Assessment of Radon, Radium, and Uranium Concentrations in Decorative Materials (Used to walls) Samples in Iraqi Markets

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Abstract: - In present study, natural alpha emitters (^{222}Rn, ^{226}Ra, and ^{238}U) were tested in decorative materials used as walls collected from different Iraqi local markets by CR-39 detectors that it was purchased from TASTRAK Analysis System. Annual effective dose and radon exhalation rate were calculated. The results obtained showed that the range and average value of ^{222}Rn concentrations in air container were 7.94 – 738.10 Bq/m^3 and 252.38±37.63 Bq/m^3, while ^{222}Rn concentrations in in sample were 45.73– 4252.99 Bq/m^3 and 1454.25±216.84 Bq/m^3. The ranged of ^{226}Ra and ^{238}U concentrations were 0.010- 1.000 Bq/kg, with an average value 0.278±0.04 Bq/kg and 0.01- 1.24 ppm, with an average value 0.344±0.05 ppm, respectively. The results obtained showed that the range and average value of annual effective dose in mSv/y were 0.2 –18.62 and 6.29±0.95. Also, it is found that the mass exhalation rate as well as surface exhalation rate were 8.38±1.33 mBq/kg.day and 240.77±36.56 mBq/m^2.day, respectively. The data of ^{222}Rn concentration (in air container), and annual effective dose in some samples of the present study were higher than the global limit range (200-300 Bq/m^3), and (3-10 mSv/y) according to ICRP. While, all results of ^{226}Ra, and ^{238}U concentrations as well as the mass with surface exhalation rate were within the permissible limits that suggested by UNSEAR, and ICRP. Then, it can be concluded that the most samples of decorative materials which used as walls according to radiation scope no causes health risk.

Key-Words: - Alpha emitters, decorative materials, building materials, and CR-39 detectors.

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1 Introduction

Human exposure to ionizing radiation is the most important subject that tends to public attention. Natural radiation is responsible for the total radiation exposure of the human population [1]. A large part of all exposure for most people is natural radiation of exposure, possibly forming a baseline upon and man-made source. The dose of natural radioactivity is very important to discuss in both ways: it affects health and the percentage of man-made radiation because natural radiation is the main source of exposure for humans. There are naturally occurring radionuclides materials (NORMS) that can be present in rocks and soils, For example, ^{238}U and ^{232}Th which are radionuclides, and ^{40}K which is originated in the primordial era [2]. These materials are still more than billions of years in the soils and rocks, so that simply can be called permanent. Furthermore, the cosmic radiation, internal radiation, and building materials contained NORMS, which are the main source of exposure [3]. These materials can be studied and tend to determine the amount of public by estimate the natural radioactivity. There is no way to avoid being exposed to these natural sources, which, in fact, cause most of the radiation exposure of the world's population. The global average effective dose per person is about 2.4 mSv and ranges from about 1 to more than 10 mSv depending on where people live [1]. Buildings may trap a particular radioactive gas, called radon, or the building material itself may contain radionuclides that increase radiation exposure. Radon is a rare natural element as it is found in gas form, noble and radioactive in its isotopes. Radon gas can be gathered in buildings, especially in closed regions, such as under roofs and basement. It is found in some spring waters and hot springs too [4]. Radon gas may be a problem for human health. The presence of ^{226}Ra in the ground of the facilities and the building materials is considered the main radon source of radon [3]. The outside air also has a role in radon concentration indoors, through the air ventilation. It was noticed that high indoor radon levels are created from radon that is in the underlying rocks and soils [5]. However, once it percolates into an enclosed space, such as a building, it can accumulate to dangerous levels, depending on the concentration of radon in the underlying soil and the construction details of the building. Both building and decorative materials contain a little number of alpha emitters (such as Uranium-238, Radium-226, and Radon-222) are mostly damaging if they are ingested or inhaled into the lungs. These building materials have a high level of natural radionuclides those results from a huge dose inside than outside. Moreover, public radiation possibly increases, if people live in buildings constructed or their houses. This increase in radiation may because using the materials that already have radiation doses more than normal levels in the area. For example: using red clay, fly-ash, rock, soil, sand, and other building materials [6,7]. The radon emissions from building materials are important to consider in certain cases. For other building materials, the emission of radon can be prevented by the selection of materials with low radium concentrations [6]. Iraq has no guidelines prescribing or standard levels of radioactivity in decorative building materials to be acceptable in our country. There are many ranges of building materials used for indoor and outdoor decoration purposes. Some studies are using CR-39 detectors to investigate decorative materials [7-9]. This study aims to establish measurements for natural alpha emitters (such as Uranium-238, Radium-226, and Radon-222) concentration in the decorative materials used as walls in Iraqi buildings. In the present study, measurements of alpha emitters were carried out for decorative materials using the TASLIMAGE System with CR-39 detectors.

2 Materials and Methods

2.1 Collection of the Samples

Thirty kinds of decorative materials samples that comment used for walls by Iraqi building during November 2020 from different locations of Iraqi markets. Decorative materials are divided into categories based on their intended use such as walls, floors, ceilings, and roofs. In the present study, exterior and interior of walls samples were collected which written according to name , type of samples, and origin of samples, as shown in Table (1).

Table 1. Information about the decorative materials samples used to walls that available in Iraqi markets and made from different countries.

No.	Type of Sample	Sample Name	Sample Code	Origin	No.	Type of sample	Sample Name	Sample Code	Origin
1	White Cement	Saten	W1	Germany	**16**	Rock	Camelot	W16	Iran
2	White Cement	Turbo	W2	Germany	**17**	Rock	Adlit	W17	Iran
3	White Cement	Fuga	W3	Germany	**18**	Rock	Tabco	W18	Jordon
4	White Cement	Tchno	W4	Bulgaria	**19**	Alabaster	AMB	W19	Iran
5	White Cement	Almalij	W5	Iraq	**20**	Alabaster	Merdo	W20	Turkey
6	Rock	Isfahan	W6	Iran	**21**	Alabaster	Yunfu	W21	Iran
7	White Cement	Darsaan	W7	Iran	**22**	Alabaster	ECF	W22	Turkey
8	Mirror (2mm)	Iranglassco	W8	China	**23**	Rock	Berdan Kazemi	W23	Iran
9	Mirror (4mm)	Iranglassco	W9	Iran	**24**	Rock	Iran zamil	W24	Iran
10	Mirror (4mm)	Sinoy	W10	China	**25**	Cement	Benoid	W25	Iran
11	Rock	Volcavo	W11	Brazil	**26**	Cement	Cimsa	W26	Turkey
12	Rock	Wand	W12	Iran	**27**	Cement	Shargh	W27	Iran
13	Rock	Belcar	W13	Spain	**28**	Cement	Cement	W28	Iran
14	White Cement	Almaein	W14	Iran	**29**	Rock	Hans	W29	China
15	Rock	Tratonen	W15	Iran	**30**	Cement	Ferana	W30	Turkey

2.2 Preparation of the Samples

After the collection of decorative materials samples, all samples are packed in polyethylene bags, and transferred to the laboratory in the physics department, Faculty of Science, University of Kufa. Samples were prepared for measuring by drying and placed by 6 hr at 100 ^{0}C in the oven. Next, using a mill model FT102 supplied by

TAISITE to crush all samples and reach a suitable homogeneity. Then, the samples were filtered through a 0.8mm pore size diameter. The weight of the respective net is measured and recorded with a high sensitive digital weighing balance (±0.01%). The specific activity of sample is defined as its activity per unit mass, so used the high sensitive digital weighing balance for getting the very high accuracy security of the specific activity radon, radium, and uranium. About 70 gm packed in a standard container (radius 2.75 cm, and length 11.5 cm) which is sealed and dry weighed. Finally, samples were kept for a couple of 4 weeks before measuring to recorded the secular equilibrium between ^{222}Rn and ^{226}Ra [10].

2.3 Measurement Methods of the Samples

Poly allyldiglycol carbonate (PADC) plastic, also known as CR-39 ($C_{12}H_{18}O_7$), serves as a solid-state nuclear track detector which is highly sensitive to a particles. It is manufactured by Track Analysis Systems Ltd. at Bristol University under the trade name TASTRAK. Alpha-particle detection with TASTRAK carried out in this laboratory provides spectroscopic information. The shape and dimensions of α-particle tracks are measured, either manually or by automated image analysis, and these measurements are used to derive for each track the dip angle and range in the plastic. CR-39 detector was used in present study that produced from TASTRAK company (Ltd., UK: TASTRACK) and developed by TASL. These detectors have many properties such as density (1.32 g/cm^3), thickness (1 mm), dimensions (2.5cm×2.5 cm), and each detector has special code and number to fit TASL image system. After end the time secular equilibrium, radon concentrations were measured for decorative materials samples are placed in the bottom of this tube then, CR-39 detector was installed by adhesive tape at the bottom of tube cover, then stored for 90 days (the exposure time) to ensure the radionuclide that exist in the samples reach equilibrium state. To prevent radon gas from escaping out, the containers' covers were fastened with a layer of adhesive tape. The current investigation used a long-term irradiation approach. After the irradiation period has ended, the detectors are removed from the containers and the chemical etching procedure begins. Next 90 day, CR-39 detectors were placed in a solution of NaOH at 6.25 N and temperature at 85˚C within 3 hours [11]. Then, detectors have been removed from the solution and extensively washed by a distilled water, and dried by soft tissue papers. At last, a microscopic treatment was performed to calculate of track density (ρ) in unit Track/mm^2 by TASLIMAGE system which determined track density by automatically [12]. The TASLIMAGE is a track analysis device based on a microscope that uses high quality Nikon optics to accomplish remarkable track and background feature discrimination. The technology is one-of-a-kind in that it analyzes and classifies any single track before calculating the dosage. The algorithm that distinguishes an etched track from a background feature , whether it's a scratch, a hair, or anything else, does so by analyzing 31 distinct criteria related to track characteristics. The method might be used in research as a totally automated readout system or for single plastic analysis. To create a dosage measurement for any slice of plastic in the automated mode, that is generally used for dosimetry services, simply a single click is required Individual plastics can also be analyzed using a user interface that offers a variety of choices for in-depth research. The scan data is translated to a dosage measurement automatically, and the findings are stored in a database. The system can read "Auto scan" type plastics or be adapted for any size plastic, including automated ID scanning, in addition to our own unique TASTRAK format detectors.

2.4 Calculations

2.4.1 ^{222}Rn, ^{226}Ra, and ^{238}U concentrations

^{222}Rn concentrations in the airspace of container (C) in Bq/m^3 was calculated by [13]:

$$C\left(\frac{\mathrm{Bq}}{\mathrm{m}^3}\right) = \frac{\rho}{\mathrm{K\,t}} \qquad (1)$$

where, K is the calibration factor of the CR-89 detector which calibrated using (^{222}Rn source) ^{226}Ra source ($A_{226_{Ra}}$= 6600 Bq). ^{222}Rn activity ($A_{222_{Rn}}$) in Bq was calculated according to ^{226}Ra activity by equation (2) [14]:

$$A_{222_{Rn}}(Bq) = A_{226_{Ra}}\left(1 - e^{-\lambda t}\right) \qquad (2)$$

Where, λ is decays constant for ^{222}Rn, and t is irradiation time in day which used the present study was 0.5, 1, 1.5, 2, 2.5, and 3 day.

The radon exposure (E_{Rn}) in cylindrical container (volume V) can be determined by [15]:

$$E_{Rn}\left(Bq.day.m^{-3}\right) = \left[A_{222_{Rn}}(Bq)/V(m^3)\right] * t(day) \qquad (3)$$

Slope of curve (calibration factor) was calculated using the relation between E_{Rn} (at different irradiation time 0.5, 1, 1.5, 2, 2.5, and 3 day) and ρ (track density in CR-39 detectors that corresponding to irradiation time), which equal, as follows:

$$Slope = \frac{\rho}{E_{Rn}} \quad (4)$$

At last, the calibration factor in present study was (0.28±0.043)Track.cm^{-2} /Bq.m^{-3}.day which it was similarity or nearly for previous studies that reported by [16- 22].

^{222}Rn concentration (C_{Rn}) in sample according to C (^{222}Rn in airspace), λ (^{222}Rn decay constant), h (thickness of the sample which equal 3cm), and L (distance between sample to detector which equal 8.5 cm), as following [23, 24]

$$C_{R_n}\left(\frac{Bq}{m^3}\right) = C\left(\frac{\lambda ht}{L}\right) \quad (5)$$

^{226}Ra activity content in the sample (C_{Ra}) in Bq/kg was calculated by equation (6) [25] which depend on many parameters such as ρ (track density), h (thickness of the sample which equal 3cm), A (area of container), M (mass of sample in kg), k (calibration factor of track detector), and T_{eff} (time of actual exposure) which can be determined by following [26]

$$C_{Ra}(Bq.Kg^{-1}) = \left(\frac{\rho}{k.T_{eff}}\right)\left(\frac{hA}{M}\right) \quad (6)$$

T_{eff} was calculated using equation (7), as following [26]

$$T_{eff} = \left[T - \lambda_{Rn}^{-1}\ \left(1 - e^{-\lambda_{Rn}\,T}\right)\right] \quad (7)$$

^{238}U concentrations of uranium (C_U) in ppm was calculated using equation (8) [27], which depend on W_U (weight of uranium in samples), and W_S (weight of sample).

$$C_U(ppm) = \frac{W_U}{W_S} \quad (8)$$

Weight of uranium in samples was calculated according to the number of uranium (^{238}U) atoms in the sample. The number of uranium in sample was determined using the property of the secular equilibrium between ^{222}Rn and ^{238}U.

2.4.2 Radiological Risk

Annual effective dose (AED) based on ^{222}Rn concentrations for decorative materials samples which depend on many parameters such as C (^{222}Rn in airspace), F (the equilibrium factor that equal 0.4), H (occupancy factor that equal 0.8), T (time in hours that equal 8760 h/y), and D (dose conversion factor that equal 9×10^{-6} mSv /Bq.h.m^{-3}) can be calculated by [28, 29].

$$AED\left(\frac{mSv}{y}\right) = C \times F \times H \times T \times D \quad (9)$$

There is considerable public concern about radon exhalation from building materials, especially those used for interior decoration like ceramic tile which considered as an important source that contributes to indoor radon concentration through exhalation from walls and floors. Exhalation rate from decorative materials is another potential source of radon in the indoor environment. Mass and surface exhalation rate (E_M) and (E_A) of the samples based on C (^{222}Rn in airspace) with another parameters were calculated using equations (10), and (11), respectively [30].

$$E_M = \frac{CV\lambda}{MT_{eff}} \quad (10)$$

$$E_S = \frac{CV\lambda}{AT_{eff}} \quad (11)$$

3 Results and Discussion

Results of alpha emitters (^{222}Rn, ^{226}Ra, and ^{238}U) in decorative materials that used as walls shown in Table (2). From Table (2), it is found that the values of C were ranged from 7.94 Bq/m^3 to 738.10 Bq/m^3, with an average 252.38±37.63 Bq/m^3, the values of C_{Rn} were ranged from 45.73 Bq/m^3 to 4252.99 Bq/m^3, with an average 1454.25±216.84 Bq/m^3, the values of C_{Ra} were ranged from 0.010 Bq/kg to 1.000 Bq/kg, with an average 0.278±0.04 Bq/kg, and the values of C_U were ranged from 0.01 ppm to 1.24 ppm, with an average 0.344±0.05 ppm. The higher concentrations of radon in airspace container as well as radon in sample are recorded for samples W30 (decorative cement, name Ferana, and made in Turkey), while the lowest can be observed for the samples W26 (decorative cement, name Cimsa, and made in Turkey). Also, The higher and lower of radium content as well as concentrations of uranium were recorded for samples W30 (decorative cement, name Ferana, and made in

Turkey), and for the samples W26 (decorative cement, name Cimsa, and made in Turkey), respectively. The main reason for large differences in alpha emitters for the sample (decorative materials) seems to be due to the type of natural materials that origin samples based on the geological formation and abundant radioactive element under concentration for these materials. There are no obvious guidelines and regulations for decorative materials alpha emitters in Iraq and the world, while found in indoor air, soil, and building materials. From the radiological protection point of view, the health and environmental protection agencies have recommended a safe limit of radon indoor air for human beings. The International Commission on Radiological Protection(ICRP) has proposed that the range allowed maximum contamination level for radon concentration indoor air is 200-300 Bq /m^3 [30]. As well, the world average soil radon concentration suggested by many studies is 7400 Bq/m^3 [31, 32]. It is noticed that from data (Table 2) ^{222}Rn concentrations (C) indoor air due to decorative materials that used as walls were within those proposed by international organizations since the higher value (300 Bq/m^3), except some samples (W2, W3, W9, W14, W15, W16, W21, W29, and W30), while ^{222}Rn concentrations (C_{Rn}) in all samples (decorative materials) were lower than the safe limit (7400 Bq/m^3) recommended by ICRP. Also, it is noticed that from data (Table 2) the values of radium content (C_{Ra}) and uranium concentrations (C_U) in all samples of the present study (decorative materials) were less than the level permissible limit of C_{Ra} (35 Bq/kg) [34], and C_U (11 ppm) [35] which were recommended by UNSCEAR 2000 and 1994, respectively. The results of annual effective dose (AED), mass and surface exhalation rate (E_M), and (E_S) of the samples based on ^{222}Rn concentrations in the airspace of the container are presented in table (3). The results of AED were ranged between 0.20 mSv/y in sample code W26 to 18.62 in sample code W30, with an average value of 6.29±0.95 mSv/y. It can be found from the results of AED in samples of decorative materials, most samples were less than the acceptable levels of (3-10) mSv/y which was recommended by(ICRP) [36], except samples W3. W9, W14, W15, W16, W21, W29, and W30 were higher than acceptable levels. Figure (1) shows the comparing percentage of AED between samples under study and maximum acceptable level that recommended by ICRP 1993. From figure (1), it is found that the values of percentage of samples W3 (6%), W9(6%), W15(7%), W16(8%), W21(8%), W29(9%), and W30(9%) were larger than the parentage value of acceptable levels (5%) recommended by(ICRP). The results of E_M in-unit mBq/kg.h (Table 3) were ranged from 0.29 to 30.24, with an average value of 8.38±1.33, while The results of E_S in-unit mBq/m^2.h were ranged from 7.67 to 713.13, with an average value of 240.77±36.56. While the values of E_S (surface exhalation rate of ^{222}Rn concentrations) in all samples of the present study was much lower than the world average of (57.6 Bq/m^2.h) [34]. Table (4). The average value of C and AED in decorative materials of the present study at different origin countries were shown in table (4). it can be seen that, the higher average value of C was 315.76 Bq/m^3 for decorative materials that made in Iran, while the lower was 79.37 Bq/m^3 for decorative materials that made in Brazil. Also from table (4), the higher average value of AED was 7.81 mSv/y for decorative materials that made in Germany, while the lower was 2 mSv/y for decorative materials that made in Brazil. The average value of C and AED in different types of decorative materials in present study were shown in table (5). it can be seen that, the higher average value of C was 296.032 Bq/m^3 for decorative materials type of decorative cement, while the lower was 201.27 Bq/m^3 for decorative materials type decorative white cement. Also from table (5), the higher average value of AED was 7.47 mSv/y for decorative materials type cement, while the lower was 5.09 mSv/y for decorative materials type white cement. These variation depending on the type of raw materials used in the manufacture of each one. But the average value of C and AED in all courtiers were lower than the world average limit, except some samples have high ^{222}Rn concentrations that shown above which may be due to the kind and nature of soil as well as natural materials from which the cement is manufactured. Finally and according to light of the study, it can be shown that the data obtained in this study improve the suitability of the CR-39 detector technique for such complex samples.

Table 2. Results of alpha emitters in decorative materials (walls).

No.	Sample code	C (Bq/m^3)	C_{Rn}(Bq/m^3)	C_{Ra}(Bq/kg)	C_U(ppm)
1	W1	91.27	525.91	0.092	0.11
2	W2	333.33	1920.71	0.506	0.63

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3	W3	503.97	2903.92	0.683	0.84
4	W4	111.11	640.24	0.187	0.23
5	W5	95.24	548.77	0.131	0.16
6	W6	253.97	1463.39	0.250	0.31
7	W7	75.40	434.45	0.108	0.13
8	W8	182.54	1051.82	0.156	0.19
9	W9	527.78	3041.12	0.477	0.59
10	W10	103.17	594.50	0.106	0.13
11	W11	79.37	457.31	0.091	0.11
12	W12	11.90	68.60	0.013	0.02
13	W13	107.14	617.37	0.110	0.14
14	W14	424.60	2446.61	0.671	0.83
15	W15	543.65	3132.58	0.421	0.52
16	W16	619.05	3567.03	0.652	0.81
17	W17	103.17	594.50	0.126	0.16
18	W18	83.33	480.18	0.075	0.09
19	W19	218.25	1257.61	0.200	0.25
20	W20	186.51	1074.68	0.134	0.17
21	W21	623.02	3589.89	0.577	0.71
22	W22	134.92	777.43	0.123	0.15
23	W23	39.68	228.66	0.037	0.05
24	W24	115.08	663.10	0.103	0.13
25	W25	246.03	1417.66	0.263	0.33
26	W26	7.94	45.73	0.010	0.01
27	W27	265.87	1531.99	0.284	0.35
28	W28	222.22	1280.47	0.296	0.37
29	W29	523.81	3018.25	0.457	0.56
30	W30	738.10	4252.99	1.000	1.24
Average±S.E		252.38±37.63	1454.25±216.84	0.278±0.04	0.344±0.05

Table 3. Results of AED, E_M, and E_S in decorative materials (walls).

No.	Sample code	AED (mSv/y)	E_M (mBq/kg.h)	E_S (mBq/m^2.h)
1	W1	2.30	2.79	88.18
2	W2	8.41	15.30	322.06
3	W3	12.71	20.65	486.92
4	W4	2.80	5.66	107.35
5	W5	2.40	3.97	92.02
6	W6	6.41	7.57	245.38
7	W7	1.90	3.26	72.85
8	W8	4.61	4.71	176.37
9	W9	13.32	14.42	509.93
10	W10	2.60	3.20	99.69
11	W11	2.00	2.76	76.68
12	W12	0.30	0.40	11.50
13	W13	2.70	3.32	103.52
14	W14	10.71	20.30	410.24
15	W15	13.72	12.73	525.26
16	W16	15.62	19.73	598.11
17	W17	2.60	3.82	99.69
18	W18	2.60	3.82	99.69
19	W19	2.60	3.82	99.69
20	W20	4.71	4.04	180.20
21	W21	15.72	17.43	601.95
22	W22	3.40	3.73	130.36
23	W23	1.00	1.11	38.34
24	W24	2.90	3.11	111.19
25	W25	6.21	7.95	237.71
26	W26	0.20	0.29	7.67

27	W27	6.71	8.59	256.88
28	W28	5.61	8.94	214.71
29	W29	13.22	13.81	506.09
30	W30	18.62	30.24	713.13
Average±S.E		6.29±0.95	8.38±1.33	240.77±36.56

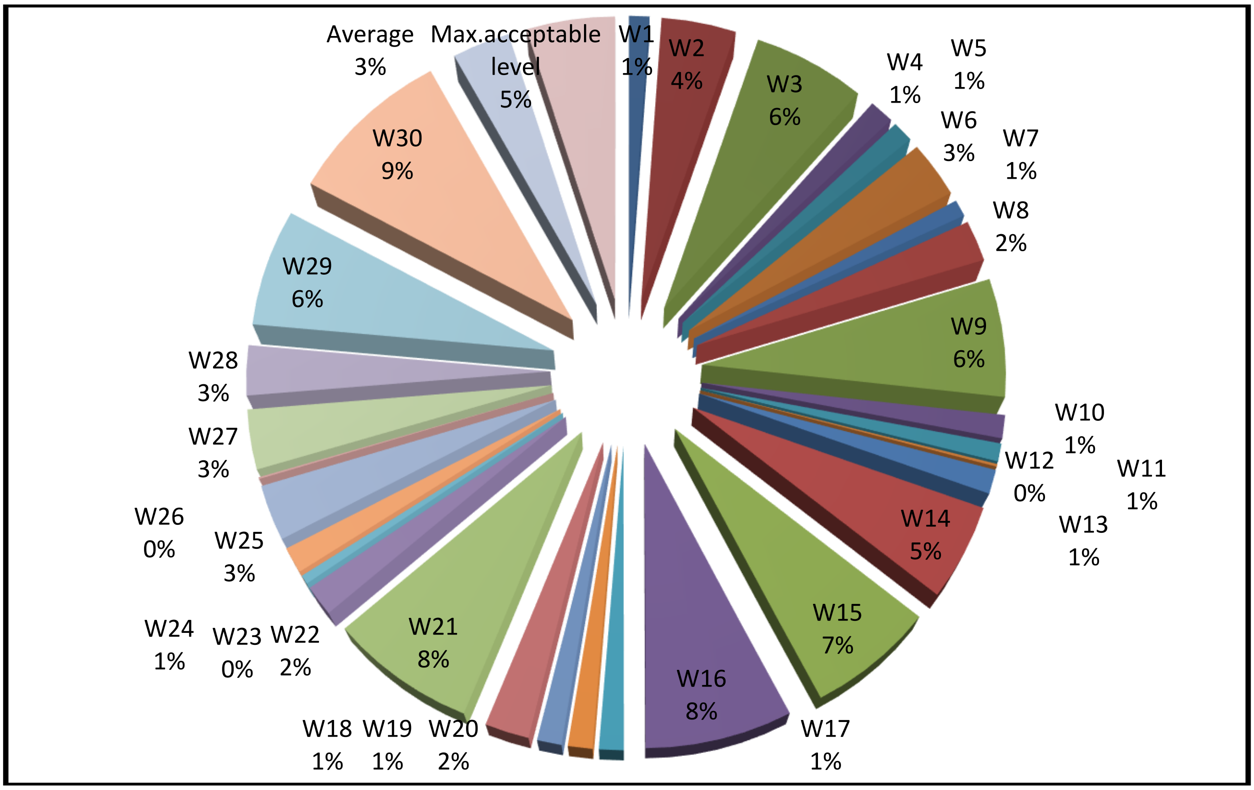

Fig. 1: The comparing percentage of AED between samples under study and maximum acceptable level that recommended by ICRP 1993.

Table 4.The comparing of C and AED in decorative materials at different origin countries.

No.	Origin Country	C (Bq/m^3)	AED (mSv/y)
1	Germany	309.52	7.81
2	Bulgaria	111.11	2.8
3	Iran	315.76	7.76
4	China	269.84	6.81
5	Brazil	79.37	2
6	Spain	107.14	2.7
7	Jordanian	83.33	2.6
8	Turkey	266.87	6.73
9	Iraq	95.24	2.4

Table 5. The comparing of C and AED in different types of decorative materials in present study.

No.	Type of decorative material	C (Bq/m^3)	AED (mSv/y)
1	White cement	201.72	5.09
2	Rock	225.47	5.73
3	Mirror	271.16	6.84
4	Alabaster	290.675	6.61
5	Cement	296.032	7.47

4 Conclusion

The decorative materials in the present study are found to contain natural alpha emitters at different rates according to origin countries and types of decorative materials. Therefore, it can be concluded that the results of ^{222}Rn concentrations in samples, ^{226}Ra activity content and concentrations of uranium, as well as mass and surface exhalation rate of the thirty kinds of decorative materials samples that comment used for walls by Iraqi building in the present study, were lower than the acceptable of the permissible limit. While radon concentrations in air space of the container and annual effective dose for some samples such as W3. W9, W14, W15, W16, W21, W29, and W30 were greater than the acceptance of the permissible limit. Accordingly, It should be careful in using it as decorative materials.

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