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Investigation of radon and its progeny in ceramic cooking dishes

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► ***Original article***

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ABSTRACT

Background: Recently, the concentration of radioactivity in the ceramic cooking dishes is found to exceed the reference level values. Therefore, there is a need to measure the radon concentration in ceramic cooking dishes, especially radon gas inhalation which is considered to be the second largest cause of lung cancer. ***Materials and Methods:*** In this work, a collection of 19 different kinds of ceramic cooking dishes is measured using CR-39 SSNTD (Solid State Nuclear Track Detector). ***Results:*** The obtained results showed that the highest value of radon concentration is found to be 23.07 ± 1.44 Bq/m^3, 148.41± 5.99 Bq/m^3 and 783.03± 35.96 Bq/m^{3}for high quality, low quality and handmade ceramic cooking dishes, respectively. Indicating that the radon concentration for 18.19% of handmade ceramic cooking dishes is exceeded the internationally accepted levels 600 Bq/m^3. ***Conclusions:*** All values of radon concentration for handmade ceramic cooking dishes are found to be higher than the measured values for high quality and low-quality manufactured ceramic cooking dishes. In addition, the mean values of annual effective doses of handmade ceramic cooking dishes are found to be varied from 2.20 to 22.47mSv/y, exceeding the internationally accepted action level from ICRP 2007 agency [1]. Hence, this study recommends that care must be taken when using the handmade ceramic cooking dishes.

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INTRODUCTION

Radon isotope Rn-22 is a naturally radioactive gas and is the main cause for lung cancer [2, 3]. Therefore, international organizations have established guidelines for occupational safety from radon protection and have developed international regulations for determining radon doses [1, 4-6]. Ceramics are naturally radioactive materials due to the rocks and soils from which they are made always containing natural radio nuclides of the Uranium (^{238}U), and Thorium (^{232}Th) series [7]. The radon exhalation rates for all ceramic tiles companies in the vicinity of ceramic surface were showed to be from 2.0 to 4.8 Bq.kg^{-1}.h^{-1} and the assessment of radium activity for ceramic tiles was found to be from 16-38 Bq.kg^{-1} for glaze surface to 23-64 Bq.kg^{-1} for clay surface [8]. Also, the average equivalent dose in contact to the ceramic surface was found to be 22 mSv.y^{-1} while the exposure in working level in the vicinity of ceramic tile was found in the range 2.4-3.8 WL [8]. Hence, radon is a health hazard [9-11], especially the radon concentration in soil goes widest from 10,000 to 50,000 Bq/m^3 [12].

Recently, ceramic materials have been used in the kitchen cooking dishes for many years. Ceramic materials, are made from ground materials and the possibility of having radioactive nuclei is imperative. So, the goal of this work is to determine the concentration of radon and radioactive dose in the low quality, high quality, and handmade ceramic cooking dishes using in kitchen. Previous research has studied the natural radioactivity level of ceramic cooking dishes in Saudi Arabia [13] without considering the handmade ceramic cooking dishes. Many studies of radon [14-23] have been investigated. However the result of this study has showed that high values for radon concentration of handmade ceramic cooking dishes, exceeding the internationally accepted action level from ICRP 2007 agency. Therefore, there are still a number of gaps in investigating the radon concentration of handmade ceramic cooking dishes and would benefit from further research.

MATERIAL AND METHODS

The samples

In the present work, a collection of 19 household ceramic cooking dishes samples is collected and divided into three types. The first type is called high quality manufactured household ceramic cooking dishes and is manufactured from a known company.

i.e. the dishes are inspected and are tested for safety from any toxins by manufacturers. The second type is called low quality manufactured household ceramic cooking dishes and is manufactured from unknown company. i.e. the dishes are not inspected and are not tested for safety from any toxins. The third type is called handmade ceramic cooking dishes and is made by hand not manufactured, see table 1. The samples of dishes are prepared by crushing and grinding by an electric grinder (Grinding Machines Gristmill Home).

The measurement by CAN

The CR-39 detector TASTRAK type, (Track Analysis System, Ltd., UK) is high sensitivity, optical transparency uniformity. The CR-39 detectors of size 1.5 × 1.5 cm^2 for detecting the radon gas are fixed to the shelf on a cup of 8 cm height (to ensure that the alpha particles do not reach the detector) where the samples occupied 200 ml, see figure 1.The cups are completely sealed and left for a period of 30 days. At the end of the 30 days period, the CR-39 detectors are collected and etched in a Sodium Hydroxide solution 6.25N (NaOH), and maintained at a temperature of 70°C for 6 hours in a water bath. After that the etched detectors have washed under water flow and then washed again under distilled water. To count the track densities (track/cm) on CR-39 detector, an optical microscope (Nikon microscope Eclipse LV100DA-U – Japan) is used [24-25].

Table 1. Sample codes and countries of origin.

Sample Code	Sample type	Sample Code	Sample type	Country
S1	Manufactured (High Quality)	H1	Handmade Ceramic	KSA (Abu al-Qayyed)
S2		H2		Yemen (Sa'da)
S3		H3		Yemen (Hajjah)
		H4		Pakistan
S4	Manufactured (Low Quality)	H5		China 1
S5		H6		China 2
		H7		KSA (Al-Darb)
S6		H8		Egypt 1
S7		H9		Egypt 2
S8		H10		Turkey
		H11		Indonesia

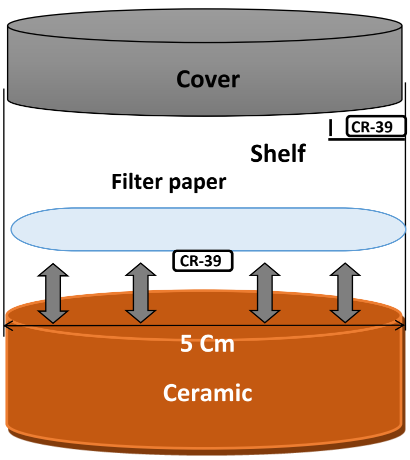

Figure 1. The CAN device.

Theoretical techniques

The equilibrium concentration of radon (C_{eq}) (Bq/m^3) is (10,26), determined by using track density according to the equation 1

$$C_{eq} = \rho / K t_{ef} \quad (1)$$

where ρ is track density, t_{ef} is the effective exposure time in hourswhich calculate from the relation $t_e = t - \frac{1-e^{-\lambda t}}{\lambda}$ where t is the exposure time and λ is the constant decay of radonand the calibration factor K of the SSNTDs which determined by Rn-chamber K = 0.045987 tracks cm^{-2} d^{-1}/Bqm^{-3}.

The surface exhalation rate (E_a) ($Bqm^{-1}\cdot h^{-1}$) for the radon [27-28] and the mass exhalation rate (E_m) ($Bqkg^{-1}\cdot h^{-1}$) are determined by using the relations 2&3 respectively.

$$Ea = C_{eq} V l / A \quad (2)$$

Where A is the cross section area of cup (m^2), V is the effective volume of the cup in m^3, and λ the decay constant for radon in h^{-1}

$$E_m = C_{eq} V l / M \quad (3)$$

Where M is the mass of sample (kg). The relationship 4 is used to determine the radium activity (R_{ac}) (Bq/Kg)

$$R_{ac} = E_m / \lambda \quad (4)$$

Finally the annual effective dose AED (mSv/y) was calculated from equation 5.

$$AED = C \times D \times F \times T \quad (mSv/y) \quad (5)$$

where, C in B /qm^3 is the measured mean radon activity concentration in air, F is the indoor equilibrium factor between radon and its progeny (F=0.4), T is time (T=7000 hy^{-1}) and D is the dose conversion factor (D=9 nSv/h per Bq/m^3) [9,29].

Statistical software

In this paper, the density of radon was measured using a high-efficiency optical microscope as mention above. Then, 50 fields are measured for each sample to ensure that all sample points are scanned. Afterwards, the average and the standard deviation were calculated using the excel program. However, equations from 1 to 5 are used to calculate the concentration of radon and radium, their absorbed dose and the radon exhalation. The average and standard deviation are calculated using Excel program. The level of considered significances is set to be 0.05.

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RESULTS

Measurement of radon concentration

In the present work, the measured radon concentrations of manufactured and handmade ceramic cooking dishes samples are shown in figures (2-3). From figure 2, the concentrations of radon in high quality manufactured ceramic cooking dishes samples are varied from 10.63±1.39 Bq/m³ to 23.07 ± 1.44 Bq/m³ with an average value of 16.27± 1.26Bq/m³. These values are lower than the obtained radon concentrations of low quality manufactured ceramic cooking dishes samples which are found to be varied from 94.77± 1.99 Bq/m³to 148.41± 5.99Bq/m³ with an average value of 116.34± 2.24 Bq/m³.

Also, the radon concentrations for handmade ceramic cooking dishes samples are determined by using a CR39 detector, and the results are shown in figure 3. The highest average concentration of radon was found in Hajjah from Yemen (H3) with value 783.03± 35.96 Bq/m³. This value is the average for eight samples which were taking from Hajjah. The lowest average concentration was found in sample from Indonesia (H11) with value 129.19± 9.69Bq/m³. Hence, one can notice that all values are found to be higher than the measured values for high quality and low quality manufactured ceramic cooking dishes samples.

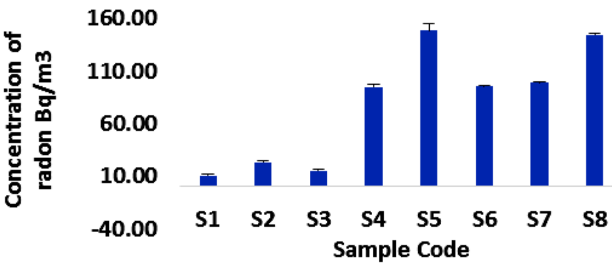

Figure 2. The radon concentration for high quality (S1-S3) and low quality (S4-S8) manufactured ceramic cooking dishes samples.

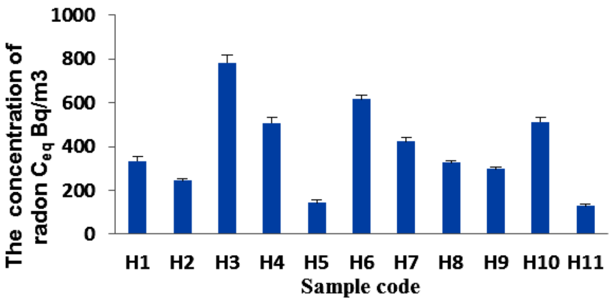

Figure 3. The radon concentration of handmade ceramic cooking dishes samples.

The exhalation rate and radium activity

The surface exhalation rates (E_a) and mass exhalation rate (E_m) of radon for all samples are calculated and reported as shown in table 2. The highest values of the radon exhalation rates and mass exhalation rate are for low quality manufacture ceramic cooking dishes samples and are found to be E_a = 3.03±0.12 Bqm^{-2}h^{-1} and E_m = 0.19 ±0.01 BqKg^{-1}h^{-1} with mean value E_a =2.38±0.05 Bqm^{-2}h^{-1} and E_m=0.15±0.003 BqKg^{-1}h^{-1}, respectively, see table 2. The radon exhalation rates for handmade ceramic cooking dishes samples are also calculated and are reported in table 2. The radon exhalation rates and mass exhalation rates are found to be varied from 23.95±3.28Bqm^{-2}.h^{-1} and 1.20±0.16 BqKg^{-1} h^{-1} to 2.34±1.48 Bqm^{-2}.h^{-1} and 0.12±0.07 BqKg^{-1} h^{-1} with a mean value 8.00±1.19 Bqm^{-2}.h^{-1} and 0.40±0.06 BqKg^{-1}h^{-1}, respectively. According to the results in table 2, the highest values for radium content are found to be 0.31±0.02Bq/Kg,1.31±0.05 Bq/Kg and 6.80±1.81 Bq/Kg for high quality, low quality and handmade ceramic cooking dishes, respectively.

Figure 4, shows the annual effective dose of radon in manufactured cooking dishes samples. The annual effective dose of radon for high quality manufactured ceramic cooking dishes samples has mean value of 0.29± 0.02mSv/y while for the low-quality manufactured ceramic cooking dishes samples have the mean value of 2.09± 0.04mSv/y. Also, the radon annual effective dose for handmade ceramic cooking dishes samples are calculated and shown in figure 5. The radon annual effective dose are found to be yielded from 13.31±3.70mSv/y for sample H3 to 2.20±1.00mSv/y for sample H11 with mean value of 7.50 ± 1.81mSv/y.

Table 2. The radon exhalation rate and radium activity for manufactured and handmade ceramic cooking dishes samples.

Samples	Exhalation rate		Radium activity
	E_a/ (Bqm^{-2}·h^{-1})		Ra/(Bq /Kg)
S1	0.32 ±0.04	0.02±0.003	0.14±0.02
S2	0.70 ±0.04	0.04±0.003	0.31±0.02
S3	0.46±0.02	0.03±0.002	0.20±0.01
S4	1.94±0.04	0.12±0.003	0.84±0.02
S5	3.03±0.12	0.19±0.008	1.31±0.05
S6	1.90±0.03	0.12±0.002	0.84±0.01
S7	1.98±0.02	0.13±0.001	0.87±0.01
S8	2.97±0.02	0.19±0.001	1.27±0.01
H1	6.04±0.28	0.30±0.01	1.49±0.98
H2	4.43±1.29	0.22±0.06	1.26±0.54
H3	23.95±3.28	1.20±0.16	6.80±1.81
H4	9.18±1.25	0.46±0.06	2.61±1.25
H5	2.59±1.20	0.13±0.06	0.74±0.56
H6	11.17±0.02	0.56±0.01	3.17±0.89
H7	7.65±0.14	0.38±0.01	2.17±0.94
H8	5.91±1.34	0.30±0.07	1.68±0.53
H9	5.40±1.48	0.27±0.07	1.53±0.49
H10	9.29±1.29	0.46±0.06	2.64±1.26
H11	2.34±1.48	0.12±0.07	0.66±0.49

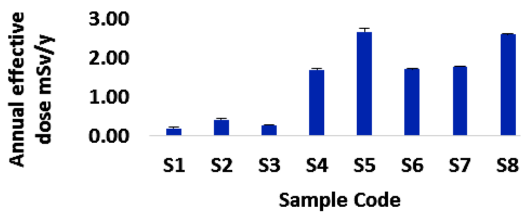

Figure 4. The radon annual effective dose for high quality (S1-S3) and low quality (S4-S8) manufactured ceramic cooking dishes samples.

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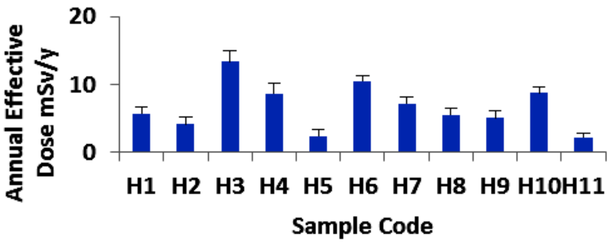

Figure 5. The radon annual effective dose for handmade ceramic cooking dishes samples.

DISCUSSION

The obtained results showed that the values of radon concentrations measured for samples of high-quality ceramic cooking dishes were lower than radon concentrations that were obtained for samples of low-quality industrial ceramic cooking dishes. These results indicate that all the values obtained are lower than the values reported by ICRP 2007 and UNSCEAR, 2000, 2008 [1,6,9].

Based on the results of the radon concentrations for handmade ceramic cooking dishes, the sample with the highest average concentration of radon was found in Hajjah city in Yemen. Hajjah city is close to a mountainous region, and it is well known for making handmade ceramic cooking dishes. Hence, the nature of Hajjah city indicates that the presence of radioactive nuclei in the soil areas. In addition, Hajjah city is also close to weapons and explosions places. Detonation may upturn the soil and increase the proportions of radioactive cores. About 81.81% of handmade ceramic samples have radon concentrations in the range of allowed level [1,9]. However, 18.19 % of samples have radon concentrations higher than the allowed level.

As shown in table 2, the results obtained for surface exhalation rates (E_a) and mass exhalation rate (E_m) for all radon samples indicate that radon exhalation rates differ significantly from one sample to another. The value of exhalation rates depends on the ratios of the radioactive nuclei in the sample, which in turn vary according to the locations where the samples are collected, i.e. the environment to which the ceramic components belong.

According to the observed results of the radium values of low-quality, high-quality hand-made cooking dishes, it was found that all the radium content values were in agreement with the previously reported values for soil [30] and within the permissible limits of 370 Bq/kg as reported by OECD, 1979 [31]. Figure 4 shows that the annual effective dose for all samples of ceramic cooking dishes is within the recommended safe limits according to the range (10 mSv/year), [1]. While the results shown in Figure 5 show that the effective dose of radon samples of handmade ceramic cooking dishes was mostly within the limits established by the International Committee for Radiation Protection [1], from 3 to 10 mSv/year. It was also found that the total annual effective doses of all manufactured ceramics and 81.81% of the samples of handmade ceramic cooking plates were within the limits of ICRP recommendation. However, only 18.19% of ceramic handmade cooking plate samples exceeded the recommended safe limit for ICRP.

CONCLUSIONS

The radon gas concentrations in 18.19% handmade ceramic cooking dishes samples were found to be higher than the allowed limit from (ICRP) agency (600 Bq/m^3) while 81.81% of the samples of handmade ceramic cooking plates were within the limits of ICRP recommendation. The annual effective dose in the handmade samples were higher than the allowable limits (10 mSv/y) recommended. Consequently, the use of handmade ceramic cooking dishes should be avoided and we recommend a more detailed study of handmade ceramic cooking dishes has to be investigated.

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Ethical considerations: Research does not involve human participants or animals (This paper is an analysis of radon and its progeny in ceramic cooking dishes analysis) Informed consent: none

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Conflicts of interests: Declared none.

Authors contribution: Authors have studied the radon gas concentrations in ceramic cooking, especially handmade ceramic cooking dishes and they concluded that the use of handmade ceramic cooking dishes should be avoided and more detailed study of handmade ceramic cooking dishes has to be investigated.

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