty-nine building materials samples (bricks, cement, ceramic, granite, gravel, marble, paint, plaster, sand and soil) were collected from the local market in Iraq. The samples were sieved with 500 mesh sieve, and then dried at 100 C. The dried samples were put in a polyethylene Marinelli beaker and sealed for about 4 weeks in order to establish secular equilibrium between $^{232}$Th and $^{226}$Ra with their progeny.

Gamma spectrometry

Activity measurements have been performed by the gamma ray spectrometer at the central laboratory for radioactivity measurements (CLERMIT) at Nuclear and Radiological Regulatory Authority, Cairo using vertical HPGe detector of relative efficiency 40% and full width at half maximum (FWHM) of 1.95 keV for $^{60}$Co gamma energy line at 1.33 MeV. The detector was operated with Canberra Genie 2000 software for gamma acquisition and analysis. The HPGe detector was contained in a 5-cm thickness free standing lead castle providing a low background environment. To shield the detector from lead fluorescent x-rays and bremsstrahlung, the lead is lined with 1.5 mm iron and 1.0 mm Cu metals. Before performing the spectroscopic measurements, the spectrometer has been calibrated using multi-nuclides standard sources distributed in a reference materials which have certified concentration of natural radioactivity provided by the IAEA. The specific activity calculations of $^{226}$Ra, $^{232}$Th, and $^{238}$U were obtained indirectly from the gamma rays emitted by their progenies which are in secular equilibrium with them while K-40 activity was determined from the 1460.7 keV gamma line ($^{40}$K).

CALCULATIONS

Activity concentrations

The specific activity (in Bq kg$^{-1}$), $A_{Ei}$, of a nuclide i and for a peak at energy E, is given by:

$$A_{Ei} = \frac{NP}{t \times I(E) \times \varepsilon(E) \times M}$$

(1)

Where:

NP is the number of counts in a given, peak area, corrected for background peaks of a peak at energy E, $\varepsilon(E)$ the detection efficiency at energy E, t is the counting lifetime, $I(E)$ the number of gammas per disintegration of this nuclide for a transition at energy E, and M the mass in kg of the measured sample. If there is more than one peak in the energy analysis range for a nuclide, then an attempt to average the peak activities is made. The result is then the weighted average nuclide activity.

Radiation hazard index

The gamma ray radiation hazards due to the specified radionuclides will be assessed by three different ways: radium equivalent ($Ra_{eq}$), the absorbed dose rate (D) and the annual effective dose rate (AED).

Radium equivalent activity

$Ra_{eq}$ is a widely-used and can be calculated using the following equation (2) (12):

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

(2)

Where:

$A_{Ra}$, $A_{Th}$ and $A_{K}$ are activities of $^{226}$Ra, $^{232}$Th and $^{40}$K respectively in Bq kg$^{-1}$. The maximum value of $Ra_{eq}$ must be < 370 Bq kg$^{-1}$ in order to keep the external dose < 1.5 mGy y$^{-1}$ (13).

Radium equivalent activity index ($Ra_{eq}$), defined according to the estimation that 1 Bq kg$^{-1}$ of $^{226}$Ra, 0.7 Bq kg$^{-1}$ of $^{232}$Th and 13 Bq kg$^{-1}$ of $^{40}$K produce the same ray dose (14).
Hazard Indices (H_ex and H_in)

The two indices are those representing the external and internal radiation hazards. These indices are calculated as shown in equations 3 and 4 (15).

\[ H_{\text{ex}} = \left( \frac{C_u}{370} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810} \right) \leq 1 \] ……(3)

\[ H_{\text{in}} = \left( \frac{C_u}{185} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810} \right) < 1 \] ……(4)

Where:

\( C_u \), \( C_{\text{Th}} \) and \( C_{\text{K}} \) are the mean activity concentrations of \(^{238}\text{U} \), \(^{232}\text{Th} \) and \(^{40}\text{K} \) in Bq Kg\(^{-1} \) respectively.

The value of this index must be less than unity for the radiation hazard to be negligible.

Representative level index (I_{\gamma})

The representative level index (I_{\gamma}) is used to estimate the level of \( \gamma \)-radiation hazard associated with the natural radionuclides in specific building materials. The value is calculated using equation 5 derived by the European Commission (EC) (10) and NAE-OECD, 1979 (13).

\[ I_{\gamma} = \frac{C_{\text{Ra}}}{300} + \frac{C_{\text{Th}}}{200} + \frac{C_{\text{K}}}{3000} \leq 1 \] ……(5)

Where:

\( C_{\text{Ra}} \), \( C_{\text{Th}} \) and \( C_{\text{K}} \) (Bq kg\(^{-1} \)) are the concentrations of \(^{226}\text{Ra} \), \(^{232}\text{Th} \) and \(^{40}\text{K} \), respectively. This is the absorbed gamma dose rate and annual effective dose.

The absorbed dose rate (D)

This is the absorbed dose rate in air at 1 meter above the ground surface, assuming uniform distribution of the naturally occurring radionuclides. This has been calculated according to UNSCEAR 2000 (10) as follows:

\[ D_{\text{out}} \left( \frac{\text{nGy}}{\text{h}} \right) = (0.427A_{\text{Ra}}) + (0.662A_{\text{Th}}) + (0.042A_{\text{K}}) \] ……(6)

Where: \( A_{\text{Ra}} \), \( A_{\text{Th}} \) and \( A_{\text{K}} \) are the activity concentrations of \(^{226}\text{Ra} \), \(^{232}\text{Th} \) and \(^{40}\text{K} \) in Bq kg\(^{-1} \), respectively.

The annual effective dose equivalent AEDE

The annual effective dose (outdoor) is a useful concept that enables the radiation doses from different radionuclides and from different types and sources of radioactivity to be added. It is calculated based on the risks of radiation induced health effects. The annual effective dose rate AED (mSv y\(^{-1} \)) can be calculated using the following equation 7:

\[ \text{AED}_{\text{(out)}} = D \times 8760 \times 0.7 \times 10^{-6} \times 0.2 \text{ (mSv y}^{-1} \text{)} \] ……(7)

Where: \( D \) is the absorbed dose rate in (nGy h\(^{-1} \)), 0.7 Sv Gy\(^{-1} \) is the conversion coefficient from absorbed dose to effective dose and 0.2 the fraction of time spent outdoors. In most of samples under investigation.

RESULTS AND DISCUSSION

The specific activity concentrations of the radionuclides \(^{226}\text{Ra} \) (\(^{238}\text{U} \) and \(^{232}\text{Th} \) series and \(^{40}\text{K} \) in various building materials examined in this work are given in Table (1). From the data obtained, it can be observed that the \(^{226}\text{Ra} \), \(^{232}\text{Th} \) and \(^{40}\text{K} \) series concentrations vary in the range (7.33 ± 1.6 - 161.39 ± 7.8), (2.37 ± 0.88 – 139.6 ± 7.9) and (85.12 ± 11.5 – 1062.9 ± 130.4) Bq kg\(^{-1} \) respectively.

The mean values for activity concentrations of natural radionuclides in the samples under investigation were 35.26±4.65, 26.99±3.45 and 321.18±28.06 for \(^{238}\text{U} \), \(^{232}\text{Th} \) and \(^{40}\text{K} \) respectively. The results are lower than the world average (50 Bq kg\(^{-1} \) for \(^{226}\text{Ra} \) and \(^{232}\text{Th} \) and 500 Bq kg\(^{-1} \) for \(^{40}\text{K} \)) present in building materials except those for \(^{238}\text{U} \) and \(^{232}\text{Th} \) obtained from marble and granite. Fig.(1)
shows a comparison between the mean activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in building materials under investigation.

Radiological hazards for different samples under this investigation are presented in Figs. (2,3,4 and 5) associated with the world standard values. As shown from these figures, the results of the hazard parameters indicate that radium equivalent values for all building materials samples (Fig.2) vary from 8.65±3.5 Bq kg$^{-1}$ in PAN-20 to 406±30.14±30.14 Bq kg$^{-1}$ in sample CRM-11 with an average value 98.59±1.75 Bq kg$^{-1}$. As can be seen, all building brand samples studied in this work, the Ra$_{eq}$ values are well below the upper limit values (370 Bq kg$^{-1}$) suggested for building material by UNSCEAR, 2000$^{(10)}$ except sample CRM-11 due to the relatively high content of quartz and feldspar in the composition.

The external and internal gamma indices for all building samples considered in this work are less than unity except samples CRM-11 and GRN-12. Fig. (3) shows that the external and internal gamma indices vary from 0.05 in PNT-20 to 1.1 in CRM-11 for external hazard and from 0.07 in PNT-20 to 1.51 in CRM for internal hazards.

The representative level index (I$_{\gamma}$) values obtained for all the building materials range from 0.07 Bq kg$^{-1}$ in paints samples to 1.45 Bq kg$^{-1}$ in ceramic and Granite with an average value 0.25. Fig.(4) shows that the (I$_{\gamma}$) values of all samples are within the world standard value except samples GRN (13,14) and CRM-11 that have values higher than one.

The absorbed dose rate varies from 9.02 nGy h$^{-1}$ in PNT-20 to 184.6 nGy h$^{-1}$ in CRM-20, with an average value of 45.99 nGy h$^{-1}$. This indicates that the obtained data for absorbed dose rate for most of samples are still below the standard limit (59 nGy h$^{-1}$) except for samples MRB-17, GRN (13,14 and 12) and CRM-11. All AEDE were lower than the recommended limit of 0.07 mSv yr$^{-1}$(10).

This indicates that the building materials under study, which are used in Iraq, are radiologically safe. The radiological assessment studies of this type are highly needed as they provide necessary baseline information on radiation level in any environment that may be related to radioactive contamination specially after the gulf war.

![Fig. (1)](image1.png) The specific activity Bq/kg of U-238, Th-232 and K-40 in the building materials in Iraq

![Fig. (2)](image2.png) Radium equivalent activity Bq/kg of the samples and the limit values (370 Bq/kg) suggested for building material
The Internal and external Radiation hazard resulted from different building materials available in Iraq.

**Table (1):** Specific activity concentration and radium equivalent in (Bq kg\(^{-1}\)) for \(^{226}\)Ra, \(^{232}\)Th and \(^{40}\)K in different building materials used in Iraq.

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Activity Concentration (Bq/kg)</th>
<th>U-238</th>
<th>Th-232</th>
<th>K-40</th>
<th>(Ra_{eq})</th>
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<tbody>
<tr>
<td>BLK 1</td>
<td></td>
<td>15.71 ± 3.4</td>
<td>10.65 ± 2.5</td>
<td>138.15 ± 15.2</td>
<td>41.58 ± 8.15</td>
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<td>BRK 2</td>
<td></td>
<td>34.21 ± 8.2</td>
<td>21.91 ± 3.4</td>
<td>448.23 ± 31.3</td>
<td>100.06 ± 15.47</td>
</tr>
<tr>
<td>BRK 3</td>
<td></td>
<td>27.15 ± 2.99</td>
<td>29.52 ± 3.08</td>
<td>609.12 ± 42.63</td>
<td>116.27 ± 10.68</td>
</tr>
<tr>
<td>CEM 4</td>
<td></td>
<td>13.68 ± 2.8</td>
<td>4.41 ± 0.89</td>
<td>145.68 ± 16.07</td>
<td>31.20 ± 5.31</td>
</tr>
<tr>
<td>CEM 5</td>
<td></td>
<td>32.97 ± 3.93</td>
<td>14.82 ± 1.77</td>
<td>296.9 ± 26.7</td>
<td>77.02 ± 8.52</td>
</tr>
<tr>
<td>CEM 6</td>
<td></td>
<td>57.76 ± 7.47</td>
<td>13.62 ± 3.22</td>
<td>245.91 ± 36.9</td>
<td>96.17 ± 14.92</td>
</tr>
<tr>
<td>CEM 7</td>
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<td>37.21 ± 4.36</td>
<td>12.79 ± 2.1</td>
<td>227.85 ± 18.01</td>
<td>73.04 ± 8.75</td>
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<tr>
<td>CEM 8</td>
<td></td>
<td>58.01 ± 9.8</td>
<td>12.12 ± 3.1</td>
<td>108.8 ± 13.9</td>
<td>83.72 ± 15.3</td>
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<tr>
<td>CRM 9</td>
<td></td>
<td>28.67 ± 4.2</td>
<td>8.08 ± 1.6</td>
<td>250.61 ± 17.7</td>
<td>59.52 ± 7.85</td>
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<tr>
<td>CRM 10</td>
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<td>36.03 ± 5.68</td>
<td>52.33 ± 6.8</td>
<td>538.43 ± 49.4</td>
<td>152.32 ± 19.21</td>
</tr>
<tr>
<td>CRM 11</td>
<td></td>
<td>150.42 ± 17.7</td>
<td>139.86 ± 7.9</td>
<td>734.39 ± 14.9</td>
<td>406.97 ± 30.14</td>
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<tr>
<td>GRN 12</td>
<td></td>
<td>161.39 ± 7.8</td>
<td>60.93 ± 6.9</td>
<td>101.15 ± 11.2</td>
<td>256.31 ± 18.53</td>
</tr>
<tr>
<td>GRN 13</td>
<td></td>
<td>68.91 ± 7.55</td>
<td>107.26 ± 10.66</td>
<td>776.35 ± 41.18</td>
<td>282.07 ± 25.96</td>
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<tr>
<td>GRN 14</td>
<td></td>
<td>21.89 ± 2.83</td>
<td>117.91 ± 12.7</td>
<td>1016.4 ± 78.05</td>
<td>268.76 ± 27.00</td>
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<tr>
<td>GRV 15</td>
<td></td>
<td>15.27 ± 3.33</td>
<td>9.62 ± 1.6</td>
<td>212.56 ± 14.13</td>
<td>45.39 ± 6.71</td>
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<tr>
<td>MRB 16</td>
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<td>17.72 ± 1.9</td>
<td>4.83 ± 1.3</td>
<td>108.29 ± 13.1</td>
<td>32.97 ± 4.77</td>
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<tr>
<td>MRB 17</td>
<td></td>
<td>59.34 ± 5.9</td>
<td>55.96 ± 4.7</td>
<td>1062.9 ± 130.4</td>
<td>221.21 ± 22.66</td>
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<tr>
<td>PNT 18</td>
<td></td>
<td>10.25 ± 1.5</td>
<td>3.72 ± 1.05</td>
<td>127.46 ± 14.02</td>
<td>25.38 ± 4.08</td>
</tr>
<tr>
<td>PNT 19</td>
<td></td>
<td>9.25 ± 1.08</td>
<td>3.32 ± 0.98</td>
<td>127.31 ± 15.27</td>
<td>23.80 ± 3.66</td>
</tr>
<tr>
<td>PNT 20</td>
<td></td>
<td>7.33 ± 1.6</td>
<td>2.91 ± 0.78</td>
<td>93 ± 10.2</td>
<td>18.65 ± 3.50</td>
</tr>
<tr>
<td>PNT 21</td>
<td></td>
<td>11.53 ± 1.2</td>
<td>2.37 ± 0.88</td>
<td>85.12 ± 11.5</td>
<td>21.47 ± 3.34</td>
</tr>
<tr>
<td>PNT 22</td>
<td></td>
<td>8.59 ± 1.2</td>
<td>2.51 ± 0.77</td>
<td>120.62 ± 10.8</td>
<td>21.47 ± 3.13</td>
</tr>
<tr>
<td>PLS 23</td>
<td></td>
<td>14.25 ± 3.4</td>
<td>7.7 ± 1.53</td>
<td>170.72 ± 12.7</td>
<td>38.41 ± 6.57</td>
</tr>
<tr>
<td>PLS 24</td>
<td></td>
<td>19.21 ± 2.8</td>
<td>10.93 ± 1.9</td>
<td>155.34 ± 17.07</td>
<td>46.80 ± 6.83</td>
</tr>
<tr>
<td>SND 25</td>
<td></td>
<td>19.76 ± 3.8</td>
<td>14.32 ± 3.65</td>
<td>331.13 ± 17.4</td>
<td>65.73 ± 10.36</td>
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<tr>
<td>SND 26</td>
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<td>28.34 ± 6.04</td>
<td>8.38 ± 2.1</td>
<td>196.43 ± 11.74</td>
<td>55.45 ± 9.95</td>
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<tr>
<td>SND 27</td>
<td></td>
<td>15.35 ± 2.3</td>
<td>13.91 ± 1.99</td>
<td>303.84 ± 55.2</td>
<td>58.64 ± 9.40</td>
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<tr>
<td>SND 28</td>
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<td>8.69 ± 4.42</td>
<td>11.5 ± 5.84</td>
<td>193.07 ± 22.19</td>
<td>40.00 ± 14.48</td>
</tr>
<tr>
<td>SOL 29</td>
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<td>33.65 ± 5.8</td>
<td>24.76 ± 4.47</td>
<td>388.69 ± 45.12</td>
<td>98.99 ± 15.67</td>
</tr>
</tbody>
</table>
CONCLUSION

Most of the analyzed samples have radioactive concentrations within the accepted range which gives a radium equivalent of less than 370 Bq kg\(^{-1}\) dry weight except a ceramic (CRM-13) sample that showed high concentrations. Most of the samples have high concentrations of \(^{40}\text{K}\) which contribute to the absorbed dose. The present building materials such as bricks, cement, marble, gravel, paint, plaster, sand and soil have low concentrations for all the radioactive nuclides than the other samples ceramic, and granite that used as decorative materials. The obtained annual effective dose equivalent (outdoor) for ceramic, marble and granite were between 0.3 to 1 mSv y\(^{-1}\) and according to UNSCEAR 2000\(^{10}\) and NAE-OECD 1979\(^{13}\) the regulatory control should be considered for that materials and the materials lower than 0.3 mSv y\(^{-1}\) should be exempted.

REFERENCES


