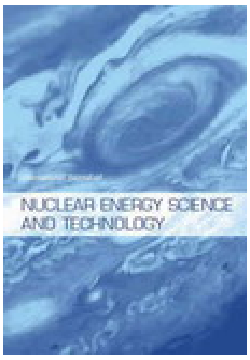

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Determination of radiological hazards due to alpha emitters from ceramic used in Iraq

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Abstract: The approach of sealed can was utilised in this work to determine the amount of the radioactivity (alpha emission) of imported ceramic tiles that are used in different kinds of buildings kinds in Iraq. The resulted data showed that the radon concentration varied from 22.105 to 302.482 Bq/m^3 with an average of 162.293 Bq/m^3. The effective radium content ranged from 0.079 to 1.087 Bq/kg with an average value of 0.583 Bq/kg. The uranium concentration varied from 1.192 to 16.313 Bq/kg with an average value of 16.313 Bq/kg. After obtaining those results and comparing them with the global average and permissible limits recommended by international scientific agencies such as ICRP and UNSCEAR, it was found that the considered ceramic samples are safe for local use.

Keywords: alpha emitters; ceramic, radiological hazards; closed-can technique.

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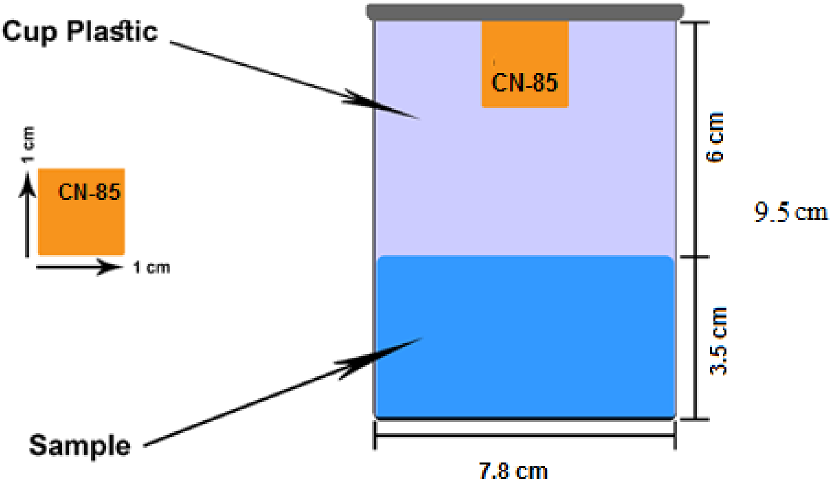
1 Introduction

Naturally, radiation is prevalent in our environment since the creation of the Earth. Hence, life has evolved in an environment that has significant levels of ionising radiation. Radiation comes from outer space (cosmic), the ground (terrestrial) and even within our bodies. It exists in the air, food, water and the construction materials used to build our houses (Somlai et al., 2007). Three common types of radioactive processes are alpha decay, beta decay and gamma-ray emission. Radon, which simplified as (^{222}Rn) is a radioactive noble gas, with a half-life of 3.82 days that is naturally occurring. It is known to belong to the well-known ^{238}U decay series (Somlai et al., 2007). Radon, together with its decay products (short-lived) e.g., ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po, has been recognised as an indoor major source of public exposure to the natural radioactivity. In this context, it is contributing to around fifty percent of the mean effective dose to the general public (Abojassim et al., 2021). Further to this, the kind of soil, building materials and water which are used for variable human purposes (i.e., drinking) can contribute to the indoor radon level (Sohrabi, 1998). The relevant knowledge on radon contamination reveals that the major source of the indoor radon is the underneath building soil (UNSCEAR, 1993). Nevertheless, it should be noted that some radium contaminated building materials together with using the domestic radon contaminated water can contribute markedly to indoor radon exposure (Kearfott, 1989). The most representative health risk aspect of human exposure to radon is ‘lung cancer’ (Li et al., 2006). For a few decades, great attention has been paid in the studying the impact of natural radiation activity by researchers. Nevertheless, a particular attention was given to the radon gas since it contributes to more than 50% of the natural ionising radiation dose that the people receive ever year (Folger et al., 1994). It is worth mentioned that alpha radiation yield occurs by the natural disintegration of radon products (i.e., ^{218}Po and ^{214}Po). So, this type of radiation is able to interact with lung cells after being inhaled with air. This can, as a result, damage DNA (Khan, 2000). The transition of radon gas from the pore into the atmosphere is a process that is known by exhalation. The radon exhalation process depends on the abundance of radium in a given material (Choudhary, 2014). Different approaches have been adopted to measure the radon. Using the Solid-State Nuclear Tracks Detectors (SSNTD) is one of the commonest and widely applied techniques for long term radon measurements. It had been used for determination of the radon emanation in a limestone cave (Yousef et al., 2016). Additionally, this kind of detector is being used for identification of radon activity and uranium content in various geological studies (Yousef et al., 2019; Abojassim, 2021; Ibrahim et al., 201a, 2021b; Al Rmahi and Abojassim, 2021). Having known that the uranium and radium exist in the soil, rocks and building materials, they are considered as the main sources of indoor radon. The current research work aimed at investigating the radiological hazards caused by radon exposure, together with identifying the effective radium content and uranium concentrations in fifteen samples of imported ceramics available in Iraqi market.

2 Materials and methods

The approach of sealed can and the CN-85 detector was used to estimate the radiation hazards from radon in addition to the effective radium content and uranium concentration in fifteen samples of ceramics available in the Iraqi market. The detector of ‘Cellulose Nitrate’ – CN-85 (12 μm thickness), is a very helpful detector for direct monitoring of alpha radiation tracks imparted on the sensitive area of the detector that is facing the samples. The experimental setup diagram can be seen in Figure 1. The generated tracks on the detector surface are not directly visible, therefore they must be exhibited via adopting an appropriate chemical processing technique. Following this, the demonstrated tracks must be magnified using a light microscope. The types of ceramics which are allocated for homes construction were collected from the local Iraqi market. The collected samples were dried in a controlled furnace (oven) adopting a temperature of 100±0.1°C for 3 hrs. This was to ensure that the moisture is certainly eliminated. After that, every sample was crushed into a fine powder when they were sieved via utilising a fine mesh to eliminate the large grains, and therefore obtain sample homogeneity. An amount of about 85 g of sample was put in a plastic can whose dimensions are: 9.5 cm height and 7.8 cm in diameter. A (1×1) cm^2 size detector was put on the top of inner side of the can surface. By the latter setting, the detector sensitive surface will always be facing the ceramic sample. After that, this can was sealed with adhesive tape, and then kept to be exposed to alpha radiation for around 90 days. During this period of alpha exposure, the detector sensitive side is exposed freely to the emergent radon from the sample in the can where it would able to record alpha particles resulted from the usual decay of radon in the remaining volume of the can. The detectors were later collected and treated by etching process in a solution of 6.25N NaOH, at 60°C. Next, these detectors were kept in the bath for 3 hours. The detectors were, then, washed and dried, and the tracks of alpha particles were counted using a 400X magnification (40X objective and 10X eye piece) optical-mission microscope.

Figure 1 An investigation tube technique used in the study



3 Theory

The density of radon C_a in the can air which is above the sample can be determined by identifying the track density in the detector using the following formula (Najam et al., 2019):

$$C_a = \frac{\rho}{KT} \tag{1}$$

where ρ is the identified track surface density of the irradiated detectors (track/cm^2),.T refers to the exposure time. The track densities are. related to the level of radon concentration utilising a calibration factor value of 0.256. track.cm^{-1}.day^{-1} /Bq.m^{-3} (Hashim and Nayif, 2019).

The dissolved radon concentrations (C_{Rn}) in the samples can be estimated using the connection between the radon concentrations emitted from the samples into the air surrounding the samples as follows (Hameed et al., 2020):

$$C_{Rn} = \frac{C.\lambda.h.T}{L} \tag{2}$$

where, λ: a constant of radon decay, *h*: refers to the distance between the surface of the sample and the detector, L: refers to the sample height. Alpha disintegration can be employed to calculate the effective radium content, the radon concentration is expected to increase together with increasing time after the closure of the canister that included the samples in which an effective equilibrium of (98%) between radium and radon can be attained in the radiation decay series within a period of approximately 1 month. Once the required equilibrium has been reached, the concentration of radium of the samples will be calculated using the following equation (Hashim et al., 2019a):

$$C_{Ra}\left(Bq.\mathrm{kg}^{-1}\right)=\left(\frac{\rho}{KT_e}\right)\left(\frac{hA}{M}\right) \tag{3}$$

where. *M* represents the sample's mass (kg), A. represents the can area of cross-section (m^2.);.*h* refers to the distance between the detector and top surface of the samples in meter. T_e refers to the effective exposure time that is given by. Hashim et al. (2019b):

$$T_e=\left[T-\lambda_{Rn}^{-1}\left(1-e^{-\lambda_{Rn}T}\right)\right] \tag{4}$$

.For the surface exhalation rate of the sample to be calculated, this expression can be used (Hashim et al., 2019a).

$$E_S\left(\mathrm{mBq\ m^{-2}h^{-1}}\right)=\frac{CV\lambda}{A\left[T+\lambda^{-1}\left(e^{-\lambda T}-1\right)\right]} \tag{5}$$

where E_S refers to the radon exhalation rate in terms of area expressed in ($\mathrm{mBq\ m^{-2}h^{-1}}$), *C* refers to the radon exposure integration expressed that is in Bq $\mathrm{m^{-3}h}$,. *V* refers to the effective volume of the cup in m^3,.*T* represents the exposure time (hour),.λ refers to the decay constant for ^{222}Rn radon (h^{-1}), and *A* refers to the area of the cup (m^2).

The rate of mass exhalation of the sample for the radon can be calculated using the expression. below. (Hashim et al., 2019b).

$$E_M\left(\mathrm{mBq\ kg^{-1}h^{-1}}\right)=\frac{CV\lambda}{M\left[T+\lambda^{-1}\left(e^{-\lambda T}-1\right)\right]} \tag{6}$$

where E_M is the radon exhalation rate, in terms of mass expressed in ($\mathrm{mBq\ kg^{-1}h^{-1}}$) and *M* refers to the mass of the sample.(kg).

For the uranium concentrations, $\left(C_U\right)$ (part per million-ppm) to determined, the following equation can utilise (Al-Saadi et al., 2013):

$$C_U\left(ppm\right)=\frac{W_U}{W_s} \tag{7}$$

where W_S: refers to the weight of sample.

W_U represents the uranium weight in considered sample, and can be calculated via the following equation (Al-Saadi et al., 2013).

$$W_U\left(gm\right)=\frac{N_U\ W_{mol.}}{N_{Av.}} \tag{8}$$

where $W_{\text{mol.}}$ represents the molecular weight of the uranium. $N_{Av.}$: Avogadro's number (i.e., 6.023×10^{23}atom/mol).

The International Atomic Energy Agency (IAEA) adopts the below conversion factor from concentration unit to activity unit in Bq.kg^{-1} (AC01022355, 1989; IAEA, 2003):

$$1\,\text{ppm of Uranium} = 12.35\frac{\text{Bq}}{\text{kg}}\text{of}\,^{238}\text{U} \tag{9}$$

4 Radiologic hazard parameters

4.1 Annual effective dose

The annual effective dose AED (mSv y^{-1}) levels resulted from Rn-222, exposure can be obtained using the following equation (Abdalla and Al-Naggar, 2019):

$$AED = \left(\left(0.17 + 9F\right)C_a\right)\times 0.8\times 8760\times 10^{-6} \tag{10}$$

where (F = 0.4) which represents the equilibrium factor among radon and its progeny, C_a : represents the concentration of radon, number of hours per year = 8760 h, and 0.8 is the indoor occupancy factor.

4.2 Alpha index

The index of alpha was employed as an indicator for excessive alpha radiations exposure caused by the inhalation of radon gas emanated from construction materials as an example; this index thus can be calculated as follows: (Prot, 1999; Moharram et al., 2012; Omeje et al., 2018) :

$$I\alpha = \frac{C_{Ra}}{200\text{Bq kq}^{-1}} \tag{11}$$

where C_{Ra} refers to the concentration of Ra-226. (Bq kg^{-1}) in the building material. Once the concentration of Ra-226 of a given building material exceeded the level of 200 Bq kg^{-1}, it is probably that the radon exhalation from an aforementioned material may lead to that indoor radon concentrations also exceeding. 200 $\text{Bq m}^{-3.}$ (Prot, 1999; Moharram et al., 2012; Omeje et al., 2018). Therefore, a high level of radon concentration. $\left(I_\alpha\right)$ is expected to equal to 1 (Omeje et al., 2018; Tufail and Hamid, 2007).

4.3 Excess lifetime cancer risk (ELCR)

It is one of the radiologic variables, that can be identified through the following formula (Shoeib, M.Y. and Thabayneh, 2014; Taskin et al., 2009):

$$ELCR = AED\times DL\times RF \tag{12}$$

where AED, DL and RF represent the total annual effective dose equivalent (mSv y^{-1}), the duration of life (70 years) and risk factor (0.05 Sv^{-1}) for stochastic effects, that has been recommended by ICRP 60 for general public (Thabayneh, 2013; ICRP, 2012).

5 Results and discussion

The obtained results (i.e., radon concentrations, annual effective dose, alpha index and excess lifetime cancer risk-ELCR) for varying kinds of ceramics that are imported to the Iraqi market can be seen in Table 1. By contrast, Table 2 presents the results of the effective radium content, radon exhalation rate and uranium concentration for the same samples. The results of the aforementioned samples were arranged in ascending order as shown in Tables 1 and 2, where the lowest values were recorded in Egyptian ceramics 1 and the highest values in Spanish ceramics 2. The air space's radon concentration that amid the sample surface and the detector varied between 22.105 Bq/m^3 and 302.482 Bq/m^3 with an average of 162.293 Bq/m^3. The resulted data demonstrated that the radon concentration for all samples in current research work is within internationally permissible values that is consistent with the permissible value (300 Bq/m^3) recommended by ICRP (2012). The annual effective dose caused by the emitted radon from the considered samples were ranged from 0.584 to 7.992 mSv/y with an average 4.287 mSv/y as shown in Table 1. It can be noticed that whole values of annual effective dose are below the permission level. (10 mSv/y) (Thabayneh, 2013). Concerning the alpha index findings, it was seen that they were ranged from 0.397×10^{-3} to 5.434×10^{-3}, with a mean of 2.915×10^{-3}. Almost all of the samples' mean of the alpha index values were observed to be well below one, which is the recommended value for $I\alpha$. The latter recommended value is resulting from the recommended marginal concentration of ^{226}Ra is 200 Bq/kg (Rafique et al., 2011). The ELCR were seen to be ranged from 2.044×10^{-3} to 27.97×10^{-3}, with a mean of 15.006×10^{-3}. It should be noted that whole ELCR findings were larger than the global value (0.29×10^{-3}) (UNSCEAR, 2000a, Scientific Annexes). By considering these findings, the risk of developing cancer increases with increased exposure time or via the prolonged survival in places containing this material. The values of effective radium content for samples are ranged from 0.079 Bq/kg to 1.087 Bq/kg with an average 0.583 Bq/kg. The whole values of the effective radium content were seen to be less than that of the recommended limit of 370 Bq/kg (OECD, 1979). The mass and surface exhalation rate of radon for samples are ranged from 0.6 to 8.215 mBq/kg.h and 10.679 $mBq/m^2.h$ to 146.126 $mBq/m^2.h$ with an average 4.407 mBq/kg.h and 78.402 $mBq/m^2.h$, respectively. All values of exhalation rate for samples are relatively lower than the permission value (57.600 $mBq/m^2.h$) set by UNSCEAR organisation (UNSCEAR, 2000b, Annex B).

The uranium concentrations for samples ranged from 1.192 to 16.313 Bq/kg with an average 8.752 Bq/kg. The whole values of uranium concentration for samples were seen to be less than that of the reported limit of 35 Bq/kg (UNSCEAR, 2000b, Annex B).

It can be said that the results of our current study do not present a risk to human health in comparison with global levels and with the results of previous local studies in which the CN-85 was used.

Table 1 Track density (ρ), radon concentration (*C*), annual effective dose (AED), alpha index $(I\alpha)$, excess life-time cancer risk (ELCR) for different ceramic sample in Iraqi market

Code sample	*Ceramic*	$\rho\times10^{2}$ *Track/cm*^{2}	*C Bq/m*^{3}	*AED mSv/y*	$I\alpha\times10^{-3}$	*ELCR*$\times10^{-3}$
S1	Egyptian1	5.092	22.105	.584	0.397	2.044
S2	Chinese 1	9.707	42.132	1.113	0.757	3.895
S3	Egyptian 2	14.321	62.159	1.642	1.117	5.747
S4	Iranian 1	18.935	82.186	2.171	1.476	7.599
S5	Emirati 1	23.549	102.213	2.700	1.836	9.451
S6	Italian 1	28.164	122.240	3.230	2.196	11.303
S7	Emirati 2	32.778	142.267	3.759	2.556	13.155
S8	Syrian 1	37.392	162.293	4.288	2.916	15.007
S9	Iranian 2	42.006	182.320	4.817	3.275	16.859
S10	Syrian 2	46.620	202.347	5.346	3.635	18.711
S11	Italian 2	51.235	222.374	5.875	3.995	20.563
S12	Saudi	55.849	242.401	6.404	4.355	22.414
S13	Spanish 1	60.463	262.428	6.933	4.715	24.266
S14	Chinese 2	65.077	282.455	7.463	5.074	26.118
S15	Spanish 2	69.691	302.482	7.992	5.434	27.970
Max.		69.691	302.482	7.992	5.434	27.97
Min.		5.092	22.105	0.584	0.397	2.044
Mean		37.391	162.293	4.287	2.915	15.006

Table 2 The dissolved radon concentrations (C_{Rn}), effective radium content (C_{Ra}), mass (E_M) and surface (E_S) exhalation rate for radon, and uranium concentration in various ceramic sample

Code sample	$C_{Rn}\times10^{3}$ *Bq/m*^{3}	C_{Ra} *Bq/kg*	E_M *mBq/kg.h*	E_S *mBq/m*^{2}*.h*	*CU Bq/kg*
S1	0.618	0.079	0.600	10.679	1.192
S2	1.179	0.151	1.144	20.353	2.272
S3	1.739	0.223	1.688	30.028	3.352
S4	2.300	0.295	2.232	39.703	4.432
S5	2.860	0.367	2.776	49.378	5.512
S6	3.421	0.439	3.320	59.053	6.593
S7	3.981	0.511	3.864	68.728	7.673
S8	4.542	0.583	4.407	78.402	8.753
S9	5.102	0.655	4.951	88.077	9.833
S10	5.663	0.727	5.495	97.752	10.913
S11	6.223	0.799	6.039	107.427	11.993

Table 2 The dissolved radon concentrations (C_{Rn}), effective radium content (C_{Ra}), mass (E_M) and surface (E_S) exhalation rate for radon, and uranium concentration in various ceramic sample (continued)

Code sample	*C_{Rn} ×10^3 Bq/m^3*	*C_{Ra} Bq/kg*	*E_M mBq/kg.h*	*E_S mBq/m^2.h*	*CU Bq/kg*
S12	6.784	0.871	6.583	117.102	13.073
S13	7.344	0.943	7.127	126.777	14.153
S14	7.905	1.015	7.671	136.451	15.233
S15	8.465	1.087	8.215	146.126	16.313
Max.	8.465	1.087	8.215	146.126	16.313
Min.	0.618	0.079	0.6	10.679	1.192
Mean	4.541	0.583	4.407	78.402	8.752

6 Conclusions

The mean radon concentration level (Bq/m^3) for the majority of the studied ceramics are below the permission limit reported by ICRP. The values of each of the effective radium content and uranium concentrations in ceramic materials belonging to all countries are lower than those of the recommended limits of 370 Bq/kg and 35 Bq/kg, respectively. The values of annual effective dose are below the permission level. The alpha index levels for all samples of ceramics are seen to be almost lower than unity. The ELCR findings for ceramic samples are almost higher than that of the world value. Finally, the considered ceramic samples are safe to exploit as building materials.

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