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Study and calculation of the specific natural radioactivity of samples of Portland cement in Iraq using a high-purity germanium detector

Estudio y cálculo de la radioactividad natural específica de muestras de cemento Portland en Irak utilizando un detector de germanio de alta pureza

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.ABSTRACT

In the current work, we used a high-purity germanium (HPGe) detector to assess particular activity concentrations in Seventeen cement samples from various origins. The results have shown that the specific activity, for ²²⁶Ra, ranged from 10.6 ± 0.7 Bq/kg) in Sample No. 11 to(59.1 ± 2.4 Bq/kg)in Sample No 12, for ²²⁸Ac ranged from (6.6 ± 1 Bq/kg) in Sample No.9 to(10.5 ± 1 Bq/kg) in Sample NO.7, for ⁴⁰K was ranged from (81.4 ± 7.6 Bq/kg) in Sample NO.12 to (209.5 ± 14.1 Bq/kg) in Sample No.7. The research demonstrates that the cement samples are safe to use in the construction of homes and do not offer any substantial sources of radiation hazard.

Keywords: Portland cement, radioactivity, germanium detector

RESUMEN

En el presente trabajo, utilizamos un detector de germanio de alta pureza (HPGe) para evaluar las concentraciones de actividad específica en diecisiete muestras de cemento de diversas procedencias. Los resultados han mostrado que la actividad específica para el 226Ra osciló entre 10.6±0.7 Bq/kg en la Muestra N.º 11 y 59.1±2.4 Bq/kg en la Muestra N.º 12; para el 228Ac, varió desde 6.6±1 Bq/kg en la Muestra N.º 9 hasta 10.5±1 Bq/kg en la Muestra N.º 7; y para el 40K, se situó entre 81.4±7.6 Bq/kg en la Muestra N.º 12 y 209.5±14.1 Bq/kg en la Muestra N.º 7. La investigación demuestra que las muestras de cemento son seguras para su uso en la construcción de viviendas y no representan fuentes sustanciales de peligro por radiación.

Palabras clave: Cemento Portland, radioactividad, detector de germanio

1. INTRODUCTION

Since the beginning of time, our planet has been radioactive. Nature includes more than 60 radionuclides. Because we are products of our environment, radionuclides can also be detected in us in addition to the air, water, and soil. Every day, the air we breathe, the food we eat, and the water we drink include nuclides that we consume or inhale. Our planet's rocks and soil, the seas and oceans, and even the materials used to construct our homes and buildings all contain radioactivity. The radioactive isotope of potassium (40 K) and

varied levels of naturally occurring radionuclides from the uranium and thorium series are present in all raw materials and finished building products originating from rock and soil , Some building materials are naturally more radioactive than others, which has long been known. Even very low levels of radioactivity naturally occurring in building materials can result in exposure both internally and externally(Abbas, 2015).Gamma radiation from the decay chains of uranium and thorium, as well as from potassium-40, is what causes the external radiation exposure. In contrast, the internal radiation exposure, which primarily affects the respiratory system, is brought on by the short-lived radon daughter products that are exhaled into the space by construction materials. Therefore, it's critical to understand the radioactivity of building materials in order to calculate the radioactive risks to human health (Tuo et al., 2020) As previously indicated, (²²⁶Ra), (²²⁸Ac), and (⁴⁰K) are the three most significant naturally occurring radionuclides found in cements(Abbas, 2015)

2. MATERIALS AND METHOD

In the present study, we used a high purity germanium (HPGe) detector to detect and measure the specific activity of radioactive elements in some types of cement samples. The cement samples were broken down into small pieces and then ground into a fine powder using an electrical mill.1 kilogram of roughly (300 mm) grain with the use of specialized sieves (mesh). In order to achieve the secular equilibrium for ²²⁶Ra and ²²⁸Ac with their respective progenies, the samples were dried at(50 °C) for one hour before being packaged in a Marinelli beaker. The sealed Marinelli beaker was then maintained for one month prior to measurements.(C33-78)

Each cement sample under investigation weighed about 450gm, was washed in distilled water to remove the formalin liquid (conservator substance), cut into uniform pieces, and then placed in a Marinelli beaker for three hours' worth of testing using a high-purity germanium detector. By using a few standard radioactive sources with known energies, the energy calibration of the germanium detector system was discovered. (Tuo et al., 2020) These sources need to be counted for a long enough time to produce clearly defined photo peaks, and then their energies should be calibrated. The system calibration in this study used a standard radioactive source (60 Co) with a period of (37 00 s.); the source's gamma-ray spectra are displayed in Figure1

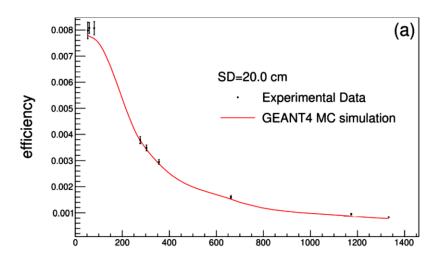


Figure 1. represents the detector efficiency diagram

The following equation describes the efficiency of the scintillation detector as the ratio of the number of pulses (counts per unit time) recorded by the detector to the number of radiation quanta (photons) emitted by the source (disintegration per unit time).(Karim et al., 2010)

Efficiency (%)=
$$\frac{cps}{dps} \times 100\% \dots (1)$$

Where cps : Count per second

dps : disintegration per second (the source activity)

The detector accumulates spectra of conventional radioactive sources with established energy and activity for a sufficient amount of time to yield well-defined photo peaks. The high purity germanium detector's efficiency in the current experiment was 50%. The following equation has been used to determine the precise activity for each radionuclide (radioactive element) found(Cember, 2009)

Specific Activity= $\frac{Net area under the peak}{W \times I_{\gamma} \times Eff \times T} \dots \dots \dots (2)$

Where T: Measuring time (s)
Eff.: Percentage efficiency
Iγ: Percentage intensity of gamma-ray
W: Mass of the sample (kg)
Net area under the peak: (Total counts –Background)

Measurement Of Parameter

1-Radum equivalent Activity

A Raeq it given by the formula .it's used to verify equality in the distribution Radiological index that has been established in order to describe the activity concentration of 226Ra ,228Ac ,and 40K by single quantity that takes into Concentration to them The index is known the radium equivalent activity of natural radionuclide 226Ra ,228Ac and 40K).Estokova and Palascakova, 2013(

$$Ra_{eq}(Bq Kg) = A_{Ra} + 1.43 A_{Ac} + 0.077 A_{k}$$
.....(3)

Where, ARa, AAC and AK are the specific activity concentrations of 226Ra,228Ac and 40K in (Bq/kg)respectively.

2-Absorbed Gamma Dose Rate

The following relation can be used to calculate the outdoor air gamma absorbed dose rate (D)in (nGy/h) due to terrestrial gamma rays at Hight (1 m)above the ground surface, which can be calculated from specific activity ARa, AAc and AK of 226Ra, 228Ac and 40K in (Bq/kg). respectively

$$D\left(\frac{nGy}{h}\right) = 0.462A_{Ra} + 0.604A_{Ac} + 0.0417A_k \dots \dots \dots (4)$$

3-Annual Effective Dose Rate

A member's expected annual effective dose equivalent was determined by converting the absorbed rate to human effective dose equivalent using a conversion factor of (0.7Sv/Gy), with an indoor occupancy of 80% and a 20% outdoor occupancy(Krane, 1991)

AED indoor(mSv/y) =
$$D_Y(nGy/h) \times 10^{-6} \times 8760 h/y \times 0.8 \times \frac{0.7Sv}{G} \dots (5)$$

AED Outdoor (mSv/y) = $D_Y(nGy/h) \times 10^{-6} \times 8760 h/y \times 0.2 \times \frac{0.7Sv}{G} \dots (6)$

4- External Annual Dose (EAD)

The following equation was used to calculate the external yearly effective dose (Estokova and Palascakova, 2013)

EAD= (0.92ARa+1.1AAc+0.08AK) ×(10-9 Gy/h) ×(0.7 Sv/Gy) ×(24×365) h/y×0.8.....(7)

5-Activity Concentration Index (Iy)

The activity index (Iy) for cement samples was estimated by applying the following solution(Al-Jundi et al., 2006)

$$I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Ac}}{100} + \frac{A_{K}}{1500} \dots \dots \dots \dots (8)$$

6- External (Hex) and Internal (Hin) Hazard Indices

Internal (Hin) and external (Hex) Hazard Indices Two further indices that represent internal and external radiation dangers were developed by Beretka and Mathew (Atyotha et al., 2023). The external hazard index is derived from the expression for (Raeq) on the basis that its permitted maximum value (equal to unity) corresponds to the upper bound of Raeq (370 Bq/kg).Following that definition, the external hazard index (Hex) is can then be defend is (Atyotha et al., 2023)

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \le 1 \dots \dots \dots (9)$$

Internal hazard index (Hin), as shown below, controls exposure to 222Rn and its) Ahmed, 2016(radiological offspring.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \le 1 \dots (10)$$

To maintain a negligible radiation risk, this index value must be smaller than unity.

3. RESULT AND DISCUSSION

Our current study is based on a review of (seventeen) samples from various cement kinds that are sold locally; all of the samples were produced in Iraq by various companies and are organized in accordance with the types utilized in construction. The specific activity content of cement sample is shown in Table (1).

Table (1) The specific activities of Radionuclides and Parameters in cement samples

| Sample | C\Ra226 Bq\Kg | C\AC228 Bq\Kg | C\K40 Bq\kg | Raeq Bq\Kg | D₅ (nGy\h) | Indoor Ein | Outdoor Eout | EAD (mSv\y) | Ix | H _{In} | Hex |
|------------|------------------|------------------|----------------|---------------|---------------|---------------|-----------------|----------------|--------|-----------------|----------|
| S 1 | 17.1 | 9.5 | 160.2 | 43.0204 | 20.31854 | 0.099675 | 0.024919 | 0.02618 | 0.3158 | 0.162418 | 0.116201 |
| S2 | 28.1 | 8 | 120.1 | 48.7877 | 22.82237 | 0.111957 | 0.027989 | 0.0346 | 0.3474 | 0.207749 | 0.131803 |

| S 3 | 13.1 | 10.4 | 143.9 | 39.0523 | 18.33443 | 0.089941 | 0.022485 | 0.02349 | 0.287267 | 0.140882 | 0.105477 |
|----------------|---------|-------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| S4 | 16.5 | 9.3 | 121.5 | 39.1545 | 18.30675 | 0.089806 | 0.022451 | 0.02541 | 0.284 | 0.150356 | 0.105762 |
| S5 | 25.1 | 9.1 | 141.3 | 48.9931 | 22.98481 | 0.112754 | 0.028189 | 0.03310 | 0.352533 | 0.200187 | 0.132349 |
| S 6 | 13.5 | 10.3 | 126.5 | 37.9695 | 17.73325 | 0.086992 | 0.021748 | 0.03275 | 0.277333 | 0.139041 | 0.102554 |
| S 7 | 39.2 | 10.5 | 209.5 | 70.308 | 33.18855 | 0.16281 | 0.040702 | 0.04761 | 0.506 | 0.295988 | 0.190042 |
| S 8 | 29.8 | 7.1 | 153.5 | 51.7725 | 24.45695 | 0.119976 | 0.029994 | 0.03522 | 0.372 | 0.220407 | 0.139866 |
| S9 | 27.3 | 6.6 | 116.8 | 45.7316 | 21.46956 | 0.105321 | 0.02633 | 0.032376 | 0.325867 | 0.197333 | 0.123549 |
| S10 | 31.2 | 7.3 | 127.2 | 51.4334 | 24.12784 | 0.118362 | 0.02959 | 0.0367 | 0.3658 | 0.223279 | 0.138955 |
| S11 | 10.6 | 7.2 | 120.8 | 30.136 | 14.28336 | 0.070068 | 0.017517 | 0.01767 | 0.2232 | 0.110211 | 0.081562 |
| S12 | 59.1 | 7 | 81.4 | 75.3778 | 34.92658 | 0.171336 | 0.042834 | 0.06207 | 0.518267 | 0.36341 | 0.20368 |
| S 13 | 23.8 | 5.7 | 99.5 | 39.6125 | 18.58755 | 0.091183 | 0.022796 | 0.02816 | 0.282 | 0.171342 | 0.107018 |
| S14 | 14.5 | 9.7 | 119.6 | 37.5802 | 17.54512 | 0.086069 | 0.021517 | 0.0240 | 0.2734 | 0.140695 | 0.101506 |
| S15 | 22.5 | 8.2 | 135.4 | 44.6518 | 20.99398 | 0.102988 | 0.025747 | 0.0297 | 0.322267 | 0.181432 | 0.120621 |
| S16 | 13.7 | 9.1 | 102.2 | 34.5824 | 16.08754 | 0.078919 | 0.01973 | 0.02261 | 0.250467 | 0.130437 | 0.09341 |
| S17 | 33.6 | 9.3 | 158.4 | 59.0958 | 27.74568 | 0.136109 | 0.034027 | 0.04114 | 0.4226 | 0.25046 | 0.15965 |
| Orval Range | 24.6294 | 8.488 | 131.6353 | 46.89762 | 21.9948 | 0.107898 | 0.026974 | 0.03199 | 0.336835 | 0.193272 | 0.126706 |

It is seen from Table (1) and several companies that: The cement with the highest specific activity concentration (226 Ra) The maximum value of radium content, 59.1±2.4 Bq/kg, was discovered. The particular concentration of activity (226 Ra) in cement has the lowest value (10.6±0.7Bq/kg), as shown in Figure 2, while the average value is 24.62 Bq/kg, The most recent findings demonstrated that the study's values for the specific activity concentrations (226 Ra) were lower than the target activity value. The global limit is (226 Ra), or (370 Bq/kg), at that concentration.(Abdallah et al., 2022) . (228 Ac) has the greatest specific activity concentration value. The quantity equivalent to (10.5±1 Bq/kg), which was measured in sample number 7, contrasts with The cement samples with the lowest specific activity concentration (226 Ac) Sample No. 13, which has a weight of 5.7±1.7 Bq/kg, contained it. An average value of (8.48 Bq/kg) is shown in Figure 3. The current findings demonstrate that the activity concentration values for (226 Ac) found in cement samples in the research locations were less than the value , value from the (228 Ac) global limit, which is equal to 35 Bq/kg of particular activity concentration(Kadum et al., 2013) The cement samples had a range of specific activity concentrations of (40 K), with sample 7 having the highest value at (209.517.8 Bq/kg) and

sample NO. 12 having the lowest value at (81. 4 ± 7.6 Bq/kg), as shown in Figure (4), and an average value of (131.6352 Bq/kg). The current findings demonstrate that the values of specific activity concentrations of (⁴⁰K) in cement samples were less than the worldwide limit, which is equal to (400 Bq/kg). (Commission, 1999)The average value of the radium equivalent activity (Raeq) in the cement samples was 46.89762.3Bq/kg, with Sample No.11 having the lowest value (Raeq) at (30.219Bq/kg) and Sample No.12 having the highest value (75.3778Bq/kg). According to the current findings, the global limit for radium equivalent activity, which is equal to (370Bq/kg), was not reached by the cement samples (Symons, 2001) The absorbed gamma dose rate in cement samples ranged from (16.08nGy/h) in Sample NO.16 to(34.9265 nGy/h) in Sample NO.12, with an average value of (21.9948 nGy/h.). Sample NO.5 had the highest value of the absorbed gamma dose rate (D) at (34.9265 nGy/h). The current findings demonstrate that the value of the so-absorbed gamma dose rate cement samples was less than the value of the global limit for the absorbed gamma dose rate (0.1713mSv/y), and sample No.12 had the highest indoor yearly effective dose rate (0.1713mSv/y). The average indoor annual effective dose rate was 0.107898 mSv/y.

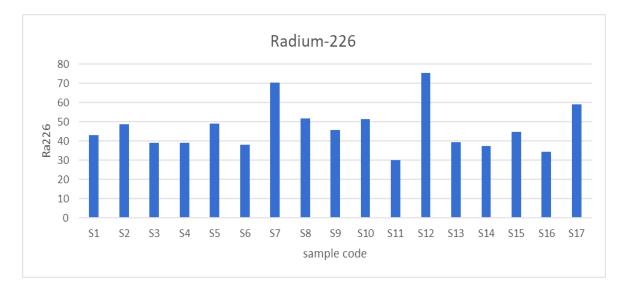


Figure 2. histogram illustrating the change specific activity Concentrations of 226 Ra for all cement sample

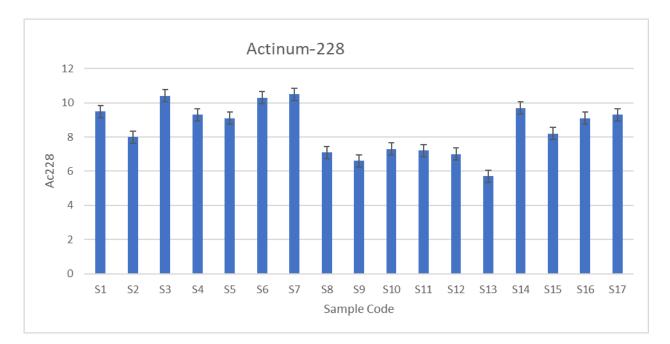


Figure 3. histogram illustrating the change specific activity Concentrations of 228AC for all cement sample

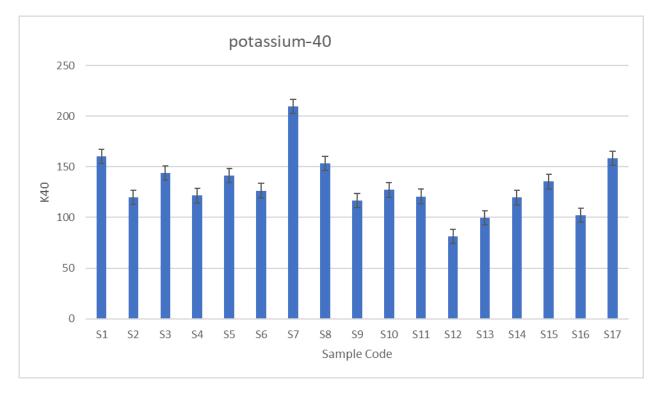


Figure 4. histogram illustrating the change specific activity Concentrations of 40K for all cement sample

The results of the current study demonstrate that the indoor annual effective dose rate of the cement samples under study was less than the indoor annual effective dose worldwide limit, which is equal to 1mSv/y(Symons, 2001). The cement samples had an average annual effective dose rate (AED)out of (0.026974 mSv/y), with Sample No.11 having the lowest value (0.0175mSv/y) indoors and Sample No.12 having the highest outdoor annual effective dose rate (AED)_{out} of (0.042834mSv/y). The results of the present study demonstrate that the outdoor annual effective dose rate values in cement samples were less than the outdoor annual effective dose rate worldwide limit, which is equivalent to 1mSv/y(Symons, 2001). With an average value of (0.03199mSv/y), cement sample No.12 had the greatest external annual effective dose (EAD) value, which was equivalent to. (0.062072mSv/y), while sample No.11 had the lowest(EAD) value, which was equal to (1.5mSv/y). According to the current findings, the outdoor annual effective dose worldwide limit, which is equal to (2.017672mSv/y). According to the current findings, the outdoor annual effective dose worldwide limit, which is equal to (2.017672mSv/y), was not reached by cement sample values for the external annual effective dose(Symons, 2001).

The activity concentration index (I) for cement samples ranged from (0.2232) for Sample No.11 to 0.518 267 for Sample No.12, with an average value of (0.336835). Sample No.12 had the greatest value, while Sample No.11 had the lowest value. The current findings demonstrate that the activity concentration index values in cement samples were less than the global limit for the activity concentration index, which is equal to1(Symons, 2001). The internal hazard index (Hin) for cement samples was found to have a range of values, with Sample No.12 having the highest value at (0.363) and Sample No.11 having the lowest value at (0.1102), with an average value of (0.193272). The current findings demonstrate that internal hazard index values in cement samples were lower than their respective global limits, which are both equal to one. With an average value of (0.126706), cement samples had an external hazard index (Hex) range of 0. 081562 to 0.20368, with Sample No.11 having the lowest value at 0.08152. Sample No.11 had the highest value of Hex, which was equivalent to (0.2031), while Sample No.12 had the lowest value at (0.081562).

The current findings demonstrate that cement sample external hazard index values were less than the global external hazard index limit, which is less than unity(Symons, 2001)

5. CONCLUSION

This study demonstrated the use of high-purity germanium (HPGe) detectors for the analysis of various cement sample types. The investigation demonstrates all the cement samples do not constitute any significant source of radiation hazard and are safe for use in the construction of houses since the specific activity concentrations of (226Ra,228Ac, and 40K) are below the specific activity concentration of the worldwide limit.

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