Indian Journal of Pure & Applied Physics
Vol. 49, March 2011, pp. 157-161

Radiological significance of Egyptian limestone and alabaster used for construction of dwellings

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Received 18 March 2010; revised 26 December 2010; accepted 11 January 2011

The natural radionuclides in limestone and alabaster found in Assuit Governorate in Upper Egypt have been investigated by passive gamma-ray spectrometry. From the measured γ-ray spectra, specific activities were determined. The measured activity concentrations for these natural radionuclides were compared with the reported data from other countries. The radiation hazard parameters related to the exposure of limestone and alabaster have also been calculated. The obtained results of radium equivalent Raeq, level index $I\gamma$r, the external hazard index H_{ex} and absorbed dose rate in limestone (90.44, 0.63, 0.17, 39.94) and (70.86, 0.50, 0.13, 31.55) are lower than the acceptable level 370 Bqkg^{-1} for radium equivalent Ra$_{eq}$, 1 for level index $I\gamma$r, the external hazard index $H_{ex} \leq 1$ and 59 (nGy.h^{-1}) for absorbed dose rate. So limestone and alabaster can be used as building construction without exceeding the proposed radioactivity criterion level.

Keywords: Natural radioactivity, Limestone, Alabaster, Radiation hazards

1 Introduction

Natural radioactivity generates from extra-terrestrial sources as well as from radioactive elements in the earths crust. About 340 nuclides have been found in nature of which about 70 are radioactive and found mainly among the heavy elements. The natural radioactivity is common in rocks, soils, water and building materials. There has been increased interest of the public at large in knowing information about the presence of naturally occurring radionuclide such as U, Th and K and their decay products in the construction materials such as bricks, marble[1].

The ancient Egyptian used alabaster as subsidiary building material, mainly for the lining of passages and rooms. Alabaster and limestone contain some radioactive materials which emit a constant stream of gamma-rays. Passive detection systems measure these spontaneous emissions without applying any external radiation or particle beams. The radiological hazards linked with natural radioactivity are not so harmful, however some radiation protection problems can occur in industrial processes involving the treatment of large quantities of slightly radioactive materials. At Luxor city in Upper Egypt, alabaster is used in industrialization of some states, vases. Large amount of alabaster ore samples is used in construction of the most famous Egyptian mosque such as Amro Ebn El-ass which includes large alabaster volumes[2].

Due to health risks associated with the exposure to indoor radiation, many International Commission on Radiological Protection (ICRP), the World Health Organization (WHO) etc, have adopted strong measures aimed at minimizing such exposures. In many countries, limits have been set on the concentration of radionuclides in various building materials and the use of materials with high levels of activity has been banned.

The assessment of natural radioactivity for (^{226}Ra, ^{232}Th and ^{40}K) in limestone and alabaster found in Assuit Governorate, Upper Egypt, has been studied in the present paper. The radiological parameters (radium equivalent activity Ra$_{eq}$, level index $I\gamma$r, external hazard index H_{ex} and absorbed dose rate) have been calculated which are related to the external γ-dose rate. The results of analysis have been compared with world reported value for the same studies in other countries.

2 Experimental Details

2.1 Sampling and sample preparation

Thirty representative limestone and alabaster samples have been collected from Assuit Governorate in upper Egypt for investigation: limestone samples from El-Nawawrra, El- Wady El Assuity and beside Assuit cement factory; Alabaster samples from Arb El-Attyat El-Baharia, El-Wady El-Assuity and Al-Matar. Each sample about 1 kg in weight was washed in distilled water and dried in an oven at about 110°C to ensure that moisture is completely removed, The samples were crushed, homogenized, and sieved through a 200 mesh, which is the optimum size enriched in heavy minerals. Weighted samples were placed in polyethylene beaker, of 350 cm^3 volume each. The beakers were completely sealed for 4 weeks to reach secular equilibrium where the rate of decay of the daughters becomes equal to that of the parent. This step is necessary to ensure that radon gas is confined within the volume and the daughters will also remain in the sample.

2.2 Instrumentation and calibration

Activity measurements were performed by gamma ray spectrometer, employing a scintillation detector 3"×3". It is hermetically sealed assembly which includes a NaI(Tl) crystal, coupled to PC-MCA Canberra Accuspes. To reduce gamma ray background, a cylindrical lead shield (100 mm thick) with a fixed bottom and movable cover shielded the detector. The lead shield contained an inner concentric cylinder of copper (0.3 mm thick) to absorb X-rays generated in the lead. In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 43200 s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program[3] from Canberra has carried out the online analysis of each measured γ-ray spectrum.

The efficiency calibration curve was made using different energy peaks covering the range up to ≈2000 keV. Measurements were performed with calibrated source samples, which contain a known activity of one or more gamma-ray emitters of the radionuclides ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1 keV), ^{137}Cs (661.9 keV) and ^{226}Ra (1764.49 keV). With certified accuracies of ≤2% supplied by PTB Braunschweig, Germany. We used Eq. (1) for calculating the absolute efficiency.

$$E_{\mathrm{eff}} = \frac{100 \cdot N_{\mathrm{p}}}{I_\gamma \cdot TOC \cdot A_{\mathrm{BOC}}} \quad \ldots(1)$$

where N_{p} is the net peak area (count/S) at E_γ, I_γ the intensity of emitted γ-ray (%), *TOC* is the time of counting(S), and A_{BOC} the activity (Bq) of the standard source at beginning of counting (*BOC*). A_{BOC} was calculated by Eq. (2):

$$A_{\mathrm{BOC}} = A_{\mathrm{DOC}} \cdot \exp\,[-\lambda \cdot (BOC - DOC)] \quad \ldots(2)$$

where A_{DOC} is the activity (Bq) of the standard source at date of calibration *DOC*, and $\lambda(s^{-1})$ is the decay constant.

Daily efficiency and energy calibrations for each sample measurement were carried out to maintain the quality of the measurements. The absolute efficiency of the detector was calculated at the specific energy of the standard sources for the same geometry of the samples. But, γ-spectra of the samples have different γ-energies. So, we need some fitting function to calculate the absolute efficiency for any considered γ-energy. A function is used for calculating the absolute efficiency[4] at any gamma-energy of interest in the energy range below 2000 keV, which is in the following form:

$$\eta = a - b \times \exp(-c \times E_\gamma^{\mathrm{d}}) \quad \ldots(3)$$

where E_γ represents energy in MeV, *A,b,c* and *d* are coefficient data. By Eq (3), the absolute efficiency, η, at any specific energy E_γ, the energies and coefficient data have been determined. From the experimental efficiency curves, the coefficient data were determined by using the curve-fitting program[5].

2.3 Uncertainty of efficiency

The combined standard uncertainty of absolute efficiency $u(EFF)$ consists of $u(N_{\mathrm{p}})$, $u(I_\gamma)$, $u(TOC)$ and $u(A_{\mathrm{BOC}})$ so,

$$\left(\frac{u(EFF)}{EFF}\right)^2 = \left(\frac{u(N_{\mathrm{p}})}{N_{\mathrm{p}}}\right)^2 + \left(\frac{u(I_\gamma)}{I_\gamma}\right)^2 + \left(\frac{u(TOC)}{TOC}\right)^2 + \left(\frac{u(A_{\mathrm{BOC}})}{A_{\mathrm{BOC}}}\right)^2 \quad \ldots(4)$$

Because $u(TOC) \ll TOC$, we neglected $u(TOC)$. The value of $u(A_{\mathrm{BOC}})$ was calculated by Eq. (5)

$$\left(\frac{u(A_{\mathrm{BOC}})}{A_{\mathrm{BOC}}}\right)^2 = \left(\frac{u(A_{\mathrm{DOC}})}{A_{\mathrm{DOC}}}\right)^2 + (BOC - DOC)^2 \cdot u^2(\lambda) \quad \ldots(5)$$

We got $u(N_p)$ from the code Genie 2000, while $u(\lambda)$ and $u(I_\gamma)$ were taken from Ref. (6). The calibration standards used had a certified accuracy of ≤ 2%. By measuring several times, it could be verified with a total uncertainty of the full-energy-peak efficiency of 5%.

2.4 Calculation of activity

Calculations of count rates for each detected photo peak and radiological concentrations (activity per mass unit or specific activity) of detected radionuclides depend on the establishment of secular equilibrium in the samples. The ^{232}Th concentration was determined from the average concentrations of ^{212}Pb (238.6 keV) and ^{228}Ac (911.1 keV) in the samples and that of ^{226}Ra was determined from the average concentrations of ^{214}Pb (351.9 keV), ^{214}Bi (609.3 and 1764.5 keV) decay products[7].

The activity concentration in Bqkg^{-1} (A) in the environmental samples was obtained as follows:

$$A = \frac{N_{\mathrm{p}}}{e \times \eta \times m} \qquad \ldots(6)$$

where N_{p} is the (cps) sample-(cps) BG, e the abundance of the γ-line in a radionuclide, η the measured efficiency for each gamma-line observed for the same number of channels either for the sample or the calibration source and m is the mass of the sample in kilograms.

The uncertainty of activity $u(A)$ was calculated by the following equation:

$$u(A) = A\sqrt{\left(\frac{u(N_{\mathrm{p}})}{(N_{\mathrm{p}})}\right)^2 + \left(\frac{u(\eta)}{(\eta)}\right)^2 + \left(\frac{u(m)}{(m)}\right)^2} \qquad \ldots(7)$$

Eq. (7) indicates that there are many sources of uncertainties of the activity and some may result in considerable uncertainties. The following sources of uncertainties were considered:

The uncertainty of each single net-peak area is determined by the spectrum-evaluation code. It takes into account the Poisson uncertainties of the counts in the individual channels as well as the uncertainty of the background determination. Sometimes a peak cannot be attributed unambiguously to a single nuclide. If it seems that the contributions of other nuclides to a peak are very small, no correction was applied. Due to this procedure, a maximum inaccuracy of 2% is assumed due to contributions of other nuclides but it must be pointed out that on the average this uncertainty should be smaller. By repeated measurements, it could be verified that the total uncertainty of the efficiency calibration was 5%.

2.5 Estimation of dose rate

Conversion factors to transform specific activities A_{K}, A_{Ra} and A_{Th} of K, Ra and Th, respectively in absorbed dose rate at 1 m above the ground (in nGy h^{-1} by Bq kg^{-1}) are calculated by Monte Carlo method and the values are:

$$D(n\mathrm{Gyh}^{-1}) = 0.0417A_{\mathrm{K}} + 0.462A_{\mathrm{Ra}} + 0.604\,A_{\mathrm{Th}} \qquad \ldots(8)$$

In natural environmental radioactivity situations, the effective dose is calculated from the absorbed dose by applying the factor 0.7 SvGy^{-1}

$$\text{Indoor: Dose } (n\mathrm{Gy.h}^{-1}) \times 8.760 \times 0.8 \times 0.7\ \mathrm{SvGy}^{-1} \qquad \ldots(9)$$

$$\text{Outdoor: Dose } (n\mathrm{Gy.h}^{-1}) \times 8.760 \times 0.2 \times 0.7\ \mathrm{SvGy}^{-1} \qquad \ldots(10)$$

The annual effective dose rate[8] outdoors in units of µ*Sv/y* is calculated by the following formula:

$$\text{Annual Effective Dose Rate} = D \times T \times F \qquad \ldots(11)$$

where D is the calculated dose rate (in nGy h^{-1}), T the outdoor occupancy time (0.2×24 h ×365.25 d≈1753 h y^{-1}), and F is the conversion factor (0.7×10^{-6} Sv Gy^{-1}).

2.6 γ-ray radiation hazard indexes

The natural radioactivity of building materials is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents. As 98.5% of the radiological effects of the U series are produced by Ra and its daughter products, the contribution from the ^{238}U has been replaced by the decay product ^{226}Ra. Since sand beach minerals, rejected light sands and sea beach soils can be used in industries and building constructions, the γ-ray radiation hazards due to the specified radionuclides were assessed by three different indices[9]. Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them. This first index can be calculated according to Beretka and Mathew[10] as:

$$\mathrm{Ra_{eq}} = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_{\mathrm{K.}} \qquad \ldots(12)$$

where A_{Ra}, A_{Th} and A_{K} are the specific activities of ^{226}Ra, 2^{32}Th and ^{40}K in Bqkg^{-1}, respectively. The Ra$_{\mathrm{eq}}$

is related to the external γ-dose and internal dose due to radon and its daughters. The maximum value of Ra_{eq} in building materials must be less than 370 Bqkg^{-1} for safe use. Beretka and Mathew[10] defined two other indices that represent the external and internal radiation hazards. The external hazard index is obtained from Ra_{eq} expression by assuming maximum value allowed (equal to unity) which corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}). This index value must be less than unity in order to keep the radiation hazards insignificant; i.e. the radiation exposure due to the radioactivity from construction materials is limited to mSv y^{-1}. Then, the external hazard index can be defined as:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad \ldots(13)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}, respectively.

3 Results and Discussion

The average values of activity concentration of ^{226}Ra,^{232}Th and ^{40}K in Bqkg^{1} for limestone samples from El-Wady El Assuity, beside Assiut cement factory and Kaw El-Nawawrra and alabaster samples collected from area near Assuit airport, El-Wady El-Assuity and Arab El-Attyat El-Baharia were measured and presented in Table 1.

From the obtained results, following observations can be recorded:

For limestone (1) — The lowest values of ^{226}Ra activity concentrations are found in samples collected from area beside the cement factory while the highest values are found in El-Wady El Assuity samples. (2) The lowest values of ^{232}Th activity concentrations are found in limestone samples collected from area beside cement factory while the highest values are found in El-Wady El Assuity samples, and (3) For ^{40}K, the lower values are found in limestone samples collected from area beside cement factory and the higher values in El-Wady El Assuity samples.

For alabaster (1) — The lowest values of ^{226}Ra activity concentrations are found in El-Wady El-Assuity samples while the highest values are found in El-Attyat samples. (2) The lowest values of ^{232}Th activity concentrations are found in El-Wady El-Assuity samples while the highest values are found in El-Attyat samples and (3) for ^{40}K, the lower values are found in El-Wady El-Assuity samples and the higher values in Al-Matar samples.

The average values of the measured radionuclide in limestone and alabaster are below the world averages for building materials 50, 50 and 500 (Bq.kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively[9,11]. The activity concentrations due to ^{226}Ra, ^{232}Th and ^{40}K for limestone in the present study have been compared with other studies which are presented in Table 2. The radioactivity in limestone varies from one country to another. The activity concentration of ^{232}Th in the present study is higher than the most corresponding values in other countries. While the activity concentrations of ^{226}Ra and ^{40}K, in the present study are comparable with the corresponding values of other countries. The radionuclide concentrations of our samples are below than the world averages for building materials 50, 50 and 500 (Bq.kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively[9,11].

3.1 Calculation of radiological effects

The average values of calculated radium equivalent activity, level found in index $I\gamma r$, the external hazard index Hex and absorbed dose rate for the limestone samples collected from El-Wady El Assuity, beside Assiut cement factory and Kaw El-Nawawrra and alabaster samples collected from area near Assuit

Table 1 — Average values of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{1} for limestone and alabaster

Sample location	Type of rock	^{226}Ra	^{232}Th	^{40}K
El-Wady El-Assuity	Limestone	27.83±4.02	46.64±2.34	66.49±3.32
Beside Assuit cement factory	Limestone	19.73±2.88	39.04±1.96	61.16±3.05
El-Nawawrra	Limestone	25.93±4.81	43.35±2.17	62.22±3.11
Almater	Alabaster	17.42±2.90	30.82±1.57	81.26±4.06
El-Wady El-Assuity	Alabaster	15.85±3.36	12.34±0.69	60.76±3.03
El-Attyat	Alabaster	26.06±3.61	53.81±2.70	65.84±3.29

Table 2 — Comparison between the activity concentrations of ^{226}Ra,^{232}Th and ^{40}K for limestone in the present study with that of other countries

Country	Activity (Bqkg^{1})			Reference
	^{226}Ra	^{232}Th	^{40}K	
Egypt (Assiut)	24.50	43.01	63.2	Present work
Australia	----	11.1	----	(12)
Austria	9.0	2.8	34.0	(12)
Italy	11	2	22	(13)
Brazil	24.3	7.0	205	(14)
India	73.9	—	64.6	.(15)
Bangladesh	68	106	1660	(16)
Algeria	16	13	36	(17)

Table 3 — Average values of radiation hazard parameters for limestone and alabaster

Sample location	Type of rock	Ra_{eq} (Bq.kg^{-1})	Dose rate (nGy.h^{-1})	H_{ex}	Iγr
El-wady El-Assuity	Limestone	99.19	43.80	0.19	0.69
Cement Factory	Limestone	79.85	35.25	0.16	0.56
Kaw El-Nawawrra	Limestone	92.28	40.76	0.18	0.64
Al-Matar	Alabaster	67.19	30.06	0.13	0.47
El-Wady El-ssuity	Alabaster	37.76	17.31	0.06	0.26
Arb El-Attyat	Alabaster	107.63	47.29	0.22	0.75

airport, El-Wady El-Assuity and Arab El-Attyat El-Baharia are presented in Table 3.

From the results the following observations can be recorded:

For limestone — The highest values of radiation hazard parameters (99.19 Bq.kg^{-1}, 0.69, 0.19 and 43.80 nGy.h^{-1} of radium equivalent activity Ra_{eq}, level index $I\gamma$r, the external hazard index H_{ex} and absorbed dose rate, respectively) are found in El-Wady El-Assuity samples. The lowest values parameters (79.85 Bq kg^{-1}, 0.56, 0.16 and 35.25 nGy.h^{-1}) are found in limestone from area near Assiut factory. The obtained results show that the averages of radiation hazard parameters for all samples under investigation (radium equivalent Ra_{eq}, level index $I\gamma$r, the external hazard index H_{ex} and absorbed dose rate (90.44, 0.63, 0.17, 39.94) are lower than the acceptable level 370 Bq/kg for radium equivalent Raeq, 1 for level index $I\gamma$r, The external hazard index $H_{ex} \leq 1$ and 59 (nGy.h^{-1}) for absorbed dose rate[18-].

For alabaster — The highest values of radiation hazard parameters (107.62 Bq.kg^{-1}, 0.75, 0.22 and 47.29 nGy.h^{-1} of radium equivalent Ra_{eq}, level index $I\gamma$r, the external hazard index H_{ex} and absorbed dose rate, respectively) are associated with Arb El-Attyat samples. The lowest values of radiation hazard parameters (37.76 Bqkg^{-1}, 0.26, 0.06 and 17.31 nGy.h^{-1}) are found in El-Wady El-Assuity samples. The obtained results show that the averages of radiation hazard parameters for Alabaster samples under investigation (70.86, 0.50, 0.13, 31.55) are lower than the acceptable level 370 Bq.kg^{-1} for radium equivalent Ra_{eq}, 1 for level index $I\gamma$r and 59 (nGy.h^{-1}) for absorbed dose rate [24-27].

4 Conclusions

The activity concentration of natural radionuclides and its radiological hazards in limestone and alabaster rock samples found in Assuit Governorate in upper Egypt were measured. The average values of ^{226}Ra and ^{232}Th for limestone are higher than that of alabaster. While the average highest values of ^{40}K in alabaster are higher than that of limestone. The measured values of natural radionuclides in (limestone and alabaster) are less than the acceptable values of the world averages for building materials 50, 50 and 500 (Bq.kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K, respectively[9,11]. The obtained results show that the averages of radiation hazard parameters for limestone (90.44, 0.63, 0.17, 39.94) and for alabaster (70.86, 0.50, 0.13, 31.55) are lower than the acceptable level 370 Bq.kg^{-1} for radium equivalent Ra_{eq}, 1 for level index $I\gamma$r and 59 (nGy.h^{-1}) for absorbed dose rate. Therefore, it can be used in building construction without exceeding the proposed radioactivity criterion level.

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