STUDIES ON NATURAL RADIOACTIVITY OF SOME EGYPTIAN BUILDING MATERIALS

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Using high-resolution γ-rays spectrometry, the natural radioactivity of 14 samples of natural and manufactured Egyptian building materials have been investigated. The samples were collected from local market and construction sites. From the measured γ-ray spectra, specific activities were determined. The radium equivalent activity in each sample was estimated. Radiological evaluations of these materials indicate that all materials meet the external γ-ray dose limitation. Calculation of concentration indices by assuming a Markkanen room model is constructed from these materials, to find the excess γ-ray dose taken over that received from the outdoors. The Austrian Standard ÖNORM S 5200 is used in testing the building materials.

INTRODUCTION

The concentrations of the natural radionuclides in building materials vary significantly from country to another and from place to place in the same country. Precise measurement of radioisotope concentrations in building materials is vital in the assessment of possible radiological hazards to human health. This requires standardized equipment, which has good calibration and quality assurance programs as well as manufactured standards as references for these materials. Restricting the use of certain building materials might have significant economical, environmental, or social consequences locally and nationally. Such consequences, together with national levels of radioactivity in building materials, should be assessed and considered when establishing building regulations.

This paper presents the analysis of γ-ray spectra of ^{238}U, ^{232}Th and, ^{40}K in major building materials used in Egypt. For the sake of comparison, the results of the present work are compared with those from other countries [1, 2, 3] and the building materials were tested according to the requirements of the Austrian ÖNORM S 5200 standard [3].

THEORETICAL HINTS

When a radioactive series achieves secular equilibrium, the activity A in Bq of a parent radioisotope and any member of the series becomes constant:

$$N_P \lambda_P = N_D \lambda_D = A \qquad (1)$$

where N_P and N_D are the number of parent and any of its daughter radioisotopes of this series and λ_P and λ_D are their decay constants in second^{-1}, respectively.
Equation (1) can be written in terms of the element masses in grams of the parent and daughter radioisotopes m_P and m_D, respectively as

$$m_P\, f_P\, N_A\, \lambda_P / M_P = m_D\, f_D\, N_A\, \lambda_D / M_D = A, \quad (2)$$

where N_A is Avogadro's number, M_P and M_D are the gram atoms of the elements containing the parent and any of its daughter radioisotopes with elemental isotopic abundance f_P and f_D, respectively.
Or it can be writhen as

$$m_P\, R_P = m_D\, R_D = A, \quad (3)$$

where R_P and R_D are the activities in Bq due to the parent and any of its daughter radioisotopes per gram of their elements, respectively. If we refer to either R_P or R_D by R_i, then

$$R_i = \frac{f_i N_A \ln 2}{M_i T_{i1/2}} = \frac{A}{m_i}, \quad (4)$$

where M_i is the gram atom of the element containing the radioisotope i, f_i is its isotopic abundance, $T_{i1/2}$ is the relevant half-life time of this radioisotope, and m_i is the mass of the element containing the radioisotope i in the radioactive series.

Dividing equation (3) by the global sample mass m_s, the sample specific activity S is obtained. Then the concentration of the element containing the parent radioisotope $C_P = m_P / m_s$ can be estimated in terms of the concentration of the element containing the daughter radioisotope $C_D = m_D / m_s$ as follows:

$$C_P R_P = C_D R_D = A / m_s = S. \quad (5)$$

Thus the γ-ray specific activity $G_{i,e}$ is the product of the specific activity S and the intensity $\gamma_{i.e}$ of a γ-ray of energy e emitted by the radioisotope i a can be written as

$$G_{i,e} = S\, \gamma_{i.e} = C_i\, R_i\, \gamma_{i.e}. \quad (6)$$

In the uranium series, the decay chain segment starting from radium ^{226}Ra is radiologically the most important and, therefore, reference is often made to radium instead of uranium [1]. Since 98.5% of the radiological effects of the uranium is due to radium and its progeny product series. The contribution of ^{238}U and the other ^{226}Ra precursors are normally ignored [2,8]. The energy-dependent detection efficiency has been determined by means of a standard monazite sample provided by the Egyptian Nuclear Material Authority. The uranium and thorium concentrations C_i in mg/g of the standard monazite sample are 2.86 and 39.4, respectively. From equation (4), one may calculate the activity R_i of the parent radioisotopes ^{238}U, ^{232}Th and ^{40}K, present in 1 g of their natural elements as 12405, 4034.61, and 304.5 Bq/g, respectively. Then $A_{i,e}$ can be calculated for the observed γ-ray lines by substituting for C_i, R_i, and $\gamma_{i.e}$ in equation (6). Therefore, the absolute efficiency of the detector $\varepsilon_{i,e}$ for the radioisotope i at the γ-ray energy e is estimated by

$$\varepsilon_{i,e} = \frac{N_{i,e}}{G_{i,e}\, m_s\, t}, \quad (7)$$

where $N_{i,e}$ is the net gamma-ray counting under a peak at energy *e* emitted in *t* seconds live time by a radio nuclide *i* due to m_s g of the standard monazite sample.

The efficiency values were used in calculating the activity concentration S (in Bq kg^{-1}) from the counting rates for each of the detected full-energy peaks of a radioisotope i and for a gamma-ray energy e, is given by

$$S = \frac{N_{i,e}}{\varepsilon_e \times t \times \gamma_{i,e} \times m_s} \quad , \tag{8}$$

where $N_{i,e}$ is the total net peak area of the radioisotope i at energy e, m_s is the mass of sample in Kg, and t is the measurement live time in seconds.

EXPERIMENTAL

A high-resolution spectroscopic system is used for the measurement of the energy spectrum of the emitted gamma-rays in the energy range between 50 *keV* and 3000 *keV*. The system consists of a hyper-pure germanium (HPGe) detector with a relative efficiency of 40%, 1.9 keV FWHM at 1.3325MeV of ^{60}Co and peak to Compton ratio of 60:1 at the same gamma-ray line energy. Experimental arrangement for spectra collection includes the high-voltage detector bias supply and signal processing electronics. The latter include a spectroscopy main amplifier, which incorporates an efficient pile-up rejecter, and a Multi-Channel Buffer (MCB) which is a PC-based plug-in PCI card consisting of an 8k ADC. Advanced Multi-Channel Analyzer (MCA) emulation software (Genie-2000) allows data acquisition, storage, display, and online analysis of the acquired γ-spectra. The HPGe is shielded by surrounding it by a Canberra low background chamber model 747. It consists of a graded cylindrical shield efficient for suppressing the gamma-ray background at the laboratory site. 293.9 g of the standard monazite sand was sufficient to fill a plastic cup similar to that used in sample preparation. The cup was sealed, and stored for more than four weeks before counting in order to allow the achievement of secular equilibrium for ^{238}U and ^{232}Th with their respective progeny [4, 5, 6, 7]. A total of 14 samples of Egyptian building materials have been collected and are listed in Table (1).

The samples were crushed, pulverized to a fine powder (<300 mesh) and homogenized in preparation. A dry weighed quantity of powdered sample sufficient to fill a 200 ml plastic cup. The cup was sealed and left to stand-alone for a month to let the sample to attain secular equilibrium. The gamma-ray spectrum of each sample was measured for 24 hours. The environmental gamma-ray background at the laboratory site was measured for an empty plastic cup under identical measurement conditions. This was subtracted from the measured gamma-ray spectrum of each sample.

Table 1. Samples of building materials analyzed and their sources.

Sample no.	*Sample name*	*Sample source*
1	Portland cement !	Suez company
2	Portland cement 2	Helwan company
3	Sulfite resistance cement	Asyut company
4	White Portland cement 1	Helwan company
5	White Portland cement 2	Senai company
6	Concrete blocks	Construction site
7	Gypsum	Ballah company
8	Ceramic1	Cleopatra company
9	Ceramic 2	El-Gawhara company
10	Ceramic 3	Alpha company
11	Burnt clay bricks	Construction site
12	Burnt mud bricks	Construction site
13	Building sands	Construction site
14	Mud (river sands)	Nile river shore

RESULTS AND DISCUSSION

Building materials contribute to environmental radioactivity in two ways. First, by gamma radiation, mainly ^{226}Ra, ^{232}Th, and ^{40}K as well as their progenies to a whole body dose. In some cases by beta radiation to a skin dose, e.g., from tiles, glazed with uranium. Second, by releasing the noble gas radon, the radioactive daughters of which are deposited in the human respiratory tract [3].

Specific activities and concentrations of uranium, thorium, and potassium in each sample were determined from equations (8) and (5), respectively. The gamma-ray lines 609.3 keV and 583.1 keV gamma–lines of ^{214}Bi and ^{208}Tl [8] for ^{226}Ra (or ^{238}U) and ^{232}Th, respectively, were used. Alternatively, one could use the ^{214}Pb (351.9 keV) and ^{228}Ac (911.1 keV) gamma–lines. For ^{40}K the gamma–ray line 1.46 Mev [2, 9, 10] was used.

The activity concentration of a building material, which contain Ra, Th, and K, can be

estimated by a common index that called the radium equivalent activity Ra_{eq}. The radium equivalent activity is a weighted sum of activities of the above three radionuclides based on the estimation that 370 BqKg^{-1} ^{226}Ra , 259 BqKg^{-1} ^{232}Th, or 4810 BqKg^{-1} ^{40}K produce the same γ–ray dose rate [8]. Ra_{eq} is given by

$$Ra_{eq} = 370 \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \tag{9}$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, in BqKg^{-1}. To limit the external radiation dose from the building materials to 1.5 mSva^{-1}, various workers have suggested a number of models. One such model, proposed to serve a criterion in the Federal Republic of Germany at the beginning of the1980s [2], was

$$\frac{1}{37}\left[\frac{A_{Ra}}{10} + \frac{A_{Th}}{7} + \frac{A_K}{130}\right] \le 1 \, . \tag{10}$$

This criterion, which only considers the external hazard due to gamma-rays, corresponds to a maximum radium equivalent activity of 370 $BqKg^{-1}$ for material.

Another index called the activity concentration index (I) take, into account typical ways and amounts in which the material is used in building [1]. A methodology, which can be used to derive such index, for Markkanen model reference room (5mx4mx2.8m; wall, floor and ceiling thickness=0.2m and density=2320$kg.m^{-3}$) is described in [11]. The following activity concentration index is derive for identifying whether a dose criterion is met:

$$I = \frac{A_{Ra}}{300} + \frac{S_{Th}}{200} + \frac{A_K}{3000} \quad . \tag{11}$$

The activity concentration index (I) does not exceed the following values depending on the dose criterion and the way and the amount the material is used in a building.

Dose criterion	0.3 mSv a^{-1}	1 mSv a^{-1}
Materials used in bulk amounts, e.g. concrete	I≤0.5	I≤1
Superficial and other materials with restricted Use: tiles, board, etc.	I≤2	I≤6

This criterion was proposed to apply within the European Union, and only considers the external hazards due to gamma-rays. Doses exceeding 1$mSva^{-1}$should be taken into account from radiation protection point of view. Building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 $Sv.a^{-1}$. ^{222}Rn and its daughters have additional dose rate, to that from building materials [12], caused by inhalation. The Austrian standard ÖNORM S 5200, prepared and used in Austria since 1996, provides the criteria to assess the total radiation dose (external and internal). Gamma radiation of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are taken into account, as well as the dose due to the noble gas radon, which is released from the building materials after the decay of ^{226}Ra. A type of building material is considered acceptable if its yearly effective dose in a room does not exceed 2.5 m $Sv.a^{-1}$.

1. Building Material Testing by ÖNORM S 5200

For gamma dose rate and radon inhalation the standard ÖNORM S 5200 formulates three criteria, A, B, and C. In this study the test C was ignored since, this test concerned with the activity of beta particles of building material (e.g., uranium glazed tiles, etc.). To prove, a certain type of building material meets the requirements of the standard, the following tests are performed.

Test A

The measured values of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K for building material or building element/component parts are inserted into equation (9) leading to

$$\frac{A_K}{10000} + \frac{A_{Ra}}{1000} \times \left(1 + 0.15 \cdot \Sigma \cdot \rho \cdot d\right) + \frac{A_{Th}}{600} \le 1 \ , \tag{12}$$

where

ρ is the density of building material in $kg.m^{-3}$ with value not more than the precondition value 2000 $kg.m^{-3}$, Σ is the real emanation factor with value not more than the precondition value 0.1, and d is the real thickness with value not more than the precondition value 0.3m

If the inequality in (12) is fulfilled, the building material or building elements/component parts comply with the standard.

Test B

If the building material or building elements/component parts of a part of the surface in a room are known but fail to meet the requirements of test *A* and the building material of the other parts of the surface are also known and meet the requirements of test *A*, then test *B* can be applied.

Calculate the value of inequality (12) for the building material of every part of the surface of the room according to test *A*. No value is allowed to be more than 2, according to (12). The values have to be weighted and added up corresponding to the ratio of the surface of the room. If the result does not exceed 1, then material meet the regulation.

By using the precondition of values of the d, ρ, and Σ the building Egyptian materials had been tested by ÖNORM S 5200. Table 2 lists the measured activities of ^{226}Ra, ^{232}Th, and ^{40}K for collected samples calculated using equation (8). Ra_{eq} were calculated using equation (9) and the results of the application of the Austrian standard ÖNORM S 5200 by using inequality (12). For the sake of comparison, Table 2 includes values of the radioactivities in similar samples (serial numbers, 15 to 20) reported in literature. Although two samples, especially if they are from different locations, are unlikely to be identical, it may be seen that values of radioactivity found in this work are comparable to reported results. For example, the activities and concentrations (ppm) of our Portland cement (serial number 1 and 2) are within the range of values reported for cement (serial number 15). The same situation for our concrete (serial number 6) and the values reported for concrete (serial number 16). The burnt mud bricks being a mixture from mud (river sand) as a major component and straw as a minor one.

Thus the Ra_{eq} of the burnt mud bricks (serial number 12) and mud (serial number 14) are comparable.

Again, from Table 2 it is apparent that the Egyptian building materials meet two criterions, the first which is based on the Ra_{eq} concept and the second which is based on activity concentration index I. The second one takes into account typical ways and amounts in which the material is used in a building. All Egyptian building materials have Ra_{eq} lower than 370 Bq Kg^{-1} and as a result, the external radiation doses from these materials are less than 1.5 $mSva^{-1}$ [2]. It should, however, be pointed that addressing the radiation hazard to respiratory organs due to radon and its progeny requires reduction of the acceptable maximum Ra_{eq} to at most 185 Bq Kg^{-1} [2,13]. As shown in Table 2, of the fourteen types of samples analyzed only ceramic1, ceramics2, and ceramics3 have Ra_{eq} greater values than 185 Bq Kg^{-1}.

The values of the activity concentration index I, for materials used in bulk amounts in Table 2 are 0.18, 0.48, and 0.55 for concrete blocks (serial no. 6), burnt clay bricks (serial no. 11), and burnt mud bricks (serial no. 12), respectively.

Thus the external dose delivered by these materials are less than 0.3 $mSva^{-1}$ for concrete and burnt mud bricks and less than 1 $mSva^{-1}$ for burnt clay bricks. Moreover, the concrete and burnt mud bricks [having $I<0.5$] can be exempted from all controls concerning their radioactivity [1]. All superficial and other materials with restricted use in Table, have I value less than 2, consequently they have the exemption level [1].

Table 2. The concentration activities in Bq/Kg of building materials, the radium equivalent, the concentration index I, and the results of building material testing are in agreement with ÖNORM S 5200 Standard for the first twelve samples.

Material	*Uranium-238*		*Thorium-232*		*Potassium-40*		*Raeq*	*I*
	Bq/Kg	*ppm*	*Bq/Kg*	*ppm*	*Bq/Kg*	*ppm*		
1-Portland cement1	39±1	3.16	20±2	4.92	128±7	0.495	78±9	0.27
2-Portland cement 2	50±1	4.07	16±1	3.94	177±6	0.685	87±6	0.31
3- sulfate resistance Cement	25±1	2.03	10±1	2.46	136±6	0.526	50±6	0.18
4-White Cement 1	20±1	1.62	23±1	5.66	64±4	0.248	58±5	0.20
5- White Cement 2	25±1	2.03	16±1	3.94	66±5	0.255	53±6	0.19
6-Concrete blocks	12±1	0.97	10±1	2.46	269±6	1.04	61±8	0.18
7-Gypsum	6±1	0.49	7±1	1.72	112±6	0.433	25±6	0.09
8-Ceramic1	99±1	8.03	70±1	17.22	629±3	2.43	248±5	0.89
9-Ceramic2	101±2	8.19	69±2	16.98	1375±19	5.32	305±12	1.14
10- Ceramic3	114±2	9.25	86±3	21.16	1540±2	5.96	356±21	1.32
11-Brunt clay bricks	42±.1	3.41	43±.1	10.58	576±27	2.23	147±.02	0.55
12-Burnt mud bricks	32±1	2.60	33±1	8.12	638±11	2.47	128±.5	0.48
13-Sands	9±.4	0.73	8±.4	1.97	270±6	1.04	41±.07	0.16
14-Mud (river sands)	12±1	0.97	43±1	10.58	738±12	2.86	130±.08	0.50
15-Cement[14]	46	3.4	21	5.1	237	0.8		
16-Concrete[14]	11	0.8	8.5	2.1	385	1.3		
17-Gypsum[14]	15	1.1	7.4	1.8	148	0.5		
18-Gypsum[1]	10	0.8	10	2.5	80	0.31		
19-Clay bricks[14]	111	8.2	44	10.8	666	2.3		
20-Clay bricks[1]	50	4.1	50	12.3	670	2.6		

The results of the application of the Austrian standard show that all Egyptian "building material" or building materials/component parts in room," agree with ÖNORM S 5200 standard. Only ceramics materials are "accepted" by applying test B, whereas the other materials "pass" through test A.

Table 3. Comparison of mean radium equivalent activities (Bq/kg) of Egyptian building materials with those from other countries.

Material	*Egypt (2003)*	*Zambia (1994) [8]*	*Australia (1985) [2]*	*Germany (1981) [2]*	*Finland (1979) [2]*	*Sweden (1979) [2]*	*UK (1977) [2]*	*Norway (1976) [2]*
Portland Cement	82.5	79	115	70	100			
Concrete block	61	94	85	155				133
Clay brick	147		218	207	240	352	170	274
Red mud – clay brick	128	180	833	640				
Gypsum	25	63	15	41	11	7	44	15
Sand	41	135	70	59	177		19	

Table 3 has been compiled to show the radioactive status of Egyptian building materials in comparison to those of other countries. As stated earlier and shown in Table 3, the radioactivity in building materials does vary from one place to the other.

If we consider the case of clay bricks, the Ra_{eq} value is 147 for Egypt, 218 for Australia, 207 for Germany, 240 for Finland, 352 for Sweden, 170 for UK, and 274 for Norway. It should be stressed here that even these values are not the representative values for the countries mentioned but for the locations from where the samples had been collected.

CONCLUSIONS

All building materials contain different portions of natural radioactive nuclides. Materials derived from rock and soil contain mainly natural radioisotopes of the uranium ^{238}U and thorium ^{232}Th series, and radioactive isotope of potassium ^{40}K.

High-resolution γ-ray spectrometry is a powerful experimental tool in studying natural radioactivity and determining elemental concentrations in various building materials. A combination of the experimental results and mathematical models makes its possible to assess and limit the external, the internal, and the total dose rates caused by building material if a building constructed using them.

The obtained results show that the most majority of the Egyptian building materials have the exemption level, thus they can be exempted from all controls concerning their radioactivity.

The results also show that Egyptian building materials agree with ÖNORM S 5200 standard. Thus, from the radiation safety, these materials are below the recommended limits for their gamma dose rates; therefore, they can be used for all kinds of republic buildings.

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