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DETERMINATION OF RADON AND URANIUM CONCENTRATION IN THE PETROLEUM PRODUCTS AT THE ALQAYARA AND ALKASIK NINEVAH GOVERNATE REFINERIES

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Abstract:

Determination of radon gas (Rn222), uranium (U238) concentrations, and the effective activity of (ARn222) in crude oil and petroleum products (asphalt sial, solid asphalt, heavy gas oil, light gas oil, kerosene, naphtha, RCR (Ramsbottom carbon residual), diesel, light naphtha, and fuel oil) using CR-39 detector at the Al-qayara and Al-kasik refineries. It was found that the radon gas concentration in crude oil (8.418 Bq/m3) and petroleum products ranged from (9.329 Bq/m3) to (1.663 Bq/m3) with an average value of (5.496 Bq/m3) at Al-qayara refinery, the higher and lower values of radon gas effective activity in the samples of products of Al-qayara refinery (10.728 ± 2.469 Bq/kg) with an average of (6.598 Bq / kg), while for Al-kasik refinery was the radon gas concentration in crude oil is (10.937 Bq/m3) and in petroleum products ranged from (8.581 Bq/m3) to (1.931 Bq/m3) with an average of (5.256 Bq/m3). As for the concentration limits of Uranium-238 from (0.928 ppm) to (0.232 ppm) with an average value of (0.58 ppm) at Al-qayara refinery, and from (0.862 ppm) to (0.206 ppm) with an average value of (0.534 ppm) at the Al-kasik refinery site. The higher and lower values of the radioactivity of radon gas at the Al-kasik refinery samples (12.07 ± 2.91 Bq/kg) with an average of (7.49 Bq/kg). By comparison, we found all results in the limitations of ICRP regulations.

Keywords: Radon, Uranium, CR-39 detector, petroleum products, TENORM

1. INTRODUCTION

Nuclear methods are one of the modern techniques in environmental studies and geological studies. It gives good and important results due to its ability to sense low concentrations of radiation and to analyze and study radioactive isotopes and their radioactive products one of the causes of environmental pollution is the spread of radioactive materials in the surface soil, rocks, and water, whether that is It spreads naturally or as a result of external contamination. Sources of radioactive contamination increased due to wars and nuclear explosions of nuclear weapons testing carried out by the great powers. So radiological studies and surveys of air and soil have led to an increase in rocks, water, food, etc. to measure the dose level ^{222}Rn the radioactivity to which humans are exposed. Radon is one of the sources of radioactive contamination. The presence of radium 226Ra in an area in nature depends on the presence of uranium ^{238}U, which is estimated and its quality in the earth's crust is within (4-3) ppm. Radon is the direct descendant of radium ($t_{1/2}$ = 1620 years) 226Ra in the uranium ^{238}U series in nature, where radium is found in soil, rocks, seawater, and ocean sediments. All elements of the radium decay chain are solid except for radon, which is the only gaseous element among them and behaves like other gases that humans inhale. A colorless, tasteless, and odorless radioactive gas previously discovered by the scientist (F.E. Dorn, 1900) with a density of 9.73kg.m^{-3}boiling point (-61.8) oC and freezing point (-71) oC and half-life (3.825) days. As a result, it diffuses over a wide range of distances in the atmosphere, despite being emitted from the soil in smaller quantities than 220Rn [1]. Because it is about nine times heavier than air, it tends to stay close to the ground on the ground floors [2]. Radon gas decomposes by emitting alpha particles with energy (5,485) Mev, which poses a threat to public health. Radon and its offspring, ionizing alpha

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particles, emit up to 50% of the background radiation in room air [3]. The release of these radionuclides is dangerous to workers' health and the environment. Therefore, we conducted a study on two of the North Oil Company's refineries, namely the qayara refinery and the Al-Kasik refinery, to determine the degree to which crude oil and petroleum derivatives have been contaminated with radioactivity.Radon gas from the decay of uranium chains were measured, and since these elements have a very long half-life, and the half-life depends on the geological formations of the Earth, [4]. These radionuclides are released during oil and gas extraction processes, as well as during product The measurement of radon levels in the Zubair oil field in southern Iraq using the CR-39 solid nuclear trace detector and the LR-115 type detector was the focus of research (Al-Jim, 2012). (Radon and tauron gas). The results revealed a difference in radon gas concentrations between oil wells, where the highest value (993.4 Bq.m^{-3}) was found in the Hamar field well and the lowest value (195.95 Bq.m^{-3}) was found in the well [10].The quantities of uranium and radon gas were evaluated in samples taken from the qayyarah refinery and field south of Mosul in the research (Mustafa A. Mahmoud, Rashid Muhammad Yusuf, 2021). The samples included petroleum-related soil and water, as well as sludge and crude oil. The radioactivity of the samples was detected using the

2. MATERIALS AND METHODS

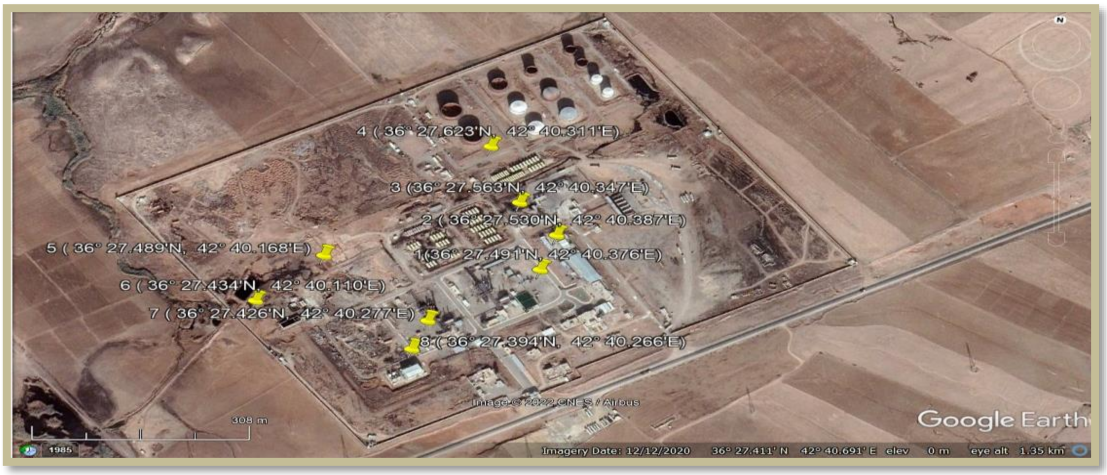
As indicated in Figures (1,2) this study included the collection of samples from two refineries in the northern area that are inside of the Nineveh

refining [5]. Several studies and reports related to NORM have been published by the International Atomic Energy Agency (IAEA) [6], which included security reports for dealing with NORM, as well as the International Oil and Gas Association (OGP) [7]. The United Nations Committee on the Scientific Effects of Atomic Radiation (UNSCEAR) [8] has published instructions for the production of petroleum and petroleum products. Recent studies have been conducted in Yemen and Riyadh on the radioactive contamination of internationally known petroleum products [7,8,9].

CR-39 solid-state nuclear track detector. Radon levels in soil samples ranged from 10.79 to 19.67 Bq/kg, with the range 7.25 Bq/kg falling within internationally permissible limits. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has set the value of uranium in soil at 32 Bq/kg and in water at 50 Bq/l, while the International Atomic Energy Agency has set the value of the value of uranium in slag is (8-5 × 105) Bq/kg, the International Association of Oil and Gas Producers (IOGP) has determined the value of uranium in crude oil (800-4×105) Bq/kg and therefore does not pose any danger to the lives of workers in the refinery, the field and the environment [11].

2.1 Location of study:

Governorate, namely (Al-qayara refinery, 60 kilometers south of Mosul) (and Al-Kasik refinery, 70 kilometers northwest of Mosul).



Fig(1): A map shows the Location of AL.Kasik refinery

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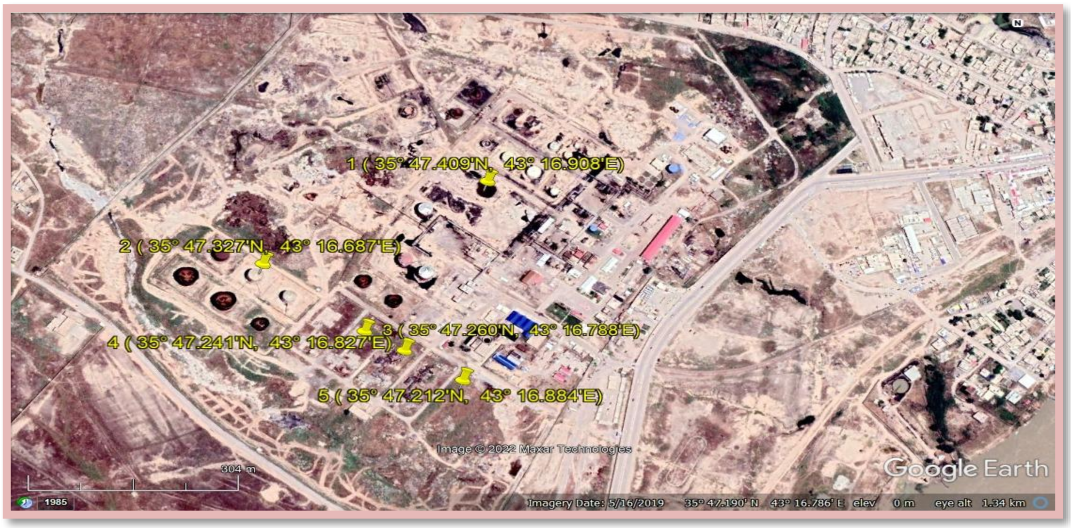

Fig(2): A map shows the Location of AL.qayara refinery

2.2 Samples Collection:

(Thirteen) different samples were collected from two Refineries, including, Al-Kasik and Al-qayara. The samples included petroleum products and crude oil. They were taken from production units, tanks, and crude oil samples. They were collected in one-liter glass bottles and glass containers with a label. Each sample has the type of sample, the location, and the date it was taken printed on it.

2.3 Sample Preparation :

The samples were taken to the University of Mosul, where they were prepared in the College of Environmental Sciences and Technologies for the study. The samples were prepared by placing them in 4 cm high plastic cans which were tightly closed, calculate the weight of the sample, and leaving it for 30 days to obtain the optimal balance between radium and radon, which was calculated to be 98 percent of the ideal equilibrium state. through the radioactivity equilibrium relationship[12,13].

$A_{Rn=}A_{Ra}(1 - e^{-\lambda Rn\, t})$

Where A_{Rn} effective radon and $A_{R\alpha}$ and represents the effectiveness of radium and λ_{Rn} represents the radon decay constant 0.1814 d^{-1} and t is the time required to reach equilibrium.

3. MEASUREMENTS:

The uranium concentration and specific radioactivity of radon gas were measured for all samples using a solid-state CR-39 nuclear track detector. The samples were placed in an irradiation system consisting of two plastic cups identical in shape and size, and filter paper was placed between them and closed with adhesive tape tightly. CR-39 detectors were left inside the chambers for 30 days [14,15] then we prepared the corrosion solution, the chemical substance 6.25 N NaOH. After the irradiation period was completed, the reagents were removed and placed inside the chemical erosive solution, and then they were placed in a water bath at a temperature of 70 degrees Celsius for 4 hours. The radiation background was 130 $Tr.Cm^{-2}$ and it was removed from the intensity of particulate effects. Alpha came from the samples under study, and the effect of Alpha was measured (using an optical microscope with a magnification of 400x). Figure 2 shows the irradiation system that was used.

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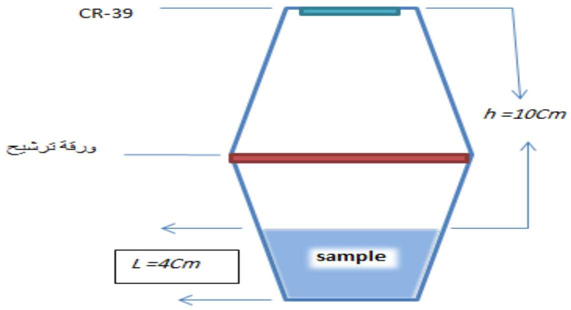

Fig(3): Explains the irradiation system

The following equations numbered (1) to (7) were used for calculation[16,17]:

1-Measure the concentration of radon gas through the following relationship

P=KC_(α) T ………(1)

Where P (Tr. cm^{-2}) represents track density, K (Tr. $\frac{Cm^{-2}}{h^{-1}Bq.m^{-3}}$) represents the diffusion constant, T represents the irradiation time in hours and C_a represents the radioactivity of radon in the air space in units (Bq. m^{-3}).

2- Determination of the propagation constant k

$$K = \frac{1}{4}r(2Cos\,\theta - \frac{r}{R\alpha}) \text{ .... (2)}$$

Where r represents the radius of the cup and is equal to 2.58 cm and Rα represents the range of alpha particles in the air produced by radon and is equal to 4.16 cm and represents the θ angle of 35 °

3- Determining radon concentration in samples (C_s) :

The radon concentration in the samples was calculated by the following relationship:

$C_s = C_\alpha\, T\, h\, \text{Rn}\lambda\, /L$ ....(3)

Where C_s represents the radon activity within the samples in units (Bq. m^{-3}),), h represents the height of the detector from the sample surface in cm units, T represents the irradiation time in days, λ_{Rn} represents the radon decay constant and L represents the thickness of the sample in cm.

4- The radioactivity of radon gas produced from samples in the (Bq) unit:

$A_{Rn} = C_s V$................ (4)

$V = L\,\pi\, r^2$...................... (5)

where A_{Rn} is the radioactivity of radon gas in the samples (Bq) unit and V volume samples (m^3) and r radius equal 2.58 (cm).

5- Counting the number of radon atoms N_{Rn} in the samples:

$N_u\lambda_u = N_{Rn}\lambda_{Rn}$ .... (6)

After determining the number of radon atoms, the number of uranium atoms is found through the law of radiation balance.

$N_{Th}\,\lambda_{Th} = N_{Rn}\lambda_{Rn}$ .... (7)

6- Determining the mass of uranium in the samples:

$W_{U=N_u A_u/A_{av}}$ .... (8)

Where represents the mass of uranium in the samples, A_U represents the mass number of uranium, and N_{av} represents the number of Avogadro, N_U number of uranium atoms in the samples.

7- Determining the uranium concentration (C_u) in samples in ppm units:

$pp_m = \frac{W_U}{W_S}$ .... (9)

Where W_S represents the mass of the sample in (gm).

4. RESULTS AND DISCUSSION:

In the present study, we determine the concentration of radon gas and uranium in 13-samples of crude oil and petroleum products (bitumen (asphalt sial & solid asphalt), heavy gas oil, light gas oil, kerosene, naphtha, RCR, diesel, light naphtha, and fuel oil) at two locations (Al-qayara & Al-kasik refineries). A bove determination was carried out through the measurements of the track density (ρ) on the CR-39 detector which used. Table(1) shows the sample identification (ID), type of sample, location, and position coordinates,while table (2) include concentrations of radon gas in can's air, can's with samples and the weight of samples. The results show that the higher radioactivity (concentrations of radon gas) and uranium-238 cocentration are (10.728 Bq/kg) and (0.862 ppm), respectively, in the asphalt sial sample at the Al-qayyarah refinery, while the lower radioactivity and concentration of uranium are (2.478 Bq/kg) and (0.145 Bq/kg) in the light gas oil sample

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at the Al-qayyarah refinery product which shown in table (3). Soil texture and composition were very important in determining the radon concentration, so the difference in values of concentration are function of the exact density, the porosity of the samples, and the type of soil. The lower value of concentrations are attributed to the kind of soil which fine dence and low porosity (lower diffusion coefficient) which lead to reduce the radon concentration that is a function of uranium concentration. With same above reason,we found that the results also showes that the higher radioactivity (concentrations of radon gas) and uranium were (11.627 Bq/kg) and (0.928 ppm), respectively, in the RCR sample in Al-kasik refinery, while the lower radioactivity and concentration of uranium are (2.91 Bq/kg) and (0.232 ppm) in the diesel sample at Al-kasik refinery, as shown in table (3). When we compare our data with the International Atomic Energy Agency [19], and the ICRP- committee guide for crude oil (800-4x10^5) Bq/kg[18], we found that we are in the limitations of maximum permissible concentrations. It was also compared with refs. [20,21] which state the permissible concentrations of uranium-238 at another sites in Iraq for crude oil (1.59-6.63 Bq/kg), the permissible concentration of radon (31.6 Bq/kg) in ref. [22], so we noticed that have no effect on the working attach with process of refineries, the lifes of workers, public, the arrounding and the environment, as well as for insurance, the results of our research within the limits of publications the Arab Atomic Energy Authority for Uranium-238, is 5.5 Bq/gm and for radon contents is 1.1 Bq/gm [23,24]. The higher concentrations of uranium-238 in the products were in Bq/gm in our current study (0.928 Bq/gm) for Al-kasik refinery and (0.862 Bq/gm) for Al-qayara,refinery.

Table1- shows the (ID & type) of samples and their locations in the refineries and coordinates for each location.

Samples ID	Type of samples	Location	the coordinates of the location
O1	Crude Oil	Qayara refinery	(35° 47.409'N 43° 16.908'E)
O2	sial asphalt	Qayara refinery	(35° 47.327'N 43° 16.687'E)
O3	hard asphalt	Qayara refinery	(35° 47.260'N 43° 16.788'E)
O4	light gas oil	Qayara refinery	(35° 47.241'N 43° 16.827'E)
O5	heavy gas oil	Qayara refinery	(35° 47.212'N 43° 16.884'E)
O6	Crude Oil	Alkasik refinery	(36° 27.491'N 42° 40.376'E)
O7	Kerosene	Alkasik refinery	(36° 27.530'N 42° 40.387'E)
O8	Naphtha+water	Alkasik refinery	(36° 27.563'N 42° 40.347'E)
O9	RCR	Alkasik refinery	(36° 27.623'N 42° 40.311'E)
O10	Diesel	Alkasik refinery	(36° 27.489'N 42° 40.168'E)
O11	Light Naphtha	Alkasik refinery	(36° 27.434'N 42° 40.110'E)
O12	Diesel	Alkasik refinery	(36° 27.426'N 42° 40.277'E)
O13	Fuel Oil	Alkasik refinery	(36° 27.394'N 42° 40.266'E)

Table 2- illustrates the sample location, number of impacts, and radon concentration in the air and samples.

Sample ID	Location	Samples Type	p ($Tr.cm^{-2}$)	Ca ($Bq.m^{-3}10^3$)	Cs ($Bq.m^{-3}10^3$)	Sample W_U (gm)
O1	Qayara refinery	Grud oil	1049	0.619	8.418	70.1
O2	Qayara refinery	Petroleum product	1162	0.686	9.329	71.02
O3	Qayara refinery	Petroleum product	280	0.165	2.250	52.3

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O4	Qayara refinery	Petroleum product	207	0.122	1.663	74.11
O5	Qayara refinery	Petroleum product	642	0.379	5.159	52.3
O6	Alkasik refinery	Grude oil	1361	0.804	10.937	58.98
O7	Alkasik refinery	Petroleum product	293	0.173	2.352	53.84
O8	Alkasik refinery	Petroleum product	438	0.258	3.508	47.9
O9	Alkasik refinery	Petroleum product	1068	0.631	8.581	60.2
O10	Alkasik refinery	Petroleum product	1033	0.610	8.301	56.07
O11	Alkasik refinery	Petroleum product	582	0.343	4.677	49.4
O12	Alkasik refinery	Petroleumproduct	241	0.142	1.931	53.9
O13	Alkasik refinery	Petroleum product	946	0.559	7.602	54.9

Table 3- the radioactivity of radon (Bq), the radioactivity of radon (Bq / L), the number of uranium atoms, the mass of uranium in the samples, the concentration of uranium(ppm)in the samples, and the alpha index.

Sample ID	**Location**	**Samples type**	**ρ ($Tr.cm^{-2}$)**	**A_{Rn}^{222} (Bq)**	**A_{Rn}^{222} Bq/kg**	**N_{Rn}^{222} (Bq/Kg)**	**N_U $*10^{17}$**	**W_U $*10^{-6}$**	**C_W PPm**
O1	Qayara refinery	Grud oil	1049	0.687	9.8	0.327	1.401	55.36	0.789
O2	Qayara refinery	Petroleum product	1162	0.761	10.728	0.362	1.55	61.24	0.862
O3	Qayara refinery	Petroleum product	280	0.183	2.581	0.087	0.374	14.77	0.282
O4	Qayara refinery	Petroleum product	207	0.135	2.469	0.064	0.247	10.82	0.145
O5	Qayara refinery	Petroleum product	642	0.421	7.099	0.200	0.857	33.86	0.57
O6	Alkasik refinery	Grud oil	1361	0.893	15.141	0.425	1.734	68.51	1.161
O7	Alkasik refinery	Petroleum product	293	0.192	3.566	0.091	0.39	15.41	0.286
O8	Alkasik refinery	Petroleum product	438	0.286	5.97	0.136	0.582	22.99	0.451
O9	Alkasik refinery	Petroleum product	1068	0.7	11.627	0.333	1.414	55.87	0.928
O10	Alkasik refinery	Petroleum product	1033	0.677	12.07	0.322	1.37	54.13	0.922
O11	Alkasik refinery	Petroleum product	582	0.381	7.71	0.181	0.775	30.62	0.603
O12	Alkasik refinery	Petroleumproduct	241	0.157	2.91	0.074	0.317	12.52	0.232
O13	Alkasik refinery	Petroleum product	946	0.620	11.29	0.295	1.264	49.94	0.909

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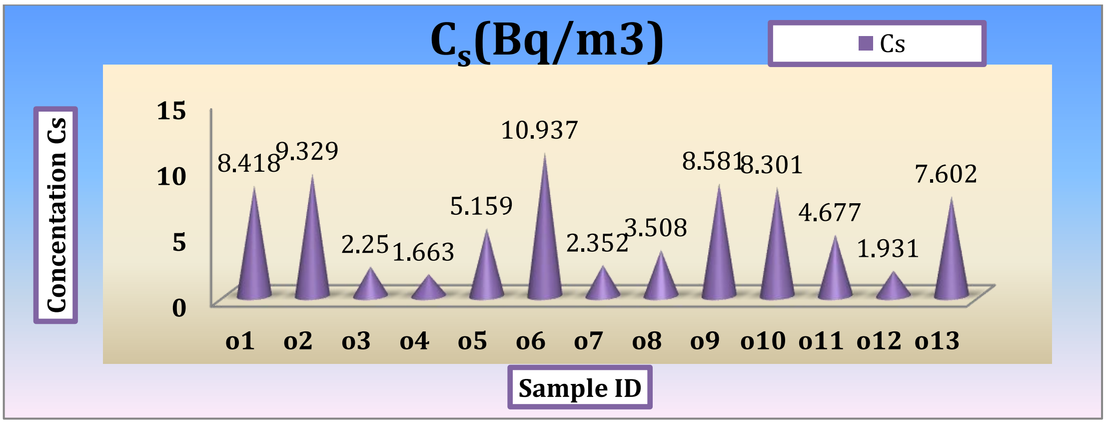
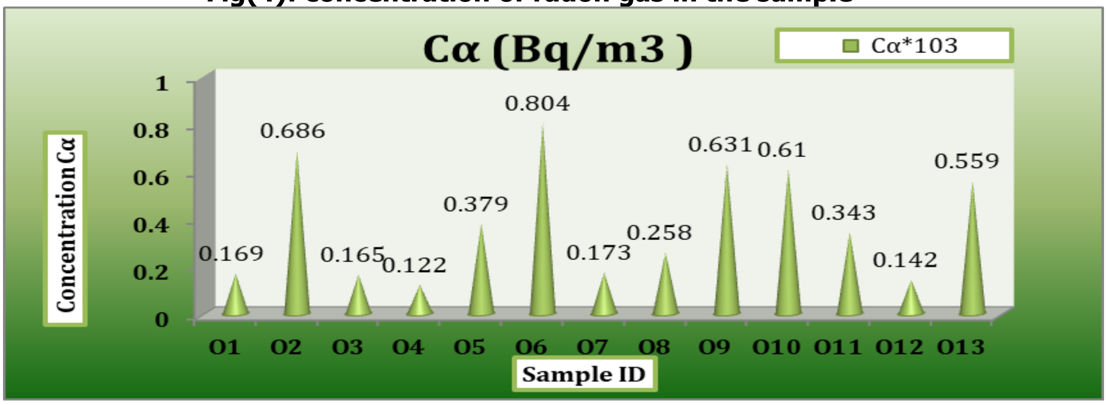
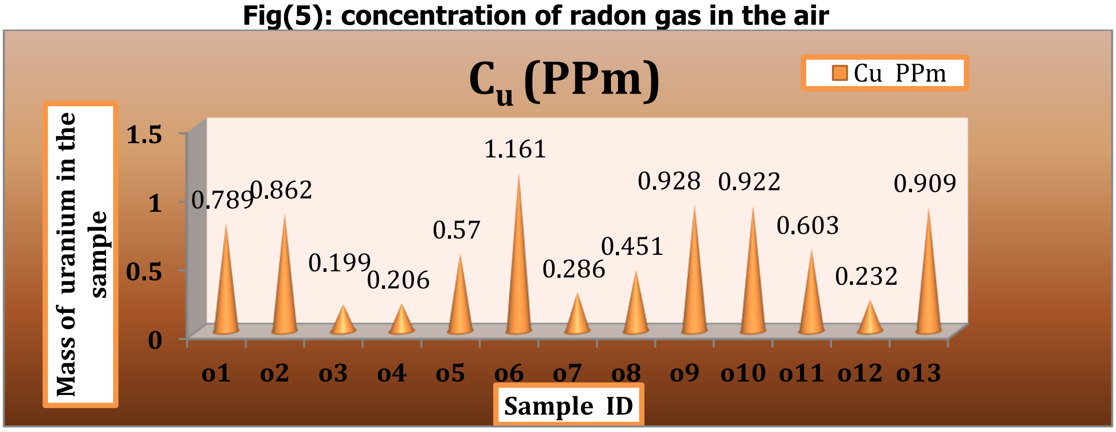


Fig(4): concentration of radon gas in the sample



Fig(5): concentration of radon gas in the air



Fig(6): The concentration of uranium in the sample

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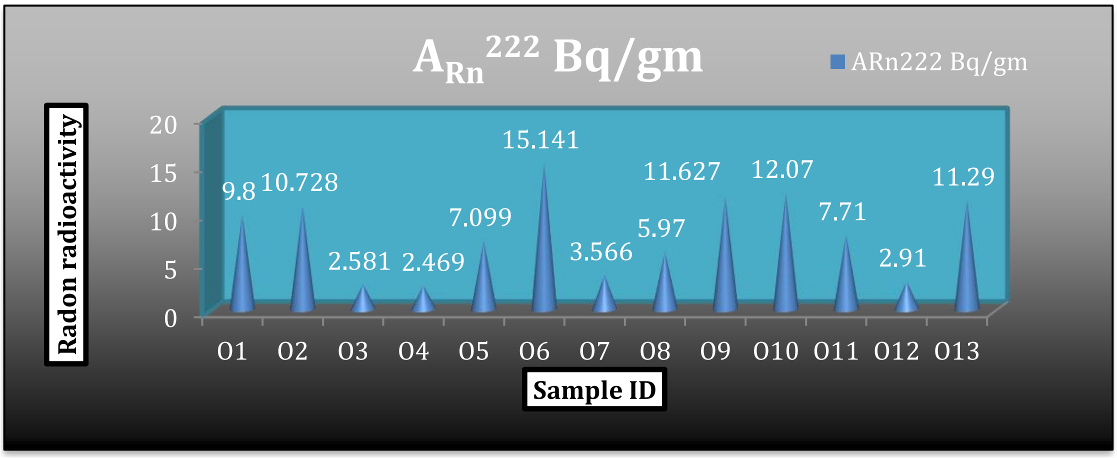

Fig(7): The radioactivity of radon gas in the sample

CONCLUSIONS:

The radioactivity and concentration of uranium radon gas in crude oil and petroleum products after refining were detected using a CR-39 detector at the qayara and Al-kasik refineries. The results show that it is within the worldwide limit. Maintaining the amounts of Naturally Occurring Radioactive Materials, however, need continual monitoring.

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