Measurement of Radon Exhalation Rate from Pottery Meal Dishes in Erbil City by using Passive and Active Techniques

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Abstract

Solid State Nuclear Track Detectors (SSNTDs) of type CR-39 has been used in this study as a passive method to measure the radon activity concentration, radon exhalation rate (in terms of area and mass), and effective radon content in the pottery meal dishes of different origins (Germany, England, China, and Iran) in Erbil city. The RAD7 solid state detector has also used as an active method for measuring the radon activity concentrations from the measurement of alpha-particle track density. The results reveal that the highest exhalation rate and effective radium content of 0.69 ± 0.035 mBq.m^{-2}.h^{-1} and 0.258 ± 0.013 Bq.Kg^{-1}, respectively, were found in England meal dishes; while the lowest exhalation rate and effective radium content of 0.196 ± 0.0098 mBq.m^{-2}.h^{-1} and 0.106 ± 0.005 Bq.Kg^{-1}, respectively, were found in China meal dishes. It is concluded that the radon exhalation rate from the pottery dishes has significant contribution to the indoor doses that should be taken into account.

Introduction

Since its discovery in 1958 (Young,1958) and 1959 [Silk *et al.*,1959], the technique now generally known as solid state Nuclear Track Detector (SSNTD) has, over the last few decades, becomes a popular and well-established method of measurement in a large number of fields involving different aspects of radioactivity or nuclear interactions.

^{226}Ra (t1/2 =1602 years) is a naturally occurring radioisotope of the ^{238}U decay series. Earth, Marine, and environmental scientists often require analysis of ^{226}Ra because of public health concerns (Aupiais, *et al.*,1998). The measurements of radium has become a matter of interest because its one of the most hazardous elements with respect to internal radiation exposure (Higuchi *et al.*, 1984). There are three natural isotopes of this radioelement in the environment – ^{222}Rn, ^{220}Rn,and ^{219}Rn (actinon) originated from the so-called decay series of ^{238}U (uranium), ^{232}Th(thorium), and ^{235}U (actinium), respectively (Banjanacl, *et al.*, 2006).

^{222}Rn (half life =3.8٢٥ days) is an inert noble gas and the immediate daughter product of ^{226}Ra. Natural radiation accounts for the majority of human exposure to radiation, and ^{222}Rn and its short-lived daughter

products are the largest contributors to this radiation dose. Radon has long been identified and as a cited, hazard in indoor air. Radon gas emanates from the soil into homes from cracks in flooring or through basement floors and the other radium content home constituents by molecular diffusion or pressure-driven flow and may also be present in building materials (Tso *et al.*, 1987). When radon is inhaled into the lungs it decays by means of alpha – emission and which causes ionization damage when it strikes the lung tissue. Over time, this damage causes lung cancer. For example smoking is responsible for 87% of the lung cancer cases. The connection between radon and lung cancer in miners has raised cancer that radon in homes might be causing lung cancer in the general population (Shafi ur-Renman, 2005).

Because of the radon health concern, several methods have been developed to monitor for radon and its daughters in air. Radon measurements are one of the most widely used applications of SSNTDs today. These include alpha track activated charcoal absorption, continuous radon monitoring, grab radon sampling, and radon progeny integrated sampling (Passo & Floeckher, 1991).

It is well known that the inhalation of short lived ^{222}Rn daughters contribute about 40% of the total exposure of population (EL-sersy *et al.*, 2004) and the other (water and food, Earth gamma radiation and cosmic rays) contribute about 60%.

In the last 30 years, more attention has been paid to the measurement of radon exhalation from building materials worldwide (Abu-Jarad *et al.*, 1980, Mustonen,1984, Al-Jarallah, 2001, Ching-Jiang *et al.*, 1993, Savidon *et al.*, 1996, Turhan 2008, SAAD,2009 and Aziz,2010).

The International commission on Radiological protection (ICRP, 1993) recommended radon concentration value ranges of (50-1500) and (200-600) Bqm^{-3} for work places and dwellings, respectively; those concentrations do not pose a significant risk for residencies and workers.

In this study, and due to the lack of studies concerning the assessment of radium contents and radon exhalation rates of materials having daily live house usages in Erbil, it is necessary to start preliminary study to measure the radon exhalation rates, and radium contents from pottery meal dishes where they may constitute a significant source of radon in houses. For this two integrating measurement methods with (CR-39) as detectors were attempted; first: a passive method, and second: an active method using the RAD7 instrument.

Experimental procedures

Different types of pottery meal dishes having different origins and wide usages in Erbil city were studied for assessing the radium content and radon exhalation rates.

Radon concentration is generally measured using either passive solid state nuclear track detectors (SSNTDs) technique or using active technique (Al-Kofahi,1992 and Awawdeh, 2001). The structure of passive dosimeters have been developed and described elsewhere by several workers (Cartwright et al., 1987,Al-Kofahi,1992 and Hassan, 1996). The dosimeter used in present work consisted of a CR-39 detector (Pershore Moulding, U.K. (Cartwright *et al.,* 1987)). For the active method, the RAD7 solid state detector has been used for measuring activity concentration and exhalation rate of radon. The collected 20 samples (five samples from each origin with different shapes and sizes) were powdered in to fine grains and dried at $110^{o}C$ for 24 hr to remove the moisture contents. About 120 g of each dried sample were poured in plastic containers made of PVC materials of radius 3.5 cm having similar volumes. These sample containers were inserted in a flat position at the bottom of an inverted cylindrical PVC cup of 3.5 cm radius and 30 cm high form the sample surface in order to count only the contribution of ^{222}Rn and its progenies and to exclude the role of thoron from the counting. Twenty pieces (1.5cm x 1.5 cm) of CR-39 radon detectors were fixed at the top center of the inverted cup attached the stopping rubber surfaces as shown in Fig. (1). Within the same condition and procedure two empty dosimeters were prepared and left for the same counting time in order to estimate the rate of background which is later subtracted from each of the sample counting data to obtain the net track density.
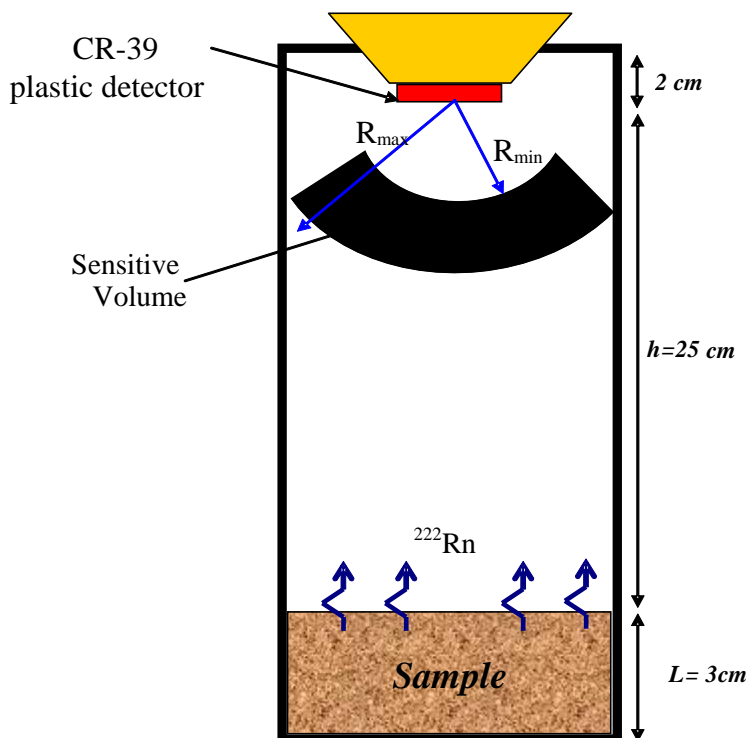
Fig.(1): Experimental arrangement for radon exhalation measurement. (Durrani, 1997)

All dosimeters were left for almost two months. After this period, the system was brought to:

1. The active method measurement, using the RAD7 detector as configured with the tube as shown in Fig. (2). A close loop configuration is connected and the valves are opened. A high voltage of 2500V is applied to the chamber walls. The solid state detector converts the alpha radiation directly to an electrical signal using an alpha technique. The procedure was performed under dry condition (relative humidity of 5-10%) and room temperature of $(25\pm 5)^{o}C$.

2. The passive method: After performing the active method, the dosimeters were collected and the detectors were taken out of the dosimeters. The detectors were then chemically etched in 6N-solution of NaOH at a temperature of $70\pm1\ ^{o}C$ maintained by a water bath for 7 hours.
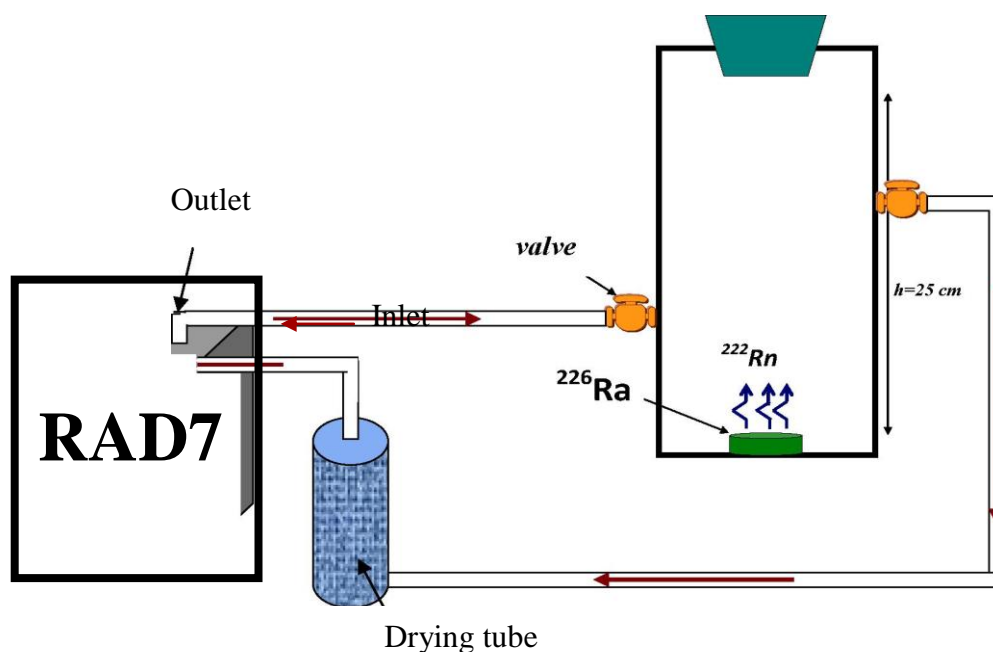
**Fig.(2): Experimental set-up of the Active Method for Measuring the Radon Concentration using the RAD7 radon monitor.
(RAD 7 Manual)**

During the etching process, the solution has to be stirred constantly. Detectors were then washed with distilled water and dried. After etching, the trials become tracks which were visible by optical microscope. The etched tracks were counted using an optical microscope with nominal magnification of 400 X. the calculated track density was converted into radon concentrations in Bq/m^3 using the calibration factor obtained and adopted in references (Al-Kofahi *et al*.,1992 and Hassan,1996). The overall uncertainly in the counting system has been estimated to be 5%.

The radon activity concentrations obtained in the detector of different cups were measured quantitatively for each dish type the mean values are calculated by averaging over the five depended samples. The radon exhalation rates and radium contents of the samples were calculated from the relevant formulae.

Calculations

If we consider only cases in which the gas is well mixed throughout the volume, the Activity of Radon concentration is given by (Hafez, 2001):
$$C_{Rn} = \rho / T\, K_i \qquad \qquad \dots(1)$$

where T is the exposure time, K_i is the calibration factor with the dimension of length or equivalent to (tracks.m^{-2}.d^{-1} per Bq.m^{-3}). ρ is a Track Density and is given by:

$$\rho = \text{Track/ Area} \qquad \qquad \dots(2)$$

1- Radon Exhalation Rate:

At the equilibrium state, the final activity of radon exhalated (A_t)from each sample inside the can is given by (Ching-Jiang *et al.*, 1993):

$$A_t = A_o (1-e^{-\lambda t}) \qquad \qquad \dots(3)$$

Where λ is the decay constant of the radon, t is exposure time and A_o is the final value of the activity concentration.

The radon exhalation rate per unit energy (E_A) and per unit mass (E_M) of the studied samples are calculated using the following relations, respectively [Surinder *et al.*, 2006, Barooah *et al.*, 2009 and Joga *et al.*, 2009):

$$E_A = \frac{\rho V \lambda}{KAT_e} \qquad \qquad \dots(4)$$

$$E_M = \frac{\rho V \lambda}{KMT_e} \qquad \qquad \dots(5)$$

Where ρ is the track density in (track/cm^2),

V is the effective volume of the chamber $= 1000 \text{cm}^3$,

K is the detector efficiency$=(0.27 \pm 0.02 \text{ Track. cm}^{-2}/ \text{ Bq . m}^{-3})$,

A is the surface area of the sample $=0.01539 \text{ cm}^2$

M is the mass of the sample materials$= 120$ g

T_e is the effective exposure time $= 1594.7$ h, which is given by

$$T_e = T - 1/\lambda(1-e^{-\lambda t}) \qquad \qquad \dots(6)$$

2- Effective Radium Content:

The radium content of the sample with reach equilibrium with the released radon inside the close can , and the effective radium content could be calculated from the formula (Hafez, *et al.*, 2001):

$$C_{Ra} = \frac{\rho h A}{KMT_e} \qquad \qquad \dots(7)$$

Where ρ is the track density recorded in the upper CR-39 plastic track detector, and h is the distance between the CR-39 detector and the top of sample surface,

Results and Discussions

The radon concentration data were obtained from 20 dosimeters collected after approximately two months and the track densities within the detectors are estimated using eq.(2), the Radon concentration are calculated by the passive (using eq.1) and active method (using RAD 7 detector), and the effective Radium contents are evaluated using eq.(7) as presented in Table (1).

Table (1): Radon Concentration and Effective Radium Contents of the Studied Samples.

Sample No.	Sample Code	Radon Concentration (Bq/m^3)		Effective Radium content (Bq/Kg)
		Passive	Active	
1	E(A)	28.6 ± 1.573	25.779±1.279	0.258±0.013
2	G(A)	27.9±1.535	28.655±1.87	0.234±0.012
3	Irn(A)	19.05±1.048	19.485±1.00226	0.181± 0.009
4	CH(A)	9.267± 0.509	9.012±0.6101	0.106±0.005

E(A) : England origin samples (Average)
G(A) : Germany origin samples (Average)
Irn(A) : Iran origin samples (Average)
CH(A) : China origin samples (Average)

For each type of the pottery dishes the mean value has been adopted by averaging over the five assessed samples. The depended calibration factor used in the calculations of present work was 0.27 ± 0.02 tracks. $cm^{-2}.d^{-1}(Bq. M^{-3})^{-1}$.

The radon exhalation rate (Area and Mass) for each type of pottery dishes have been calculated using eqs.(4 and 5, respectively), and presented in Table (2).

Table (2): Radon Exhalation Rate (in terms of Mass and Area) from the Studied Samples.

Sample No.	Sample Code	Effective Radium content (Bq/Kg)	Radon exhalation Rate	
			Mass (mBq.Kg^{-1}.h^{-1})	Area (mBq.m^{-2}.h^{-1})
1	E(A)	0.258±0.013	2.263±0.125	0.690±0.035
2	G(A)	0.234±0.012	2.075±0.114	0.636±0.032
3	Irn(A)	0.181± 0.009	1.606±0.088	0.403±0.020
4	CH(A)	0.106±0.005	0.802±0.044	0.196±0.009

Throughout the results, it has been observed that samples originated from England reveal higher values of radon concentration (which may ascribed to the higher Radium content of the original soil from which the potteries are made), radium content, and radon exhalation rates than the other samples, while the China samples record a lowest value; these variations altogether with the results of other samples have been shown in Figures (3) through (5). Radon exhalation rates (in terms of mass and area) and radon concentrations are always inherent to the radium content of the

studied samples; this relation has been approved through the plot shown in Figures (6) and (7).

The overall variations noticed within the obtained results may ascribed to the soil and mud from which the pottery dishes have been made, Comparing the resulted radon concentration with the standard values (ICRP,1993) indicates the consistency within the safe level, but it should be mentioned that the existence of the pottery dishes which constitutes an important requirement for every house in our region and every where will contribute by a significant amount in the indoor doses.

Fig. (3): Effective radium Content within the studied Samples

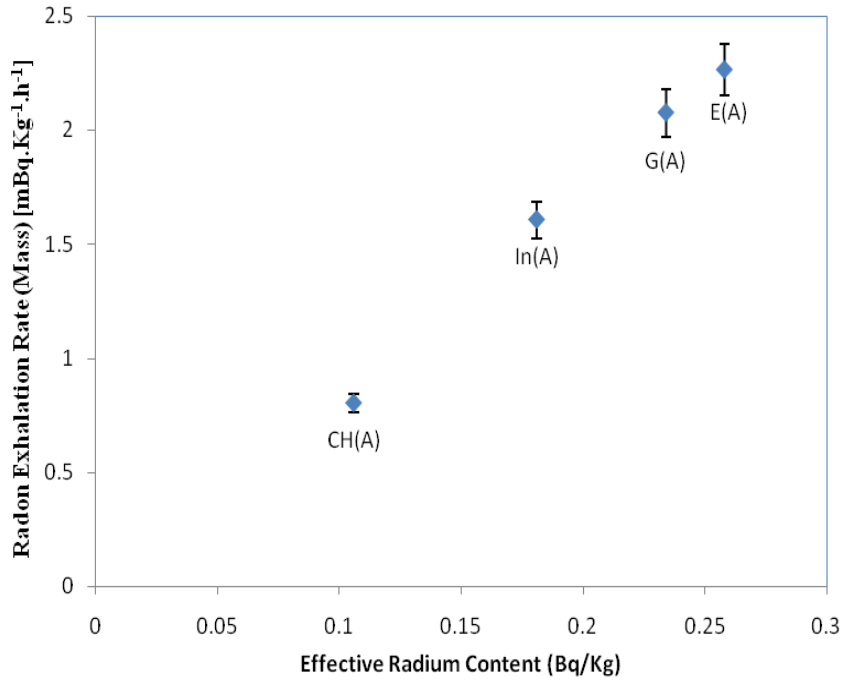**Fig. (5): Radon Exhalation Rate (Area) from the Studied Samples**
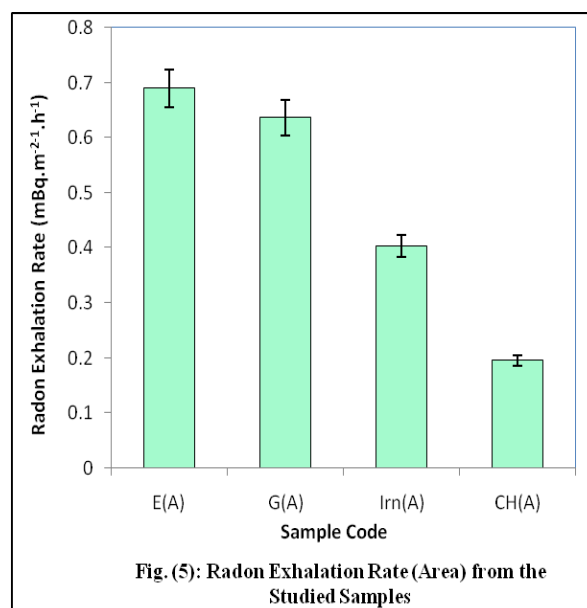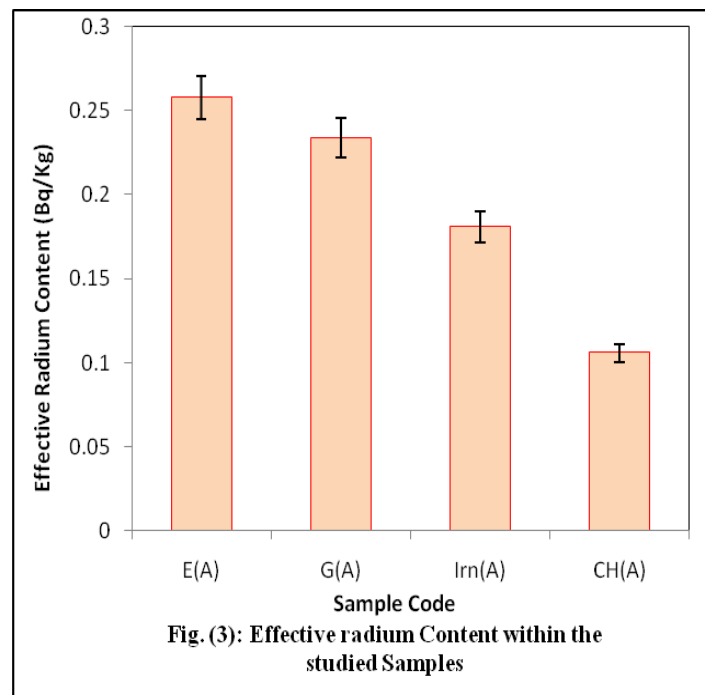
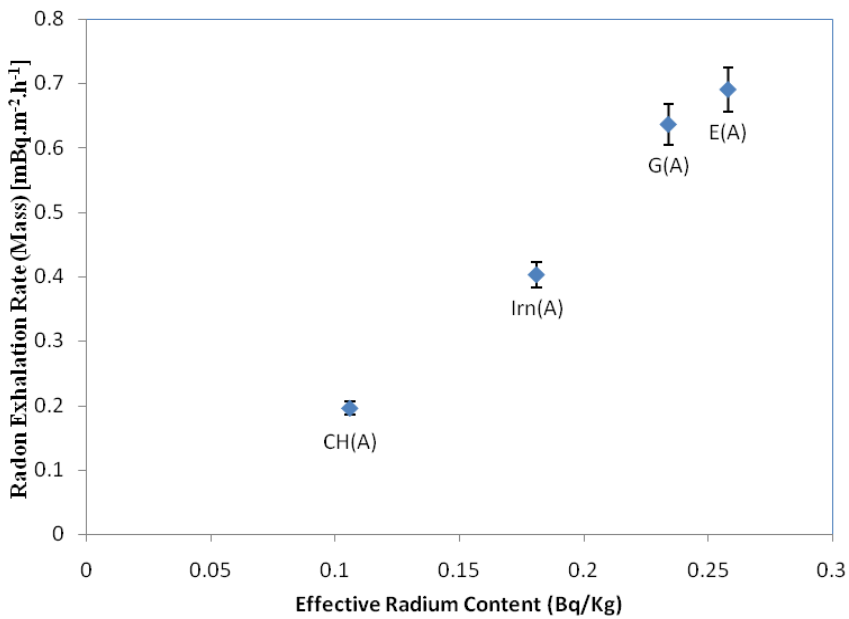Figure (6): Radon Exhalation Rate (Mass) against Radium Content of the Studied Samples.

Figure (7): Radon Exhalation Rate (Area) against Radium Content of the Studied Samples.

Conclusions

The results show that the pottery meal dishes originate from England has greater radium content and radon exhalation rate than the other samples while the China samples reveal the lowest values. However, the study reveals the fact that the pottery meal dishes possess a measurable amount of radioactivity that should be taken into account in the estimation of the indoor doses.

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قياس معدل تحرر غاز الرادون من أواني الطعام الفخارية المستعملة في مدينة أربيل باستخدام طريقتي طويل الامد والسريعة

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الخلاصة

استخدمت في هذه الدراسة كاشف الحالة الصلبة SSNTD من نوع CR-35 كطريقة طويلة الأمـد لحساب تركيز فعالية غاز الرادون، نسبة تحرر الرادون (كدالة للمساحة و الكتلة)، و محتوى عنصر الراديــوم الفعال في نماذج صحون الطعام الفخارية من المناشئ المختلفة (ألماني، أنكليزي، صيني و أيراني) والمستعملة في مدينة أربيل. تم استخدام تقنية كاشف الحالة الصلبة RAD7 كطريقة سريعة لحساب تركيز فعالية الـرادون بقياس كثافة أثر جسيمات ألفا. أظهرت النتائج أن أعلى نسبة تحرر غاز الرادون و محتوى عنصـر الراديــوم سجلت في النموذج الأنكليـزي 0.69 ± 0.035 mBq.m^{-2}.h^{-1} و 0.258 ± 0.013Bq.Kg^{-1} كدالـة للمسـاحة والكتلة على التوالي، بينما كانت أقل نسبة لتحرر غاز الرادون و محتوى عنصر الراديوم سجلت في النمــوذج الصيني 0.196 ± 0.0098 mBq.m^{-2}.h^{-1} و 0.106 ± 0.005 Bq.Kg^{-1} على التوالي.

أظهرت النتائج المستحصلة أن معدل تحرر غاز الرادون من النماذج المدروسة يجب ان تؤخذ فــي الحســبان ضمن قياس كميات الجرع الداخلية.