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Measurement of Radon (^{222}Rn) Concentrations in the Basaltic Rocks of Yarmouk River, Jordan

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Abstract

This article was conducted to measure radon concentration levels in the Yarmouk River Basalt (YRB), North-Jordan. The YRB is of Quaternary in age and occurred as a successive sheet with a total thickness of 122 meters. The Yarmouk River Basalt is classified into four major phases. Namely, Yarmouk Sheet basalt, Yarmouk blocky basalt, Yarmouk massive basalt and Yarmouk exfoliated. Furthermore, each major phase is subdivided into several flows. Sixteen samples are measured by using Nuclear Track Detector (Columbia Resin CR-39) in the laboratory. The results show that the concentration of radon is gradually decreased from the lower basalt first phase towards the upper fourth phase in considerable amounts. The values range from 12413.12 to 4137.71 Bq/m³, with an average value of 6635.74 Bq/m³. The results of this investigation indicate that the uranium isotopes element decay is the origin of radon emitted from the interstitial glass in the basalt rocks, rather than ^{222}Rn emission along with fractures or major faults.

Keywords: ^{222}Rn ; R-Model; Mineralogy; Fault; Basalt; River

1. Introduction

In the natural environment, there are three isotopes of radon occurring naturally as ^{222}Rn (radon), ^{220}Rn (thoron) and ^{219}Rn (actinon). These isotopes are direct decay products of radium isotopes. Radon is the most important isotope in environmental studies (NCRP, 1984; Alshahri, 2014). It has a half-life equal to 3.82 days which equals 91.68 hr. The second isotope is thoron ^{220}Rn , comes from the decay of thorium series ^{232}Th , and it has a half-life (55.6 s). While, the third isotope actinon ^{219}Rn is part of the decay chain of ^{235}U , a relatively low abundance in nature and has an extremely short half-life (3.96 s) (Muhassar, 2009). Radon can be considered to be of the most dangerous radioactive elements in the environment (Mansur et al., 2005; Al-qadi et al., 2016; Hamed, 2017). Risk increases as an individual is exposed to higher levels of radon over a longer period time (Al-Malabeh and Hamed, 2020; Al-Malabeh et al., 2008).

Basaltic, limestone and sandstone rocks, as well as granite rock, are used in building materials now. It can be said that the essential sources of radon in the outside locations are soil and water (Al-Malabeh and Hamed, 2020; Al-Malabeh et al., 2017). Nearly 80% of the pop-up radon gas to the outside media is produced in the earth's crust (crustal rocks). It diffusing through the soil depending on the amount of egress of granular soil material, and spreading through the pores of the soil into the outside

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media, in general the amount of emitted radon from rock is larger than that from the soil and minerals (Al-Malabeh et al. 2002). Moreover, high concentration of radon gas in the rock content leads to groundwater pollution during the recharge process to groundwater (Kullab et al. 2001; Ahmad et al, 2020). The aim of the current research is to measure the radon concentration in the YRB. It can be seen that no comprehensive study has been carried out on the basaltic rocks in Jordan until 2017 in general and in Yarmouk basalt in particular. According to the literature survey no measurements of ^{222}Rn were yet reported for basaltic rock world-wide which make this study as the first of its type globally.

2. Study Area

Yarmouk River basalt YRB is a part of the Harat Al-Shaam (Fig.2) and lies on both sides of the Yarmouk Valley. The age of the basaltic rock ranges from 2.6 million years to 100,000 years (Al-Malabeh, 2003). It is located 100 km north of Amman, Jordan. It can be approached from the capital via Jarash. The area is bounded by the longitudes $35^{\circ} 40' 43.12'' \text{ E}$ to $35^{\circ} 42' 19.34'' \text{ E}$ and the latitudes $32^{\circ} 40' 43.07'' \text{ N}$ to $32^{\circ} 43' 22.08'' \text{ N}$, more details of the coordination are given in (Figs. 1 and 3).

Fig. 1. Location map the study area (Araydah, 2019)

Basalt flows in the study area are underlain by Wadi Shallalah Chalk Formation (WSC), which is the main exposure formation in the area. The sampling locations divide according to the four phases of the Yarmouk River (Table1).

Fig. 2. Location of Harrat Al-Shaam and the study area (Al-Malabeh, 2011 in Al-Qadi et al., 2020)

Fig. 3. Geological distribution of basalt rock in North Jordan (Al-Malabeh, 2015 in Hamed, 2017)

3. Methods and Materials

3.1. Experimental Technique

The laboratory measurements of radon concentration are carried out on basalt powder by applying following procedure following different authors (El-Farrash et al., 2009; Abbas et al., 2020):

- The sample was crushed into small pieces or granules that ranges between few mm's up to 1 cm's in long diameter.

- The crushed basalt samples were powdered by using Timma mill machine (steel alloy balls) at workshop of earth and environmental Department in Yarmouk University.
- The homogeneous size powders basalt samples were sieved in a mesh into 0.1 mm.
- The samples powders were dried at hot air oven at a temperature of $110 \pm 1^\circ\text{C}$ for 24 hours.
- The samples powders were weighed by sensitive balance the weight of each sample equals to 100 gm.
- These powders were put in cups, Fig.4.
- The cups with powders were covered by Radon dosimeters.
- The whole setup of dosimeters was put inside vacuum plastic bags.

The structure and the analysis of the passive dosimeters are described in a previous work (alqadi et al., 2016). The Detectors were treated following the procedure proposed by Abbas et al. (2020).

Fig.4. Schematic diagram of cup and Radon Dosimeter

3.2. Radon Measurement

The concentration of radon gas inside the powder samples can be determined using the following equation (Al-Bataina et al., 1997):

$$C_{\text{sample}} = C_{\text{air}} \left(\frac{\lambda h t}{L} \right) \quad (1)$$

where:

C_{sample} : Radon concentration in the powder sample (Bq/m^3).

C_{air} : Radon concentration in the dosimeter air above the sample (Bq/m^3).

λ : The decay constant of radon.

h : The distance between the detector and the surface of the sample.

t : The exposure time for powder samples in the Laboratory.

L : Thickness of the sample in the cup.

3.3. XRF Analyses

Representative basalt rocks from the YRB were crushed and powdered using geochemical techniques. SiO_2 concentration from different flows were analysed on fused glass disks at the Geological Institute, University of Wuerzburg, Germany, using XRF spectrometry. Powdered samples were dried for two hours at 110°C . Samples were then mixed with sodium tetra-borate and fused in crucibles over

gas burners for hours. Melts were poured into a mold creating 32 mm diameter glass disks. Analytical data are given in Table 1 (Al-Malabeh, 1994).

3.4. EDX Analyses

Chemical analyses of uranium in the basalt samples of YRB were carried out by using EDX (Energy Dispersive X-ray spectroscopy) at the Erlangen Nurnberg University, Germany and Darmstadt University, Germany. The resulted of the article data are presented in Table 4.

3.5. Polarized Microscope

The petrographic description of the basalt is presented as a typical thin section represents for the basalt in the area (Fig. 8). For the purposes of comparison model analyses for all basaltic phases are counted for 35 thin sections, the averages are presented in Table 3. Model analyses are determined using Nikon polarized optical microscope housed at Department of Earth and Environmental sciences, Hashemite University.

4. Results

The Yarmouk River basalt occupies 100 Km² and classified into four major phases. Namely, Yarmouk Sheet basalt (YS), Yarmouk blocky basalt (YB), Yarmouk massive basalt (YM) and Yarmouk exfoliated (YE). Yarmouk Sheet basalt is mainly exposed in the southern part of Yarmouk River. The most important characteristics of this phase is the arrangements of the basalt in sheet form (henceforth S) from. Seven main flows or sheet are recognized and named YS1 to YS7, and have a total thickness of about 35 m. The blocky basalt phase is overlain the YS-Basalt a layered sediment plan marked by the occurrence of the disconformity plan. It is subdivided into 5 flows, named YB1 through YB5. They have a total thickness of 27 m. The massive basalt is seen in the field to overlie the lower two phases from which is distinguished by different lithological and stratigraphical features. The total thickness of this phase equals to 25 m. The Yarmouk exfoliated basalt (YE) forms the last phase of basalt outcrop in the area. The basalt is distinguished by the exfoliation feature and looks like onion like structure. This phase has 6 flows in all, and measure about 35 m in total thickness (Hamed, 2020).

Furthermore, each major phase is subdivided into several flows. Each flow is given specific name and the GPS locations are determined. Moreover, the thickness of each flow is measured. The collected data and the detailed information are given in Table 1 for the purposes of ²²²Rn concentrations in the laboratory selective samples from each phase were collected. Those samples are marked by red color in Fig.5. Four Samples were selected from each phases of Yarmouk River basalt to determine the radon concentration inside their powder samples. The basis of collection is the state of basaltic rocks in the field (fresh or weathered), the radon concentration obtained from each flow in the field, the structural and tectonic sitting of each basaltic phase and interior basaltic flows. The laboratory radon concentrations in the investigation basaltic rocks range from (12413.12 ± 1.60) Bq/m³ to (4137.71 ± 1.39) Bq/m³ with an average value of (6635.74 ± 1.76). Samples from the lower phase (Yarmouk Sheet basalt) are relatively enriched in ²²²Rn, with averages (10233.22 ± 2.05) Bq/m³ and ranges from (12413.12 ± 1.60) Bq/m³ for the top flow (YS1) to (8275.41 ± 1.53) Bq/m³. On the other hand, the samples from phase two (Yarmouk Blocky basalt) are slightly have high: ²²²Rn concentration, which averages (6448.66 ± 2.14) Bq/m³ and ranges between (5986.47 ± 2.40) for the top flow (YB5_L) and (7307.01 ± 2.13) Bq/m³ for the bottom flow (YB1_L). The values of : ²²²Rn in phase three (Yarmouk Massive basalt) ranges from (5722.36 ± 1.30) Bq/m³ for the first flow (YM1_L) in the bottom and (4930.03 ± 3.79) Bq/m³ for the fifth flow (YM5_L) at the upper with an average of (5458.25 ± 1.48) Bq/m³. Lastly, phase four (Yarmouk Exfoliated basalt) shows the lowest radon concentration ranges from (4665.92 ± 1.42) Bq/m³ for the bottom flow (YE1_L) to Bq/m³ for the

upper flow (YE_{6L}) with an average of (4401.82 ± 1.36) Bq/m³. It's important to notice that a distinction can be made among from all Yarmouk River basaltic phases in which the concentration of each phases are clearly decreases from lower phase (Yarmouk Sheet basalt) to the upper (Yarmouk Exfoliated basalt). The details of these results are given in Table1 and shown in Fig.5.

Table 1. Radon concentration (Bq/m³) of all phases for Yarmouk basalt measured in the laboratory

Phase	Flow (m)	Total Thickness (m)	Radon Concentration (Bq/m ³)	Sample No.	S.D (Bq/m ³)	Latitude (N)	Longitude (E)	Elevation (BMSL) (m)
Four	6	122	4137.71	YE6 _L	1.39	32° 41' 56.75" N	35° 41' 17.76" E	-25
	4	110	4313.78	YE4 _L	1.36	32° 41' 59.37" N	35° 41' 15.75" E	-37
	3	105	4489.85	YE3 _L	1.28	32° 42' 2.09" N	35° 41' 15.22" E	-43
	1	92	4665.92	YE1 _L	1.42	32° 42' 3.33" N	35° 41' 12.42" E	-50
Three	5	87	4930.03	YM5 _L	1.50	32° 42' 6.74" N	35° 41' 11.82" E	-55
	3	78	5546.29	YM3 _L	1.39	32° 42' 08.06" N	35° 41' 10.23" E	-60
	2	73	5633.32	YM2 _L	1.72	32° 42' 09.48" N	35° 41' 09.98" E	-69
	1	67	5722.36	YM1 _L	1.30	32° 42' 10.32" N	35° 41' 08.97" E	-75
Two	5	62	5986.47	YB5 _L	2.40	32° 42' 11.80" N	35° 41' 07.63" E	-80
	4	58	6073.50	YB4 _L	2.36	32° 42' 12.07" N	35° 41' 07.15" E	-84
	2	46	6426.65	YB2 _L	1.68	32° 42' 13.60" N	35° 41' 05.29" E	-96
	1	41	7307.01	YB1 _L	2.13	32° 42' 14.01" N	35° 41' 03.47" E	-101
One	7	35	8275.41	YS7 _L	1.53	32° 42' 03.77" N	35° 40' 54.05" E	-107
	5	23	9595.96	YS5 _L	2.58	32° 42' 03.68" N	35° 40' 49.99" E	-119
	3	15	10652.39	YS3 _L	2.48	32° 42' 07.00" N	35° 40' 43.10" E	-127
	1	5	12413.12	YS1 _L	1.60	32° 42' 06.07" N	35° 40' 40.52" E	-137

Fig. 5. ²²²Rn concentration (x-axes) versus total thickness (y-axes) for all phases of Yarmouk River basalt

5. Discussion

5.1. Source of Radon in the Studied YRB

This discussion is carried out by means of two models proposed by this study:

- T-Model, where T: Tectonics.
- R- Model, where R: Rock.

5.1.1. Tectonic model (T-model)

This model proposes that the high radon emissions are used as Precursory phenomenon related to earthquakes and as an indicator of underlying geological faults (Yousef et al., 2015). Fractures are not visually obvious on the surface probably because they have been covered by basalt flows. The YRB lies in Wadi Al-Yarmouk which extends from Al-Maqarn area in the east exact in the Jordanian-Syrian boundary. Several field evidences such as tilting of the strata on both sides of the valley toward the interior of the valley and the occurrences of several large to medium faults; indicate the occurrence of graben. This may lead to the conclusion that the unlimited macro- and micro-fractures (Fig. 6) occurred in the Valley. These fractures allow radon to escape from the underlain ^{222}Rn -bearing rocks that have different ages starting from the Precambrian to recent, such as Uranium in Apatite minerals (Ap: $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Cl}, \text{OH})$) that forms phosphatic rocks and in Zircon (ZrSiO_4) from sandstone and granite rocks (Al-Malabeh, 2009; Al-Malabeh et al., 2017). This conclusion demands further investigation and evidences; therefore, a second model (R-model) is proposed to determine the exact source of ^{222}Rn emission source.

5.1.2. Rock model (R-model)

- Basalt rocks

Basaltic rocks have considerable amounts of radioactivity due to the presence of minerals that containing radioactive elements such as uranium (^{238}U) and thorium (^{232}Th and ^{234}Th) (Cox et al., 1979). Silicate minerals in basalts such as forsterite (olivine), labradorite (plagioclase), augite and diopside (pyroxene) have radioactive elements (Al-Malabeh et al., 2008; Al-Malabeh and Hamed, 2020). Also, magnetite and apatite contain considerable amount of the most ^{238}U (Hughes, 1982 and Al-Malabeh, 2010). Furthermore, basaltic rocks have microscopic features or fractures: Intact, deformed and failed. These features play important roles in amounts of: ^{222}Rn emission. For that, they should be taken into consideration and well discussed.

- Basaltic rocks microscopic features

The four phases of YRB exhibit microscopic features or fractures: Intact, deformed and failed. These features in hand specimens increased the possibility of radon emission from the sources. The microscopic features were measured on hand samples (Fig. 6) and their accurate percentages were determined by using optical microscope carried out in thin sections (Fig. 7). The obtained results are listed in Table 2

Table 2. Microscopic percent of intact (coherent) and fractured (deformed and failed) and massive minerals in the YRB

Phase	Intact (vol. %)	Fractured (vol. %)	
		Deformed	Failed
Sheet	77	15	8
Blocky	82	12	6
Massive	90	4	6
Exfoliated	86	6	8

Fig. 6. Basaltic sample from YRB shows vesicles, of intact (coherent) and fractured (deformed and failed) phenomena

Fig. 7. Photomicrograph showing a presentative thin section of the YRB. The percentages of the component are given in Table 3, Mag. CN 10X

Table 3. Model analyses (vol. %) given for all phases of YRB

Mineral phase (vol. %)	Olivine	Plagioclase	Pyroxene	Oxide minerals	Interstitial glass	Vesicles	Secondary minerals
Sheet	23	35	7	8	10	7	10
Blocky	27	32	5	6	7	5	18
Massive	30	28	4	5	4	14	15
Exfoliated	35	25	10	6	2	12	10

Radon measurements have evidenced that the samples of our phases of YRB showed that the fractured sample have significant variation in radon emanation with respect to the intact sample (Fig. 6), where, the basalt with micro-fractures, radon emanation drastically increased. Therefore, major finding from this study is that, in the case of low porosity (vesicles vol.%) and relatively moderate deformed basalt rocks (Table 2), the development of a macroscopic fracture provides new large exhaling surface resulting in a substantial increase in radon emission rate (Figs. 6 and 7) mainly from the interstitial glass. Moreover, the comparing of field: ^{222}Rn concentrations with similar samples measured in the laboratory clear that the ^{222}Rn concentrations in laboratory sample is higher than those measured in the field (Hamed, 2017 and Al-Malabeh et al., 2017) which is consistent with the interstitial glass source of ^{222}Rn .

5.2. Petrographical Results

The basaltic rocks of YRB are composed of olivine, pyroxene and plagioclase with oxide minerals. Interstitial glass presented in considerable amounts and it is important to stress out that there are gradual decrements of interstitial glass from the first phase (10 vol. %) to the fourth phase (2 vol. %). Interstitial glass in basalt is composed of non-crystalline elements of the residual magma (Table 3).

5.3. Uranium Content in YRB

Uranium (U) occurs in different oxidation states. The most common cases are 4, 5 and 6 and have high ionic radius (r) ranges from U^{+4} , $r = 1.56 \text{ \AA}$ to U^{+6} , $r = 0.81 \text{ \AA}$ with high ionic potential (I.P) which ranges from 2.56 (U^{+4}) and 7.41 (U^{+6}), (Dana et al., 2002). According to their distribution coefficient (DC), which is defined as the mass ratio of U in solid phase (crystals), (Sc) and in the silicate liquid (magma), (Lc):

$$DC = Sc/Lc \quad (2) \quad (\text{Lamarchand et al. 1987; Al-Malabeh, 1993})$$

According to DC calculation ^{238}U has DC less than one in the basaltic melt (Al-Malabeh, 1994). The pyrogenic interstitial glass is formed from the residual melt. The glass is distributed between the mineral phases (Cox et al., 1979 and Dupuy and Dostal, 1984). Interstitial glass is fresh and has isotropic indicate figure. It normally has no color and seen as a matrix between crystals. Interstitial glass in the studied basalt (Fig. 7) ranges from 10 vol.% in YS (Sheet basalt) and decreases to 7, 4, and 2 vol. % in the YB, YM, YE, respectively (Table 3). The analyzed data show that the YRB interstitial glass (Figs. 7 and 8) have considerable contents of ^{238}U decreases from 5 ppm in the lower first basaltic phase (YS) to values of 3, 2 and 1ppm in the second through four phases, respectively (Table 4).

Table 4. Selected element analyses: SiO_2 and uranium, in the interstitial glass

Basalt phase	SiO_2 (wt. %)	^{238}U (ppm)
Sheet	49.43	5
Blocky	47.18	3
Massive	46.59	2
Exfoliated	45.01	1

Fig. 8. EDX image of the interstitial glass of YRB

6. Conclusions

This study aims to investigate natural gas concentration ^{222}Rn Jordan. Detailed field investigations show that the basaltic rock in the area of Yarmouk River occupies an area of about 100 km². Four major basaltic phases are recognized, sheet, blocky massive and exfoliated, respectively, in the area with a total thickness of 122 m. All basaltic phases are composed of several flows with a total of 23 flows. R-model proposed by this shows that the highest: ^{222}Rn are recorded in basaltic rocks (12413.12 – 4137.71) Bq/m³, with an average value of (6635.74) Bq/m³. The source of radon ^{222}Rn in the studied rocks may attribute to the basalt interstitial glass and associated pyrogenic origin. However, the main mineral phases are olivine, plagioclase, pyroxene and oxide minerals magnetite could assist as a source of ^{222}Rn . The major result of the present investigation is the ^{222}Rn enrichment in the area especially the basalt flows through the fractures long major faults cannot be considered in this research as the source of the ^{222}Rn . These results are consistent with those obtained by Al-Malabeh and Hamed (2020) and Abu-khalaf (2014) where the radon concentration was 981-1241.8 Bq/m³ and 379-636.6 Bq/m³ repressively. Radon gas (^{222}Rn) concentration increases gradually upwards in the YRB. This may indicate the rate and the level of concentration of the parent isotope (^{238}U) and their decay rate.

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