ISSN : 0974 - 7451 Volume 7 Issue 7

Trade Science Inc.

Environmental Science
An Indian Journal

Current Research Paper

ESAIJ, 7(7), 2012 [256-262]

Measurements of the natural radioactivity in building materials (raw and manufactured), other than granites in KSA

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Received: 15th May, 2012 ; Accepted: 6th August, 2012

ABSTRACT

In this work we have measured the natural radiation content of the building materials (raw and manufactured) using HPGe detector. The intensities of γ-ray emitted by radioactive nuclides present in the samples were determined. Efficiency of the detector at different energies was determined using standard sources, so that radiation content of the samples can be estimated. Samples from different areas in KSA were collected. These samples were prepared in a form of fine powder suitable to be used by Marinelli beakers. Data for every sample were collected for 36000 s. Soft ware equipped with the detector electronic system had been used to analyze the data, hence the results were recorded. The highest measured activity concentrations in the samples are: 48, 42 and 971 Bq.kg^{-1} for Th- 232, Ra - 226 and K-40, respectively, which are in the range of the corresponding typical worldwide values. The absorbed dose rates, effective dose rates, radium equivalent activities as well as the radiation hazard indices were estimated. The maximum radium equivalent activity (Ra_{eq}) was 186 Bq kg^{-1}, which is lower than the limit of 370 Bq kg^{-1} set by the Organisation for Economic Cooperation and Development (Exposure to radiation from the natural radioactivity in building materials. Report by a Group of Experts of the OECD, Nuclear Energy Agency OECD, Paris, 1979)

KEYWORDS

Natural radioactivity;
Building materials;
Gammy-ray spectrometry.

INTRODUCTION

All building materials contain various amounts of radioactive substances. Building materials originating from rock and soil contain mainly natural radio nuclides of uranium (U -238), thorium (Th-232) series and the radioisotope of potassium (K-40). In the uranium series, the decay chain starts by the most important radioisotope Ra-226, therefore, reference is often made to radium instead of uranium. In addition to natural radioactive substances, some industrial by-products also contain radionuclides, Caesium (Cs-137) in particular, which are spread into the environment due to fallout from nuclear weapon tests and industrial nuclear accidents such as Chernobyl and recently Fukoshima in Japan[1].

If such by-product is incorporated into building material; the final product will also contain these artificial

radionuclides[2]. Ash, which is generated during combustion of peat, coal, wood, forest processed chips, field biomass, by-products of wood industry or other comparable materials, contains both natural radioactive substances and artificial radionuclides originating from fallout. Ash is used as landscaping, and as an additive in concrete. It is also used as bulk material under roads, taken to dumps, or mounded[2]. The world wide concentrations of radium, thorium, potassium and Caesium-137 in earth crust ranging from 27-40 Bq kg^{-1}, 46-62 Bq kg^{-1}, 63-629 Bq kg^{-1} and 3-5.15 Bq kg^{-1}, respectively, depending on the location on earth[3]. The reported values of Th in rocks was 1.6-20 ppm, while in crust it is 10.7 ppm in average. The reported values of U are 0.5-4.7 ppm in common rock types[4]. Typical activity concentrations in common building materials and industrial by-products used for building materials in EU ranging from 10 up to 390 Bq kg^{-1} for Ra-226, from 10 up to 100 Bq kg^{-1} for Th – 232 and from 60 up to 670 Bq kg^{-1} for K-40[5]. While maximum activity concentrations of these materials were ranging from 25 up to 2600 Bq kg^{-1} for Ra – 226, from 30 up to 340 Bq kg^{-1} for Th -232 and from 200 up to 4000 Bq kg^{-1} for K -40[5]. Worldwide average annual effective doses of radiation at year 2000 from natural sources (2400 μ Sv) and Inhalation (mainly radon) 3600 μ Sv)[6].

Many studies have been done to measure the concentrations and the doses resulting from the natural radioisotopes present in the building materials, all over the world. Samples of cement and ceramic bricks used as building materials in Poland have been analyzed for Ra-266, Th-232 and K-40 using a high resolution HPGe gamma ray spectrometer shows a dose rate ranging from 99- 102 nGy h^{-1}[7]. The radioactivity concentration of Ra-266, Th-232 and K-40 (Bq kg^{-1}) in soil samples taken from different locations in State of Qatar ranging from 65-301,11-25 and 160-296, respectively[8]. While the reported values in other countries were as follows[8]:

Algeria: 2-127, 2-144, 35-1405, respectively, Egypt: 5-24, 10-20, 293-660 Respectively, Jordan: 15-60, 4-29, 99-379, respectively, India: 5-71, 15-76, 20-854, respectively, Bangladesh: 13-43, 3-81, 402-750, respectively, China: 40-443, 33-68, 442-913, respectively and Worldwide: 40, 40, and 580, respectively.

The radioactivity concentrations of Ra-266, Th-232 and K-40 (Bq kg^{-1}) were also measured for various cement samples used for building materials in the State of Qatar, the reported values ranging from 27-32, 8-14 &54-98 respectively[9]. The activity concentrations of natural radionuclides in various building materials in Vietnam were found to be ranging from 5-170, 20-190 and 20-1980 (Bq/m^3) for Ra-266, Th-232 and K-40, respectively[10]. An investigation on the natural radioactivity of building materials in central Turkey, show that the average values resulting from Ra-266, Th-232 and K-40 were: 632.2 Bq kg^{-1} for fly-ash, 4.4 Bq kg^{-1} for brick, 73.3 Bq kg^{-1} for soil, 306.6 Bq kg^{-1} for cement, 302 Bq kg^{-1} for gypsum and 83.6 Bq kg^{-1} for solvent-based paint[11].

The investigation on the natural radioactivity of bricks in India, show that the average values resulting from Ra-266, Th-232 and K-40 vary from 9.89-23.48 Bq kg^{-1}, 25.35-62.02 Bq kg^{-1} and 342.48-405.24 Bq kg^{-1}, respectively[12]. In Ghana, the calculated average activity concentrations of Ra-266, Th-232 and K-40 in the cement samples were 35.9 Bq kg^{-1}, 25.4 Bq kg^{-1} and 233.3 Bq kg^{-1}, respectively[13]. The activity concentrations of Ra-266, Th-232 and K-40 in Bq kg^{-1} of different Egyptian building materials were as follows: sand (9.2,2.9,54.5), cement (22.5, 3.9,68.7), limestone (13.4,3.3,61.6), yellow clay (27.2,19.3, 188.7), respectively[14]. The concentrations of Ra-266 and Th-232 in building materials used in Saud Arabia were found to be less than those reported in different countries, while the concentrations of K -40 were higher than those reported from other countries[15]. Measurements of the concentrations of naturally occurring radionuclide (Ra-266, Th-232 and K-40) in rocks and sand samples taken from Taif province in Saudi Arabia using GRS-2000 and PGR gave lower values than acceptable value of 1 mSv y^{-1}[16], thus they concluded that accurate measurements on the collected samples using HPGe detector are highly recommended[16]. Saudi Arabia is considered a large market for local and foreign building materials. The aim of this present work is to measure the radiation doses resulting from these radionuclides. These doses can be hazardous to those who live in houses and buildings constructed from these building materials, if they exceed a limited value known as MPD reported in ICRP[17] which is about 600 μ Sv/ y. Ra-

dioactive Radon (Ra-222) gas is emitted from Uranium series and can be accumulated in houses, thus causing lung cancer if exceeds allowed levels. Thus certain precautions can be taken to protect humans from radiation, by using selected building materials having lower concentrations of radioactivity. Several countries have adopted their own policies for limiting the radioactivity concentrations of building materials. If these materials have high concentration of radioactive nuclides, they can be used in other constructions, like roads instead houses. Even though this work can not cover all samples of building materials used in the kingdom, it can be a pointer for similar studies if the findings are significant. It may also help to adopt policies with respect to regulating the use of building materials that might be hazardous to public. Authorities in areas such as minister of commerce and industry and the Saudi Arabian standards organization (SSO) can benefit from the outcome of this work. High resolution γ- ray spectroscopy employing an HPGe detector was adopted in this work. The radioactivity concentrations of Ra-266, Th-232 and K-40 in all the building material samples were determined. A comparative study of our data with different results in other areas of the world has been done. Thus some conclusions can be derived, so that certain policies can be adopted to minimize the hazards of radiation

EXPERIMENTAL

Acquisition of samples

Local and imported Samples of different building materials were collected from different areas in the kingdom. We focus on the widely and mostly used materials in various constructions and buildings. Each sample was classified by its origin (local or imported) and the place of origin.

Processing of samples

Brick and tiles samples were crushed in the laboratory in two stages in sequence to convert them into fine powder. The first grinder breaks the hard samples to small pieces. The second grinder crushes the samples to dust. The samples were then sieved through 40 mesh sieve and then heated at 110° C in an oven for 24 hours to get rid of moisture, if any. The samples were then cooled and transferred to plastic Marinelli beakers of volume 500 cc each and then sealed by epoxy. The net mass of each sample was recorded. Also vinyl tapes were wrapped along the edges of the lids of the beakers. This kind of sealing by epoxy and tapes is expected to keep the radon gas that emanates from the crushed samples to be confined to the beaker, as much as possible. These sealed Marinelli beakers were then kept for a period of about one month in order to allow the daughters of U-238 and Th – 232 to reach secular equilibrium with each other.

Energy calibration

The multichannel analyzer of our electronic system was calibrated for energy using standard radioactive source of Eu -152. Eu -152 being a multi-peak gamma emitter provides a reasonable coverage of the gamma spectrum, the lowest energy is 121.78 keV and the highest is 1408.11 keV.

To have the same geometry as the samples, standard Eu -152 sources should be prepared in Marinelli beaker. Eu -152 in liquid form with known activity was the standard radioactive source used for the preparation of such extended source. For this purpose, clean Marinelli beaker was weighted empty and then was filled with selected sample. The sample was then counted for the background for a reasonable period of time (about 7000s). The region of interests (ROIs) in the MCA spectrum corresponding to the Eu -152 energies where marked previously, and the counts for all ROIs for this time period were noted. The sample was now emptied to a clean glass bowl. Using pipette, 0.2 ml of Eu -152 solution was pipette drop by drop to the sample. This was mixed thoroughly and carefully to insure homogeneity of the radioactivity in the sample. The radioactive standard was then transferred to the Marinelli beaker and counted for 7000 s.

Efficiency calibration

The detector efficiency is known as: the ability of the detector for recording interactions within its effective volume. If spectral quantitative analysis is required, the detector efficiency must be determined over the range of energies of interest. The general method is to collect a spectrum from a known source and determine the ratio of the number of events counted to the number

of actual events. This was done for several peaks in the spectrum to allow the plotting of the efficiency curve. In this work we will use the so called absolute efficiency. The absolute efficiency is defined as the ratio of the number of pulses recorded by the detection system to the number of radiation quanta emitted by the source; it depends on the geometry of the experiment and the detector properties. The absolute efficiency ($\in$) is given by the following equation[18]:

$$\varepsilon = \frac{c_p}{A.I_\gamma} \quad (1)$$

Where cp is the total net counts per second, representing the number of photons detected under the photopeak; Is the activity of the standard source in Bq A; I_γ is the intensity of gamma line (relative abundance) The error can be calculated using the relation:

$$\sigma\varepsilon = \varepsilon \times \sqrt{\frac{\sigma^2 c_p}{c_p^2} + \frac{\sigma^2 A}{A^2}} \quad (2)$$

Gamma-ray spectrometer

The high-resolution gamma spectrometry system used for analyzing the samples consisted of Canberra high-purity germanium HPGe detector, of 50 % relative efficiency coupled to Canberra Digital Spectrum Analyzer DSA2000 (GENIE-2000) with an FWHM of 2 keV for the 1.332 MeV gamma ray of ^{60}Co.

The HPGe' detector is equipped with model 747 Canberra lead shield system (composed of 10-cm thick low-background lead, and 1-mm tin and 1.6-mm copper graded liner to prevent interference by lead X rays)

The radionuclides under focuses were ^{226}Ra, ^{232}Th and ^{40}K. The ^{226}Ra activities were estimated from the gamma rays of ^{214}Pb (295.2, 351.9 keV) and ^{214}Bi (609.3 and 1120.3 keV). The ^{232}Th activities were estimated from the gamma rays of ^{212}Pb (238.6 keV), ^{228}Ac (338.4, 911 keV) and ^{208}Tl (583.2 keV).

Several peaks from ^{226}Ra and ^{232}Th daughters were also monitored. The K -40 activities was determined from its own gamma ray (1460.8 keV).

Once the absolute efficiency curve is plotted, the efficiency of the detector at any value of gamma energy can be determined; hence the activity concentrations (A_p in Bq kg^{-1}) for the natural radionuclide in the given sample can be calculated using the relation:

$$A_p = \frac{C_p}{\varepsilon P_r M_s} \quad (3)$$

Where C_p is the net peak counting rate of gamma ray (counts per second), ε is the detector efficiency of the specific gamma ray, and Ms is the mass of the sample (kg). P_r represents the absolute transition probability of the specific gamma decay. When the activity concentration is evaluated from several gamma peaks, the weighted average is considered after taking into account all necessary corrections as previously indicated.

Assessment of radiological risk

In order to assess the radiation hazard associated with the building materials used, the Ra_{eq} and the absorbed dose rate (***) have been evaluated, where it is assumed that all the decay products of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their precursors. The Ra_{eq} is calculated according to the following formula[19,20]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

Where, A_{Ra}, A_{Th} and A_K are the specific activities (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. This formula is based on the estimation that 1 Bq kg^{-1} of ^{238}U, 0.7 Bq kg^{-1} of ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose rates.

The outdoor absorbed dose rate (nGy h^{-1}) in air from terrestrial gamma radiation at 1 m above the ground is calculated after applying the conversion factors (in nGy h^{-1} per Bq kg^{-1}) to transform specific activities A_{Ra}, A_{Th} and A_K into absorbed dose rate according to the following formula[21]:

$$\dot{D} = 0.429A_{Ra} + 0.666A_{Th} + 0.042A_K \quad (5)$$

In addition, the annual effective dose rate indoors ($\dot{E}$) (measured in mSv y^{-1}) was calculated, assuming a value of 0.7 Sv Gy^{-1} for the conversion factor from absorbed dose in air to annual effective dose received by adults and a 0.8 factor for the indoor occupancy[22,23]. The formula used is:

$$\dot{E} = \dot{D}(nGy\,h^{-1}) \times 24h \times 365.25d \times 0.8 \times 0.7\,Sv\,Gy^{-1} \times 10^{-6} \quad (6)$$

To estimate the gamma-radiation dose expected to be delivered externally from building materials, a model was suggested by various researchers to limit the radiation dose from building materials to 1.5 mSv y^{-1}. In

this model the external hazard index has been defined as[19]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (7)$$

The radiation risk is negligible when the maximum value of the external hazard index is less than unity ($H_{ex} \leq 1$), which is equivalent to a maximum value of the Ra_{eq} activity <370 Bq kg^{-1}. In terms of dose equivalent, this index must be less than unity so that the annual effective dose due to radioactivity in the material will be ≤ 1.5 mSv h^{-1}.

Internal exposures arise from the inhalation of radon (^{222}Rn) gas and its short-lived decay products as well as from the inhalation or ingestion of other radionuclides. Radon is part of the radioactive series of ^{238}U, which is present in building materials. To assess the internal exposure to ^{222}Rn gas, the internal hazard index has been defined as[19]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (8)$$

The use of a material in the construction of dwellings is considered safe when the maximum value of the internal hazard index is less than unity ($H_{in} \leq 1$).

RESULTS AND DISCUSSION

The activity concentrations of most commonly used building materials in the kingdom of Saudia Arabia were measured, employing the high – resolution gamma ray spectrometry. In TABLE 1 we show the various ^{232}Th, ^{226}Ra and ^{40}K activity concentrations (Bq kg^{-1}) for the building materials samples under investigation. It is seen from this table that activity concentration of Th- 232 ranges from 0.2 Bq.kg^{-1} (for Kumryy gypsum) to 48 Bq.kg^{-1} (for Kishani China) The activity concentration of Ra-266 ranges from 0.7Bq.kg^{-1} (for Yanbu gypsum) to 42 Bq/kg (for Kishani China). The activity concentration of K-40 ranges from 1.9 Bq.kg^{-1} (for Teyma sand) to 971 Bq.kg^{-1} for (Kishani China, also). Thus Kishani China seems to have the highest activity concentration among all the investigated samples, but it is still below the acceptable values.

In TABLE 2, we show the Absorbed Dose rate (nGy.h^{-1}), Ra_{eq} (Bq.kg^{-1}) and the indoors effective dose rate (mSv.y^{-1}) for all samples. This Table shows that the absorbed dose rate (nGy.h^{-1}) ranges from 0.8 for Yanbu gypsum to 90 (nGy.h^{-1}) for Kishani China, while the Ra_{eq} (Bq.kg^{-1}) ranges from 1.7 for Yanbu gypsum to 185 (Bq.kg^{-1}) for Kishani China, which is less than the accepted value of 370 Bq.kg^{-1}[24]. The effective dose rate (μSv.y^{-1}) ranges from about 0 for Yanbu gypsum to 450 μ Sv.y^{-1} for Kishani China, also.

TABLE 1 : ^{232}Th, ^{226}Ra and ^{40}K activity concentrations (Bq / kg) for the building materials samples under investigation.

Sample no.	Sample Name	Th-232	Ra-226	K-40
1	Yanbu gypsum	0.4±0.1	0.7±0.1	6.3±0.5
2	ceramic china	34.9±0.2	26.0±2.7	660.2±0.02
3	red brick	6.2±0.3	6.6±0.7	223.1±0.02
4	teyma sand	7.6±0.0	5.6±0.6	1.9±0.0
5	bathat khomra	3.7±0.2	3.9±0.5	508.8±0.0
6	kishani china	48.0±1.5	42.3±4.7	971.7±20
7	concrete block	4.8±0.4	11.1±1.9	505.4±0.6
8	white cement Egypt	6.0±0.0	11.2±1.0	2.2±0.0
9	white block	2.5±0.1	5.1±0.2	436.9±0.4
10	kishani local	6.3±0.4	10.6±0.8	303.3±0.09
11	Saudi ceramic	30.9±0.4	36.8±4.1	734.1±0.0
12	Yanbu cement	4.9±0.0	17.8±2.1	165.2±0.06
13	breman gravel	2.3±0.1	3.2±0.5	262.7±0.43
14	Saudi mozaic	3.6±0.2	7.4±0.6	313.1±0.32
15	bahra sand	5.4±0.4	4.9±0.6	638.2±0.32
16	kumreyyeh sand	2.6±0.0	3.1±0.3	8.9±0.0
17	kumryyah badhaa	5.8±0.2	4.2±0.7	522.6±7.8
18	kumryy gypsum	0.2±0.1	7.2±1.8	47.3±0.0
19	kumryy soil	6.1±0.1	7.9±1.0	763.7±0.14

In TABLE 3, we show the calculated values in this work of both H_{ex} and H_{in} indices for all samples, it is seen clearly that their values are less than unity, which indicate that the radiation doses are below the hazardous limits. It is clear that Yanbu gypsum has the smallest values of these indices, while the highest values are reported in case of Kishani China, Ceramic China and Saudi ceramics. But all these values are ≤ 1, which means that the radiation risk is negligible. The lowest concentration values were reported for Yanbu gypsum,while the highest concentration values were reported for Kishani China. From the reported values, it is cleare that the concentrations of K-40 are higher than the values reported for Th- 232, Ra -226, as expected. The measured activity concentrations in the samples are in

TABLE 2 : The absorbed dose rate (nGy.h^{-1}), Ra_{eq} (Bq.Kg^{-1}) and the indoors effective dose rate (mSv.y^{-1}) for all samples.

Sample number	Sample Name	Absorbed Dose Rate	Ra_{eq}	Indoors effective Dose Rate
1	Yanbu gypsum	0.8	1.7	0.00
2	ceramic china	62.1	126.8	0.30
3	red brick	16.3	32.6	0.08
4	teyma sand	7.6	16.7	0.04
5	bathat khomra	25.5	48.4	0.13
6	kishani china	90.9	185.7	0.45
7	concrete block	29.2	56.9	0.14
8	white cement Egypt	8.9	19.9	0.04
9	white block	22.2	42.4	0.11
10	kishani local	21.5	43.0	0.11
11	Saudi ceramic	67.2	137.5	0.33
12	Yanbu cement	17.8	37.5	0.09
13	breman gravel	13.9	26.6	0.07
14	Saudi mozaic	18.7	36.7	0.09
15	bahra sand	32.5	61.8	0.16
16	kumreyyah sand	3.4	7.5	0.02
17	kumreyyah badhaa	27.6	52.8	0.14
18	kumreyyah gypsum	5.2	11.1	0.03
19	kumreyyah soil	39.5	75.4	0.19

TABLE 3 : Values of both H_{ex} and H_{in} calculated for all investigated samples in this work.

Sample number	Sample Name	H_{ex} Index	H_{in} Index
1	Yanbu gypsum	0.00	0.01
2	ceramic china	0.34	0.41
3	red brick	0.09	0.11
4	teyma sand	0.04	0.06
5	bathat khomra	0.13	0.14
6	kishani china	0.50	0.62
7	concrete block	0.15	0.18
8	white cement Egypt	0.05	0.08
9	white block	0.11	0.13
10	kishani local	0.12	0.14
11	Saudi ceramic	0.37	0.47
12	Yanbu cement	0.10	0.15
13	breman gravel	0.07	0.08
14	Saudi mozaic	0.10	0.12
15	bahra sand	0.17	0.18
16	kumreyyeh sand	0.02	0.03
17	kumreyyah badhaa	0.14	0.15
18	kumreyyah gypsum	0.03	0.05
19	kumreyyah soil	0.20	0.22

the range of the corresponding typical world values which are: 50, 50 and 500 Bq.kg^{-1} for Th-232, Ra - 226 and K-40, respectively[5]. Thus, one can conclude that:

1. The highest radiation content was detected for each of : Kishani China, Ceramices China and Saudi Ceramics samples.
2. The lowest radiation content was detected for each of : sand samples, gypsum and cement samples. For sand samples, we can see that the Tayma sand has the lowest radiation content. The Yanbu gypsum gave the lowest radiation content. Egyption white cement containd the lowest radiation level.
3. 3- The effective dose rates from all the samples are all at the acceptable level with maximum value of 450 μ Sv.y^{-1}.

According to the previous conclusion, we recommend the using of Tyma sand, the Yanbu gypsum and the Egyption white cement from among other building materials. While we recommend littele use of Kishani China and Ceramics in building materials, because of their relatively higher radiation content.

ACKNOWLEDGEMENTS

Thanks to ALLAH Most Gracious, Most Merciful, first and last who enables us to do this work. The investigators would like to acknowledge the financial support provided by Taif University, Saudi Arabia, Project number 1-432-1058. The acknowledgments would be also extended to all those unknowns who helped to bring this work to light.

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