ENRICHMENT OF RADON (^{222}Rn) CONCENTRATION IN REMAH VOLCANO, NE-JORDAN

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ABSTRACT

Remah Volcano (RV) is one of the numerous volcanoes that are distributed in three volcanic fields in the Jordanian Harrat, northeast Jordan. The RV is about 4 km². It is about 80 m high. They are arranged in three main horizons: lower, middle, and upper and show planar stratification. ^{222}Rn content has been measured by using Nuclear Track Detector (Columbia Resin CR-39) studied rocks show that the range from 945 – 1283 (Bq/m³) and the average of 1104.46 (Bq/m³), the highest content of 1175 – 1283 (Bq/m³), is recorded in the samples from the upper horizon from RV. ^{222}Rn ranges between 1049 – 1125 (Bq/m³) as shown in the middle horizon and have contents of 945 – 999 (Bq/m³), in the lower horizon. The ^{238}U concentrations increase gradually upwards, respectively. This study concludes that the correlating of contents of ^{238}U contents with ^{222}Rn concentrations results obtained in this study it's clear that the ^{222}Rn -concentration shows a distinct increase as ^{238}U contents increases. This can lead to the conclusion that the ^{238}U -decay is the main source that is produced ^{222}Rn from the interstitial glass which resulted from residual melt in tephra rather than from emission of inferred source in cracks and fractured of inferred faults.

Keywords: ^{238}U ; ^{222}Rn ; Remah volcano; Concentrations; Jordan.

INTRODUTCION

Pyroclastic and basaltic rocks are used in building materials now. It can be said that the essential sources of radon in the outside middle. Nearly 80% of the pop-up radon gas to the outside middle produces the crustal rocks, emitted from the soil depends on the amount of egress of granular soil material, and to spread through the pores of the soil to the outside middle, Al-Fugha and Al-Malabeh (2019) it was found that generally emitted from rock larger

than from the soil and minerals. so a high concentration of radon gas in the rock content leads to pollution of groundwater (Henshaw et al., 1990). The radon gas has an impact on health, it is dissolved in a quantity of less than of the recommended standard of United States Environmental Protection Agency, from the potential source of uranium-rich hydrocarbons (Farhan et al., 2020). Harrat Al-Shaam has 50,000 km² total area and occurs in the Arabian plate. It spreads from Syria to Saudi Arabia (Al-Malabeh et al., 2008). In Jordan is called as Jordanian Harrat and covers an area 12,000 km². The Jordania Harrat consists of thick basalt flows and pyroclastic volcanic cones. The volcanic cones are clutered in three fields, namely: Remah, Ashqaf and Dhriwa (Al-Dafneh) (Al-Malabeh et al., 2002 and Smadi et al., 2018). 200 Tephra cones have occurred in Jordanian Harrat (Al-Malabeh, 1994). These cones are poorly known. Some research has been carried out on the volcanology, mineralogy, chemistry and radioactivity of the volcanoes.

The aim of the present study is to carry out detailed investigations of radioactive concentrations of scoria in RV for selected isotopes namely: ²³⁸U and ²²²Rn. This research represents the first of its type world-wide since no ²²²Rn concentrations in scoria rocks have been carried out elsewhere globally.

STUDY AREA

Remah Volcano RV is considered in this study. The RV is considered as an outstanding volcano because of its variety of scoria colors and sizes. RV lies in Remah Volcanic Field, about 70 km east of the Mafraq city, Jordan, near Dair Al-Gen town (Fig. 1). RV has an area of 4 km², and one of the well-established cinder volcanos in Harrat Al-Shaam.

Fig. 1. Location map of the Remah Volcano, and selected structural and tectonics in Jordan (Modified from different sources, e.g., Al-Malabeh, 1993)

MATERIALS AND METHODS

Field Study and Sampling

The field study aims at collecting representative fresh samples from the three horizons of RV: Lower, Middle and Upper horizons following the classification of Al-Malabeh (2010) (Fig. 2). For Radon and Uranium concentrations measures five samples from each horizon were collected.

Fig. 2. (a) Field photograph of the three horizons in Remah volcano (b) Volcano cross-section through the volcanic succession of Remah volcano

Radon Measurements Tools

Radon concentrations are measured by CR-39 dosimeter detectors. This method is highly recommended by CR-39 producers (British company). Due to its accurate results, several scientists are published recent articles using the CR-39 dosimeter method for example köç, et al., (2017) and Jebur et al. (2019).

The procedure was done, as follows:

1. The sample was crushed by using jaw crusher into small pieces or granules that a range between few mm up to 1 cm in a long diameter.
2. The crushed samples were powdered by using Timma mill machine (steel alloy balls) housed at workshop of earth and environmental Department at Hashemite University in Jordan.
3. The resulted homogeneous size powders (0.2 mm) by using U.S standards sieve sets (ASTM E-11) sieve number 200 were put in cups.
4. The samples powders were weighed by sensitive balance the weight of each sample equals to 100 gm.

5. The cups with powders were covered by converted Radon dosimeters (Al-Bataina et al., 1997 and Alqadi et al., 2016)
6. Finally, the whole setup of dosimeters was put inside vacuum plastic bags.

Chemical Etching of CR-39 Detectors

The dosimeters were left for 70 days as exposure time for each sample in Laboratory. During the exposure time, α particles which were emitted by radon and their daughters were registered on the CR-39 detector's surface as nuclear tracks. the CR-39 detectors were removed from the cups Then, were chemically etched by immersing them in a beaker containing: 7 molar KOH aqueous solution all surrounded by heat water bath at $70 \pm 0.2^\circ\text{C}$ for 8 hours. Then the detectors were taken out and washed in distilled water to clean them from any chemicals and then dried (Kullab et al., 2001 and Al-Bataina et al., 1997)

Counting the Tracks of Alpha Particles on the Surface of CR-39 Detectors

A Normal light (M1 9000) microscope (MEIJI TECHNO Co. LTD) with a suitable magnification of 40x for the objective lens was used to count the number of tracks per cm^2 recorded on each CR-39 detector surface. The nuclear tracks were viewed and counted over 20 different views for each CR-39 detector, and then the average number and standard deviation of tracks on each detector were calculated using two the following:

$$\bar{X} = \frac{x_1 + x_2 + \dots + x_n}{n} = \frac{\sum_{k=1}^n x_k}{n} \quad (1)$$

$$\sigma = \sqrt{\frac{\sum_{k=1}^n (x_k - \bar{X})^2}{n}} \quad (2)$$

Where

$\bar{X}$: the average number of tracks per view on CR-39

x_k : the number of tracks in view k

n : the number of views measured on each CR-39 detector.

σ : the standard deviation of the average value of the number of tracks per view

The track number density per sample ρ was calculated by dividing the average number of tracks ($\bar{X}$) by the area of a view (A) as shown below (in units of track/ cm^2)

$$\rho = \frac{\bar{X}}{A} \quad (3)$$

Measuring the Radon Concentration

By using the average number of tracks density, the radon and its progeny concentration in the air inside the dosimeter is calculated by the following calibration equation (Al-Bataina et al., 1997):

$$C_{air} = \frac{C_0 t_0 \rho}{\rho_0 t} \quad (4)$$

where:

C_{air} : the Radon concentration in air inside the dosimeter in units of (Bq.m⁻³).

C_0 : the Radon concentration of the calibration chamber (90 kBq.m⁻³). Calibration for CR-39 is carried out by (Al-Bataina et al., 1997)

t_0 : the calibration exposure time (2 days).

ρ : the measured track number density per cm² on the CR-39 detectors inside the dosimeters used in the study.

ρ_0 : the measured track number density per cm² on those of the calibrated dosimeters (3.302x10⁴tracks/cm²).

t : time of exposure for the detectors.

The concentration of radon gas inside the powder can be determined by Al-Bataina et al. (1997):

$$C_{sample} = C_{air} \left(\frac{\lambda h t}{L} \right) \quad (5)$$

where:

C_{sample} : ²²²Rn concentration in the powder sample (Bq/m³).

C_{air} : ²²²Rn concentration in air above the sample (Bq/m³).

λ : The decay constant of radon.

h : space between the CR-39 detector and the surface of the sample (m).

t : Exposure time for samples (hrs).

L : Thickness of the sample in the cup (cm).

Polarized Microscope

The petrographic description of the scoria is presented as a typical thin section represents the scoria in the area (Fig. 4). For the purposes of comparison, modal analyses for all scoria horizons are counted for 15 thin sections, the averages are presented in (Table 1). Modal analyses are determined using Nikon polarized optical microscope.

EDX Analyses

Chemical analyses of Uranium in the fresh glass of RV were carried out by using EDX (Energy Dispersive X-ray spectroscopy) housing at the Friedrich-Alexander University (FAU), Germany. The analysed elements' analytical results are given in (Table 2).

RESULTS

Petrographic Features

The scoria from the three horizons of RV has slightly similar petrographic features, and may be classified as scoriaceous glassy olivine-phyric basalt (Fig. 3). The glass in the RV shows little evidence of alteration, is transparent to light green, and constitutes averages of 31, 34 and 37 vol. % in rocks from the lower, middle and upper horizons, respectively (Table 1). Olivine forms dominantly subhedral crystals and needles them average 15, 16 and 18. Pyroxene phenocryst average range from 2, 2 and 6 vol.%. Finally, plagioclase phenocrysts and microlites have a total content 3, 4, and 7 vol. %. opaque grains (magnetite) occur as inclusions within phenocrysts and the fresh glass. The vesicles are circular to sub-circular, and account from 48, 42 and 29 vol. % of the whole rock. Representative scoriaceous glassy olivine, plagioclase and pyroxene-phyric basalt from the RV (Fig. 3).

Fig. 3. Sketch photomicrograph, represents scoriaceous glassy olivine, plagioclase and pyroxene-phyric basalt from the RV

Table 1. Averages of modal proportions of the different components given in vol.% for the scoria samples from RV

Horizon	Minerals vol. %					
	Olivine	Pyroxene	Plagioclase	Interstitial glass	Opaque minerals	Vesicles
Upper	18	6	7	37	3	29
Middle	16	2	4	34	2	42
Lower	15	2	3	31	1	48

Radon Measurements

^{222}Rn content of the studied rocks has an average value equal to 1104.46 (Bq/m^3) and ranges between 945 and 1283 (Bq/m^3) (Figs. 4 and 5). The highest content of 1175 - 1283 (Bq/m^3),

is recorded in the samples from the upper horizon from RV. ^{222}Rn concentration ranges from 1049 – 1125 (Bq/m^3) in the middle horizon and have 945 and 999 (Bq/m^3) in the lower horizon. The obtained results for analysed ^{222}Rn and ^{238}U are presented in Table 2.

Uranium Concentrations

^{238}U concentration in the investigated rocks ranges from 412- 903 ppm with average 705.13 ppm as shown in Figs. (6 and 7). The highest content of 793-903 (Bq/m^3) is reported in the scoria from the upper horizon of RV (Table 2). ^{238}U ranges from 691-781 ppm in the middle horizon and have contents of 412-643 ppm in the lower horizon.

Fig.4. ^{222}Rn concentration according to horizon in RV samples

Table 2. Uranium (^{238}U) and Radon (^{222}Rn) concentrations in scoria samples horizons

Horizon	Sample No.	Height (m)	Uranium ^{238}U (ppm)	Radon ^{222}R (Bq/m^3)
Upper	URV5	49	903	1283
	URV4	46	886	1254
	URV3	44	847	1250
	URV2	41	831	1247
	URV1	38	793	1175
Middle	MRV 5	36	781	1125
	MRV4	34	768	1105
	MRV 3	30	746	1098
	MRV 2	27	721	1076
	MRV 1	25	691	1049
Lower	LRV 5	22	643	999
	LRV4	17	611	997
	LRV 3	11	485	988
	LRV2	7	459	976
	LRV1	2	412	945

Fig. 5. ^{222}Rn minimum, maximum and average concentrations according to horizon in RV samples

Fig. 6. ^{238}U concentration according to the horizon in RV samples

Fig. 7. ^{238}U minimum, maximum and average concentrations according to horizon in RV samples

DISCUSSION

Pyroclastic rocks in general and scoria of different sizes, in concern, contain considerable concentrations of ^{238}U , ^{234}Th and ^{222}Rn . Most of the minerals in scoria such as olivine, plagioclase pyroxene and opaque minerals contain some radio-elements: ^{238}U and ^{234}Th (Raymond, 2002 and Al-Malabeh et al., 2008). Moreover, the interstitial glass which resulted from the residual glass. This glass is the rest of magma. During the process of fractional crystallization, melts become enriched in incompatible elements Hence, knowledge of the crystallization sequence is critical in understanding how the residual melt compositions evolve the most ^{238}U (Dobosi et al., 1991; Hughes, 1982 and Al-Malabeh, et al., 2017).

^{238}U has a distribution coefficient (KD) $\text{KD} < 1$, this low value of KD will allow low percent partition ($\leq 1\text{ppm}$) in olivine, pyroxene and plagioclase during the partial melting and fractional crystallization processes (Wilson, 1989; Al-Malabeh and El-Hasan, 2009). The rest from U-content in the basaltic magma will concentrate in residual melt and solidified inside the interstitial glass (Cox et al., 1979). The interstitial glass is defined as an original glass that is solidified from the rest (residual) of the magma and occurs between the crystals (Al-Malabeh and Kempe, 2005). It is normal, fresh isotropic and mostly colourless to light green. This glass is the residue of melts and contains elements that have not been crystalline to minerals. It serves as groundmass for the associated minerals. In the following, an attempt will be made to propose a genetic model for the ^{222}Rn enrichment in RV. This model will

concentrate on comparing the ^{238}U content and the parent isotope ^{222}Rn depend on the scoria component and by comparing the IG averages and the concentration of ^{238}U and ^{222}Rn .

The interstitial glass in RV samples from three horizons exhibit gradual increment is in consisting with the increasing of ^{238}U of the ^{222}Rn with the IG may indicate the that Interstitial glass in the RV scoria (Fig. 8) ranges from 10 vol.% in RV scoria and increase upwards as seen in the (Table 3). The analyzed data show that the RV interstitial glass have considerable contents of ^{238}U increases upwards in RV. The ^{238}U average ratios for the samples of RV increase from the lower, middle and upper horizons, respectively. This is consistent with the increase of IG vol. % the upwards increase IG vol. % through the RV different horizons moreover, the $^{238}\text{U}/\text{IG}$ average ratios for the samples of RV range from 16.84, 21.80 and 23 from the lower, middle and upper horizons, respectively. ^{238}U has a clearly positive correlation with IG, $R^2 = 0.96$ (Fig. 8). On the other hand, the $^{222}\text{Rn}/\text{IG}$ average ratios for the samples of RV range from 31.64, 32.08 to 33.56 from the lower, middle and upper horizons, respectively. ^{222}Rn has a distinct positive correlation with IG, $R^2 = 0.99$ (Fig.9).

Finally, the comparing of the average ^{222}Rn concentrations which are measured in laboratory sample in the present study (Table 3) with the field-measured ^{222}Rn concentrations by Abu Khalaf (2014), it is clear that the concentrations found by this study are higher than those measured in the field by the later which is consistent with the interstitial glass origin of ^{222}Rn rather than emission rates along faults, fractures and tectonic complex source. Also, the averages all horizons are lower than the averages that range from 4051-16396 Bq/m³ reported by Yousef et al. (2015) in Granite rocks, Um Ara eastern desert/Egypt. They may attribute to the enrichment of Granit rocks by radioactive elements such as ^{238}U .

Table 3. Comparison between laboratory and field measurements of ^{222}Rn in RV

Horizon	Laboratory ^{222}Rn (Bq/m ³)	Field ^{222}Rn (Bq/m ³)
Upper	1241.8	636.6
Middle	1090.6	484.4
Lower	981	379

Fig. 8. ²³⁸U- Interstitial glass correlation diagram

Fig. 9. ²²²Rn - Interstitial glass correlation diagram

CONCLUSIONS

This research aims at a detailed investigation of natural gas concentration (²²²Rn) in Remah Volcano RV, NE-Jordan. Field investigation and chemical analysis have led to the following conclusions, which are presented as point summary:

1. Scoria in RV has a thickness of 49 m. The volcano stratigraphy shows three distinct horizons.
2. Petro-graphically, the scoria rocks of RV consist of volcanic interstitial glass (IG) vesicles, olivine, pyroxene, plagioclase and opaque minerals. IG and vesicles are the dominant components, and together may exceed 70 % of the total volume of the scoria.

3. Analyses have been performed on 15 fresh scoria samples collected systematically from the horizon. It was noted that the concentration of Radon in RV divided depending on the horizon components of the RV, and the average of concentrations was as follows. in the upper horizon average Radon concentration reached 1241.8 (Bq/m³), the rate of Radon concentration in the middle horizon reached 1090.6 (Bq/m³) and the average Radon concentration in the lower horizon reached 981(Bq/m³). This shows that the upper horizon has the highest Radon concentration, and through previous results it is clear that the concentrations of Radon increases from the lower to the middle up to the upper horizons.
4. The comparing of field ²²²Rn concentrations with similar samples measured in the laboratory clear that the ²²²Rn concentrations in laboratory sample is higher than those measured in the field which is consistent with the interstitial glass source of ²²²Rn. This can lead to the conclusion that the ²²²Rn is mainly produced as a result of ²³⁸U-decay in interstitial glass rather than from emission of radon from cracks and fractured of inferred faults.

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REFERENCES

- Abu khalaf, A., 2014. Natural Gas Concentration (Rn) and its Relation to the Enrichment of Selective Low and High Field Strength Trace Elements in Jabal Remah Volcano, NE-Jordan. Unbupl. M.Sc. thesis, Hashemite University, Zarqa-Jordan.
- Al-Bataina, B., Ismail, A., Kullab, M., Abumurad, K., and Mustafa, H., 1997. Radon measurements in different types of natural water. *Radiation Measurements*, 28: 591-594.
- Al-Fugha, H., and Al-Malabeh, A., 2019. Mineralogy, geochemistry and petrogenetic of cretaceous chert from Wadi al-Wala, central jordan. *Iraqi Geological Journal*, 50: 1-11.
- Al-Malabeh, A., 1993. The Volcanology, Mineralogy and Geochemistry of Selected Pyroclastic Cones from NE-Jordan and their Evaluation for Possible Industrial Applications, Ph.D. thesis, Erlangen University, Germany.
- Al-Malabeh, A. & Kempe, S., 2005. Origin of iron ore nuggets ("Bohnerze") through weathering of basalt as documented by pebbles from the Herbstlabyrinth, Breitscheid-Germany. – *Acta Carsologica*, 34:459-470.
- Al-Malabeh, A., and El-Hasan, T., 2009. Mineralogy and chemistry of coronadite from middle Cambrian manganese deposits at Wadi Dana, Southern Jordan. *American Journal of Applied Sciences*, 6: 576-581.
- Al-Malabeh, A., 1994. Geochemistry of two volcanic cones from the intra-continental plateau basalt of Harra El-Jabban, NE-Jordan. *Basaltic Rocks of Various Tectonic Settings, Special Issue of the Geochemical Journal.*, 28: 542-558.
- Al-Malabeh, A., 2010. Volcanic successions of the Jebal Remah volcano, Northeast Jordan. *International Journal of Economic and Environment Geology*, 1:6-10.

- Al-Malabeh, A., Al-Smadi, Z., and Ala'li, J., 2017. Mineralogy, geochemistry and economic potential of zircon and associated minerals in Dubaydib sandstone formation, Jordan. *Iraqi Geological Journal*, 50 :1-11.
- Al-Malabeh, A., El-Hasan, T., Lataifeh, M., and O'Shea, M., 2002. Geochemical and mineralogical related magnetic characteristics of the tertiary-quaternary (Umm A-Qutein) basaltic flows from the basaltic field of harra el-jabban, northeast jordan. *Physica B-Physics of Condensed Matter*, Netherlands, 321: 396-403.
- Al-Malabeh, A., Kempe, S., Henschel, H.-V., Hoffmann, H., and Tobschall, H., 2008. The warda iron ore deposit and historic mine near ajloun, (Northern Jordan): mineralogy and geochemistry of a potential speleogene iron ore deposit. *Acta Carsologica*, 37: 241-253.
- Alqadi, M., Alzoubi, F., and Jaber, M., 2016. Assessment of radon gas using passive dosimeter in Amman and Al-Rusaifa cities, Jordan. *International Journal of Radiation Research*, 14: 367-371.
- Cox, K., Bell, J. and Pankhurst, R., 1979. *The Interpretation of Igneous Rocks*, fourth ed. Allen and Unwin, London.
- Dobosi, G., Schultz-Guttler, R., Kurat, G., and Kracher, A., 1991. Pyroxene chemistry and evolution of alkali basaltic rocks from Burgenland and Styria, Austria Mineral Petrol., 43: 275 – 292.
- Farhan, A. O., Ahmed, A. M., Awadh, S. M., and Al-Sultani, A. H., 2020. Radon gas and effective dose in groundwater in Abu-Jir Village in Anbar, Western Iraq. *Iraqi Geological Journal*, 2C:2020.
- Henshaw, D., Eatough, J., and Richardson, R., 1990. Radon as a causative factor in induction of myeloid leukemia and other cancers. *The Lancet*, 335:1008-1012.
- Hughes, C., 1982. *Igneous Petrology, Development in Petrology*, first ed. Elsevier, New York.
- Jebur, J. , Subber, A. R. H., and Saadon, W., 2019 .Calibration of CR-39 for different heights of radon dosimeters and application of radon concentration in Mahajran River sediment in Basra Governorate, Iraq. *Journal of Physics Conference Series*, 1294: 55-67.
- Köç, P., Ekinci, N., Cinan, P., and Kavaz, E., 2017. Determination of radon concentration by using CR-39 Plastic track detectors in dwellings of Bingöl and Mus Provinces of Turkey. *Asian Journal of Chemistry*, 30: 226-230.
- Kullab, M., Al-Bataina, B., Ismail, A., and Abumurad, K., 2001. Seasonal variation of radon-222 concentrations in specific locations in Jordan. *Radiation Measurement*, 34: 361-364.
- Raymond, L., 2002. *Petrology: The Study of Igneous, Sedimentary, and Metamorphic Rocks*, McGraw-Hill. USA.
- Smadi, A., Al-Malabeh, A., and Odat, S., 2018. Characterization and origin of selected basaltic outcrops in Harrat Irbid (HI), northern jordan. *Jordan Journal of Earth & Environmental Sciences (JJEES)*, 9:185 -196.
- Yousef, H., El-Farrash, A., Saleh, G. and Khalf, M., 2015. Estimation of radon concentration for granite rocks samples using LR-115 detector. *International Journal of Recent Scientific Research*, 6.