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Determination of natural radioactivity and radiological hazards for Cement and Gypsum in Sulaimani Area (Kurdistan Region – Iraq)

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Abstract: Activity concentration of 238U, 232Th and 40K in twelve samples of cement and gypsum is used as a building material in Sulaimani Governorate-Kurdistan Region-Iraq, these samples were measured using γ-spectrometry based on scintillation detection. Also activity concentration of 222Rn was determined using solid state nuclear track detector (SSNTD) technique through the CR-39 detector and radium equivalent activities (Raeq), absorbed dose rate (D), external and internal hazard indices (Hex and Hin) and level index of γ-radiation hazard (Iγ) were calculated to assess the potential radiological hazard associated with these samples. The activity concentration of 238U, 232Th and 40K for the cement samples were found to range between (4.458 ± 0.304 - 51.573 ± 11.955, 6.330 ± 1.253 – 19.188 ± 6.612 and 54.950 ± 6.050 -81.648 ± 10.958) Bq/Kg, respectively. Values of average radium equivalent activities (Raeq), absorbed dose rate (D), external and internal hazard indices (Hex and Hin) and γ- index hazard (Iγ) are ranged as (36.74-78.028) Bq/Kg, (32.093 ± 4.235-69.369 ± 13.110) nGy/hr, (0.099-0.21), (0.111-0.349) and (0.131-0.267) but the radon concentration are ranged inside the air of the tube between (75.514±3.876 - 286.082±28.583) Bq/m3, while inside the studied sample materials is ranged between (30618.211±295.7 - 8081.946±40.101) Bq/m3, the natural radioactivity concentrations. The higher values appeared in the cement samples and the lower one in the gypsum except in thorium concentration, fortunately they are not higher than the limit of worldly values, therefore the studied samples (cement and gypsum) in Sulaimani building materials can be used in construction safely from the radioprotection.

Keywords: Natural radioactivity, Cement, Gypsum, γ-spectrometry, SSNTD.

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1. Introduction:

Natural radiation sources can be classified into [1]:

1- Gamma and radioactive nuclides present in the crust of earth, building materials and air.

2- Internal sources, comprising the naturally occurring radionuclides which are taken into the human body.

The main radioactive materials in naturally occurring radioactive materials are long –lived radionuclides such as ^{238}U, ^{235}U, ^{232}Th and ^{40}K. Natural radioactivity and terrestrial gamma dose originated from naturally occurring radioactive in the building materials depend essentially on geological and geographical conditions, therefore concentrations of natural radioactivity in the material especially in soil vary from one region to another in the world [2, 3].

Studies of natural radioactivity in building material such (Cement and Gypsum) are necessary not only because of their radiological effect, but they also act as an excellent biochemical and geochemical tracer in the environment. The presence of radium associated with the emanation of radon, the inhalation of radon and short-lived daughter products decay is a major contributor to the total radiation dose in an exposed subject. Therefore the lung dose due to radon may be cause to increase the lung cancer [4]. Among all the radon isotopes, only ^{222}Rn is important which produced from the decay product of ^{226}Ra, deriving from the uranium series of natural radionuclides and has a half-life of 3.8 d [5]. Indoor radon comes from several major sources, especially from the soil underlying and surrounding building foundations and building materials.

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All types of building materials like cement, gypsum, concrete, brick, sand, granite, limestone, etc. cause direct radiation exposure because of their uranium, thorium and potassium content [6].

In the present work, the concentrations of natural radionuclide were measured in 12 samples of cement and gypsum, which considered as a building material samples that, is used commonly in Sulaimani Governorate and its surroundings by means of gamma-ray spectrometry. The potential radiological hazards associated with those materials (cement and gypsum) were assessed by calculating the radium equivalent activity (Ra_{eq}), indoor absorbed gamma dose rate (D), external and internal hazard indexes (H_{ex}, H_{in}) and level index of gamma radiation hazard (I_{γ}). In addition, the same samples were analyzed for the radon concentrations using passive radon detector (CR-39). The obtained results were compared with the recommended values to assess the radiation hazards to human due to building materials (cement and gypsum) and also compared to the corresponding values of the building materials from different countries which done through using the same technique.

2. Experimental procedure:

2.1 Sampling:

The majority of building materials (cement and gypsum) is produced in Sulaimani (Kurdistan Region-Iraq) from both factories Tasluja and Bazyan and also from the other neighbor countries Turkey and Iran used in sulaimani.

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The samples weight were about 1Kg putting in a Marnelli beaker, hermetically sealed and stored for 1 month to take place the equilibrium between ^{226}Ra and its decay products of short half-life before being taken in gamma spectrometry analysis [7].

The measurement of radon concentrations was carried out by using (SSNTD) solid state nuclear detector (CR-39) samples with 130 g were prepared after drying and placed on the bottom of a sealed polyethylene bottle (cylindrical plastic) and the (CR-39) detector with pieces of area 1cm x1. 5cm on the other side of cylindrical plastic suspended, then stored for two months to allow radioactive equilibrium between ^{222}Rn and ^{226}Ra. Exposed detector was collected and etched chemically using 6.25 M NaOH at 70C^{o} for 6 hrs after washing these detectors in distilled water, the track density on CR-39 (tr. /cm^2) was counted using an optical microscope with magnification 400x [8].

2.2 Gamma-ray measurements:

In gamma-ray spectrometry measurement, a (2 inch x 2 inch) NaI (TI) detector was used with energy resolution 9.46% at the energy of 662 keV for ^{137}Cs line. The detector is surrounded by an 8cm thick lead shield to reduce the gamma-radiation background. Each sample of cement and gypsum was measured for 6 hrs. The photo peak of ^{238}U was measured from 609 keV gamma-line of ^{214}Bi, ^{232}Th from 911 keV gamma-line of ^{228}Ac and ^{40}K from 1460 keV gamma-energy. The concentrations of ^{238}U, ^{232}Th and ^{40}K in each sample by ppm were calculated form the calibration curve which done for each of the above nuclides alone from the obtained standard sources were counted with the NaI(TI) detector and their spectrums yield, when they are used in the establishment of calibration curve, through their net area and by fitting these calibration curves we obtained these relations as shown in equations (1),(2) and (3) [9]:

$$Y = 0.0022x - 0.2492 \quad \text{----------- (1)} \quad \text{for } ^{238}U$$

$$Y = 0.0223x - 1.617 \quad \text{----------- (2)} \quad \text{for } ^{232}Th$$

$$Y = 9\times10^{-6}\, x - 0.0004 \quad \text{----------- (3)} \quad \text{for } ^{40}K$$

Then to measure the specific activity by using the conversion factor for each radionuclides (^{238}U, ^{232}Th and ^{40}K) can be used for 1 ppm of U corresponds to 12.35 Bq/Kg for Th corresponds to 4.1 Bq/Kg and for K corresponds to 259.2 Bq/Kg [10].

2.3 Radon concentration measurement:

The concentrations of ^{222}Rn in the air of tube, as shown in Fig.(1), that contain cement and gypsum samples by using CR-39, can be calculated from the equation (4) [11,12]:

$$C_a = \varrho / \eta T \quad \text{------------- (4)}$$

Where ϱ is the track density on the exposed detector CR-39 (Tr/cm^2)

T is the exposure time of the samples (60 d)

η is the detection efficiency and can be calculated from this relation eq.(5) using the dimensions of the tube [8]:

$$\eta = \frac{1}{4} r \left(2\cos\theta_t - \frac{r}{R_\alpha}\right) \quad \text{----------- (5)}$$

where r - Tube radius for the diffusion volume (3.6 cm)

θ_t - Threshold angle for the CR-39 detector (35^{o}) [13]

R_α - Range of α- particle in air from Rn

R_α can be calculated from the equation (6) [14]:

$$R_\alpha = (0.005\, E_\alpha + 0.285)\, E_\alpha^{3/2} \quad \text{------------ (6)}$$

$$= 4.019 \text{ cm} \quad (\text{for energy } E_\alpha = 5.49 \text{ MeV})$$

The value of diffusion constant (η) according to dimensions of the present system equals to (0.057744) Tr.cm^{-2}.d^{-1} / Bq . m^{-3}

To calculate ^{222}Rn concentration in the samples, equation (7) can be used [12]:

$$C_s = \frac{\lambda_{Rn} C_a H T}{L} \quad \text{-------------- (7)}$$

Where C_s - Rn^{222} concentration in the samples (Bq\m^3)

C_a – Rn^{222} concentration in air space inside the tube (Bq\m^3)

λ_{Rn} - Decay constant of Rn^{222} (0.1814 day^{-1})

H – Height of air space in the tube (29.5 cm)

L – Thickness of the sample in the tube (3 cm)

T - Time of irradiation (60 days)

2.4 Estimation radiation hazard index:

The most widely used radiation hazard index is called the radium equivalent activity Ra_{eq} which is a weighted sum of activities of the 3 radionuclides based on following relation as showing in equation (8) [5,7]:

$$Ra_{eq} = 1.43\, A_{Th} + A_{Ra} + 0.077\, A_k \quad \text{-------------- (8)}$$

Where: A_{Th} is the activity concentration of thorium

A_{Ra} is the activity concentration of radium

A_k is the activity concentration of potassium

2.5 Estimation of dose rate (D):

The average absorbed dose rates D (nGyh^{-1}) in air 1m from terrestrial sources of gamma- radiation in the samples can be calculated from the equation (9) based on Gide lines provided by UNSCEAR 2000 [5]

$$D(\text{nGyh-1}) = 0.462\, A_{Ra} + 0.621\, A_{Th} + 0.0417\, A_K \quad \text{------------ (9)}$$

Where: D is the dose rate in (nGyh^{-1})

2.6 Estimation of (External and Internal) hazard index (H_{ex} and H_{in}):

The external hazard index H_{ex} is defined as shown in eq. (10) [5,15]:

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \quad \text{--------------- (10)}$$

Another hazard due to radon effect on respiratory organs called internal hazard index H_{in} can be determined from the eq. (11):

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \quad \text{-------------- (11)}$$

2.7 Estimation of level of gamma hazard index (I_γ):

But the level index of gamma-radiation hazard which was associated with natural radionuclides in specific investigated can be calculated from equation (12) [5]:

$$I_\gamma = 0.0067\, A_{Ra} + 0.01\, A_{Th} + 0.00067\, A_K \quad \text{------------- (12)}$$

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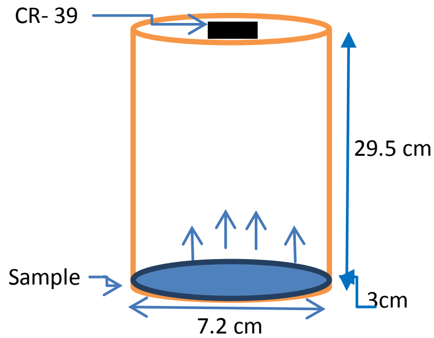

Fig. (1) Schematic Diagram of Long-Tube for SSNTD Technique.

3. Result and discussion:

3.1 Radon measurements:

The obtained radon and uranium concentrations of the studied samples through using SSNTD technique are given in Table (3-1). The radon concentration is ranged inside the air in the tube between (75.514±3.876 - 286.082±28.583) Bq/m^3 and the studied samples ranged between (30618.211±295.7 - 8081.946±40.101) Bq/m^3. The highest value of the radon concentration appears in the cement-5 sample from Sulaimani (Kurdistan Region-Iraq) and the lower value of the Gypsum-1 sample from Iran as showing in Fig.(2). Table (3-2) shown the radon concentrations values in the cement and gypsum samples. Fortunately, these values approximately lower than the world values with comparison to the other works [16], also the highest values appear in the cement-5 and gypsum-3 samples, the mentioned samples were located in the same area of Sulaimani governorate, furthermore these regions enriched with marlstone and sandstone which contain a concentration of uranium according to geology information [9].

3.2 Radionuclide concentrations:

Table (3-3) shows the radionuclide concentrations of ^{238}U, ^{232}Th and ^{40}K, using gamma- spectroscopy technique, the lowest specific activity of ^{238}U was (4.45835±0.304) Bq/Kg appeared in the gypsum-1 sample from Iran. The ^{232}Th specific activity value record 6.3304±1.253 Bq/Kg appeared in the cement-2 sample from Sulaimani (Kurdistan Region – Iraq). The ^{40}K predicted the lowest activity (54.9504±6.050) Bq/Kg, which appeared in the gypsum-3 sample from Sulaimani (Kurdistan Region – Iraq), while the highest value is recorded to these radionuclide; the ^{238}U in the cement-6 sample from Iran, the ^{232}Th in gypsum-1 sample from Iran and the ^{40}K in the cement-3 sample from Sulaimani (Kurdistan Region – Iraq), as shown in Figures (3 & 4) for cement and gypsum, respectively. Although the distribution of radionuclides U, Th and K in building materials, especially in cement and gypsum is not uniform they could be considered a bit higher in the cement samples while it's less in the gypsum sample by comparing to the other works [7] while it's vice versa in the other [16] as shown in Table (3-3).

3.3 Dose Calculations:

The calculated values of the radium equivalent activity, total absorbed dose rate (D) and external, internal with gamma hazard indices are given in Table (3-4), from the obtained results the lowest value of Ra_{eq} was 36.74 Bq/Kg calculated in (Gypsum1) sample from Iran while the highest value is 78.028 Bq/Kg in (Cement White 8) from Turkey. The calculated values of the total dose rate range from (32.093 to 69.369) nGy/hr and Fig.(5) shows all the ranges for each samples. The values of H_{ex} and H_{in} varied in the range from (0.099 to 0.21) and from (0.111 to 0.349), respectively. For the safe use of a material in the construction places, H_{ex} and H_{in} must be less than the unity [7] and all the calculated values are less than the unity. Also the calculated values of I_γ for the studied samples varied in the range between (0.131-0.267), for all types of samples it's observed they are less than the critical value of unity as showing in the Figures(6 & 7) for cement and gypsum, respectively. By comparison, these results with the other works which made in the same region they are approximately close together [7] but lower than the other [16] as shown in the Table (3-5).

Table (3-1) Radon and Uranium Concentrations in the samples

Sample	Track/cm²	C_{Rn} (Bq/m³)	C_s(Bq/m³)
Cement 1 (Iran)	706.77	204.033±17.216	21836.826±178.101
Cement 2 (Iraq)	599.875	173.174±13.462	18534.129±139.264
Cement 3 (Iraq)	418.78	120.895±7.852	12938.900±81.232
Cement 4 (Iran)	688.536	198.769±16.554	21273.457±171.253
Cement 5 (Iraq)	990.988	286.082±28.583	30618.211±295.7
Cement 6 (Iran)	670.3	193.505±15.901	20710.025±164.495
C. White 7 (Iran)	877.804	253.408±23.829	27121.204±246.516
C. White 8 (Turkey)	921.82	266.114±25.644	28481.151±265.288
Gypsum 1 (Iran)	261.58	75.514±3.876	8081.946 ±40.101
Gypsum 2 (Iraq)	293.02	84.590±4.596	9053.337 ±47.544
Gypsum 3 (Iraq)	308.112	88.947±4.955	9519.629 ±51.264
Gypsum 4 (Iraq)	299.308	86.405±4.744	9247.615 ±49.082

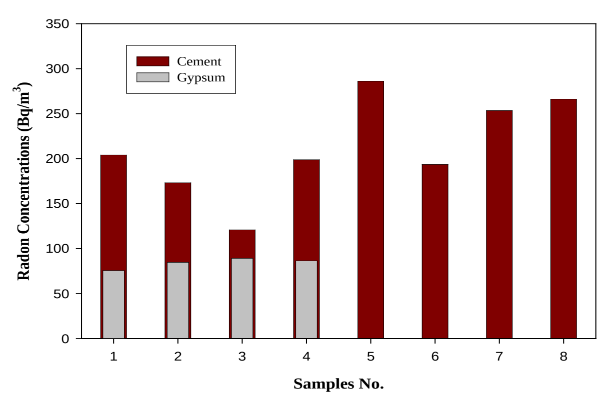

Fig.(2) Radon Concentrations (Bq/m^3) in Cement and Gypsum samples

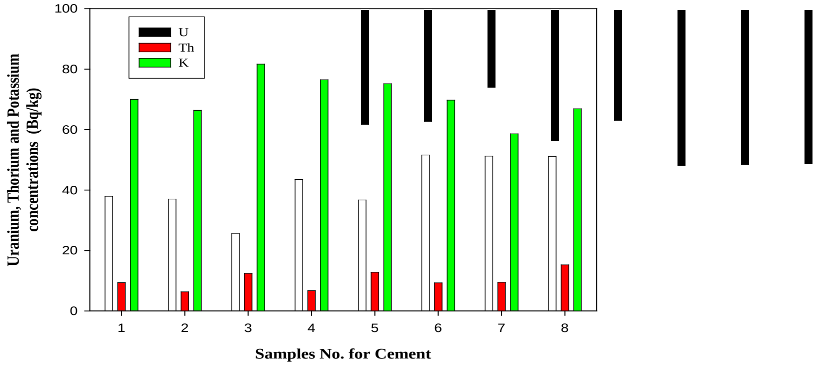

Fig.(3) Uranium, Thorium and Potassium Concentrations (Bq/Kg) in Cement samples

Table (3-2) Radon, Uranium, Thorium and Potassium concentrations in the samples Cement and Gypsum in the other work

Sample	$^{222}Rn(Bq/m^3)$	^{238}U(Bq/Kg)	^{232}Th(Bq/Kg)	^{40}K(Bq/Kg)	Reference
Gypsum	367.47	12.5 ± 1.0	2.7 ± 0.1	1141.9±36	7
Cement	453.24	24.7 ± 1.6	20.7± 1.5	2493±78.9	7
White Cement	-	49.577±0.865	16.74±2.28	32.6± 4.31	14
Bridge Cement	-	37.18±0.28	6.798±0.47	194±10.939	14

Table (3-3) Uranium, Thorium and Potassium concentrations in the samples.

Sample	Conc.(^{238}U) pmm	Conc.(^{238}U) Bq/Kg	Conc.(^{232}Th) pmm	Conc.(^{232}Th) Bq/Kg	Conc.(^{40}K) pmm	Conc.(^{40}K) Bq/Kg
Cement 1 (Iran)	3.072	37.9392±7.543	2.291	9.3931±2.265	0.27	69.984±8.696
Cement 2 (Iraq)	2.996	37.0006±7.264	1.544	6.3304±1.253	0.256	66.3552±8.028
Cement 3 (Iraq)	2.078	25.6633±4.196	3.027	12.4107±3.439	0.315	81.648±10.958
Cement 4 (Iran)	3.52	43.472±9.251	1.644	6.7404±1.377	0.295	76.464±9.931
Cement 5 (Iraq)	2.972	36.7042±7.177	3.116	12.7756±3.592	0.29	75.168±9.680
Cement 6 (Iran)	4.176	51.5736±11.955	2.268	9.2988±2.231	0.269	69.7248±8.648
C. White 7 (Iran)	4.146	51.2031±11.826	2.313	9.4833±2.297	0.226	58.5792±6.659
C.White 8 (Tur)	4.14	51.129±11.800	3.715	15.2315±4.676	0.258	66.8736±8.123
Gypsum 1 (Iran)	0.361	4.45835±0.304	4.68	19.188±6.612	0.24	62.208±7.288
Gypsum 2 (Iraq)	0.932	11.5102±1.260	4.225	17.3225±5.672	0.243	62.9856±7.425
Gypsum 3 (Iraq)	0.779	9.62065±0.963	4.16	17.056±5.541	0.212	54.9504±6.050
Gypsum 4 (Iraq)	0.932	11.5102±1.260	4.225	17.3225±5.672	0.243	62.9856±7.425

Table (3-4) Ra_{eq}, total absorbed dose rate, external, internal and gamma hazard indices calculations.

Sample	$Ra_{eq.}$ (Bq/Kg)	D (nGy/hr)	H_{ex}	H_{in}	I_{γ}
Cement 1 (Iran)	56.759	50.824±8.133	0.153	0.255	0.196
Cement 2 (Iraq)	51.161	46.012±6.960	0.138	0.238	0.176
Cement 3 (Iraq)	49.693	44.418±6.663	0.134	0.203	0.174
Cement 4 (Iran)	58.997	53.104±8.619	0.159	0.276	0.203
Cement 5 (Iraq)	55.894	49.851±7.948	0.15	0.236	0.194
Cement 6 (Iran)	55.368	49.584±7.836	0.149	0.248	0.191

C. White 7 (Iran)	69.274	61.863±10.967	0.187	0.325	0.237
C.White 8 (Turkey)	78.028	69.369±13.110	0.21	0.349	0.267
Gypsum 1 (Iran)	36.74	32.093±4.235	0.099	0.111	0.131
Gypsum 2 (Iraq)	41.13	36.168±5.017	0.111	0.142	0.145
Gypsum 3 (Iraq)	40.879	35.727±4.971	0.11	0.136	0.144
Gypsum 4 (Iraq)	41.13	36.168±5.017	0.111	0.142	0.145

Table (3-5) Ra_{eq}, total absorbed dose rate, external, internal and gamma hazard indices in the other work.

Sample	Ra_{eq}(Bq/Kg)	D(nGy/hr)	H_{ex}	H_{in}	I_{γ}	Reference
Gypsum	104.2	105.82	0.2	0.3	-	7
Cement	246.1	245.17	0.66	0.73	-	7
White Cement	75.94	-	0.205	0.229	0.259	14
Bridge Cement	61.9	-	0.166	0.185	0.223	14

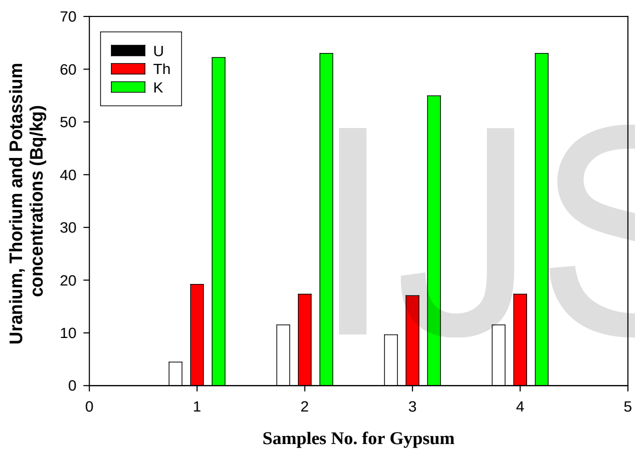

Fig.(4) Uranium, Thorium and Potassium Concentrations (Bq/Kg) in Gypsum samples

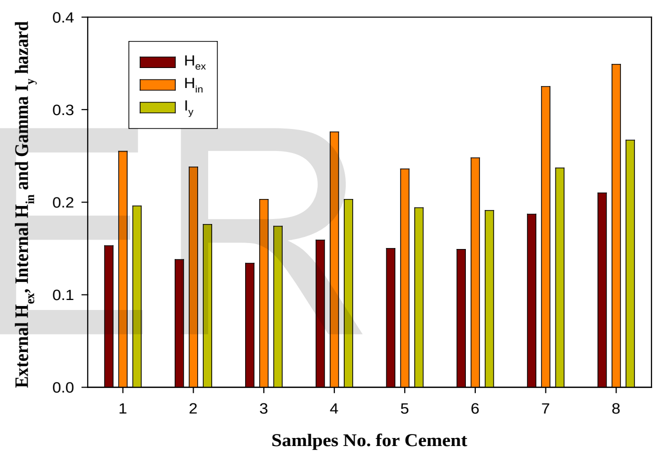

Fig.(6) Radiological hazards in Cement sample

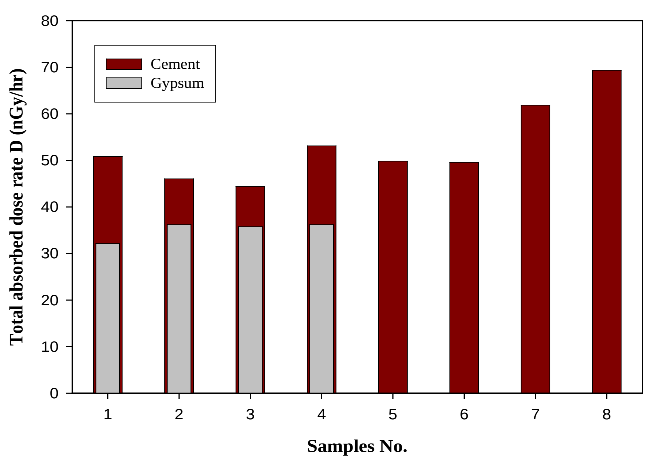

Fig.(5) Total absorbed dose rate D (nGy/hr) in Cement and Gypsum samples

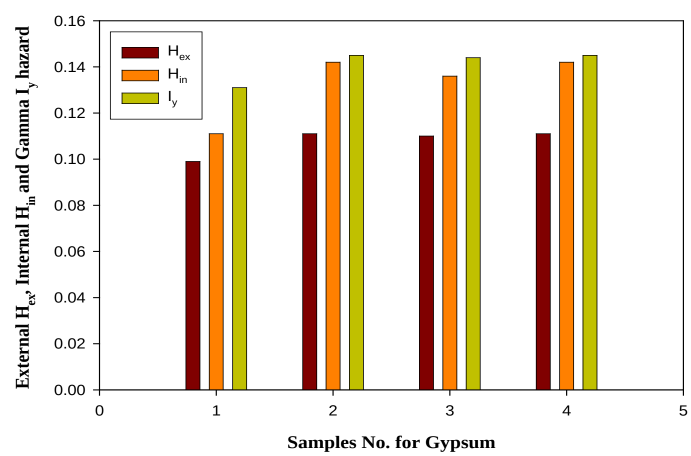

Fig.(7) Radiological hazards in Gypsum samples

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4. Conclusion

The radon concentration, natural radionuclide content, external and internal hazard indices of some building material such as cement and gypsum were determined which commonly used in Sulaimani area in Kurdistan Region-Iraq, using SSNTD and gamma spectroscopy techniques. The result of measuring obtained activity concentrations to estimate qualify and quantify radiological hazards which associated with the studied samples. It is concluded that the studied samples of Sulaimani building materials can be used in construction safely from the radioprotection.

5. References

1- Laith A. et al. (2011), Natural radioactivity levels of limestone rocks in northern Iraq using γ-spectroscopy and SSNTD. (289): 709-715

2- Poschl M. (2007), *Radionuclides concentration in food and the Environment*, Taylor & Francis Group, USA.

3- Turhan, S. (2008), Assessment of the natural radioactivity and radiological hazards in Turkish cement and its raw materials. J. Environmental Radioactivity, 99 (2), 404-414.

4- UNSCEAR (1993), Exposure from natural sources of radiation. United Nation, New York.

5- (UNSCEAR 2000), Sources and Effects of Ionizing Radiation, Report to general assembly, with scientific annexes, United Nations. (http://www.unscear.org)

6- Cetin, E. et al. (2012), Natural radioactivity levels of granites used in Turkey, Radiation Protection Dosimetry, 151(2): 299-305.

7- Oktay, B. et al. (2011), Assessments of natural radioactivity and radiological hazards in construction materials used in Elazig, Turkey. Radiation Measurement, (46): 153-158

8- Omer N. (1999), Electrochemical Etching, Msc. dissertation, University of Mousl.

9- Kamal O. and Rasheed M. (2013), Natural Radioactivity Measurements of soil and water in Sulaimani Governoraye, Ph.D. Dissertation, University of Sulaimani.

10- Samuel S. M. (2004) Introductory Nuclear Physic, 2nd Edition, University of Toronto.

11- Azam,A. et al. (1995).Radium Concentration and Radon Exhalation Detectors . Nuclear Geophysics, 9 (6) : 653- 657.

12- Ahmad M. (2011).“Employment SSNTD (CR-39) to Detect Radioactive Pollution of DU in a Particular Region from Salahadeen Governorate” Proceeding in Ist Scientific Conference to College of Science University of Tikrit.Iraq, (295-300).

13- Barillon R. et al. (1993) “Comparison of Effectiveness of three Radon Detectors (LR- 115,CR-39 and Si. Diode pin) Placed in Cylindrical Device- Theory and Experimental Techniques ,Nuclear Track” Radiation Measurement, 22(1-4) ; 281-282.

14- Michael F. Annunziata (2007),” Radioactivity Introduction and History”, Ist edition USA.

15- Sroor A. et al.(2001),Natural Radioactivity and Radon Exhalation rate of Soil in Southern Egypt, Applied Radiation and Isotopes, 55: (873-879).

16- Laith, N. et al. (2013),Measurement Of Natural Radioactivity in Building Materials used in Iraq. Australian J. of basic and Applied Science, 7(1): 56-66.