International Journal of Science and Research (IJSR)
ISSN: 2319-7064
SJIF (2022): 7.942

Assessment of Radiological Impact for Individuals Associated with Natural Occurring Radioactive Material in Ceramic Industry

T. M. Taha[1], Khaled A. Salman[2]

[1, 2]Radiation Protection Department, Nuclear Research Center, Egyptian Atomic Energy Authority Cairo.P.O.13759 Egypt
[1]Corresponding Author Email: *tahaalfawwal[at]eaea.sci.eg*
orcid.org/0000-0001-8663-0748

Abstract: ***Terrestrial radiation and radon gas are the main sources of natural background radiation. The current study aimed to assess the external exposure to gamma radiation and internal exposure to radon gas emitted from zirconium-containing material. The ceramic raw materials under investigation were collected from Ceramic Company A with selection criteria for sampling ceramic raw materials covering the ceramic industry. The specific activity of primordial natural radionuclides in ceramic components was detected using an HPGe gamma ray spectrometer. The radon gas was monitored using RAD7.The specific activity was varied from specific activity was varied from 5.03± 0.1 Bqkg^{-1} (EGC7) to 2745±60 Bqkg^{-1} (Zirconium silicates) for ^{226}Ra, ND (Lime stone) to 234± 12 Bqkg^{-1} (EGC1) for ^{232}Th and ND (EGC7) to 328±8 Bqkg^{-1}(EGC5) for ^{40}K. Radiation doses and external risks were determined. The annual effective dose resulting from uranium and thorium series and ^{40}K and inhalation of radon gas is within the public dose limit recommended by ICRP.***

Keywords: Natural radioactivity, ^{226}Ra, ^{232}Th, ^{40}K, HPGe

1. Introduction

Radiation in the ceramic industry is an issue of concern for scientists and engineers due to the potential presence of naturally occurring radioactive materials in the raw materials used in this industry. These materials include uranium, thorium, and potassium-40, which naturally occur in certain minerals such as clay, feldspar, and sand. Clay and other minerals: Raw materials such as clay, feldspar, silica, and quartz sand may contain small amounts of naturally occurring radioactive materials (NORM). These levels vary depending on the geographical source of the raw material. Synthetic additives: Sometimes, additives that may have higher radiation levels are used. For example, heavy metal oxides may be added to certain types of ceramics to enhance their physical or aesthetic properties. Human exposure: People working in the ceramic industry may be exposed to low levels of radiation when handling raw materials or finished products. Consumers: Final products (such as tiles and pottery) are generally safe, as radiation levels are minimal and do not pose a significant risk, except in cases where a high percentage of radioactive materials is used.

The main sources of terrestrial radiation are the rocks and the earth crust. Ceramics are one of the most important types of building materials. The specific activity concentrations of ceramic material samples are the preliminary steps for the calculation of radiation doses and external risk (UNSCEAR 2000). The ceramic tiles are lined by a mixture of zinc oxide, feldspar and kaolin that is called glaze as an emulsifier. The emanation radon gas from glazed surfaces may be because of external risk (Mao Yahong et al, 2014). A lot of studies determined the concentrations of natural radioactivity in cement, ceramics, and building raw materials such as A F. El-Mekawy et al, 2015 found that all annual effective dose values were below the world average of 410 μSv for the public and 20 mSv for workers [3]. Nabil M et al, 2017 were determined the specific activity of ^{226}Ra, ^{232}Th, and ^{40}K in some samples of ceramic in Egypt using a highly pure germanium detector gamma spectroscopy system [4]. Shiraz et al, 2019 measured activity concentrations for the natural radioactivity contained in ceramic materials collected from different containers in Bangladesh. Azmary Khatun et al, 2018 determined natural radioactivity in some building materials and found that the radium-equivalent activities for the studied building materials were also below the criterion limit of γ-radiation dose of 370 Bq·kg−1. The current study aimed to assess the external exposure to gamma radiation and internal exposure to radon gas emitted from zirconium-containing material

2. Materials and Methods

Sample Collection and preparation

Seven samples of ceramic components were collected randomly from company B . Samples were labeled with the date and placed in polyethylene bags. The samples were crushed, homogenized, and sieved through ≤2 mm sieve shakers. To prevent the spatial variability of radionuclide distribution associated with sample collection. Each sample was weighed (dry weight) using an electronic scale before being transferred to a clean and empty 402-cc cylindrical plastic container of that size (80 mm in height and 80 mm diameter), which was sealed for around four weeks; usually, ceramic measurements sample weight ranges from 0.250 to 0.300 kg of dry ceramic powder. This was done before gamma spectroscopy to allow radon and its short-lived offspring to reach secular radioactive equilibrium (IAEA, 2014).

Activity Assessment

All measurements were performed in the Laboratory of Low-level Counting of Radiation Protection department of the Nuclear Research Center. The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K in the ceramic raw materials samples were measured using a high-purity germanium gamma-ray spectrometer. The spectrometer consisted of a coaxial p-type

HPGe detector (GC 2520 model), a gamma with cryostat model 7905-7.5S, and a preamplifier model 2002C with a relative efficiency of 25% and shielded with lead to minimize the background. The detector bias voltage was 4500 V and the energy resolution was 1.18 keV at 122 keV and 1.9 keV at 1.33 MeV. The detector connected with the preamplifier then connected to a digital signal analyzer to acquisition analysis software, APEX. The counting time was set to 86400 sec. The Minimum Detectable Activity (MDA) of the three radionuclides under study, ^{226}Ra, ^{232}Th, and ^{40}K, was mathematically calculated using equation (1) (Currie, 1989) and presented in Table (1), for each separate measured sample considering the detector efficiency, the sample weight, and background counts at the region of interest of a certain radionuclide, the geometry information (diameter and height), and the specific density of the measured sample, where the MDA of ^{226}Ra, ^{232}Th, and ^{40}K is 2.3, 1.7 and 14 Bq kg− 1 for ceramic sample. The activity of ^{232}Th was determined using the gamma-ray photopeak of ^{228}Ac (911.1 keV) and ^{208}Tl (583.1 keV), while the activity of ^{226}Ra was determined using the photo peaks of ^{214}Bi (1120.3 and 609.3 keV) and ^{214}Pb (351.9 keV). The specific activity of ^{40}K was directly determined from its gamma-ray line at 1460.8 keV.

Calibration

The radioactivity concentrations of ceramic raw material samples were measured using a high-purity germanium gamma-ray spectrometer (HPGe) (Canberra) with 25% relative efficiency. The secondary standards were calibrated by the primary standard RGU–I with a concentration of 4940 Bqkg^{-1}. The detector was calibrated to convert channel numbers or spectra into gamma-ray energies. A spectroscopy study was conducted for a source-to-detector distance of 0.2 cm. Efficiency calibration was calculated using equation (1) and presented as shown in Figure (1).

$$\xi = \frac{N}{Y\ X\ K\ X\ A\ X\ C} \quad (1)$$

Where N is the net number of counts in a given peak area obtained from each photo peak and A is the activity value of each calibrating source, t is the counting time of a spectrum acquired, Y is the emission probability, K is the decay factor for the decay of source during counting time and C is the correction factor for self attenuation.

Minimum Detectable Activity

The minimum detectable activity, MDA for ^{226}Ra, ^{232}Th, and ^{40}K are calculated using equation (2). (Currie, 1989)

$$MDA = \frac{2.7 + 4.65\sqrt{B}}{EIt} \quad (2)$$

Where: B: background under each peak
E: absolute efficiency
I: gamma line intensity
t: counting time in second
MDA for ^{226}Ra, ^{232}Th, and ^{40}K are 2.3, 1.7, and 14 Bq/kg.

The absorbed dose rate was calculated by Eq. (3).

$$D\ (nGy/h) = R_{Ra}\ C_{Ra}\left(\frac{Bq}{kg}\right) + R_{Th}\ C_{Th}\left(\frac{Bq}{kg}\right) + R_K\ C_K\left(\frac{Bq}{kg}\right) \quad (3)$$

Where; R_{Ra}, R_{Th}, and R_k, are the conversion factors in (nGy/h)/(Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K.

Radium equivalent activity (Raeq) was calculated with Eq. (4).

$$R_{Ra}(Bq/kg) = A_{Ra}\left(\frac{Bq}{kg}\right) + 1.43\ A_{Th}\left(\frac{Bq}{kg}\right) + 0.07\ A_K\left(\frac{Bq}{kg}\right) \quad (4)$$

External radiation hazard for primordial natural radionuclides was calculated using Eq. (5) as follows:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (5)$$

The gamma annual effective dose for ^{226}Ra, ^{232}Th and ^{40}K was calculated using Eq. (6) for occupancy factor 0.2.

$$E = = Total\ Dose\ rate\left(\frac{nGy}{h}\right) x\ 2000\ h.y^{-1}\ x\ (occupancy\ factor)\ x\ 0.7\frac{Sv}{Gy}\ X10^{-3} \quad (6)$$

Radon Measurement Survey, RAD7

The DURRIDGE RAD7 Radon Monitor can detect radon concentration using half sphere with a silicon solid-state detector as shown in Figure 2. The radon gas monitored was performed for three rest offices and based on the U.S Environmental Protection Agency protocols (Environmental Protection Agency, 1992). The annual average effective dose for indoor radon is calculated using Equation 7.

$$E_{Rn}(nSv/h) = C_{Rn}\ Bq.m^{-3}\ (mean\ radon)\ x\ 0.4x\ 2000\ h.y^{-1}\ 9\text{nSv(Conversion factor)x}\ \text{Bq}.m^{-3}.\text{h}^{-1} \quad (7)$$

3. Results and Discussion

The specific activity was varied from 5.03± 0.1 Bqkg^{-1} (EGC7) to 2745±60 Bqkg^{-1} (Zirconium silicates) for ^{226}Ra, ND (Lime stone) to 234± 12 Bqkg^{-1} (EGC1) for ^{232}Th and ND (EGC7) to 328±8 Bqkg^{-1}(EGC5) for ^{40}K was presented in Table 1**.** The ^{226}Ra and ^{232}Th concentrations for the measured samples were lower than the worldwide recommended value except for sample (EGC1). Radiation doses and external risk are presented in Table 2. Table 3 presents the mean activity (Bq/kg) of ^{226}Ra, ^{232}Th, ^{40}K, and Raeq in zircon silicate samples compared with some internal publications, as presented in Table 3. The Mean radon gas concentration and total effective dose in different rest offices are presented in Table 4. The highest value of Ra_{eq} is 2745±60 Bq.kg^{-1} (EGC1), zirconium silicate, while the corresponding lowest value is 9.9±0.5 Bq/kg (EGC3) ceramic raw materials. The annual effective dose was calculated and was in good agreement with the International Atomic Energy Agency, (International Atomic Energy Agency, 2007). The highest value of the hazard index is 8.8 (EGC7), zirconium silicate while the corresponding internal radiation hazard index is

16.5. The highest value of Ra_{eq} of zirconium silicate is in good agreement with the China study (Mao Yahong, et al,2004), and the previous Egypt study (El-Afifi E et al, 2006) and disagreement with Bangladesh study (Beretka et al, 1985).

4. Conclusions

The annual effective dose associated with exposure of ceramic components was found to be 0.40 mSv/y and the total annual effective dose was found to be 0.753 mSv lower than the occupational dose limit recommended by (IAEA,2014); (ICRP-60, 1990). The attributed risk may be reduced by careful choice of raw materials and using new mechanical technology.

References

[1] UNSCEAR., 2000. Report to General Assembly, with Scientific Annexes, Sources and Effects of Ionizing Radiation, United Nations, New York, 2000.
[2] Yahong, Mao.,Yahong, Mao., Yigang, Liu., Zhang Guang, and Xiaole, Hu., 2004. Title , Health Physics DOI: 10.1097/00004032-200407000-00009
[3] El-Mekawy, A F., Badran, H M, Seddeek M K., Sharshar, T , Elnimr, T., 2015. Environ Monit Assess DOI 10.1007/s10661-015-4813-y. Hassan, Nabil M., Mansour, N. A., Fayez-Hassan, M., and Samar Fares., 2017. Indoor and Built Environment, Vol. 26(3) 317:326. M. Siraz, S. Pervin, S. Banik, A. Rahman, A.. Rahman and S. Yeasmin , title, Nuclear Science and Applications Vol. 28. No. 1&2 . (2019) . International Atomic Energy Agency, 2014. Radiation Protection and NORM Residue Management in the Zircon and Zirconia Industries.Safety Reports Series No. 51.
[4] Environmental Protection Agency,.1992. Indoor Radon and Radon Decay Product Measurement Device Protocols, Office of Air and Radiation, U.S.EPA Publication;402-R-92-004.
[5] El-Afifi, E., Hilal, M., Khalifa S., and Aly, H., 2006. Evaluation of uranium (U), thorium (Th), potassium (K-40) and emanated radon (Rn-222) in some NORM and TE-NORM samples, Radiation Measurements, ;41:627-633. Beretka, J., and Mathew, P. 1985, Health Phys, 48(1), 87:95
[6] International Atomic Energy Agency, 2014 . Radiation Protection and Safety of Radiation Sources : International Basic Safety Standards. IAEA Safety Standards Series No. GSR Part 3.
[7] ICRP,1990. Recommendations of the International Commission on Radiological Protection, ICRP Publication 60. Ann. ICRP 21 1991; (1:3) pergamon press.

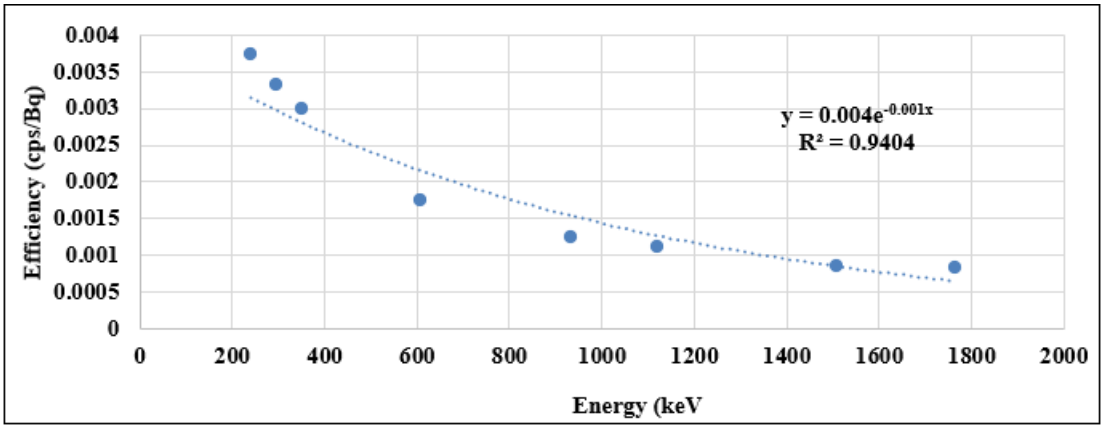

Figure 1: Energy Efficiency of HPGe using Reference Material RGU1-IAEA

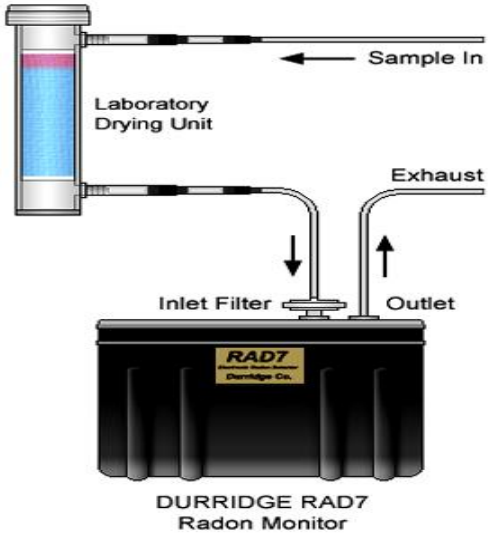

Figure 2: DURRIDGE RAD7 Radon Monitor

Table 1: Specific activity of ^{226}Ra, ^{232}Th, ^{40}K and Ra_{eq} in Ceramic Raw Materials

Ceramic material	S. No.	Activity Concentrations in Ceramic Raw Materials			Mean
		^{226}Ra	^{232}Th	^{40}K	Ra_{eq}
Z zirconium silicates	S1	2899±116	2 353 ± 11	ND	3403
	S2	2899±78	2 436 ± 09	ND	3523
	S3	2743±76	238 ±11	ND	3083
	S4	2745±60	234± 12	ND	3079
Mean±σ		2821±80	315± 11	ND	3272
Kaolinite	S5	77±5.4	ND	90±1.8	10.9
	S6	79±6.3	ND	89±1.78	11.2
	S7	81±5.7	ND	90±1.8	11.5
	S8	76±6.1	ND	88±1.78	10.8
Mean±σ		78±5.88		89 ±1.79	85
Glass Sand	S9	13.6±1.08	9.9±0.5	66 ±1.3	33
	S10	11.7±0.94	10.11±0.7	67 ±1.34	31
	S11	16±1.7	10.11±0.7	68 ±1.36	35
	S12	13.7±2	9.93±0.6	67 ±1.34	33
Mean±σ		13.8±1.43	10.01±0.63	67± 1.34	33
Silica Sand	S13	9.87±0.7	22±1.1	135±5	50
Clay	S14	45±2.3	51±1.5	328±8	41
Feldspar	S15	26±1.7	38+1.9	242±6.5	75
Lime stone	S16	5.03± 0.1	ND	111±3.3	14
Talc	S17	14±1.8	14±1.74		34

Table 2: Mean Radiation doses and external and internal risk

Samples	S. Code	Ra_{eq}	Dr	AED	Hex	H_{in}
		(Bq/kg)	$nGyh^{-1}$	µSv/y		
Zirconium silicates	EGC1	3272	1494	418	8.8	16.5
Kaolinite	EGC2	85	40	11	0.23	0.44
Glass Sand	EGC3	33	15	4	0.09	0.13
Silica Sand	EGC4	51	24	6.6	0.14	0.17
Clay	EGC5	141	65	18	0.39	0.51
Feldspar	EGC6	75	36	10	0.2	0.28
Limestone	EGC7	14	7.5	2	0.04	0.05
Talc	EGC8	37	19	5.4	0.1	0.12
Total		3708	1701	475	9.99	18.2

Table 3: Comparison between mean current activity (Bq/kg) of ^{226}Ra, ^{232}Th, ^{40}K, and Ra_{eq} of zircon with international countries.

Country					References
	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	
Bangladesh	22-515	16-131	12-91	45-705	(Beretka et al, 1985)
China	158-1987	92-1218	473-1031	> 370	(Mao Yahong, et al,2004)
El Behaira Governorate, Egypt	2249	503	326	> 370	(El-Afifi E wt al, 2006)
Present Study	2821±80	315± 11	ND	3272	

Table 4: Mean radon gas concentration and total effective dose in different rest offices

Rest offices	Radon gas Concentration, Bq/m^3
RO1	50±5.0
RO2	48±4.8
RO3	49± 5.2
Mean ±SD	49± 49± 5
Total Effective Dose	
Exposure	Annual Effective dose, mSv/y
External effective dose	0.4
Internal effective dose	0.353±0.7
Total effective dose	0.753