

Estimating the risk of exposure to internal and external radiation resulting from Portland cement

Ahmad Yahya Raham, and Prof. Dr. Hadi D. Alattabi

Department of Physics, College of Science, Wasit University, Iraq.

Abstract

The current study assessed specific activity concentrations in seventeen cement samples from varied origins using a high-purity germanium (HPGe) detector. Results shows that the specific activity for (^{226}Ra) ranged from ($10.6 \pm 0.7 \text{ Bq/kg}$) in sample number 11, to ($59.1 \pm 2.4 \text{ Bq/kg}$) in sample number 12. While (^{228}Ac) ranged from ($6.6 \pm 1 \text{ Bq/kg}$) in sample number 9 to ($10.5 \pm 1 \text{ Bq/kg}$) in sample number 7. Moreover, (^{40}K) was ranged from ($81.4 \pm 7.6 \text{ Bq/kg}$) in sample number 12 to ($209.5 \pm 14.1 \text{ Bq/kg}$) in sample number 7. The study shows that there are no significant sources of radiation threat in the cement samples, making them safe for use in home construction.

Keywords: Portland Cement, Radiation, and ^{226}Ra .

1. Introduction

Earth is radioactive from the beginning of time. There are more than 60 radionuclides in nature. In addition to air, water, and soil, radionuclides can also be found in humans since we are products of our surroundings [1]. Every day, human ingest or take in nuclides from food, water we drink, and air that we breathe [2]. Radioactivity can be found in the rocks, soil, seas, and even the building materials that utilised to build houses [3]. All raw materials contain varying concentrations of

naturally occurring radionuclides from the uranium and thorium family as well as the radioactive potassium isotope (^{40}K) [40]. Materials and completed building products made of rock and soil. It has long been recognized that some building materials naturally contain more radioactivity than others [5]. Exposure can occur both inside and externally to even extremely low quantities of radioactivity that naturally occur in construction materials [6]. The external radiation exposure is brought on by gamma radiation from the decay chains of uranium and thorium as well as from

potassium-40 [7]. Conversely, the short-lived radon generates products that are expelled into the space by construction materials cause the internal radiation exposure [8]. That mostly affects the respiratory system, therefore, it's important to comprehend a building's radioactivity materials to determine the health risks posed by radioactivity to people [9-10]. The three most important naturally occurring radionuclides in cements are (^{226}Ra), (^{228}Ac), and (^{40}K) [11].

2. Instrumentation and Calibration

Using a high purity germanium (HPGe) detector provided by Ortec EG&G. Operating at 3500 V, the detector is an n-type gammaX-ray (GMX) detector with a useable energy range of 3 keV to 10 MeV. At 1.33 MeV of ^{60}Co , the standard energy resolution was 2.02 keV, and the relative efficiency was 56.9 %. This was done by calibrating the detector's absolute efficiency using the IAEA standard "soil-6" source inside a 90 mm diameter and 10 mm thick Petri dish. For twelve hours, the spectrum was gathered. The peak efficiency curve was created by plotting the log of efficiency against the log of peak energy, using the areas under the energy peaks of interest [12]. The curve was fitted with a polynomial, and the outcome was saved for later use. Gamma ray transitions to measure

concentrations of the assigned nuclides in the series, assuming that secular equilibrium was attained between ^{226}Ra and its short-lived results, are as follows. In the studied samples, ^{238}U activity were obtained from the weighted averages of the ^{234}Th photopeak's (63.3, 92.4, and 92.8 keV). The mean activity of the three distinct photopeak's of its nuclides, ^{214}Pb at (295.2 and 352.0 keV) and ^{214}Bi at (609.3 keV), was used to calculate the activity of ^{226}Ra . To determine ^{232}Th , the photopeak's of ^{212}Pb (at 583.1 keV), ^{208}Tl (at 238.6 keV), and ^{228}Ac (at 911.1 keV) were utilized. The ^{40}K was obtained directly from the 1460.8 keV photopeak. The ambient activity concentration (A) in Bq.kg-13

3. Material and Methodology

Seventeen samples of cement types produced by Cement Iraq Factory (Portland cement, Portland Pozzolanic cement (5 %), Portland Pozzolanic cement (25 %), White cement and Sulphate Resistant Cement (S.R.C.)) were collected for this study [13-15]. The percentage values in parentheses above indicate the percent of Pozzolana in cement. For comparison with products from other factories. Eight samples were selected from the ordinary Portland cement from (Arabia Company, Ashamaliya Company, and Al-Rajhi Company). Four samples were collected from Portland Pozzolanic

cement (25 %), from Arabia Company and Al-Rajhi Company. Also, four samples collected from (S.R.C.). Moreover, for individual sample, a 1kg in weight, was dried in an oven at 110 °C for 24 hours to ensure that moisture is completely removed. Then, samples were crushed, homogenized and sieved through a 200 mesh, which is the optimum size to be enriched in heavy minerals. Weighed samples were placed in a polyethylene beaker of 350-cm³ volume. Beakers were completely sealed for four weeks to reach secular equilibrium, where the rate of decay of the radon. This step is necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample 4.

4. Equipment and Standardisation

High purity germanium (HPGe) detector provided by EG&G Ortec was used for the measurements. Operating at 3500 V, the detector is an n-type gamma X-ray (GMX) detector. a standard energy resolution of 2.02 keV, a practical energy range of 3 keV to 10 MeV, and a 56.9 % relative efficiency of ⁶⁰Co at 1.33 MeV. This was done by calibrating the detector's absolute efficiency using the IAEA standard "soil-6" source inside a 90 mm diameter and 10 mm thick Petri dish. For twelve hours, the spectrum was gathered.

The peak efficiency curve was created by plotting the log of efficiency against the log of peak energy, using the areas under the energy peaks of interest. The curve was fitted with a polynomial, and the outcome was saved for later use. Hence, gamma ray transitions to measure concentrations of the assigned nuclides in the series, assuming that secular equilibrium was attained between ²²⁶Ra and its short-lived daughters. The weighted means of the ²³⁴Th photo peaks (63.3, 92.4, and 92.8 keV) were used to calculate the ²³⁸U activity in the datasets under study. The three distinct photo peaks of its daughter nuclides, ²¹⁴Pb at (295.2, and 352.0 keV) and ²¹⁴Bi at (609.3 keV), were averaged to determine the activity of ²²⁶Ra. The photo speaks of ²¹²Pb (at 583.1 keV), ²⁰⁸Tl (at 238.6 keV), and ²²⁸Ac (at 911.1 keV) were utilized to determine ²³²Th. Using the 1460.8 keV photopeak, ⁴⁰K was directly the following equation was used to determine the activity concentration (A) in Bq.kg-1 in the ambient samples [15].

$$175 A = \frac{N_p}{exE xm} \quad (1)$$

Where E is the measured efficiency for each gamma-ray peak observed for the same number of channels. Either for the sample or the calibration source, and *m* is the sample mass in kilograms. *N_p* is the difference between counts per second of the sample and counts per second of the background [16]. A minimum number of

spectrum lines coming from naturally occurring radionuclides, which may be present in the system components and the surrounding environment of the counting facility, must be present for the counting system to have a background as low as possible. The background count rates of natural radionuclides were measured in the current investigation at least twice a week, for a total counting duration of 80,000 s. The spectrum of the measurements was recorded in a PC-based multichannel analyser (MCA). The gamma-ray spectra were automatically calculated and entered the PC-based MCA when the predetermined amount of time had passed.

4.1 Absorbed Gamma Dose Rate, D (nGy/h)

Outdoor air gamma absorbed dose rate (D_Y) in (nGy/h) due to terrestrial gamma rays at height (1 m) above the ground surface which can be computed from specific activities AU, ATh and AK of ^{238}U , ^{232}Th and ^{40}K in (Bq/kg) respectively using the following relation [17].

$$D(\text{nGy/h}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (2)$$

where C_{Ra} , C_{Th} and C_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Bq.kg-1, respectively.

4.2 Radium Equivalent Index, Ra_{eq}

It is a widely hazard index used when comparing the specific activity of the samples which containing different amounts of ^{226}Ra , ^{232}Th and ^{40}K . It is supposed that 370 Bq.kg-1 of ^{226}Ra , 259 Bq kg-1 of ^{232}Th , and 4810 Bq kg-1 of ^{40}K produce the same γ -radiation dose rate [17].

$$Ra_{eq} = \left(\frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \right) \quad (3)$$

Where, C_{Ra} , C_{Th} and C_K are the activity concentrations (Bq/Kg) of ^{226}Ra , ^{232}Th and ^{40}K , respectively. The radium equivalent calculation (Ra_{eq}) is ranged from 136.040 to 192.015 with an average 163.406 for the surface samples, while it is ranged from 139.91 to 187.53 with an average 160.45 Bq kg-1 for the deep samples [18].

4.3 Representative Level Index ($I_{\gamma r}$)

The primary purpose of another radiation hazard index was to determine the amount of γ radiation linked to various concentrations of certain radionuclides. Its definition is displayed in the formula that follows.

$$I_{\gamma r} \text{Bq. Kg}^{-1} = ((1/150) A_{Ra} + (1/100) A_{Th} + (1/1500) A_K) \quad (4)$$

where A_{Ra} , A_{Th} and A_K are the respective activity concentration values of ^{226}Ra , ^{232}Th and ^{40}K in Bq.kg⁻¹ [19].

4.4 External Hazard Index, H_{ex}

Many radioactive materials decay naturally and when these materials decay produces external radiation field which exposed humans. In terms of dose, the principal primordial radionuclides are ^{232}Th , ^{226}Ra and ^{40}K . Thorium and uranium head series of radionuclides that produce significant human exposure. The external hazard index (H_{ex}) is calculated by equation (5) [19].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (5)$$

Where, concentration of Ra, concentration of ^{228}Ac , and concentration of ^{40}K represent the levels in Bq kg⁻¹ of radium, Actinium, and potassium [10].

4.5 Internal radiation hazard (H_{in})

In addition to the external irradiation, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}) which is given by the following formula [11].

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Where, concentration of ^{226}Ra , concentration of ^{228}Ac represent the activity concentrations of ^{226}Ra , concentration of ^{228}Ac , concentration of ^{40}K in Bq kg⁻¹ [20].

5. Results and discussion

The distribution of natural radionuclides in different brands of cements is presented in (table 1). From (table 1) the activity concentration of ^{226}Ra were varies from 10.6 to 59.1 Bq.kg⁻¹. The activity concentration of AC-228 varies from 5.7 to 10.5 Bq.kg⁻¹. The activity concentration of ^{40}K varies from 81.4 to 209.5. The mean ^{226}Ra and ^{232}Th values are slightly higher than the corresponding worldwide average values which are 35 and 30 Bq.kg⁻¹.

Where, ^{40}K values are lower than the corresponding worldwide average (400 Bq.kg⁻¹) as shown in (table 2). Figure one shows the activity concentrations of natural radionuclides for the cement types. Since the distribution of the natural radionuclides in each cement type is not uniform. A common index termed radium equivalent activity (Ra_{eq}) is required to obtain the total activity and is also used to assess the gamma radiation hazards.

The radium equivalent of the total activity of each cement type is shown in (table 2). The highest value of Ra_{eq} is seen with white cement (125.49 ± 6.12 Bq.kg⁻¹) and the lowest with Portland Pozzolanic cement (5 %) with an average value of (78.08 ± 4.03 Bq.kg⁻¹) [21-22].

Table 1: Results of samples measuring in Bq/kg unit.

Number	Sample	C/Ra-226	C/AC-228	C/K-40
1	S1	17.1	9.5	160.2
2	S2	28.1	8	120.1
3	S3	13.1	10.4	143.9
4	S4	16.5	9.3	121.5
5	S5	25.1	9.1	141.3
6	S6	13.5	10.3	126.5
7	S7	39.2	10.5	209.5
8	S8	29.8	7.1	153.5
9	S9	27.3	6.6	116.8
10	S10	31.2	7.3	127.2
11	S11	10.6	7.2	120.8
12	S12	59.1	7	81.4
13	S13	23.8	5.7	99.5
14	S14	14.5	9.7	119.6
15	S15	22.5	8.2	135.4
16	S16	13.7	9.1	102.2
17	S17	33.6	9.3	158.4
18	Orval Range	24.629 4	8.488	131.635 3

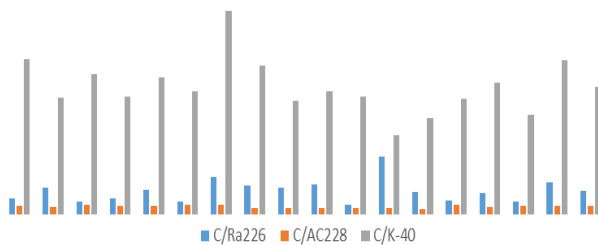


Figure 1: concentrations of ^{226}Ra , ^{228}Ac , and ^{40}K .

Table 2: Radium equivalent activity (Ra_{eq}), representative level index ($I_{\gamma r}$), gamma dose rate (D), external hazard index (H_{ex}) and internal hazard index (H_{in}), for different brands of cement.

Number	Sample	Ra226 _{eq}	H _{ex}	H _{in}	DnGy/h	I _r	Ef _{ind} mSv/y	Ef _{out} mSv/y
1	S1	43.0204	0.116201	0.162418	20.31854	0.3158	0.099675	0.024919
2	S2	48.7877	0.131803	0.207749	22.82237	0.3474	0.111957	0.027989
3	S3	39.0523	0.105477	0.140882	18.33443	0.287267	0.089941	0.022485
4	S4	39.1545	0.105762	0.150356	18.30675	0.284	0.089806	0.022451
5	S5	48.9931	0.132349	0.200187	22.98481	0.352533	0.112754	0.028189
6	S6	37.9695	0.102554	0.139041	17.73325	0.277333	0.086992	0.021748
7	S7	70.308	0.190042	0.295988	33.18855	0.506	0.16281	0.040702
8	S8	51.7725	0.139866	0.220407	24.45695	0.372	0.119976	0.029994
9	S9	45.7316	0.123549	0.197333	21.46956	0.325867	0.105321	0.02633
10	S10	51.4334	0.138955	0.223279	24.12784	0.3658	0.118362	0.02959
11	S11	30.136	0.081562	0.110211	14.28336	0.2232	0.070068	0.017517
12	S12	75.3778	0.20368	0.36341	34.92658	0.518267	0.171336	0.042834
13	S13	39.6125	0.107018	0.171342	18.58755	0.282	0.091183	0.022796
14	S14	37.5802	0.101506	0.140695	17.54512	0.2734	0.086069	0.021517
15	S15	44.6518	0.120621	0.181432	20.99398	0.32226	0.102988	0.025747
16	S16	34.5824	0.09341	0.130437	16.08754	0.25046	0.078919	0.01973
17	S17	59.0958	0.15965	0.25046	27.74568	0.4226	0.136109	0.034027
18	Range	46.89762	0.126706	0.193272	21.9948	0.336835	0.107898	0.026974

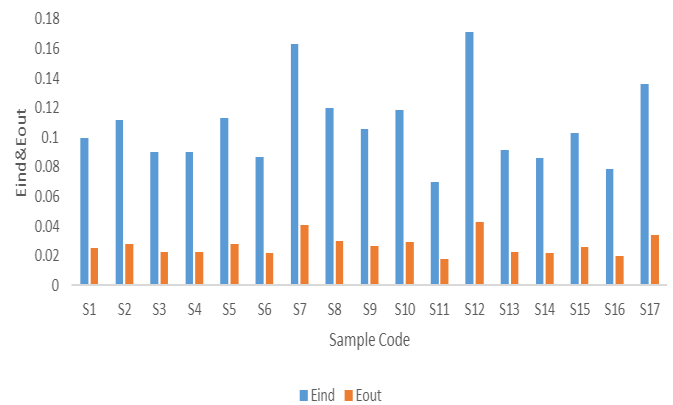


Figure 2: difference between indoor and outdoor samples radiations.

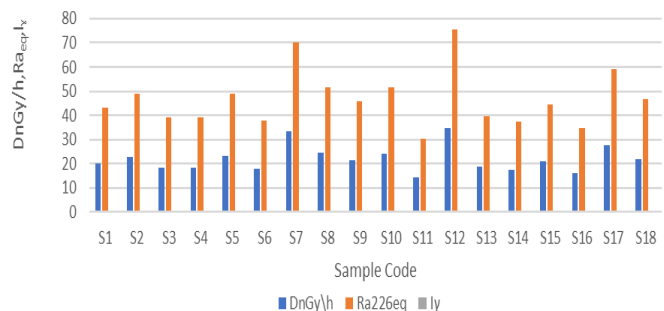


Figure 3: Difference in concentrations between DnGy/h , Ra_{eq} , and I_{γ} .

However, all cement samples, and the reported results were within the international standard values, calculations of dose rate, annual effective dose, and radium equivalent external hazard.

6. Conclusion

Specific radioactivity of ^{226}Ra , ^{228}Ac , ^{40}K , Ra_{eq} equivalent, beside the rest of the indicators, are evaluated through these values' Potential radiation risks for cement and building materials. These results determine the radiation content of buildings and the risks accompanying radiation. The specific activity of ^{226}Ra was evaluated and calculated. ^{228}Ac , ^{40}K and indicators for risk assessment associated with cement and raw materials and the results were within the permissible limits according to scientific recommendations Scientific Committee at the United Nations. In comparison with some countries in the world, results show less of concentration values were equal to some Ra_{eq} (31.7) Bq/kg which is less than the limit Allowed (370 Bq/kg).

7. References

1. Puertas F., Suárez-Navarro J. A., Alonso López M., and Gascó C., (2021). NORM waste, cements, and concretes. A review.
2. Sahoo P., and Joseph J., (2021). Radioactive hazards in utilization of industrial by-products: Comprehensive review. *Journal of Hazardous, Toxic, and Radioactive Waste*. 25, 3, 03121001.
3. Eskander S. B., Bayoumi T. A., and Saleh H. M., (2013). Leaching behavior of cement-natural clay composite incorporating real spent radioactive liquid scintillator. *Progress in Nuclear Energy*. 67, 1-6.
4. Rakhimova N., (2022). Recent advances in alternative cementitious materials for nuclear waste immobilization: A review. *Sustainability*. 15, 1, 689.
5. Collier N. C., Milestone N. B., and Travis K. P., (2019). A review of potential cementing systems for sealing and support matrices in deep borehole disposal of radioactive waste. *Energies*. 12, 12, 2393.
6. Varlakov A., Zhrebtsov A., Ojovan M. I., and Petrov V., (2021). Long-term irradiation effects in cementitious systems. In *Sustainability of Life Cycle Management for Nuclear Cementation-Based Technologies*. Woodhead Publishing. 161-180.
7. Haneefa K. M., Santhanam M., and Parida F. C., (2013). Review of concrete performance at elevated temperature and hot sodium exposure applications in nuclear industry. *Nuclear Engineering and Design*. 258, 76-88.

8. Paria S., and Yuet P. K., (2006). Solidification–stabilization of organic and inorganic contaminants using portland cement: a literature review. *Environmental reviews*. 14, 4, 217-255.
9. Phung Q. T., Maes N., and Jacques D., (2017). Current concerns on durability of concrete used in nuclear power plants and radioactive waste repositories. In *Congrès International de Géotechnique–Ouvrages–Structures*. Singapore: Springer Singapore. 1107-1121.
10. Walton J. C., Plansky L. E., and Smith R. W., (1990). Models for estimation of service life of concrete barriers in low-level radioactive waste disposal.
11. Raham A. Y., and Al-Attabi H. D., (2023). Study and calculation of the specific natural radioactivity of samples of Portland cement in Iraq using a high-purity germanium detector. *Nexo Revista Científica*. 36, 06, 1141–1149.
12. Roczniak J., Pluta J., Tudyka K., Poreba G., and Szymak A., (2023). A new fast screening method for estimating building materials hazard indices with correlated inputs. *Journal of Radioanalytical and Nuclear Chemistry*. 332.
13. Michael Dine, (2007). *Supersymmetry and String Theory: Beyond the Standard Model*. Cambridge University Press.
14. Pervin S., Dewan Md J., Apon A., Siraz M. M., and Yeasmin S., (2021). Measurement of natural radioactivity and its health hazards associated with the use of different branded cement samples collected from different manufactures in Dhaka city using gamma spectrometry. *Journal of Bangladesh Academy of Sciences*. 45, 95-104.
15. Kassi B., Boukhair A., Azkour K., Fahad M., Benjelloun M., and Nourreddine A., (2018). Assessment of Exposure Due to Technologically Enhanced Natural Radioactivity in Various Samples of Moroccan Building Materials. *World Journal of Nuclear Science and Technology*. 08, 176-189.
16. Estokova A., and Palašćáková L., (2013). Assessment of Natural Radioactivity Levels of Cements and Cement Composites in the Slovak Republic. *International journal of environmental research and public health*. 10, 7165-7179.
17. Legasu M. L., and Chaubey A. K., (2022). Determination of dose derived from building materials and radiological health related effects from the indoor environment of Dessie city, Wollo, Ethiopia. *Heliyon*. 8, 3, e09066.
18. Oleiwi M. H., Oleiwi M. R., and Oraibi A., (2020). Determination of radiological hazards resulting from

- natural radiological activity in soil samples in Al Azizia district in Wasit Governorate, Iraq. *Journal of Physics: Conference Series*. 1660.
19. Kořátková J., Zatloukal J., Reiterman P., and Kolář K., (2017). Concrete and cement composites used for radioactive waste deposition. *Journal of environmental radioactivity*. 178, 147-155.
20. Lewicka S., Piotrowska B., Łukaszek-Chmielewska A., and Drzymała T., (2022). Assessment of natural radioactivity in cements used as building materials in Poland. *International Journal of Environmental Research and Public Health*. 19, 18, 11695.
21. El-Taher A., Makhluף S., Nossair A., and Halim A. A., (2010). Assessment of natural radioactivity levels and radiation hazards due to cement industry. *Applied Radiation and Isotopes*. 68, 1, 169-174.
22. Atkins M., Cowie J., Glasser F. P., Jappy T., Kindnes A., and Pointer C., (1989). Assessment of the performance of cement-based composite material for radioactive waste immobilization. *MRS Online Proceedings Library*. 176, 1, 117.