

## **An investigation on the natural radioactivity of $Th^{232}$ , $Ra^{226}$ and $K^{40}$ in some samples of raw building materials in Governorate of Karbala.**

**اختبار النشاط الإشعاعي الطبيعي لـ  $Th^{232}$  ,  $Ra^{226}$  ,  $K^{40}$  في بعض نماذج خام مواد البناء في محافظة كربلاء.**

Hassan Issa Dawood

Physical Department-College of Education-Qadisiya University

### **Abstract:**

The study of the radiation hazards from raw building materials were performed in the present work. The measurements of the natural radioactivity of  $Th^{232}$  ,  $Ra^{226}$  and  $K^{40}$  were verified for some Karbala raw building materials (sand , cement , gypsum and thermiston powder) to assess any possible radiological hazard due to their use. The measurements were done by gamma-ray spectroscopy using Hyper-Pure Germanium (HPGe) detector. The results were found to vary from  $(18.88\pm 4.34$  to  $54.33\pm 7.37$  ,  $13.085\pm 3.617$  to  $82.80\pm 9.09$  and  $14.185\pm 3.766$  to  $140.523\pm 11.854$ ) Bq/Kg for  $Th^{232}$  ,  $Ra^{226}$  and  $K^{40}$  respectively. The concentrations of these radionuclides are compared with the available data from other countries. The measured activity concentration of  $Th^{232}$  ,  $Ra^{226}$  and  $K^{40}$  in raw building materials is higher than the world average except the activity of  $K^{40}$  show lower than the available data previously. Radium equivalent activities (Req) are calculated for the analyzed samples to assess the radiation hazard arising due to the use of these raw building materials samples in the construction of dwellings. All the raw materials have radium equivalent activities lower than the limit  $(370)$  Bq/Kg. As well as calculated values of external radiation doses in air are also lower than the world average of about 0.5 mSv per year.

### **الخلاصة:**

دراسة مخاطر الإشعاع الناجمة من خام مواد البناء أجريت في هذا العمل الحالي. تم قياس النشاط الإشعاعي لـ  $Th^{232}$  ,  $Ra^{226}$  ,  $K^{40}$  لبعض نماذج خام مواد البناء في محافظة كربلاء (رمل , سميت , جص , باوذر الترمستون) لتجنب أية احتمالية وجود مخاطر إشعاع بيولوجية نتيجة استعمال هذه المواد. تمت القياسات باستعمال كاشف مطياف أشعة كاما عالي النقاوة (HPGe). ووجدت من خلال نتائج القياس أنها تتراوح (من  $18.88\pm 4.34$  إلى  $54.33\pm 7.37$  , ومن  $13.085\pm 3.617$  إلى  $82.80\pm 9.09$  , ومن  $14.185\pm 3.766$  إلى  $140.523\pm 11.854$ ) Bq/Kg لكل من  $Th^{232}$  ,  $Ra^{226}$  و  $K^{40}$  وعلى التوالي. تراكيز النشاط الإشعاعي لهذه النويدات المشعة تمت مقارنتها مع النتائج المتوفرة لبقية الدول. ثبتت النتائج إن تراكيز هذه النويدات ( $Th^{232}$  ,  $Ra^{226}$  ,  $K^{40}$ ) في خام مواد البناء هي أعلى من معدلات القيم العالمية ماعدا تركيز النشاط الإشعاعي لـ ( $K^{40}$ ) هي أدنى من القيم المتوفرة سابقا. تم حساب النشاط الإشعاعي لمكافئ الراديوم (Req) لهذه النماذج المختارة لتلاشي خطورة الإشعاع الناجمة من نماذج خام مواد البناء هذه والمستخدمة في بناء المساكن. جميع خام مواد البناء في هذا البحث لها نشاط مكافئ الراديوم وأنها دون الحد المسموح بها (370) Bq/Kg. إضافة إلى ذلك تم حساب معدل جرعة الإشعاع الخارجية المتعرضة لها في الهواء ووجدت أنها دون الحد (0.5) msv في السنة.

### **Introduction:**

The most abundant sources of external and internal radiation exposure in homes are gamma rays ,emitted by members of the uranium ( $U^{238}$ ) and thorium ( $Th^{232}$ ) decay chains as well as the Potassium ( $K^{40}$ ) , which are exhaled by building materials. Knowledge of the radioactivity present in building materials enables one to assess any possible radiological hazard caused by the use of such materials. The study has been satisfied in most countries [1,2,3,4].

The natural radioisotopes are present at different content. The concentration of natural isotopes is common in our building materials and homes [5,6]. All building materials contain various amounts of natural radioactivity nuclides. Materials derived from cement , sand , gypsum and powder of thermiston contain mainly natural radioisotopes of the uranium ( $U^{238}$ ) , thorium ( $Th^{232}$ ) and the radioisotope of potassium ( $K^{40}$ ) [7,8].

The question of radioactivity in raw building materials is of the particular interest , as it has become clear that the use of certain raw building material components can result in significant increase of the radioactive burden for a large number of persons. Building materials contribute to environmental radioactivity in two way: firstly by gamma radiation , mainly  $K^{40}$  ,  $Ra^{226}$  and  $Th^{232}$  to a whole human body ( external exposure) and the second by releasing the invisible odourless radon gas, the radioactive daughters of which are deposited in the human respiratory tract (internal exposure) [9,10,11].

The study in the present work performed to measure the natural radioactivity of  $Th^{232}$  ,  $Ra^{226}$  and  $K^{40}$  in some samples of raw building materials (sand , cement , gypsum and thermiston powder) by high purity germanium detector (HPGe).

### **Preparation of samples for $\gamma$ -spectrometry:**

A total of 10 samples were collected from different regions of Governorate of Karbala: three samples of sand from Al-hur , Al-akhaither and Karbala center , two cement samples from factory of cement in Karbala (Ain Al-tamer) , three gypsum samples and two samples of thermiston powder. The samples were ground into a fine powder with a particle size less than 1mm. The samples were then dried in a temperature-controlled furnace at  $100C^{\circ}$  for 24 hour to remove the moisture. After moisture removal , these samples were cooled in a moisture-free atmosphere. Each sample was then filled into cylindrical plastic containers and hermetically sealed.

### **Measurement system:**

After preparation the samples of raw building materials , each sample was measured the activity concentration by Germanium spectrometer with highest resolution (HPGe) that manufactured by Telnelec company in U.S.A , with size of crystallized  $(2.9)cm^3$  , diameter (63)mm and hight (67)mm as shown in Fig(1).It working at (2100) volt with the efficiency of 40%. The viability of discrimination detector of energy (1.75) Kev at energy (1.332) Mev that returns to ( $CO^{60}$ ). The detector externally surrounded by lead shield to prevent the background radiation and internally surrounded with thin layer of (Cu) and (Ca) to attenuation the x-ray radiation. The detector was cooled to temperature of  $77K^{\circ}$  by nitrogen liquid.



**Fig(1) HPGe system for the counting of radionuclides**

**Method of calculation:**

The activity concentration (in Bq kg<sup>-1</sup>), AE<sub>i</sub>, of a radionuclide( i ) and for a peak at energy E, is given by [12]:

$$AE_i = CE_i / (ceff \cdot \gamma \cdot m \cdot t) \text{----- (1)}$$

Where

CE<sub>i</sub>: is the net area of a peak at energy E in (Bq/Kg).

Ceff: is the detection efficiency at energy E.

t : is the counting time in (sec).

γ : is the number of gammas per disintegration of the radionuclide i for a transition at energy E.

m : is the mass in (kg) of the measured sample.

If there is more than one peak in the energy analysis range for a nuclide, then an attempt to average the peak activities is made.

Radium equivalent activity; The radiation hazards associated with the radionuclides are estimated by calculating the radium equivalent activity (Raeq). It is a weighted sum of activities of Ra<sup>226</sup>, Th<sup>232</sup>, and K<sup>40</sup>; and it is based on the assumption that 370 Bq.kg<sup>-1</sup> of Ra<sup>226</sup>, 259 Bq.kg<sup>-1</sup> of Th<sup>232</sup>, and 4810 Bq.kg<sup>-1</sup> of K<sup>40</sup> produce the same gamma radiation dose rate [13,14]. To avoid radiation hazards, materials whose Raeq is greater than 370 Bq kg<sup>-1</sup> should not be used. Raeq is defined by the following formula [15]:

$$R_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \text{ -----(2)}$$

where  $A_{Ra}$ ,  $A_{Th}$ , and  $A_K$  are the activity concentrations of  $Ra^{226}$ ,  $Th^{232}$ , and  $K^{40}$ , respectively.

Air absorbed gamma radiation dose rate; Effects of gamma radiation are normally expressed in terms of the absorbed dose rate in air, which originate from radioactive sources (raw building materials). The activity concentrations in these raw materials of building correspond to the total absorbed dose rate in air above the ground level. The absorbed dose rate in air (D) for the population living in the studied area is calculated using the following equation [13,14]:

$$D = (F_{Ra} \cdot A_{Ra} + F_{Th} \cdot A_{Th} + F_K \cdot A_K) \times 10^{-6} \text{ -----(3)}$$

where D is the absorbed dose rate in air ( $nGy h^{-1}$ ).  $F_{Ra}$ ,  $F_{Th}$ , and  $F_K$  are the dose conversion factors for  $Ra^{226}$ ,  $Th^{232}$ , and  $K^{40}$  respectively. They are taken as 4.27, 6.62, and 0.43 for  $Ra^{226}$ ,  $Th^{232}$ , and  $K^{40}$ , respectively as assessed by [16].

### **Result and discussion:**

In this study, 10 samples of raw building materials were measured for the natural radionuclides ( $Th^{232}$ ,  $Ra^{226}$  and  $K^{40}$ ) they contain. The activity concentrations of these three radionuclides were calculated as shown in table (1) using the formula proposed by [12] to compare the overall biological damage. The mean activity concentrations of  $Th^{232}$ ,  $Ra^{226}$  and  $K^{40}$  were calculated in all samples that collected from Governorate of Karbala as reported in table (2). Table (3) represent the comparison of the mean values of activity concentrations of these three radionuclides in raw building materials sand and cement with the available data that reported previously, It is cleared from this table the measured values of all activities in the present study higher than the world average except the activity of  $K^{40}$  in sample of cement within the permitted limits. Later, radium equivalent activities (Req) and the Air absorbed gamma radiation dose rate (D) were calculated using the formula by [13,14,15] as pointed in table (4), from the data of external radiation doses were calculated in this table are found lower than the world average (0.5 msv/year). Table (5) verified the Comparison of the mean radium equivalent in (Bq/Kg) of raw building materials(sand and cement) of the present work with the average data exist in the other countries and it is clear from this table that the Req of this study in the acceptable range.

**Conclusions:** The present study has been carried out to establish baseline data regarding concentration levels of naturally occurring radionuclides of  $Th^{232}$ ,  $Ra^{226}$  and  $K^{40}$  in raw building materials and the corresponding radiation doses in Governorate of Karbala. Measured mean activity concentrations of the 3 radionuclides are found to be higher than the world's average values. Calculated values of external radiation doses are lower than the world average of about 0.5 mSv per year. To estimate the potential radiological health risk in raw materials, the dose rate associated with  $Cs^{137}$  in raw building materials studied should be investigated as well. However, the data generated here may be useful for the introduction of radiation safety standards by the authorized organizations for the protection of general population from radiation hazards owing to terrestrial sources.

Table (1):The activity concentrations of Th<sup>232</sup> , Ra<sup>226</sup> and K<sup>40</sup> in Bq/Kg .

Samples and locations	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>
Sand of Al-hur	41.206±6.419	13.085±3.617	140.523±11.854
Sand of Imam Ali	38.933±6.239	24.25±4.924	14.185±3.766
Sand of Al-akhaither	54.337±7.371	50.357±7.096	24.321±4.931
Cement of Karbala (1)	46.138±6.792	82.80±9.09	82.413±9.078
Cement of Karbala (2)	33.212±2.391	69.09±5.112	65.112±5.061
Thermiston powder(1)	18.88±4.345	-----	64.239±8.014
Thermiston powder(2)	19.120±3.109	-----	51.278±3.331
Gypsum of Imam Ali (1)	45.401±6.738	39.927±6.318	122.044±11.047
Gypsum of Aon (2)	33.019±2.172	41.213±2.076	119.921±5.013
Gypsum of Al-hur (3)	39.127±3.071	32.019±1.911	117.022±3.129

Table (2):Means and ranges of the activity concentrations for radionuclide's in (Bq/Kg)

Radioisotopes	Mean value	Range value
Th <sup>232</sup>	36.937	18.880 → 54.337
Ra <sup>226</sup>	44.092	13.085 → 82.80
K <sup>40</sup>	80.105	14.185 → 140.523

Table(3) Comparison the mean values of activity concentrations of Th<sup>232</sup> , Ra<sup>226</sup> and K<sup>40</sup> in(Bq/Kg) for raw building materials (sand and cement) with the corresponding values reported previously.

Type of material	Th <sup>232</sup>	Ra <sup>226</sup>	K <sup>40</sup>	Reference
Sand	44.82±6.67	29.23±5.21	59.676±6.85	Present work
	3.3±1.3	9.2±2.5	47.3±9.0	[17]
	2.91±0.1	9.26±3.28	54.52±6.64	[18]
Cement	39.67±4.59	75.94±7.1	73.76±7.06	Present work
	11.8±3.0	37.6±6.0	178.6±15	[19]
	11.1±1.1	31.3±3.6	48.6±4.0	[17]
	3.99±1.02	22.51±3.23	68.76±8.29	[18]

Table (4):The radium equivalent activity (Req) and the Air absorbed gamma radiation dose rate (D) .

Samples and locations	Req in (Bq/Kg)	D in (nGy h <sup>-1</sup> )
Sand of Al-hur	82.829	0.000389
Sand of Imam Ali	81.016	0.000367
Sand of Al-akhaither	129.931	0.000585
Cement of Karbala (1)	155.123	0.000694
Cement of Karbala (2)	121.596	0.000542
Thermiston powder (1)	31.944	0.000152
Thermiston powder (2)	31.290	0.000148
Gypsum of Imam Ali (1)	114.247	0.000523
<b>Gypsum of Aon (2)</b>	<b>97.664</b>	<b>0.000446</b>
<b>Gypsum of Al-hur (3)</b>	<b>96.981</b>	<b>0.000446</b>

Table (5): Comparison the mean radium equivalent in (Bq/Kg) of sand and cement with the average data in the worlds

Countries	Mean Req of sand	Mean Req of cement	References
Iraq(Karbala)	97.92	138.359	Present work
Germany	118	123	[20]
Bangladesh	592	348	[21]
Malaysia	152	181	[22]
Spain	59	137	[23]
USSR	70	126	[20]
Australia	70	130	[24]

**References:**

- 1- sharma, D.K., Kumar, A., Kumar, M. and Singh, S. (2003) Radiation Measurements, Vol. 36, p.363.
- 2- Petropoulos, N.P., Anagnostakis, M.J. and Simopoulos, S.E. (2002) J. Environ. Radioactivity, Vol. 61, p.257.
- 3- Keller, G., Hoffmann, B. and Feigenspan, T. (2001) The Science of the Total Environment, Vol. 272, p.85.
- 4- Khan, A.J., Prasad, R. and Tyagi, R.K. (1992) Nucl. Tracks Radiat. Meas., Vol. 20, No. 4, p.609.
- 5- Radiological Protection Principles concerning the Natural Radioactivity of Building Materials. Radiation protection 112. European Commission.1999.
- 6- Naturally occurring radioactive materials (NORM IV). Proceeding of International Conference held in Szczyrk, Poland, May 2004. IAEA 10/2005.
- 7- Nguyen Hao Quang. Radioactivity of building materials in Hanoi. Report. National Conference on nuclear physics and nuclear technique. Hanoi. 1999, in Vietnamese.
- 8- Vietnam National Radiation and Nuclear Safety Decree No. 50. 1999, in Vietnamese.
- 9- IEC 6157:2000. Radiation protection instrumentation – Radon and radon decay product measuring instrument.
- 10- Ta Minh Hoang, Luu Thi Hong, Nguyen Van Doan. Report and draft of national building standard “Natural radioactivity of building materials-Levels and measuring method”. Vietnam Institute for building materials.2007, in Vietnamese.
- 11- Ta Minh Hoang, Kieu Thi Y Sao. Report and draft of national building standard “ Natural radon activity in buildings – Levels and general requirements of measuring methods”. Vietnam Institute for building materials.2008, in Vietnamese.
- 12- Tzortzis, M., Tsertos, H., Christofides, S.and Christodoulides, G., ”Gamma-Ray Measurements of Naturally Occurring Radioactive Samples from Cyprus Characteristic Geological Rocks”, Radiation Measurement 37 (3) 221-229, 2003.
- 13- Matiullah, Ahad, A., Ur Rehman, S., Ur Rehman, S.and Fahee, M., ”Measurement of Radioactivity in the Soil of Bahawalpur Division, Pakistan”, Radiat. Prot.Dosimetry 112 (3), 443-447, 2004.
- 14- Fatima, I., Zaidi, J.H., Arif, M., Daud, M., Ahmad,S.A., Tahir, S.N.A., ”Measurement of natural radioactivity and dose rate assessment of terrestrial gamma radiation in the soil of southern Punjab, Pakistan”, Radiat. Prot. Dosimetry, 128(2), 206-212, 2008.
- 15- Yu, K.N., Guan, Z.J., Stoks, M.J., Young, E.C., 1992. The assessment of natural radiation dose committed to the Hong Kong people. J. Environ. Radioactivity 17, 31.

- 16- United Nations Scientific Committee on Effects of Atomic Radiation, "Sources and Effects of Ionising Radiation", UNSCEAR Report, New York, 1993.
- 17- Sharaf, M., Mansy, M., El Sayed, A. and Abbas, E. (1999) Radiation Measurements, Vol. 31, p.491.
- 18- Eissa, M.F., Mostafa, M.R., and Shahin, F. (2008) Natural Radioactivity of Some Egyptian Building Materials, Int.J.Radiation, Vol.5, No.1.
- 19- El-Arabi, A.M., Abbady, A., El-kamel, A.H., Nosier, A. and Moustafa, A. (2004) VII Radiation Physics & Protection Conference, Ismailia-Egypt, 27–30 November.
- 20- United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. Report to the General Assembly. New York, N.Y.: United Nations; 1977.
- 21- Mollah, A. S.; Ahmed, G. U.; Husain, S. R.; Rahman, M. W. The natural radioactivity of some building materials used in Bangladesh. Health Phys. 50:849-851 ; 1985.
- 22- Chong, C.S. Gamma activity of some building materials in west Malaysia. Health Phys. 43:272; 1982.
- 23- Garzon, L.; Fontenla, P.; Suarez, A. Radioactivity of building materials. Absorbed doses. In: Vohra, K. G.; Mishra, U. C.; Pillai, K. C.; Sadasivan, S., eds. Natural radiation environment. New Delhi: Wiley Eastern Limited; 1982:544-546.
- 24- Beretka, J.; Mathew, P.J. Natural radioactivity of Australian building materials, industrial wastes and by-products. Health Phys. 48:87-95; 1983.