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A. EL-SHERSHABY*

# ENVIRONMENTAL ISOTOPES AND NATURAL RADIOACTIVITY ASSESSMENT FOR CLAYS, PRODUCTS DERIVED FROM CLAY AND RADON EXHALATION RATE OF CLAYS IN EGYPT

Scientific investigations have long concluded that prolonged exposure to low dose radiation can induce deleterious effects in humans. The aim of this paper is to investigate the radioactivity of different types of clay and clay derived products, such as red brick and jars to preserve oils and organs, a seed pot to protect seeds from rodents' insects and mildew, a vase for decoration, a vessel for cooking, a storage jar for storing food, dough bowls for raising wheat breads and olla or bottle for storing water, which are suspected to have natural radioactivity radiation risk in Egypt. Natural radioactivity content was determined in different types of clay using a hyperpure germanium spectrometer (HPGe). The gamma dose of the radiations emitted from the clays is proportional to their U, Th, and K concentration. The concentration ranges were 6.77–70.73 Bq/kg, 7.74–99.26 Bq/kg and 172.34–776.98 Bq/kg for $^{238}U$, $^{232}Th$ and $^{40}K$, respectively. The radium equivalent activity and the absorbed dose rate were found to be from 29.9 to 247.98 Bq/kg and 15.42 to 117.6 nGy/h, respectively. The representative level index ranged from 0.2 to 0.67.

The solid state nuclear track detectors, CR-39, have been devised to measure radon exhalation rate in the samples, which were found to vary from 3.19 to 33.33 Bq $m^{-2}d^{-1}$ and these values are in agreement with the uranium concentration values measured by hyperpure germanium detector in the corresponding samples. This study has shown that the different types of clay and the fabricated clay have traces of radioactivity. However, the associated levels are not detrimental to health. In terms of the radiation safety, the natural radioactivity of the Egyptian clays is below the recommended limits of the gamma dose rate. Therefore, they can be used for all kinds of public buildings and all types of pottery.

## 1. INTRODUCTION

Naturally occurring radioactive material (NORM) is the most important source of radiation a human is exposed to. Although NORM contains low level radioactivity (LLR), the accumulation and production of large amounts of NORM wastes in industrial activities produces high doses of radiation to man. So, in recent years, the measurements of natural radioactivity have stirred up great interest due to the hazards con-

* Nuclear Physics Laboratory, Faculty of Girls, Ain-Shams University, Cairo, Egypt.

nected with of exposure to radiation. Measurements of the radiation exposure and radiation levels have become widespread.

The knowledge of the radioactivity is of primary importance for estimation of environmental data as well as for eventual radioactive contamination during the nuclear accidents. The radioactive elements are distributed everywhere and in the different concentrations in the biosphere. From the point of view of the natural "risk", it is necessary to know the dose limits of public exposures. At the same time, it is necessary to measure the natural environmental radiation level provided by ground, air, water, foods, clays, building interiors, etc., for the estimation of exposures to natural radiation sources. IAEA [1] has published data for the doses accumulated by human beings during their life activities.

The clays have good absorbing features, especially as clay colloids, they also contain natural radioactive elements and rare earth elements. As regards radioactive concentration, the "normal" clays which are used in pottery (such as a bottle for drinking, a vessel for cooking and a vase for decoration) and in construction materials for residential and public buildings, etc., contain some ppm of U, Th, and K. In case the concentration of these radioactive elements in clays is higher than 10 ppm for U, 20 ppm for Th, and higher than 5% for K, it is necessary to consider the possibilities of limiting the use of such clays in construction materials. The reason for this limitation is that human beings are exposed to high-intensity radiation created by radioactive materials. In general, the total natural radioelement concentration of clays for the residential and public use should not exceed 30 ppm of $U_e$, (uranium equivalent), see DODONA [2]. In building industry, the majority of raw materials are clays (production of bricks, tiles, ceramics, fillers, etc.).

Raw materials contribute to environmental radioactivity in two ways. First, by gamma radiation, mainly U, Th, K and their progenies to a whole body dose and in some cases by beta radiation to a skin dose, e.g., from tiles, glazed with uranium, and secondly by releasing the noble gas radon, the radioactive daughters of which are deposited in human respiratory tract, STEGER et al. [3].

For more than 60 centuries brick has been the predominant component of the walls of public buildings in Egypt. The bricks in Egypt are made mainly of clay originating from sediments deposited by the river Nile over millennia during the annual floods at the end of summer. Since 1970, the date of completion of the Aswan high dam at the very southern part of Egypt, some other types of bricks have been fabricated and used, although the percentage of these types is still low, EL-TAHAWY et al. [4].

From the age of the ancient Egyptians up to now, clays have been used in pottery (e.g., jars to preserve oils and organs, a seed pot to protect seeds from rodents' insects and mildew, a vase for decoration, a storage jar for storing food, dough bowls for raising wheat breads and olla or bottle for storing water). Today, pottery is often made by individual potters working alone. For many, pottery has become their principal source of income as well as their art.

In a few areas of the world, radiation from natural background is due to the presence of large amounts of radioactive materials in soil or building materials from which houses are constructed and different types of clays from which pottery, vessels and bricks are constructed. Most of these radionuclides are members of radioactive decay chains beginning with $^{238}U$, $^{235}U$ and $^{232}Th$. Radiation emitted from such radionuclides and their daughters is taken up by plants and consequently absorbed in the food chain. In order to study radiation background, which is changing with these developments, it is necessary to establish base-line data on radiation and radioactivity background. This makes it possible to detect any new contamination in time and take appropriate measures to protect the environment. The dose due to the natural sources of radiation has been evaluated by UNSCEAR [5].

Public exposure to radon and its radioactive daughters present in the environment results in the largest contribution to the average effective dose received by human beings, UNSCEAR [6]. In a natural radiation environment, radon gas and its airborne progency can be a signifcant internal health hazard through the inhalation, especially when uranium or radium content in the soil is high or the radon and its daughters are concentrated in enclosed area. In order to evaluate the radon risk, it is necessary to check the relation between the activity concentration of uranium and radon for the same samples.

The introduction of low-level gamma-ray spectrometry technique has become very important in various fields, especially geology and environmental science. By applying gamma-ray spectrometry, the various radionuclides in the samples can be identified quantitatively.

This study was undertaken with the purpose of measuring radioactivity in different types of Egyptian clay and fabricated clay. The activity concentration of U, Th and K was measured by gamma-ray spectroscopy. The absorbed dose rate in air, the radium equivalent activity and the representative external hazard index level in the samples investigated were also estimated. The radon concentration and the exhalation rate were measured also for the fabricated samples using solid state nuclear track detector (CR-39).

The results obtained are considered as a base-line level and as reference values that can be used in emergency action. It is also considered as data base for the Egyptian radiation monitoring network.

## 2. EXPERIMENTAL METHOD

### 2.1. SAMPLING AND SAMPLE PREPARATION

Eight types from the Egyptian clay and five types from the fabricated clay were investigated. These are: the first type is Aswanly clay from Aswan, the second type in also Aswanly clay from another location in Aswan, the third type is Tebien clay from

El-Tebien, the fourth type is Pokla clay from Aswan, the fifth type is soil clay from El-Geiza and Qalube city, the sixth type is a mixture of Aswanly and selika for fabrication of bowls, the seventh and eighth types are Safy and Taffla clay from El-Geiza city. The fabricated clays are: No. A is a bowl for storing milk, No. B is red brick, No. C is a vase for decoration, No. D is a bottle for water (olla), and No. E is vessel (tagen or peram) for cooking. Five samples from each type were measured to calculate the average concentration of U, Th, and K.

The samples, each about 1 kg in weight, were ground, homogenized and sieved to about 200 mesh, then dried at 110 °C for 48 hours to ensure that moisture is completely removed. Weighed samples were placed in polyethylene bottles of 350 $cm^3$ volume. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached between $^{238}U$ and $^{232}Th$. This step was necessary to ensure that radon gas was confined within the volume and that the daughters would also remain in the sample.

The individual samples were placed on the detector manually and each sample was analyzed for a time of 70000 seconds. The gamma emitting radionuclides recorded were: $^{238}U$, $^{232}Th$ and $^{40}K$.

## 2.2. EXPERIMENTAL SET-UP FOR GAMMA-RAY SPECTROMETRY

Radioactivity measurements were performed by gamma-ray spectrometry, employing a coaxial hyperpure germanium detector and multi-channel analyser. The detector has a resolution of 1.85 keV for the 1332.5 keV gamma-ray transition of $^{60}Co$ with 30% photopeak efficiency. To reduce the gamma-ray background, a cylindrical lead shield with a fixed bottom and a movable cover shielded the detector. The lead shield contained two inner concentric cylinders of copper and cadmium.

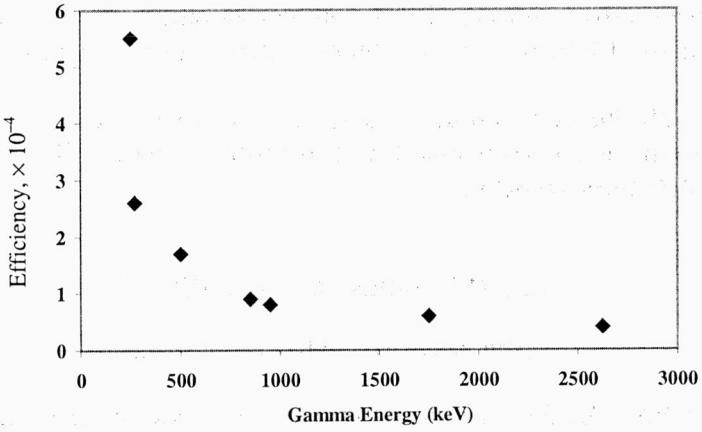


Fig. 1. The photopeak efficiency of HPGe detector

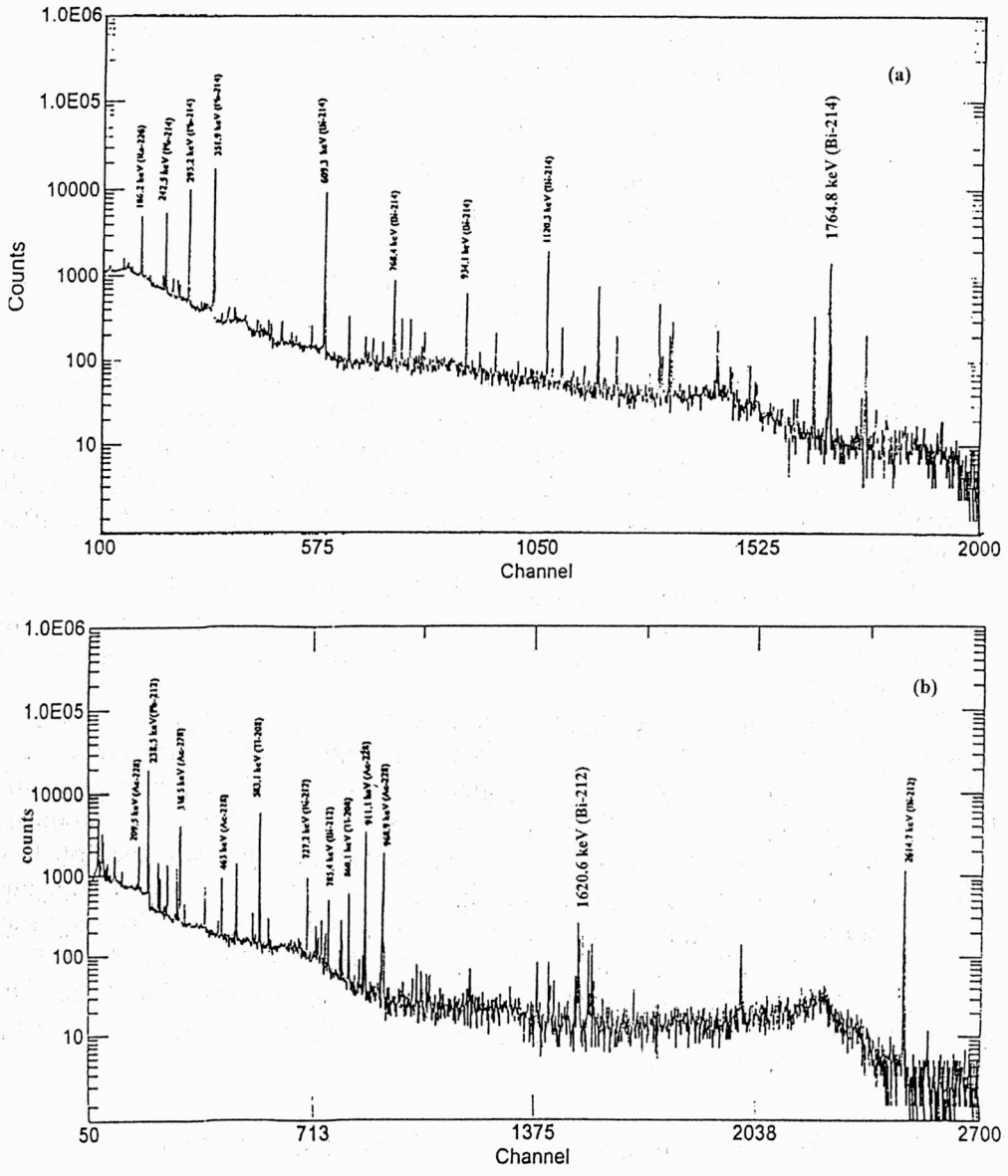


Fig. 2. The energy spectra for the standard natural U-238 series (a) and Th-232 series (b)

The detection system was calibrated using $^{60}$Co, $^{137}$Cs, $^{226}$Ra and $^{241}$Am point sources. The absolute detection efficiency of the HPGe detector was determined by using standard $^{238}$U with activity of 2120.37 Bq (420 mg) and standard $^{232}$Th with activity of 1333.96 Bq (410 mg), and KCl with activity of 15.9 Bq, IAEA [7]. Each of these standard sources was placed in the same identical polyethylene bottles with the

same volume of 350 $cm^3$. The absolute efficiency of the detector for each gamma ray energy was then calculated from the formula

$$\varepsilon = z/AI \times 100\% \quad [\text{cps/Bq}], \tag{1}$$

$\varepsilon$ is the absolute photopeak efficiency, $z$ is the net count per second for the standard, $A$ is the activity of the radionuclides in Bq and $I$ is the absolute intensity of the gamma transition.

The efficiency curve for the HPGe detector is shown in figure 1. In figure 2(a) and (b), energy spectra obtained for standard natural $^{238}U$ and $^{232}Th$ series, respectively, are given.

The gamma transitions of energies of 186 keV ($^{226}Ra$), 351.9 keV ($^{214}Pb$), 609.3 keV ($^{214}Bi$) and 1120.3 keV ($^{214}Bi$) were used to determine the concentration of $^{238}U$ series. The gamma transitions of energies of 338.0 keV ($^{228}Ac$), 583.1 keV ($^{208}Tl$) and 911.2 keV ($^{228}Ac$) were used to determine the concentration of $^{232}Th$ series, and the 1460 keV gamma transition was used to determine the concentration of $^{40}K$ in different samples.

In order to determine the background distribution in the environment around the detector, an empty bottle was counted in the same manner and in the same geometry as the samples. The back ground spectra were used to correct the areas of gamma rays of measured isotopes.

The 186 keV gamma transition gave apparent radium concentration that was too high because of the contribution of $^{235}U$ which emits gamma rays of the same energy. The 351.9 keV and 609.3 keV peaks gave apparent radium concentration that was too low because of the emanation of radon ($^{214}Pb$ and $^{214}Bi$ are radon decay products) from materials; the results were corrected for these phenomena. For the 186 keV peak, it was assumed that 58% (KAHAN et al. [8]) of the gamma-ray activity was due to $^{226}Ra$, and the concentrations were divided by 1.72. For $^{214}Pb$ and $^{214}Bi$ the mean emanation coefficient was assumed to be equal to 0.86% (KOMINEK and MRNUSTIK [9]) and the concentrations calculated from these peaks were multiplied by 1.094. The results obtained from the 186 keV peak and the 351.9 and 609.3 keV peaks were generally in good mutual agreement.

### 2.3. EXPERIMENTAL SET-UP FOR RADON MEASUREMENTS

Solid-state nuclear track detectors are widely applied because of their availability, low cost, and ease of operation. For integrated measurements of radon concentration a variety of dosimeters having different shapes are used. CR-39 detectors used in this work were supplied by Pershore Modlings, Ltd., U.K, in the form of large sheets cut into 2 cm × 2 cm squares. Each sample was placed in a glass cylinder chamber of radius 3.5 cm and 10 cm in length. Dosimeters were prepared by putting CR-39 detectors at the bottom of the chamber cover. The

cylindrical container was sealed and the samples were stored for 30 days to let the equilibrium be established between radium and radon before the start of each experiment. Alpha particles interacting with CR-39 were registered as latent damage trials. In order to amplify the latent damage trials, etching conditions were provided, KHAN et al. [10]. After etching the detectors were washed in distilled water, dipped for a few seconds in 3% acetic acid solution and washed again, then they were dried in air.

The track densities were determined manually using an optical microscope at magnification of 400×, a number of 50 fields were scanned for each detector and the average number of tracks per field was determined.

The exhalation rate can be calculated from the relation [11]:

$$Ex = P V \lambda / \varepsilon A T_{\text{eff}}, \tag{2}$$

and the effective radium content is determined from the equation [11]:

$$E_{\text{Ra}} = PV / \varepsilon M T_{\text{eff}}, \tag{3}$$

where:

$P$ is the track density ($\text{Tm}^{-3}$),

$V$ is the effective volume of the cylindrical container ($\text{m}^3$),

$\lambda$ is the radon decay constant,

$T_{\text{eff}}$ is the effective exposure time equal to $T + 1/\lambda\,(e^{-\lambda t} - 1)$,

$T$ is the exposure time,

$A$ is the area of the cylinder chamber ($\text{m}^2$),

$M$ is the mass of the sample (kg),

$\varepsilon$ is the detector efficiency.

## 3. RESULTS AND DISCUSSION

About 40 clay samples and 25 fabricated clay samples were collected from Egypt as mentioned before. About seventeen well resolved energy photopeaks were observed. Eight photopeaks from uranium series, eight from thorium series, and one for $^{40}$K as shown from the selected spectrum in figure 3 for the sample number 3.

$^{238}$U is the parent of uranium series, considered environmentally very important as it includes important radionuclides. Tables 1 and 2 list the average values of concentration of radionuclides and activity concentration for the different respective types of Egyptian clay. It can be seen that the highest mean value of $^{238}$U series concentration is 70.73 ± 3.54 Bq/kg measured in the Aswanly clay sample No. 2, while the lowest mean value for the same radionuclide is 6.77 ± 0.34 Bq/kg in the Safy clay sample No. 8.

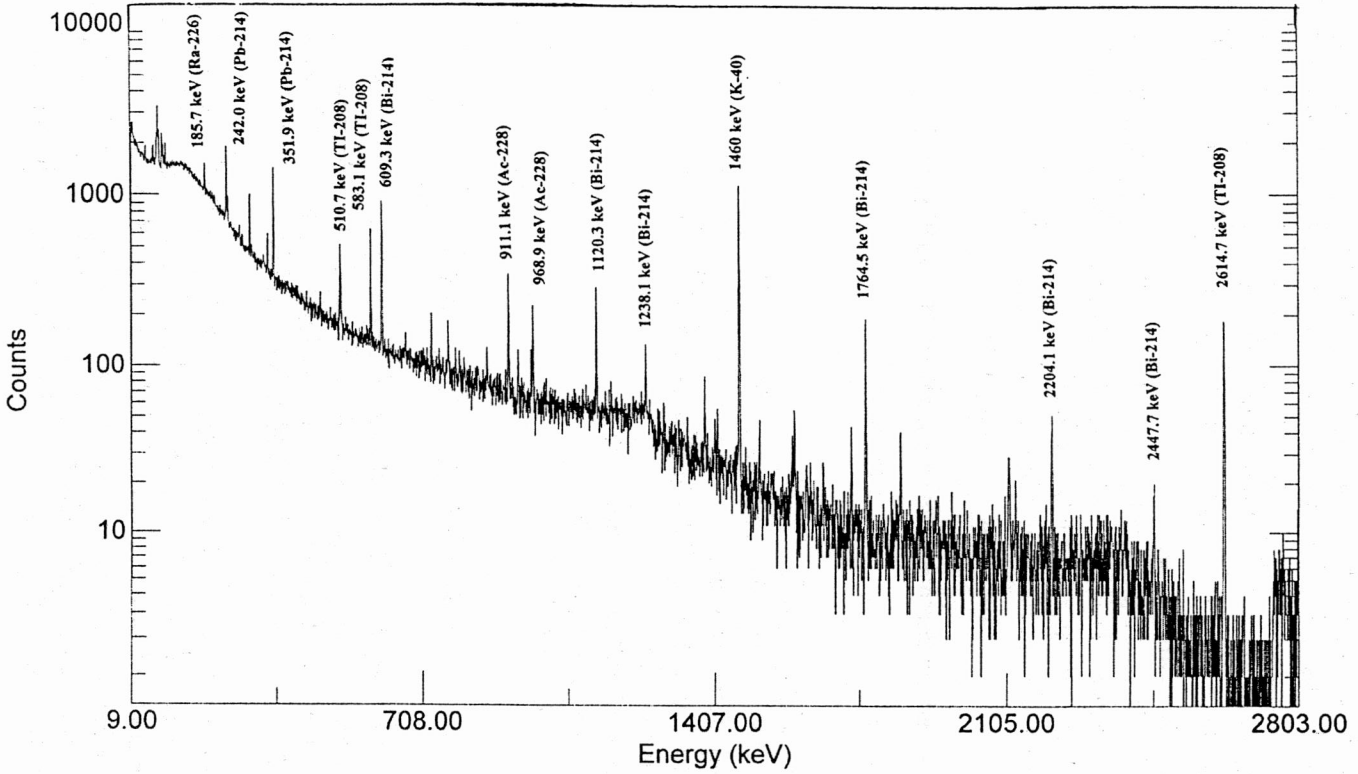


Fig. 3. The spectrum of natural radioactivity for sample No. 3

Table 1

Average radionuclide concentrations (in Bq/kg)
for the different types of Egyptian clay

| Samples | Ra-226 | Pb-214 | Bi-214 | Ac-228 | Bi-212 | Tl-208 | K-40 |
|---|---|---|---|---|---|---|---|
| 1: Mean | 57.67±2.88 | 52.79±2.64 | 67.69±3.38 | 77.92±3.89 | 73.25±3.66 | 89.77±4.49 | 448.1±22.41 |
| Range | 52.79–67.23 | 48.65–62.35 | 60.93–72.45 | 69.43–82.51 | 69.57–80.73 | 81.67–93.40 | 442.30–456.61 |
| 2: Mean | 73.75±3.69 | 61.43±3.07 | 77.01±3.85 | 81.82±4.09 | 92.53±4.63 | 123.42±6.17 | 504.49±25.22 |
| Range | 69.23–77.37 | 58.84–66.25 | 73.43–81.25 | 76.37–88.32 | 86.29–99.40 | 120.50–129.95 | 500.91–512.512.87 |
| 3: Mean | 43.06±2.15 | 13.39±0.67 | 19.31±0.67 | 11.75±0.59 | 18.21±0.91 | 23.82±1.19 | 282.77±141.14 |
| Range | 35.20–46.72 | 11.92–6.35 | 16.95–24.35 | 10.92–14.31 | 17.54–20.30 | 18.64–24.25 | 280.92–291.50 |
| 4: Mean | 48.92±2.45 | 45.18±2.26 | 57.79±2.89 | 40.33±2.02 | 53.43±2.67 | 68.94±3.45 | 172.80±8.64 |
| Range | 42.73–51.47 | 42.91–49.32 | 49.47–61.23 | 36.42–41.30 | 50.72–56.53 | 64.84–75.34 | 165.70–179.42 |
| 5: Mean | 29.26±1.46 | 18.58±0.93 | 25.45±1.27 | 30.16±1.51 | 41.27±2.06 | 39.78±1.99 | 374.33±18.72 |
| Range | 27.63–32.85 | 17.34–20.92 | 24.61–28.90 | 25.62–32.70 | 38.39–45.40 | 32.91–40.53 | 368.84–376.50 |
| 6: Mean | 55.79±2.79 | 25.23±1.26 | 51.77±2.59 | 53.68±2.68 | 61.87±3.09 | 71.63±3.58 | 428.95±21.45 |
| Range | 51.34–60.18 | 23.72–30.43 | 45.37–62.31 | 47.81–59.72 | 59.73–65.41 | 67.34–76.92 | 419.57–431.60 |
| 7: Mean | 54.66±2.73 | 44.64±2.23 | 61.98±3.09 | 58.86±2.94 | 91.15±4.56 | 95.22±4.76 | 694.27±34.71 |
| Range | 49.41–60.91 | 40.53–49.41 | 55.83–66.40 | 52.60–65.73 | 87.43–98.21 | 92.42–96.83 | 689.72–695.80 |
| 8: Mean | 8.44±0.42 | 5.42±0.27 | 6.44±0.32 | 4.24±0.21 | 3.81±0.19 | 15.16±0.76 | 172.34±8.62 |
| Range | 7.23–10.32 | 2.94–7.98 | 4.74–7.98 | 2.95–5.54 | 2.53–5.12 | 14.62–17.31 | 165.41–178.93 |

Table 2

The average activity concentration (in Bq/kg)
for the different types of Egyptian clay

| Sample | U-238 series | Th-232 series | K-40 | U-235 series |
|---|---|---|---|---|
| 1 | 59.38±2.97 | 80.31±4.02 | 448.10±22.41 | **15.97±0.79** |
| 2 | **70.73±3.54** | **99.26±4.96** | 504.49±25.22 | ~ |
| 3 | 25.25±1.26 | 17.93±0.89 | 282.77±14.14 | 6.89±0.34 |
| 4 | 50.63±2.53 | 54.23±2.71 | 172.80±8.64 | **5.02±0.25** |
| 5 | 24.09±1.20 | 37.07±1.85 | 374.33±18.72 | ~ |
| 6 | 44.08±2.20 | 62.39±3.12 | 428.95±21.45 | ~ |
| 7 | 53.76±2.69 | 81.74±4.09 | **694.27±32.21** | ~ |
| 8 | **6.77±0.34** | **7.74±0.39** | **172.34±8.62** | ~ |

The maximum and minimum values of the activities are given in bold face.

Table 3 shows that the $^{238}U$ concentration for the fabricated Egyptian clay ranged from 32.53 ± 1.63 Bq/kg to 68.29 ± 3.41 Bq/kg which is within the global value, UNSCEAR [5].

Table 3

The average activity concentration (in Bq/kg) for the fabricated Egyptian clay

| Sample | U-238 series | Th-232 series | K-40 | U-235 series |
|---|---|---|---|---|
| A: Mean | 68.29±3.41 | 66.17±3.31 | 776.98±38.85 | 2.38±0.12 |
| Range | 57.23–70.31 | 62.72–75.40 | 765.34–78.92 | 1.9–2.5 |
| B: Mean | 32.53±1.63 | 58.12±2.91 | 427.60±21.38 | 5.74±0.29 |
| Range | 29.73–35.52 | 54.65–60.83 | 422.30–432.89 | 4.76–6.24 |
| C: Mean | 50.33±2.51 | 71.19±3.56 | 244.17±12.21 | 9.39±0.47 |
| Range | 47.53–55.42 | 67.72–73.35 | 239.21–250.40 | 7.29–10.34 |
| D: Mean | 37.96±1.89 | 35.63±1.78 | 644.28±32.21 | ~ |
| Range | 30.43–42.56 | 30.72–43.38 | 636.52–649.43 | ~ |
| E: Mean | 40.08±2.0 | 63.54±3.18 | 460.57±23.03 | ~ |
| Range | 34.89–43.52 | 57.42–69.23 | 453.24–468.52 | ~ |

$^{232}Th$ is the parent of the thorium series, it is also very important from the environmental point of view, as it includes many essential radioisotopes. From table 2, the maximum level of $^{232}Th$ series is observed in clay number 2 (Aswanly clay) and the minimum is observed in clay number 8 (Safy clay), also it is clear that in the case of fabricated clay, the concentration of $^{232}Th$ ranged from 35.63 to 71.19 Bq/kg which is within the range of the control value, UNSCEAR [5].

The value of $^{40}K$ ranged from 172.34 ± 8.62 to 694.27 ± 32.21 Bq/kg for the different types of clay and ranged from 244.17 ± 12.21 to 776.98 ± 38.85 Bq/kg for the fabricated Egyptian clay. We see that the concentration of $^{40}K$ is higher than the international limit recommended by UNSCEAR [5] except in the samples number 3, 4, 7 and C.

$^{235}U$ series were detected in some samples only, the maximum value was 15.97 ± 0.79 Bq/kg in Aswanly clay and the minimum value was 2.38 ± 0.12 Bq/kg in sample number A (a bowl for milk).

It is important to assess the radiation hazards to human health associated with the different types of clay and the fabricated clay. This is done by calculating different radiation hazard indices.

The uniformity in respect of exposure to radiation has been defined in terms of radium equivalent activity ($Ra_{eq}$) in Bq/kg, this index being defined as follows:

$$Ra_{eq} = C_{Ra} + 10/7\ C_{Th} + 10/130\ C_K\,, \tag{4}$$

where $C_{Ra}$, $C_{Th}$ and $C_K$ are the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$, respectively. This formula is based on the estimation that 1 Bq/kg $^{226}Ra$, 0.7 Bq/kg $^{232}Th$ and 13 Bq/kg $^{40}K$ produce the same gamma dose rate, BERETKA et al. [12]. The values of $Ra_{eq}$ for the samples investigated are shown in table 4, where $Ra_{eq}$ activities range between 29.9 ± 1.49 Bq/kg for clay No. 8 (Safy clay) and 247.98 ± 12.39 Bq/kg for clay No. 2 (Aswanly clay). The $Ra_{eq}$ of the samples under investigation is on the permissible level. In relation to the radium equivalent some researchers, e.g., KRISIUK et al. [13], TUFAIL et al. [14], suggested the value of 1.5 mGy as the maximum annual safe radiation dose from raw materials which gives a limit value of $Ra_{eq}$ equal to 370 Bq/kg, KRIEGER [15]. Hence the annual radiation dose (radioactivity level) based on the following expression (RUIXIANG et al. [16]).

$$C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1. \tag{5}$$

Table 4

The radium equivalent activities (Bq/kg), dose rate (nGy·h$^{-1}$), radioactivity level and representative level index values ($I\gamma r$) for the samples investigated

| Sample | Req. | Dose rate | Radioactivity level | $I\gamma r$ |
|---|---|---|---|---|
| 1 | 205.59±10.28 | 97.8±4.89 | 0.56 | 1.50 |
| 2 | 247.98±12.39 | 117.60±5.88 | 0.67 | 1.80 |
| 3 | 70.68±3.53 | 34.81±1.74 | 0.20 | 0.54 |
| 4 | 140.27±7.01 | 64.95±3.25 | 0.39 | 0.99 |
| 5 | 103.3±5.17 | 50.92±2.55 | 0.29 | 0.78 |
| 6 | 163.33±8.17 | 51.56±2.58 | 0.45 | 1.21 |
| 7 | 219.24±10.96 | 106.92±5.35 | 0.61 | 1.64 |
| 8 | 29.90±1.49 | 15.42±0.77 | 0.09 | 0.24 |
| A | 217.30±10.87 | 106.37±5.32 | 0.60 | 1.64 |
| B | 145.57±7.28 | 70.76±3.54 | 0.40 | 1.09 |
| C | 169.22±8.46 | 79.11±3.96 | 0.46 | 1.21 |
| D | 134.0±6.70 | 67.50±3.38 | 0.37 | 1.04 |
| E | 163.18±8.16 | 78.97±3.95 | 0.46 | 1.21 |

According to this equation the maximum $Ra_{eq}$ must be < 370 Bq/kg if the maximum value of the radioactivity level ($H_{ex}$) is to be < 1. Is follows from this study (table 4) that $Ra_{eq}$ < 370 Bq/kg, which means that the maximum value of $H_{ex}$ is < 1.

Another radiation hazard index, that is the representative level index, may be defined as:

$$I\gamma r = C_{Ra}/150 + C_{Th}/100 + C_K/1500. \tag{6}$$

This index can be used to estimate the level of $\gamma$- radiation hazard associated with the natural radionuclides in specific raw materials. The higher limit for the representative level is 1, OECD [17]. The values of $I\gamma r$ are calculated for all the samples un-

der study and are given in table 4. The range of $I\gamma r$ is between 0.24 to 1.64. The samples which have $I\gamma r$ more than 1 are not recommended for use as raw materials.

The dose rate in nGy/h was calculated using the convenient formula given by YU et al. [18]:

$$D = 0.427\ C_{Ra} + 0.662\ C_{Th} + 0.043\ C_K \quad \text{nGy/h}. \tag{7}$$

From table 4, the values of dose rates are very high, even higher than the international limit (55 nGy/h) as given in UNSCEAR data, except for samples Nos. 3, 5, 7 and 8, for which those rates are lower than the international limit.

Table 5

Comparison of activity concentrations and radium equivalents (Bq/kg) for the fabricated clay in different countries

| Place | $C_K$ | $C_{Ra}$ | $C_{Th}$ | $Ra_{eq}$ | Ref. |
|---|---|---|---|---|---|
| U.K. | 703 | 52 | 44 | 169 | NEA-OECD (1979) |
| | 620 | 65 | 48 | 181 | Cliff et al. (1985) |
| Norway | 1058 | 104 | 62 | 276 | Stranden (1976) |
| Finland | 962 | 78 | 62 | 241 | NEA-OECD (1979) |
| China | 717 | 41 | 52 | 171 | Pan et al. (1984) |
| Australia | 681 | 40.7 | 88.8 | 220 | Beretka and Mathew (1985) |
| The Netherlands | 560 | 39 | 41 | 141 | Ackers et al. (1985) |
| Greece | 670 | 49 | 24 | 135 | Papaste Fanou et al. (1983) |
| Hong Kong | 850 | 143 | 158 | ND | K.N. Yu et al. (1992) |
| California | 527 | | 42.3 | ND | Ingersoil (1983) |
| Canada | 800 | 37 | 49 | ND | L. Zikovsky and G. Kennedy (1992) |
| Egypt | 258 | 24 | 24 | 78 | El-Tahawy et al. (1995) |
| Egypt | 276 | 134 | 68 | ND | Naim et al. (1999) |
| Banha | 427.6 | 32.53 | 58.12 | 145.57 | Present work |

Table 5 lists the average values obtained in the present study compared to others from different regions and countries for the fabricated clay No. B (red brick). From this table, the concentration of $^{238}U$ in the present study was 32.53 Bq/kg which is lower than the global average of 67 Bq/kg. The highest value of U appears to be in Hong Kong (143 Bq/kg) and the concentration of $^{232}Th$ in the present work was 58 Bq/kg, which is within the range recorded in different countries except Hong Kong (158 Bq/kg). Also, the concentration of $^{40}K$ in Egypt ranging from 258 to 427 Bq/kg which is lower than the global average of 700 Bq/kg, and is the lowest value among the values being compared, while the highest value of $^{40}K$ is shown in Norway and equals 1058 Bq/kg.

From table 6 we can see that the effective radium content was found to vary from 6.65 to 4.09 Bq/kg and the exhalation rate varied from 31.29 to 19.86 Bq/m$^2$. Figures 4 and 5 show the relation between the exhalation rate, effective radium content and uranium concentration, respectively, from which it is evident that there is a linear relation between them.

Table 6

Effective radium content, exhalation rate of radon and uranium concentration for the samples investigated

| Sample No. | Uranium con. (ppm) | (Track/cm$^2$)×10$^3$ | Eff. Ra content (Bq/kg) | Ex. rate (Bq m$^{-2}$d$^{-1}$) |
|---|---|---|---|---|
| 1 | 59.38±2.97 | 8.14±0.96 | 6.65±0.78 | 31.29±3.69 |
| 2 | 70.73±3.54 | 8.67±1.06 | 7.88±0.93 | 33.33±4.06 |
| 3 | 25.25±1.26 | 3.09±0.38 | 2.83±0.33 | 11.90±1.45 |
| 4 | 50.63±2.53 | 6.94±0.76 | 5.67±0.66 | 26.68±3.14 |
| 5 | 24.09±1.20 | 2.95±0.36 | 2.68±0.31 | 11.35±1.38 |
| 6 | 44.08±2.20 | 5.40±0.66 | 4.94±0.57 | 20.77±2.53 |
| 7 | 53.76±2.69 | 6.58±0.81 | 5.99±0.70 | 28.33±3.09 |
| 8 | 6.77±0.34 | 0.93±0.10 | 0.76±0.09 | 3.19±0.39 |
| A | 68.29±3.41 | 8.36±1.02 | 7.65±0.89 | 32.18±3.92 |
| B | 32.53±1.63 | 3.99±0.49 | 3.62±0.42 | 17.14±1.87 |
| C | 50.33±2.51 | 6.17±0.75 | 5.04±0.61 | 23.72±2.88 |
| D | 37.96±1.89 | 4.65±0.57 | 4.23±0.46 | 20.0±2.17 |
| E | 40.08±2.00 | 4.91±0.60 | 4.47±0.52 | 21.12±2.29 |

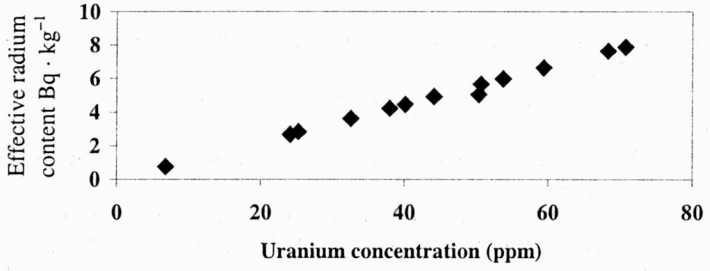


Fig. 4. Relation between uranium concentration and eff. *Ra* content for the samples under study

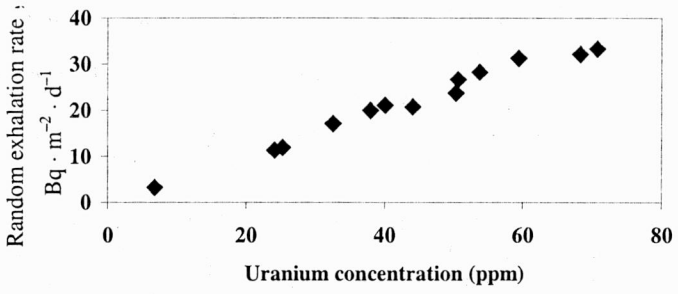


Fig. 5. Relation between radon exhalation rate and uranium concentration for the samples under investigation

## 4. CONCLUSIONS

The results obtained show that the concentration of radioelements in Egyptian clay and fabricated clay falls within the values accepted as "normal". The value of 30 ppm of $Ra_{eq}$ which is considered to be the upper concentration level (30 ppm = 370 Bq/kg) has not been exceeded for the majority of clay samples being measured.

As regards the radiation safety, the clays of Egypt are below the recommended limits for their gamma dose rate, therefore they can be used for all kinds of fabricated goods such as red brick, pottery, bottles for drinking, vessels for cooking, storage jars and vases for decoration, etc.

A combination of gamma-ray spectroscopy and solid state nuclear track detectors was recommended for radon and uranium concentrations.

The results obtained in this study will contribute to a proper and adequate use of clays and their industrial products, in connection with effective economic and public health costs.

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## NATURALNE IZOTOPY I OCENA RADIOAKTYWNOŚCI GLINY ORAZ WYROBÓW Z GLINY NA OBSZARZE EGIPTU

Badania naukowe już dawno temu dowiodły, że długotrwałe narażenie ludzi na niskie dawki promieniowania może być szkodliwe. Celem tego artykułu jest zbadanie radioaktywności różnych typów gliny pochodzącej z obszaru Egiptu i wyrobów z tej gliny takich jak: czerwona cegła, dzbany do przechowywania żywności, donice nasienne zabezpieczające nasiona przed gryzoniami i pleśnią, wazy dekoracyjne, naczynia do gotowania, misy do wypieku chleba oraz butelki do przechowywania wody. Istnieją obawy, że wymienione przedmioty wykazują naturalną radioaktywność. Stopień tej radioaktywności określono dla różnych typów gliny za pomocą spektrometru germanowego (gamma). Dawka promieniowania emitowana przez próbki gliny jest proporcjonalna do stężenia w niej uranu (U), toru (Th) i potasu (K). Zakresy stężeń wynosiły 6,77–70,73 Bq/kg, 7,74–99,26 Bq/kg i 172,34–776,98 Bq/kg odpowiednio dla $^{238}U$, $^{232}Th$ i $^{40}K$. Równoważna aktywność promieniowania i dawka pochłoniętego promieniowania wynosiły odpowiednio od 29,9 do 247,98 Bq/kg oraz od 15,42 do 117,6 nGy/h.

Jądrowy detektor śladowy CR-39 został wykorzystany do pomiaru natężenia promieniowania emitowanego przez próbki. Natężenie to zmieniało się od 3,19 do 33,33 Bq/m$^2$d. Te wartości są zgodne z wartościami stężeń uranu zmierzonymi za pomocą spektrometru germanowego w odpowiednich próbkach. Badania wykazały, że różne typy gliny oraz wyroby garncarskie charakteryzują się pewną śladową radioaktywnością. Jednakże poziom tej radioaktywności nie jest szkodliwy dla zdrowia. W odniesieniu do bezpieczeństwa promieniowania naturalna radioaktywność różnego typu glin na terenie Egiptu jest poniżej dopuszczalnej granicy natężenia promieniowania gamma. Z tego też względu egipska glina może być wykorzystywana jako surowiec w budownictwie i garncarstwie.