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Measurement of the radioactivity in raw materials used in the ceramic industry in AL-Ramadi ceramic factory using (HPGe) detector

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Abstract

In this research, we have measured specific activity concentrations in five samples for raw materials used in ceramic industry in AL-Ramadi ceramic factory by using (HPGe) detector. The results have shown that, the average specific activity, for ^{238}U, ^{232}Th and ^{40}K are equal to (18.300±6.4 Bq/kg), (17.988±6.1 Bq/kg), (167.952±63.5 Bq/kg), respectively. In order to assess the radiological hazards of the radioactivity in samples, radium equivalent activity, absorbed gamma dose rate, indoor and outdoor annual effective dose rates, gamma Index and both (external and internal) hazard effects have been calculated. All results were found to be less than the allowed global limit given by (UNSCEAR, 2000).

Keywords: radioactivity , (HPGe) detector , specific activity , ceramic industry.

قياس النشاط الاشعاعي في المواد الاولية المستخدمة في صناعة السيراميك في معمل سيراميك الرمادي باستخدام كاشف (HPGe)

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الخلاصة

في هذا البحث تم قياس النشاط الاشعاعي لخمس نماذج من المواد الاولية المستخدمة لصناعة السيراميك في معمل السيراميك في الرمادي باستخدام كاشف (HPGe). بينت النتائج ان معدل النشاط النوعي لليورانيوم–238 والثوريوم–232 والبوتاسيوم–40 يساوي (18.300±6.4 Bq/kg)، (17.988±6.1 Bq/kg)، (167.952±63.5 Bq/kg)، على التوالي. ولأجل تقييم المخاطر للنشاط الاشعاعي في النماذج، فقد تم حساب كل من النشاط المكافئ للراديوم ومعدل جرعة امتصاص كاما، ودليل الخطورة لأشعة كاما ومؤثرات المخاطر (الخارجي والداخلي). جميع النتائج وجدت أنها أقل من الحد العالمي المسموح به (UNSCEAR, 2000).

Introduction

Radioactivity was discovered in 1896 by the French scientist Henri Becquerel by studying the relationship between the phosphoric phenomenon of uranium salts when exposed to sunlight and the

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fluoridation of the surfaces of the discharge tube at the emission of X-rays. It was found that radiation emitted from these salts whether exposed to light or not exposed [1], It then shows him the similarity between his discovery and the discovery of X-rays, that the new radiation can produce an electrical discharge as X-rays do, and realize that these radiation are not the result of fluorine, but the presence of uranium metal was the reason. This characteristic of uranium has an spontaneous radiation emission called radioactivity [2].

The study of radioactive decay and nuclear reactions is a study of the movement of the nucleus any properties that change over time. The processes of decomposition can be spontaneous (radioactive decay) or artificial (nuclear reactions) and there is a great similarity between the two processes in theory. The large nucleus that emits radiation is called the parent nucleus and is called the large nucleus produced by the daughter nucleus. If the daughter nucleus is not stable, we will have the so-called radioactivity chain [3].

The current study aims at finding the concentrations of the specific radioactivity of ^{238}U, ^{232}Th and ^{40}K in the raw material samples used in the ceramic industry using the high Purity Germanium (HPGe) detector technique, as well as finding radiation doses from these concentrations to determine the extent of the increase and gravity that these concentrations may pose in the ceramic industry.

Raw materials for ceramics

The basic raw materials used in the ceramic industry are sand (silica SiO_2), feldspar, black clay, white clay, in addition to some other materials used in certain processes such as sodium hydroxide, sodium silicate, calcium carbonate, colored materials and lubricants. The constituents of the glass layer (glazes) are used to cover the surfaces of the products and the glass material is prepared using feldspar, sodium hydroxide, bonding materials, dolomite and water. Some chemicals are used in the laboratories for quality control and conducting analysis, natural gas and diesel fuel are used as fuel. Glazes is one of the most important materials used in the ceramic industry and glazes is a glass material designed to have a coefficient of thermal expansion suitable for the surface of the ceramic covered. Glazes give the product better durability and wider applications [4].

High Purity Germanium (HPGe) Detector:

A high Purity Germanium (HPGe) detector which is a semiconductor detector (P-type) made of company (ORTEC) American made model (GEM20-70) contains a (3×3) inch crystal, this detector operates with operating voltage (3800 V) and efficiently (20%) and it has energy resolution of (1.77 keV) for energy (1.33 MeV) of source (^{60}Co). These types of detectors cool down to (-196) degree when operating with liquid nitrogen. The detector is surrounded by a (12 cm) lead wall to reduce the radiative background as shown in Figure-1.

Figure 1-(HPGe) detector system.

Efficiency Calibration

The detectors efficiency is defined as the ratio between the number of pulses recorded in the detector and the number of photons emitted from the radioactive source. From this definition, we can express the efficiency equation as follows [5]:

$$\varepsilon(E_\gamma) = \frac{Net}{A \times I_\gamma(E_\gamma) \times T} \times 100\% \quad \text{.....(1)}$$

Where:

$\varepsilon(E_\gamma)$: represents the efficiency of the detector.

Net :Net Area under peak for selected energy on measurement time (3600 sec).

$I_\gamma(E_\gamma)$: The relative intensity of each energy of the irradiated a source.

The standard sources was used to calibrate the efficiency and the spectrum was collected for a period of 3600 sec and the program was used (MAESTRO, Version 7.01) to calculate the detector efficiency of the source energy used as in Table-1 and Figure-2 shows the efficiency calibration curve of the (HPGe) detector using the standard source.

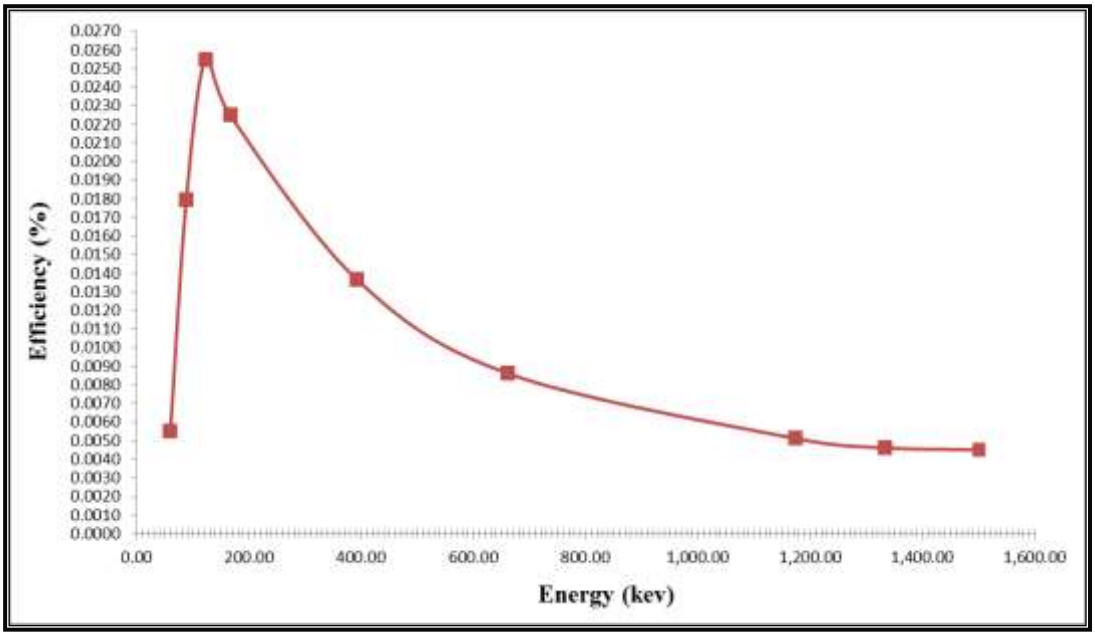

Figure 2-Curved efficiency of germanium system using the standard source.

Table 1-radioactive elements, half-life, activity, relative intensity, energy of radioactive element of standard source.

Radioactive elements	*Half-life* (*Days*)	*Activity* (*KBq*)	*Relative intensity* Iγ (%)	*Element energy* (*keV*)
Am-241	**157800.0**	**4.433**	**35.9**	**59.5**
Cd-109	**462.60**	**16.17**	**88.03**	**3.70**
Ce-139	**137.50**	**0.740**	**79.9**	**165.85**
Co-57	**271.26**	**0.855**	**122**	**85.6**
Co-60	**1925.40**	**2.659**	**99.85**	**1173.24**
Cs-137	**11019.00**	**2.439**	**85.10**	**661.66**
Sn-113	**115.10**	**3.087**	**64.97**	**392**
y-88	**106.60**	**3.995**	**93.70**	**898**
Hg-203	**46.72**	**2.064**	**279.19**	**81.56**
Sr-85	**64.78**	**4.024**	**96**	**514**

Calculation of the Specific Activity (A)

Specific activity is defined as radiation activity during the mass unit of the radioactive material and is measured in curie units per gram or Becquerel per kilogram (Bq/kg). Specific activity is calculated by (Bq/kg) unit by using the equation [6]:

$$A\ \ (Bq/kg) = \frac{N}{\varepsilon(E_\gamma)\cdot I_\gamma(E_\gamma)\cdot M\cdot t} \quad \text{……………………………………………… …….(2)}$$

Where:

N: Count of γ – ray.

$\varepsilon(E_\gamma)$:the efficiency of the γ – ray detector.

$I_\gamma(E_\gamma)$: the relative intensity of each energy of the irradiated source.

M: the mass of the sample measured (kg).

t: the time of measurement which is equal to (3600 sec).

Calculation of radiation hazard effects of γ – rays:

1. Radium Equivalent Activity (Ra_{eq})

The radium equivalent activity (Ra_{eq}) is defined as a radiological factor used to ensure the uniform distribution of natural radionuclides represented by (^{238}U, ^{232}Th, ^{40}K) and measured in (Bq/kg) unit and it can be calculated in the following equation[7]:

Ra_{eq} (Bq/kg) = $A_U + 1.43A_{Th} + 0.077A_K$ ……………………………………………………...….(3)

Where:

A_U, A_{Th}, A_K: the radiation activity of ^{238}U, ^{232}Th, ^{40}K in (Bq/kg) unit respectively.

2. Absorbed Dose Rate in Air (D_γ)

The rate of absorbed dose of γ – rays in the air (D_γ) at one meter above ground level can be calculated by compensating the specific activity values (^{238}U, ^{232}Th, ^{40}K) as in the following equation[8]:

D_γ (nGy/h) = $0.462A_U + 0.604A_{Th} + 0.0417A_K$ …………………………………………...….(4)

Where:

D_γ : absorbed dose rate in (nGy/h) unit.

A_U , A_{Th} , A_K : the specific activity of ^{238}U, ^{232}Th, ^{40}K in (Bq/kg) unit respectively.

3. Annual Effective Dose (AED)

Annual effective dose (AED) is defined as an irradiation factor used to judge the health effects of the absorbed dose and is measured by (mSv/y) unit. The annual effective dose is estimated using the conversion factor (0.7 Sv/Gy) , which converts the absorbed dose into the air to the effective dose, as well as using the outdoor works factor (0.2) and the indoor works factor (0.8) as in the following equations[9]:

$(AED)_{out}$ (mSv/y)= D_γ (nGy/h)×10^{-6}×8760 h/y×0.20×0.7Sv/Gy ……..……………………..(5)

$(AED)_{in}$ (mSv/y)= D_γ (nGy/h)×10^{-6}×8760 h/y×0.80×0.7Sv/Gy ………………………..….(6)

4. Gamma Index (I_γ)

Gamma index is a radiation coefficient in which the risk levels of γ – rays associated with natural radionuclides are estimated in the samples and can be calculated from the following equation [10]:

$$I_\gamma = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad \text{……………………………………………………………(7)}$$

5. Hazard Index (H)

A hazard index (H) is defined as a radiation factor used to determine the risk of external and internal radiation. The external hazard index (H_{ex}) and internal hazard index (H_{in}) are calculated using the following equations [11]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{…………………………………………………………..…(8)}$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{…………………………………………………………..…(9)}$$

Results and discussion

Five samples of the ceramic elements (sand, feldspar, glazes, white clay and black clay) were collected from the ceramic factory in Ramadi city in Anbar province. The specific activity of natural radionuclides of uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) was calculated using the High Purity Germanium (HPGe) detector, Radiation hazard effects for the samples were also calculated as in Table-2,

The lowest value of the specific activity of ^{238}U, ^{232}Th was equal to (8.440 , 6.920) in (Bq/kg) unit respectively in the feldspar sample, it was found that the lowest value of the specific activity of ^{40}K was equal to (83.250 Bq/kg) in the white clay sample and the highest value of the specific activity of ^{238}U, ^{232}Th and ^{40}K was equal to (26.430 , 27.030 , 267.580) in (Bq/kg) unit respectively in the sand sample as in Figure-3. That the appearance of the highest level of radioactivity in sand samples is due to the containment of these samples on the proportion of radioactive minerals such as monazite and zircon minerals, which are an important source for obtaining uranium and thorium irradiated, and the general rate of the specific activity of ^{238}U, ^{232}Th and ^{40}K is (18.300±6.4, 17.988±6.1, 167.952±63.5) in (Bq/kg) unit respectively. Current results show that the specific activity rate of ^{238}U, ^{232}Th and ^{40}K in the samples was lower than the values of the global average of the specific activity of ^{238}U, ^{232}Th and ^{40}K (35 , 30 , 400) in (Bq/kg) unit respectively[12].

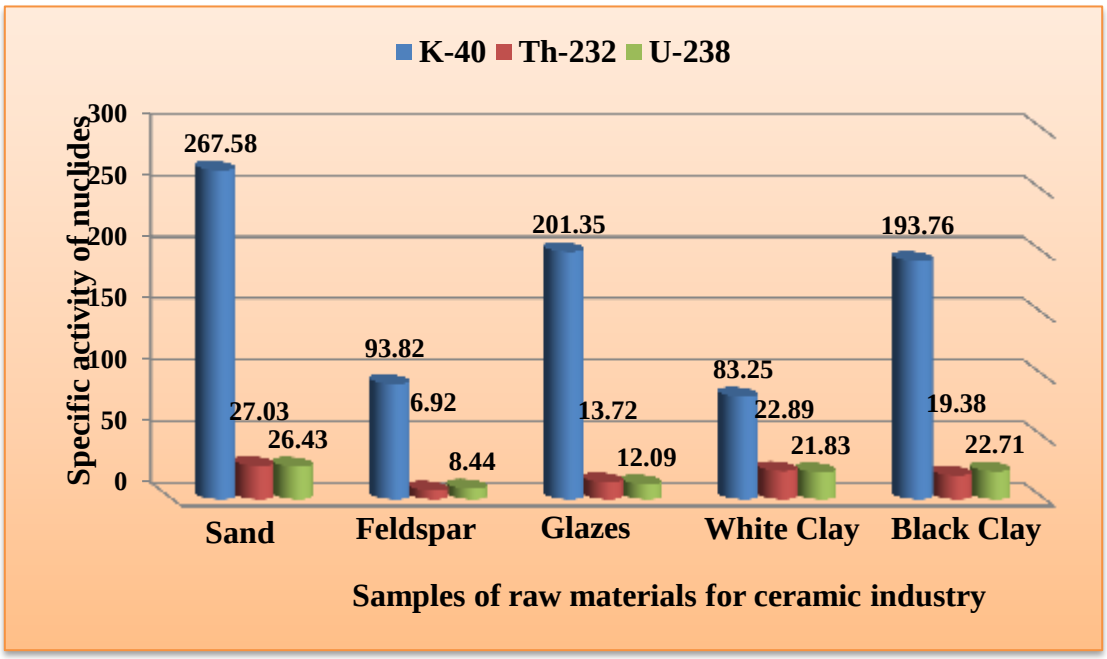

Figure 3- Concentrations of the nuclides (^{238}U, ^{232}Th, ^{40}K) respectively in samples (sand, feldspar, glazes, white clay and black clay) used in the ceramics industry.

The lowest value of the radium equivalent activity (Ra_{eq}) in the samples was equal to (25.560 Bq/kg) in the feldspar sample, and the highest value of the radium equivalent activity (Ra_{eq}) in the samples was (85.687 Bq/kg) in the sand sample and the general rate of the radium equivalent activity (Ra_{eq}) is (56.955±16.4 Bq/kg). Its current results show that the radium equivalent activity (Ra_{eq}) rate in the samples was lower than the values of the global average of the radium equivalent activity (Ra_{eq}) (370 Bq/kg)[12].

The lowest value of the absorbed dose rate in air (D_γ) in the samples was equal to (11.991 nGy/h) in the feldspar sample, and the highest value of the absorbed dose rate in air (D_γ) in the samples was (39.695 nGy/h) in the sand sample and the general rate of the absorbed dose rate in air (D_γ) is (26.323±7.35 nGy/h). Its current results show that the absorbed dose rate in air (D_γ) in the samples was lower than the values of the global average (55 nGy/h)[12].

The lowest annual effective dose of outdoor $(AED)_{out}$ and indoor $(AED)_{in}$ exposure was equal to (0.015 , 0.059) in (mSv/y) unit respectively for feldspar sample, and the highest value of outdoor $(AED)_{out}$ and indoor $(AED)_{in}$ exposure was equal to (0.049 , 0.195) in (mSv/y) unit respectively for

sand sample, and the general rate of outdoor $(AED)_{out}$ and indoor $(AED)_{in}$ is (0.032±0.009 , 0.129±0.036) in (mSv/y) unit respectively. Current results show that the annual effective dose rate of outdoor $(AED)_{out}$ and indoor $(AED)_{in}$ exposure in the samples was lower than the global average (1 mSv/y)[12].

The lowest value of the Gamma Index (I_γ) in the samples was equal to (0.094) for feldspar sample, and the highest value of (I_γ) in the samples was equal to (0.312) for sand sample, and the general rate of (I_γ) is (0.207±0.058). Its current results show that the Gamma Index (I_γ) rate in the samples was lower than the global average (1)[12].

The lowest value of external hazard index (H_{ex}) and internal hazard index (H_{in}) in the samples was equal to (0.069 , 0.092) respectively for feldspar sample, and the highest value of (H_{ex}) and (H_{in}) in the samples was equal to (0.231 , 0.303) respectively for sand sample, and the general rate of (H_{ex}) and (H_{in}) is (0.154±0.044 , 0.203±0.062) respectively. Current results show that the external hazard index (H_{ex}) rate and internal hazard index (H_{in}) rate in the samples was lower than the global average (1)[12].

Table 2-Specific activity of radionuclides and radiation hazard effects for samples (sand, feldspar, glazes, white clay and black clay) used in the ceramic industry.

Type of sample	U-238 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	Ra_{eq} (Bq/kg)	D_γ (nGy/h)	(AED) (mSv/y)		I_γ	H_{ex}	H_{in}
						(AED_{out})	(AED_{in})			
Sand	26.430	27.030	267.580	85.687	39.695	0.049	0.195	0.312	0.231	0.303
Feldspar	8.440	6.920	93.820	25.560	11.991	0.015	0.059	0.094	0.069	0.092
Glazes	12.090	13.720	201.350	47.214	22.269	0.027	0.109	0.176	0.128	0.160
White clay	21.830	22.890	83.250	60.973	27.383	0.034	0.134	0.215	0.165	0.224
Black clay	22.710	19.380	193.760	65.343	30.277	0.037	0.149	0.237	0.176	0.238
Ave.	18.300 ±6.4	17.988± 6.1	167.952±6 3.5	56.955± 16.4	26.323± 7.35	0.032 ±0.009	0.129 ±0.036	0.207 ±0.058	0.154 ±0.044	0.203 ±0.062
Min.	8.440	6.920	83.250	25.560	11.991	0.015	0.059	0.094	0.069	0.092
Max.	26.430	27.030	267.580	85.687	39.695	0.049	0.195	0.312	0.231	0.303
worldwide average[12]	35	30	400	370	55	1	1	1	1	1

Conclusions

The results obtained indicate that the radioactivity in all the raw material samples is (sand, feldspar, glazes, white clay and black clay) used in ceramics industry in the ceramic factory in the city of Ramadi is less than the international limit, and the highest radiation activity was in the sand sample. That the appearance of the highest level of radioactivity in sand samples is due to the containment of these samples on the proportion of radioactive minerals such as monazite and zircon minerals, which are an important source for obtaining uranium and thorium irradiated.

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