RADIOMETRIC ANALYSIS OF CONSTRUCTION MATERIALS USING HPGe GAMMA-RAY SPECTROMETRY

M. U. Khandaker1,*, P. J. Jojo1,2, H. A. Kassim1 and Y. M. Amin1
1Department of Physics, University of Malaya, 50603 Kuala Lumpur, Malaysia
2Centre for Advanced Research, Department of Physics, Fatima Mata National College, Kollam 691001, Kerala, India

*Corresponding author: mu_khandaker@yahoo.com

Concentrations of primordial radionuclides in common construction materials collected from the south-west coastal region of India were determined using a high-purity germanium gamma-ray spectrometer. Average specific activities (Bq kg$^{-1}$) for $^{238}\text{U}(^{226}\text{Ra})$ in cement, brick, soil and stone samples were obtained as $54 \pm 13$, $21 \pm 4$, $50 \pm 12$ and $46 \pm 8$, respectively. Respective values of $^{232}\text{Th}$ were obtained as $65 \pm 10$, $21 \pm 3$, $58 \pm 10$ and $57 \pm 12$. Concentrations of $^{40}\text{K}$ radionuclide in cement, brick, soil and stone samples were found to be $440 \pm 91$, $290 \pm 20$, $380 \pm 61$ and $432 \pm 64$, respectively. To evaluate the radiological hazards, radium equivalent activity, various hazard indices, absorbed dose rate and annual effective dose have been calculated, and compared with the literature values. Obtained data could be used as reference information to assess any radiological contamination due to construction materials in future.

INTRODUCTION

Primordial radionuclides have been distributed in the earth’s crust since its formation with a considerable degree of heterogeneity. Therefore, radioanalyses of earth-borne materials are necessary for an accurate assessment of radiation exposure. Human beings are primarily exposed to external radiation originating from the natural radionuclides of construction or building materials(1). The world, its constituent elements and environment are naturally radioactive due to the various sources of radiation such as terrestrial radiation, cosmic radiation radon gas etc(2). Construction materials such as cement, brick, soil and stone etc. are actually the transformed form of basic constituent elements of this earth. All natural materials contain radionuclides which belong to mainly $^{238}\text{U}(^{226}\text{Ra})$ and $^{232}\text{Th}(^{228}\text{Ra})$ series, and the radioactive isotope of potassium ($^{40}\text{K}$). Both the particulate and energetic form of radiation released from construction materials via the decay chain of $^{238}\text{U}$ and $^{232}\text{Th}$ together with $^{40}\text{K}$ constantly cause the environment and living beings exposure. The aforementioned materials may, therefore, have a significant radiation background and can have negative consequences for the health of humans with long-duration irradiation and exposure(3, 4).

Radiation exposure due to construction materials can be categorised into external and internal exposures. External exposure is caused by direct gamma-ray irradiation, whereas internal exposure is caused by the inhalation of the radioactive inert gases radon ($^{222}\text{Rn}$), thoron ($^{220}\text{Rn}$) and their air-borne short-lived progenies. In order to assess the radiological hazards to human health, it is important to study the radioactivity of construction materials(5). Studies of radiation exposure to human beings are currently done in many countries in the world; however, there is not more data for radiation-exposure levels inside houses than for open spaces(3–10).

Additionally, as natural materials reflect the geological variability of their sites of origin, radiometric analyses of construction materials in different regions are required for accurate assessment of radiation exposure. Thus, the objective of this work was to obtain a baseline reference data of primordial radionuclides present in the construction materials of the south-west coastal region of India, and aid in decision-making processes in setting up guidelines for the control of radiation exposure in dwellings in India.

MATERIALS AND METHODS

Preparation and detection of samples

Samples of cement, brick, soil and stone were collected from various places where houses and buildings were under construction in the south-west coastal region of India. This is well known as a high background radiation area mainly because of the thorium-rich monazite sand available in plenty in this region. Even if the sampling region is a high background region, the bricks and cement have their origin in the normal background region. The bricks are generally made from surface clay and shale. The analysed cement samples were ordinary Portland cement available in the market in different grades. The soil samples were collected from a wide region nearly on equidistant basis using a coring tool within an area of 1 m$^2$ from the upper 5-cm layer of the land. Collected soil samples were purified by
removing all contaminants such as rocks, woods, metals, vegetation residual parts etc. The analysed stone samples were mainly crushed rocks. The soil, brick and stone samples were dried in a hot air oven at 110°C for 24 h to ensure that the sample is completely moisture free, and also a constant dry weight was obtained. Dried samples were crushed into small pieces and were ground to fine grain sizes and sieved in order to homogenise them. Cement samples were also dried in a hot air oven and sieved to homogenise them. All homogenised samples were then transferred to the standard cans, sealed hermetically and left for ~6–8 weeks at room temperature in order to reach secular equilibrium of 238U and 232Th decay series with their short-lived progeny. The amount of samples taken in the sealed cans for analysis varied from 242 to 268 g in case of cements, from 188 to 222 g for bricks, from 258 to 273 g for soil and from 265 to 275 g for stones.

An n-type, coaxial high-purity germanium (HPGe) gamma-ray detector was used to collect gamma spectra emitted from the studied samples. A cylindrical multi-nuclide source (having homogeneously distributed activity with the same volume and shape of the sample cans) was used for detector efficiency determination and the same procedure was applied to the sample activity determination. Each sample was counted for 60 000 s and the background counts were deducted to obtain net activity. Minimum detectable activity (MDA) of the gamma-ray measurement system was calculated according to the following equation

$$\text{MDA} = \frac{K_a \sqrt{N_B}}{\eta(E) P_{e} T_{c} M}$$

where $K_a$ is the statistical coverage factor equal to 1.645 at 95 % confidence level, and $N_B$ are the background counts at the interest of a certain radionuclide. $M$ is the dry-weight of sample (kg), $P_e$ is the gamma-ray emission probability, $T_c$ is the counting time, and $\eta(E)$ is the photo-peak efficiency. The MDA for the radionuclides of interest was calculated as 0.8 Bq kg$^{-1}$ for 226Ra, 0.9 Bq kg$^{-1}$ for 232Th and 2.4 Bq kg$^{-1}$ for 40K.

### Data correction for interfering gamma lines

Table 1 represents that some radionuclides have very close gamma-lines which could not be resolved properly by using the typical HPGe detector. As an example, the radionuclide 212Pb has only one strong gamma-line of 238.63 keV (43.6 %) with a half-life of 10.64 h. On the other hand, the radionuclide 214Pb has several characteristic gamma-lines of 241.98 keV (7.38 %), 295.21 keV (18.5 %) and 351.92 keV (35.8 %) with a shorter half-life of 26.8 m. In case of activity counting with a decay time of >5 h, it could be considered that the activity contribution of 214Pb radionuclide from 238.63 keV (241.98 keV) gamma-ray line almost completely decayed out. However, for a gamma spectrum having decay time <5 h, it is possible to separate the contribution of 214Pb from 238.63 keV gamma-ray line by using a well-known activity distribution formula available in Khandaker et al.$^{(11)}$.

### Calculations of activity and other radiation indices

Activity concentrations of the 238U(226Ra), 232Th and 40K radionuclides were determined using the characteristic gamma-rays of their short-lived progenies presented in Table 1, assuming that secular equilibrium was attained between parents and daughters within the period of storage. The activity concentrations were calculated using the following formula:

$$A = \frac{\text{CPS} \times 1000}{\epsilon_\gamma \times I_\gamma \times W}$$

where $A$ is the specific activity in Bq kg$^{-1}$, CPS is the net counts per second of the experimental sample, $W$ is the weight of the sample in gm, $\epsilon_\gamma$ is the efficiency of the HPGe detector at respective gamma-ray energy and $I_\gamma$ is the intensity of the corresponding gamma-ray energy. However, it is more practical to know ‘effective specific activity’ instead of ‘specific activity’ which can be calculated using the following equation$^{(12)}$

$$A_{\text{eff}} = A_{\text{Ra}} + 1.31 A_{\text{Th}} + 0.085 A_{\text{K}}$$

where $A_{\text{Ra}}$, $A_{\text{Th}}$ and $A_{\text{K}}$ are the specific activities of 226Ra, 232Th and 40K radionuclides, respectively. In practice, $A_{\text{eff}}$ should not exceed a value of 370 Bq kg$^{-1}$ for the materials which are used for newly built dwellings and public buildings.

### Radium equivalent activity

The distribution of 226Ra, 232Th and 40K radionuclides in the investigated materials were not found to be uniform. Uniformity with respect to the exposure of radiation can be defined in terms of radium equivalent activity ($R_{\text{aeq}}$)$^{(13)}$, which can be calculated using the following equation

$$R_{\text{aeq}} = C_{\text{Ra}} + \left(\frac{10}{7}\right) C_{\text{Th}} + \left(\frac{10}{130}\right) C_{\text{K}}$$

where $C_{\text{Ra}}$, $C_{\text{Th}}$ and $C_{\text{K}}$ are the specific activities of 226Ra, 232Th and 40K in Bq kg$^{-1}$, respectively.
External hazard index

External as well as internal exposures are the two pathways of radiation dose imparted to human beings from the construction materials. As more than one radionuclide contributes towards the gamma doses, radiological hazards are presented in terms of a single quantity called ‘hazard index’. Equation (5) was used to assess the external radiological hazards from the studied construction materials (13)

\[
H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810}
\]  

(5)

To keep the radiation hazard insignificant, the value of this index must be less than unity (14). It has been assumed that 370 Bq kg\(^{-1}\) of 226Ra or 259 Bq kg\(^{-1}\) of 232Th or 4810 Bq kg\(^{-1}\) of 40K produces the same gamma dose equivalent.

Internal hazard index

Radiation hazard threat to respiratory organs due to the inhalation of 222Rn and its progenies can be expressed by the internal hazard index (13) as

\[
H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810}
\]  

(6)

The construction materials would be safe if the internal hazard index, \(H_{in} \leq 1\).

Alpha index

The excess alpha radiation due to the radon inhalation originating from the construction materials can be assessed via the alpha index \((I_{\alpha})\) (15)

\[
I_{\alpha} = \frac{C_{Ra}}{200}
\]  

(7)

The safe limit of 226Ra activity concentration is 200 Bq kg\(^{-1}\) (i.e. indoor radon concentration is 200 Bq m\(^{-3}\)), for which \(I_{\alpha} = 1\).

Absorbed dose rate

The absorbed dose rate, \(D\) (nGy h\(^{-1}\)) in air (16) was calculated using the formula given in Equation (9)

\[
D = \left(\frac{0.427 \times C_{Ra} - 226 + 0.662 \times C_{Th} - 232}{C_{K} - 40} + 0.0432 \times C_{K} - 40\right)
\]  

(8)

where \(C_{Ra-226}\), \(C_{Th-232}\) and \(C_{K-40}\) are the average activity concentrations of 226Ra, 232Th and 40K, respectively. The worldwide average dose rate (1, 2) is in the range of 18–93 nGy h\(^{-1}\).

Annual effective dose

The annual effective dose equivalent (17) received by the members of the public from the radioactivity from construction materials has been calculated using the following relation:

\[
E_{eq} = D \times Q \times T \times O_{f}
\]  

(9)

where \(D\) is the absorbed dose rate in air (nGy h\(^{-1}\)) calculated from Equation (8), the value of \(Q\) is the dose conversion factor from gray to Sievert, 0.7 (Sv Gy\(^{-1}\)) (17), \(T\) is time in hours per year (h y\(^{-1}\)), \(O_{f}\) is the occupancy factor, i.e. the factor for length of time of being inside houses, considered as 80 % (0.8). The worldwide average background value of the annual effective dose for a bricks-made room is \(\sim 0.41\) mSv y\(^{-1}\) and for soil, it is 0.46 mSv y\(^{-1}\) (18).

RESULTS AND DISCUSSION

Samples of some common construction materials collected from the construction sites were analysed...
Table 2. Radiation indicators for the $^{238}$U, $^{232}$Th and $^{40}$K radionuclides in the investigated samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Code</th>
<th>Specific activities (Bq kg$^{-1}$)</th>
<th>Radium equivalent activity (Bq kg$^{-1}$)</th>
<th>Radiation indices (maximum value)</th>
<th>Absorbed dose rate</th>
<th>Annual effective dose</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>$^{238}$U</td>
<td>$^{232}$Th</td>
<td>$^{40}$K</td>
<td>$R_{aeq}$</td>
<td>$(H_{in} \leq 1; H_{ex} \leq 1; I_a = 1)$</td>
</tr>
<tr>
<td>Cement</td>
<td>Ce-1</td>
<td>57 ± 4</td>
<td>68 ± 6</td>
<td>567 ± 24</td>
<td>197.7 ± 14.4</td>
<td>0.69 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Ce-2</td>
<td>77 ± 6</td>
<td>66 ± 6</td>
<td>312 ± 22</td>
<td>195.3 ± 16.3</td>
<td>0.74 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>Ce-3</td>
<td>48 ± 4</td>
<td>80 ± 5</td>
<td>438 ± 26</td>
<td>195.9 ± 13.1</td>
<td>0.66 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Ce-4</td>
<td>62 ± 5</td>
<td>72 ± 6</td>
<td>566 ± 25</td>
<td>208.4 ± 15.5</td>
<td>0.73 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>Ce-5</td>
<td>44 ± 5</td>
<td>62 ± 6</td>
<td>410 ± 23</td>
<td>164.1 ± 15.3</td>
<td>0.56 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Ce-6</td>
<td>34 ± 4</td>
<td>44 ± 5</td>
<td>348 ± 22</td>
<td>123.6 ± 12.8</td>
<td>0.43 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>54 ± 13</td>
<td>65 ± 10</td>
<td>440 ± 91</td>
<td>180.7 ± 34.3</td>
<td><strong>0.63 ± 0.13</strong></td>
</tr>
<tr>
<td>Brick</td>
<td>Br-1</td>
<td>20 ± 3</td>
<td>20 ± 3</td>
<td>283 ± 21</td>
<td>70.3 ± 8.9</td>
<td>0.24 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Br-2</td>
<td>18 ± 2</td>
<td>18 ± 3</td>
<td>264 ± 25</td>
<td>64.0 ± 8.2</td>
<td>0.22 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Br-3</td>
<td>25 ± 3</td>
<td>22 ± 3</td>
<td>275 ± 23</td>
<td>77.6 ± 9.1</td>
<td>0.28 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Br-4</td>
<td>27 ± 3</td>
<td>19 ± 2</td>
<td>320 ± 24</td>
<td>78.7 ± 7.7</td>
<td>0.29 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Br-5</td>
<td>23 ± 3</td>
<td>27 ± 3</td>
<td>318 ± 23</td>
<td>86.0 ± 9.1</td>
<td>0.29 ± 0.03</td>
</tr>
<tr>
<td></td>
<td>Br-6</td>
<td>15 ± 2</td>
<td>21 ± 2</td>
<td>277 ± 21</td>
<td>66.3 ± 6.5</td>
<td>0.22 ± 0.02</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>21 ± 4</td>
<td>21 ± 3</td>
<td>290 ± 20</td>
<td>73.3 ± 9.8</td>
<td><strong>0.25 ± 0.04</strong></td>
</tr>
<tr>
<td>Soil</td>
<td>So-1</td>
<td>47 ± 4</td>
<td>54 ± 6</td>
<td>333 ± 22</td>
<td>149.7 ± 14.3</td>
<td>0.53 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>So-2</td>
<td>49 ± 4</td>
<td>56 ± 6</td>
<td>356 ± 23</td>
<td>156.4 ± 14.3</td>
<td>0.56 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>So-3</td>
<td>48 ± 4</td>
<td>60 ± 7</td>
<td>348 ± 23</td>
<td>160.5 ± 15.8</td>
<td>0.56 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>So-4</td>
<td>52 ± 5</td>
<td>62 ± 7</td>
<td>476 ± 29</td>
<td>177.2 ± 17.2</td>
<td>0.62 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>So-5</td>
<td>54 ± 5</td>
<td>58 ± 6</td>
<td>390 ± 23</td>
<td>166.8 ± 15.3</td>
<td>0.60 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>50 ± 12</td>
<td>58 ± 10</td>
<td>380 ± 61</td>
<td>162.1 ± 31.0</td>
<td><strong>0.57 ± 0.12</strong></td>
</tr>
<tr>
<td>Stone</td>
<td>St-1</td>
<td>48 ± 5</td>
<td>52 ± 6</td>
<td>433 ± 22</td>
<td>155.6 ± 15.3</td>
<td>0.55 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>St-2</td>
<td>46 ± 4</td>
<td>54 ± 6</td>
<td>485 ± 23</td>
<td>160.4 ± 14.3</td>
<td>0.56 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>St-3</td>
<td>44 ± 4</td>
<td>58 ± 7</td>
<td>396 ± 23</td>
<td>137.3 ± 15.8</td>
<td>0.54 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>St-4</td>
<td>48 ± 5</td>
<td>58 ± 7</td>
<td>428 ± 29</td>
<td>163.8 ± 17.2</td>
<td>0.57 ± 0.06</td>
</tr>
<tr>
<td></td>
<td>St-5</td>
<td>42 ± 4</td>
<td>62 ± 7</td>
<td>417 ± 23</td>
<td>162.6 ± 14.3</td>
<td>0.55 ± 0.05</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td>46 ± 8</td>
<td>57 ± 12</td>
<td>432 ± 64</td>
<td>160.6 ± 30.1</td>
<td><strong>0.56 ± 0.10</strong></td>
</tr>
</tbody>
</table>

The bold values indicate the determined average activities in different samples.
to obtain information on various radiation indicators, and obtained values are summarised in Table 2. The measured levels of radioactivities in cement, brick, soil and stone samples were found to be within the world range\(^1\) and also in agreement with the literature values\(^{19,20}\). The activity concentration of the anthropogenic radionuclide \(^{137}\)Cs was not found within the detection limit. Effective average specific activities for cement, brick, soil and stone samples were found to be 176.55 ± 33.83, 73.16 ± 9.63, 158.28 ± 30.28 and 157.39 ± 29.16 Bq kg\(^{-1}\), respectively. The quoted uncertainties are the standard deviations from the mean values.

**CONCLUSIONS**

We determined primordial radionuclides concentrations in construction materials with varying levels of activity using HPGe gamma-ray spectrometry. The average specific activities (Bq kg\(^{-1}\)) for \(^{238}\)U(\(^{226}\)Ra) in cement, brick, soil and stone samples were obtained as 54 ± 13, 21 ± 4, 50 ± 12 and 46 ± 8, respectively. Respective values of \(^{232}\)Th concentration were obtained as 65 ± 10, 21 ± 3, 58 ± 10 and 57 ± 12. Concentrations of long-lived \(^{40}\)K radionuclide were obtained as 440 ± 91, 290 ± 20, 380 ± 61 and 432 ± 64 in cement, brick, soil and stone samples, respectively. As the average specific activities of the studied samples were found to be within the world range\(^1\) (for \(^{226}\)Ra 17–60 Bq kg\(^{-1}\), for \(^{232}\)Th 11–64 Bq kg\(^{-1}\) and for \(^{40}\)K 140–850 Bq kg\(^{-1}\)), it is safe to use these materials for construction. Additionally, the values of several radiological hazard indices were found within the safe limit, i.e. ≤1. The results may not reflect the real situation for all types of cement and bricks available throughout India. However, the obtained data can be considered as base values for the distribution of natural radionuclides in cement, brick, soil and stone for the studied region, and could be used as reference information to carry out an approximately designed programme on environmental radioactivity monitoring aimed at minimising population exposure, which can ensure a safer living environment.

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**REFERENCES**