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Measurement of Radium and Radon Exhalation Rate in Marble Samples used in Al-Bayda City Market-Libya

Jemila Mussa Ali*, Suhayla Abdelhady Ibrahim

Department of Physics, Faculty Of Science Omar AL-Mukhtar University, Al-Bayda, Libya

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Abstract: The aim of the present study is to measure the activity concentrations of ^{226}Ra, ^{222}Rn, the mass exhalation rate of ^{222}Rn, and the annual effective dose of radon in marble samples collected from Al-Bayda city local market –Libya. Samples were measured by using a low-background NaI (Tl) detector. The average activity concentrations of ^{226}Ra and ^{222}Rn were 72.57 Bq.kg^{-1} and 597.85 Bq.m^{-3}.The radon exhalation rate in marble samples vary from 0.05-0.30 Bq.kg^{-1}.S^{-1} with an average of 0.13 Bq.kg^{-1}.S^{-1}. The annual effective dose of radon was calculated in samples under investigation. For most samples, the values were lower than the maximum permissible dose limits. It can be concluded that marble samples under investigation do not pose any radiological hazard to the dwellers of buildings used in their construction.

Keywords: Marble, NaI (Tl) Detector, Annual Effective Dose of Radon, Radon Exhalation Rate.

INTRODUCTION

The human body is naturally exposed to ionizing radiation, which can be found in soils, rocks, and water (Abo-Elmagd, 2014). In addition, artificial radiation was added to this background radiation. The background radiation arises from natural sources present in natural ores, such as some building materials. This radiation is due to primordial radionuclides of the natural radioactive series of Thorium-232 (^{232}Th) and uranium-238 (^{238}U) series and their decay products(Ghose et al., 2012). These radionuclides are widely distributed and their concentrations depend on the geological conditions. Therefore, it is important to measure the natural activity of all building materials. This step will help to assess the possible radiological risks to human health (Kumara et al., 2018). This radioactive isotope results from the disintegration of radium-226 (^{226}Ra), a decay product of the ^{238}U series and responsible for the largest source of natural radiation to which the population is exposed (Kama et al., 2011). Radon and Thoron are both generated from radium decay in the solid grains, then migrated to a significant distance from the site of generation in rock, soil, and building materials into the atmosphere (exhalation) before undergoing radioactive decay (Bala et al., 2017). Radon-222 (^{222}Rn) concentration can reach high levels in buildings depending on exhalation from the building material used, such as concrete, marble, or granite. Marble is one of the metamorphic rocks found to occur on the earth's surface. The colors of the marble depend on the mineral composition and metamorphism. Marbles are used commonly as floor laying material (Kaiser et al., 1999).

***Corresponding author:** Jemila Mussa Ali: *Jemila.mussa@omu.edu.ly* , Department of Physics, Faculty Of Science Omar AL-Mukhtar University, Al-Bayda, Libya

In this study, the gamma radiation has been measured in marble samples collected from the Al-Bayda local market in Libya to obtain an activity concentration of ^{226}Ra, radon exhalation rate, and the annual effective dose of radon. Because of health risks caused by exposure to indoor radiation, many international organizations, such as the International Commission on Radiological Protection (ICRP,. 1993), the World Health Organization (WHO,. 2021), and UNSCEAR, have adopted strong measures in order to reduce such exposure.

MATERIALS AND METHODS

Samples Collection and Preparation: Nine marble samples were collected from the local market in Al-Bayda city – Libya, to measure the radioactivity concentrations of ^{226}Ra and ^{222}Rn (Marble available in Libya, some local and some imported.). All samples were brought to the laboratory and properly cataloged, washed, and dried (at 110C^{o} for 2h) for complete removal of moisture. Then, all samples were crushed to a fine powder with a particle size ≈1 mm (This process took place in the laboratory of the Faculty of Engineering- Omer Al-Mukhtar University, Al-Bayda, Libya). Samples were packed and sealed in radon impermeable airtight cylindrical plastic containers, then stored for four weeks before counting to ensure ^{226}Ra and its short-lived daughters reached secular equilibrium (Sroor, 2013). Table (1) shows the description of the samples.

Gamma-Ray Detection System: A gamma-ray NaI(Tl) scintillation detector contains a 3"×3" crystal, with a multichannel analyzer (MCA) used for the spectral measurements of naturally occurring radionuclides. The detector was placed in the center of a two-layered shield made from stainless steel of 10 mm thickness, and lead of 30 mm thickness. The shield must be used to reduce the radioactive background, as well as the detector from unwanted background radiation, and reduce the contribution of scattered radiation. After that, the sample was placed on a detector for 7200 S. The spectra were analyzed using a software program. The samples were prepared and measured in the Advanced Nuclear Lab-Department of Physics-Faculty of Science - Omer Al-Mukhtar University, Al-Bayda, Libya.

Table (1). Description of marble samples.

Samples code	Trade Name	Country of manufacture
M1	Brak El-Shati	(made in Libya)
M2	Classic Paradiso	(made in India)
M3	Galaxy	(made in India)
M4	Monty Clad	(made in India)
M5	Ten Brown	(made in India)
M6	Oriental Cream	(made in Oman)
M7	Self-Marble	(made in Egypt)
M8	Miral Lacquer Marble	(made in Turkey)
M9	Mirwan Marble	(made in Spain)

Activity Concentration: The activity concentration (A) of a radionuclide for a peak at energy, is given by the relation (Al-Sewaidan, 2019):

$$A = \frac{N}{\varepsilon\, I_{\gamma}\, t\, m} \qquad (1)$$

Where: ε is the absolute efficiency at photopeak energy, t is the time of the sample spectrum collection in seconds, $I_{\gamma}$is the intensity of emitted gamma-ray (gamma abundance), m is the mass of the sample in (kg), N is the number of count in a given peak area corrected for background peaks of a peak at energy.

Radon Mass Exhalation Rate: The emanation rate coefficient and factor of ^{222}Rn that can diffuse through the raw and building materials is known as the emanation coefficient. The emanation coefficient (CRn) is a very important radiological index that can be used to determine the amount of the ^{222}Rn emanated fraction released from building raw materials and products containing naturally occurring radionuclides such as ^{226}Ra in radioactivity equilibrium with parents. The emanation rate is estimated by measuring gamma-rays from the radon decay daughter products, ^{214}Pb and ^{214}Bi. Assuming an equilibrium state:

$$C_{Rn} = (A_{Ra} - A_D) \times \rho \qquad (2)$$

Where A_{Ra} is the measured activity of ^{226}Ra, A_D is the measured activity of daughter element ^{214}Pb (or ^{214}Bi), which escapes into the surrounding environment. ρ is the density of radon (9.73 $kg.m^{-3}$). The introduction of the radon emanation factor F, which is defined as:

$$F_{Rn} = \frac{A_{Ra} - A_D}{A_{Ra}} \qquad (3)$$

The radon exhalation rate E_{Rn} ($Bq.kg^{-1}.S^{-1}$) is the product of the emanation factor, and the ^{222}Rn production rate was determined by used relation (Turhan & Gündüz, 2008):

$$E_{Rn} = F_{Rn} \cdot A_{Ra} \cdot \lambda_{Rn} \qquad (4)$$

Where: $\boldsymbol{\lambda_{Rn}}$ is the decay constant of ^{222}Rn (2.1×10^{-6} S^{-1}).

Annual Effective Dose of Radon: Radon concentration was converted into an effective dose, as the long-standing exposure to a high concentration of radon, and its progenies, may lead to pathological effects like lung cancer. The annual effective dose, received by workers and residents due to inhalation of radon gas and its decay products, where calculated by relation (Abd El-Halim, 2019):

$$AED_{Rn} = \frac{C_{Rn} \times 0.4 \times K \times H}{3700\ Bq.m^{-3} \times 170h} \qquad (5)$$

Where AED_{Rn} is the annual effective dose ($mSv.y^{-1}$), C_{Rn} is the emanation coefficient of radon ($Bq.m^{-3}$), K is the ICRP dose conversion factor (5 mSv $WL.M^{-1}$ for occupational workers, and 3.88 $mSvWLM^{-1}$ (effective dose per unit Work Limit in Month) for the general public), H is the annual occupancy at the location, 2160 h for workers and 7000 h for residents (80% of total time) and 170 is exposure hours taken for $WL.M^{-1}$ (ICRP, 1993).

RESULTS

The present values in Table (2) show, the activity concentration of ^{226}Ra, ^{222}Rn, radon emanation factor F_{Rn}, Radon mass exhalation rate E_{Rn}, and the annual effective dose from radon (AED_{Rn}). The activity concentrations of ^{226}Ra ranged between (26.03-148.58) $Bq.kg^{-1}$, and the average value was 72.57 $Bq.kg^{-1}$. For ^{222}Rn, the activity concentrations varied between (240.33 – 1392.85) $Bq.m^{-3}$, with an average 597.85 $Bq.m^{-3}$, as shown in Figure (1).

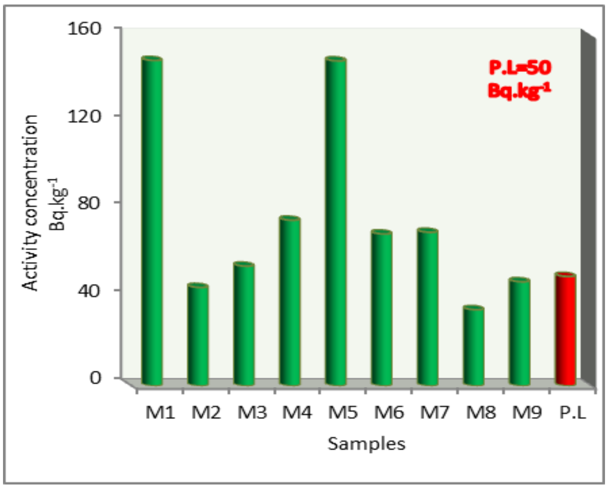

Figure (1). The activity concentrations of ^{226}Ra in marble samples (P.L: Permissible Level).

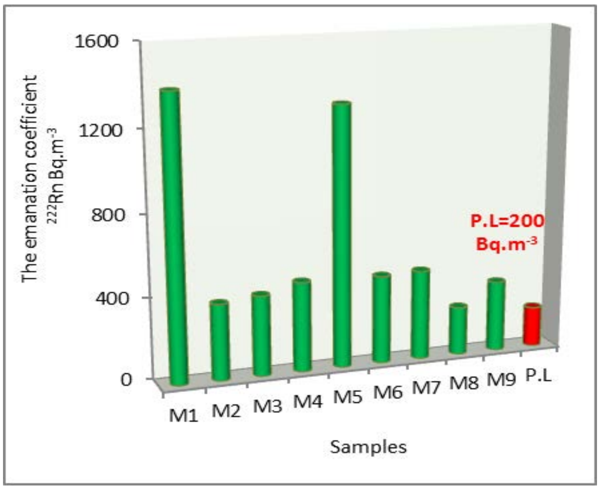

Figure (2). The emanation coefficient of ^{222}Rn $Bq.m^{-3}$ in marble samples (P.L: Permissible Level).

The results show that there is a variation in radon exhalation rate from one sample to another, depending on the geological formation of the region from which the sample is taken. The variation in values of radon exhalation rate may be due to the differences in radium content and porosity of the marble (Frutos-Puerto et al., 2020).

Table (2): The values of activity concentrations of ^{226}Ra $Bq.kg^{-1}$, emanation coefficient ^{222}Rn $Bq.m^{-3}$, E_{Rn} and AED_{Rn} in marble samples.

Samples Code	A_{Ra} $Bq.kg^{-1}$	C_{Rn} $Bq.m^{-3}$	Ern*10^{-3} $Bq.kg^{1}.S^{-1}$	AED_{Rn} $mSv.y^{-1}$	
				for worker	for residents
M1	148.58	1392.85	0.30	9.57	24.06
M2	26.03	379.76	0.08	2.61	6.56
M3	32.44	399.90	0.09	2.75	6.91
M4	75.69	446.60	0.10	3.07	7.71
M5	148.39	1286.89	0.28	8.84	22.23
M6	69.407	437.82	0.09	3.01	7.56
M7	70.17	445.05	0.10	3.06	7.69
M8	34.84	240.33	0.05	1.65	4.15
M9	47.60	351.45	0.08	2.41	6.07
Max	148.58	1392.85	0.30	9.57	24.06
Min	26.03	240.33	0.05	1.65	4.15
Average	72.57	597.85	0.13	4.11	10.33

The present values of the radon exhalation rate observed in the marble ranged between 0.05-0.30 $Bq.kg^{-1}.S^{-1}$ (Turkey -Libya) with a 0.13 $Bq.kg^{-1}.S^{-1}$ average, as shown in Figure (3).

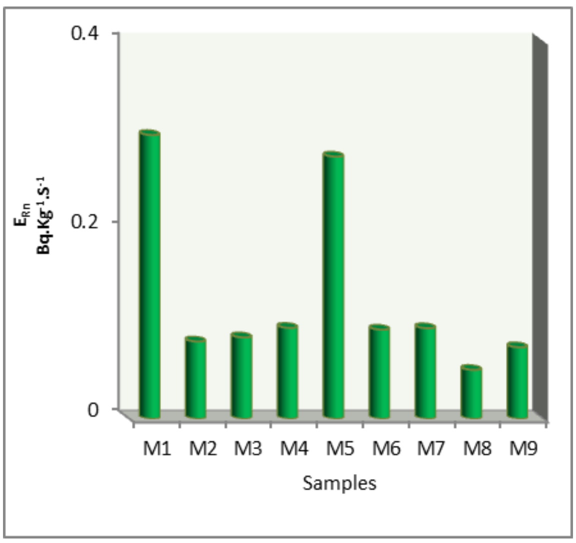

Figure (3): The mass exhalation of radon in marble samples.

From Figure (4), the variation of radon mass exhalation rate with ^{226}Ra activity concentrations shows a correlation between them. Therefore, it can be concluded that it is possible to predict the radon exhalation rate from the activity concentration of radium.

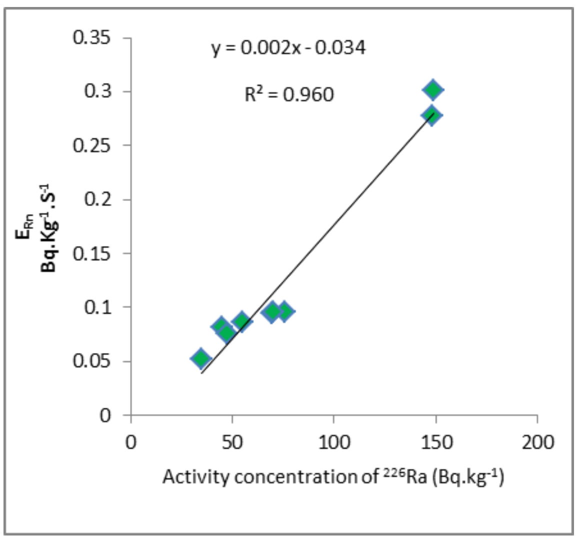

Figure (4): Correlation between ^{226}Ra activity concentration and radon exhalation rate in the marble samples.

The results indicate low levels of an annual effective dose from radon in marble samples for workers and residents, except samples M1 and M5 (made in Libya and India) for residents. This result is due to the increased exposure time for residents, as shown in Figure (5).

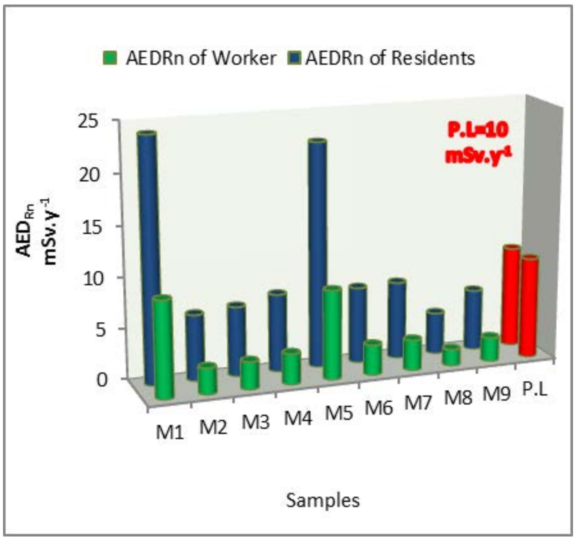

Figure (5): The annual effective dose of radon in marble samples (P.L: Permissible Level).

DISCUSSION

The results have shown that the activity concentration of ^{226}Ra for most samples is higher than the world value of 50 Bq.kg^{-1}(WHO, 2021), and for ^{222}Rn, the values are higher than the average permissible level of 200 Bq.m^{-3} (Jasaitis & Girgždys, 2007; Sharma et al., 2016). When comparing values of the radon exhalation rate between the different samples of marble, we found that the present value of the radon exhalation rate in sample M1 is higher than the values of other samples. The results indicate that low levels of an annual effective dose from radon in most marble samples were lower than the maximum permissible dose limits (10 mSv.y^{-1}) recommended by (ICRP,. 1993).

Table (3). Comparison values of radium concentration, radon exhalation rate and of marbles samples used in different countries.

Countries	Radium concentration Bq.kg^{1}	Radon exhalation rate × 10^{-3} Bq.kg^{1}.S^{-1}	References
Egypt	17.60-120.03	0.015-0.14	(Sroor, 2013)
India	31.90-44.60	53.75- 66.78	(Bala et al., 2017)
Morocco	18	1.66×10^{-3}	(Kassi et al., 2018)
Spin	4.9–40.7	(2.8-6)×10^{-3}	(Frutos-Puerto et al., 2020)
Presents work	26.03-148.58	0.05-0.30	

CONCLUSION

The obtained results showed that the average radium and radon concentration in investigation samples varies from 72.57 Bq.kg^{-1} and 597.85 Bq.m^{-3}. Also, the obtained value of radon mass exhalation rate varies between 0.05-0.30 Bq.kg^{-1}.S^{-1} (Turkey -Libya) with an average of 0.13 Bq.kg^{-1}.S^{-1}. It is recommended that the radon exhalation rate should be measured for all building materials and a standard code placed on all products. The annual effective dose of radon in marble samples for workers and residents is lower than the maximum permissible dose limit 10mSv.y^{-1} recommended by (ICRP, 1993), with the exception of samples M1 and M5 (made in Libya and India) for residents. The variation in obtained results depends on the geological formation of the region and the increased exposure time. The annual effective dose limit and the activity concentration index show that the investigated samples are within the recommended safety limit and do not pose any source of radiation hazard. Therefore, the use of these materials in the construction of dwellings is considered to be safe for inhabitants.

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قياس الراديوم ومعدل الزفير للرادون في عينات الرخام المستخدمة في سوق مدينة البيضاء – ليبيا

جميلة موسى علي *، سهيلة عبد الهادي إبراهيم

قسم الفيزياء، كلية العلوم، جامعة عمر المختار، البيضاء، ليبيا

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المستخلص : الهدف من هذه الدراسة هو قياس تراكيز نشاط الراديوم–226 والرادون–222، ومعدل الزفير للرادون–222، والجرعة الفعالة السنوية من غاز الرادون في عينات الرخام التي تم جمعها من السوق المحلي لمدينة البيضاء – ليبيا ، تم قياس العينات بواسطة باستخدام كاشف ايوديد الصوديوم المنشط بالثاليوم NaI (Tl) منخفض الخلفية. كان متوسط تركيزات النشاط للراديوم–226 والرادون– 222 72.57 بيكرل/ كجم و 597.85 بيكرل/ م3 ويتراوح معدل الزفير الشامل في عينات الرخام من 0.05– 0.30 بيكرل/ كجم. ث بمتوسط 0.13 بيكرل/ كجم. ث. تم حساب الجرعة الفعالة السنوية من الرادون في العينات قيد الدراسة، وكانت القيم بالنسبة لمعظم العينات أقل من الحد الأقصى للجرعات المسموح بها. يمكن الاستنتاج أن عينات الرخام في هذه الدراسة لا تشكل أي خطر إشعاعي على سكان المباني التي استخدم في تشيدها.

الكلمات المفتاحية : الرخام، كاشف ايوديد الصوديوم، الجرعة الفعالة السنوية للرادون، معدل الزفير للرادون.

[1]* جميلة موسى علي: *Jemila.mussa@omu.edu.ly*، قسم الفيزياء، كلية العلوم، جامعة عمر المختار، البيضاء، ليبيا