

## Measurements of Radiation Level around the Location of NORM in Solid Wastes at Petroleum Companies in Egypt

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**Abstract:** Recent concern has been devoted to the hazard arising from Naturally Occurring Radioactive Materials (NORM) which are omnipresent in the earth's crust. These scales contain mainly <sup>226</sup>Ra and its daughter products, which can cause an exposure risk. Fifteen petroleum scales samples were collected at different Oil fields in the Red Sea Refineries (Company) for Petroleum Services in the Eastern Desert of Egypt were investigated. The specific radio activities of <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra, <sup>40</sup>K, and its daughter nuclide for all samples were determined using high-resolution gamma-ray spectrometry. The radium equivalent activity, radiation hazard indices and absorbed dose rate in air for all waste samples were estimated. The radon emanation coefficient of the waste samples and the radon exhalation rate was estimated. Its values equal 0.341547312 gm m<sup>-3</sup> and 0.011153571 (111.5 × 10<sup>-4</sup> Bqm<sup>-2</sup>s<sup>-1</sup>) respectively. The gamma ray dose rates, with the associated occupational doses in the samples, and <sup>226</sup>Ra concentration in hard/soft scale samples were determined. The chemical structure of the waste samples was investigated using X-ray fluorescence technique (XRF) and Sr, Ca, Fe, Ba, Si, Pb, K, Zn, S, Ti, and Mn were found in all samples. From this study, it was noticed that the concentrations of the natural radionuclides in the petroleum scales samples were higher than that of the petroleum sludge samples exceeds the NRC limits. Results obtained are discussed and compared with the international recommended data.

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### 1. Introduction

Radioactive materials which occur naturally and expose people to radiation occur widely, and are known by the acronym 'NORM'. NORM is an acronym for Naturally Occurring Radioactive Material, which includes all radioactive elements found in the environment. Long-lived radioactive elements such as uranium, thorium and potassium.

In 1998, an attention was given to the health impacts from the uncontrolled release of naturally occurring radioactive materials (NORM) wastes on concentrated and accumulated in tubing and surface equipment in the form of scale, sand and sludge [Rood, 1998]. In the exploration and extraction processes of oil and gas, the natural radionuclides <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, as well as the radium- radionuclides (<sup>223</sup>Ra, <sup>224</sup>Ra, <sup>226</sup>Ra and <sup>228</sup>Ra) and <sup>210</sup>Pb, etc., are brought to the slurry surfaces and may contain levels of radioactivity above the surface background [API, 1992; Rood, 1998, 2001; Shawky *et al.*, 2001; Matta *et al.*, 2003; Al-Masri and Suman, 2003; Godoy and Crux, 2003; Hamlat *et al.*, 2003; Mohamad Puad and Muhd Noor, 2004; Omar *et al.*, 2004; El Afifi and Awwad, 2005; Gazineu *et al.*, 2005]. As these materials are handled, their radioactive constituents may be separated, resulting in NORM waste [Testa *et*

*al.*, 1998]. Nuclear spectroscopic analysis showed that the main radionuclides present in NORM waste associated with petroleum industries are <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K series and any of their decay products, such as radium and radon are example of NORM. Radon is a radioactive gas; therefore it follows the general gas law and the theory of radioactivity. Radon has two aspects; it induces significant health hazards to uranium miners and people living in normal buildings. Lung cancer is the principal concern associated with radon exposure [Tanner, 1980 and UNSCEAR, 1988]. Radon itself is not responsible for the hazard. Being chemically inert, it does not accumulate to a great degree in the body. Therefore, the primary concern is associated with the short-lived decay products of radon. These species are chemically reactive and so may be deposited on respiratory tract tissues when inhaled may damage cells near the deposition site, contributing to increase the risk of lung cancer [Nazaroff 1992]. The radon concentration inside the container was continuously detected by an AlphaGuard radon monitor (Genitron Instruments GmbH. Co. Frankfurt am Main Germany) for 67 hours see (Fig. 5). These elements have always been present in the Earth's crust and within tissues of all living beings. The philosophy of radiation is to limit NORM locations to a minimum and to

concentrate contaminated equipment/waste into one area such as the NORM storage. NORMS in the Petroleum Industry are present in components of both petroleum production facilities and natural gas production facilities. The mineralogical analysis by X-ray techniques (XRF) indicated the incorporation and co-precipitation of these radionuclides with the alkaline earth metals (e.g. Mn, Ca, Sr, and Ba) and some quantities of lead sulphate, carbonate and/or silicate [Shuller *et al.*, 1995]. This work was necessary since no studies have been carried out on radioactivity levels in petroleum scales and wastes in Egypt and published data are not available. The present study includes the determination of the activity concentrations of the  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$ , content in the petroleum scales and wastes in Egypt City Refinery. The radium equivalent activity, radiation hazard indices, Annual Effective Dose  $\mu\text{Sv}$  and absorbed dose rate in air for all waste samples were estimated. The radon emanation coefficient of the waste samples was estimated. The chemical structure of the waste samples was investigated using X-ray fluorescence analysis (XRF) see Fig. (7). We did not find any gamma spectroscopy data in the literature on radiation levels in petroleum products, only on their waste products. Human beings have always been exposed to natural radiations from their surrounding. The exposure to ionizing radiations from natural sources occurs because of naturally occurring radioactive elements in the soil and rocks, cosmic rays entering the earth's atmosphere from outer space and the internal exposure from radioactive elements through food, water and air. Therefore the assessment of gamma radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population [UNCEAR, 1998, California SOP, 1999]. Wherever the petroleum scales samples is used in many industrial purposes. Consequently there are specific measures to protect the working in the petroleum scales samples related industry such as reducing exposure levels and time of exposure and the use of exhaust ventilation. So the aim of this work is to determine the concentration of natural radioactivity uranium, thorium and potassium in the petroleum scales samples and to measure the surface radiation dose rate and the radium equivalent activity and radiation hazard index.

### Geological Origin

U and Th are present in the earth's crust at an average concentration of 4.2 and 12.5 ppm, respectively [Wollenberg and Smith, 1990]. When a geological formation containing  $^{238}\text{U}$  and  $^{232}\text{Th}$  has not been disturbed (closed system) for more than a million years, the members of the individual decay series will have the same activity (Bq/kg) which is known as secular equilibrium. However, when the geological

formation is not closed to radionuclide migration,  $^{226}\text{Ra}$  can migrate and be deposited somewhere outside the formation. Then secular equilibrium will not exist and the growth of  $^{226}\text{Ra}$  by radioactive decay of its ancestors will not occur.  $^{226}\text{Ra}$  is said to be unsupported. See the average radium, uranium, thorium and potassium content in sedimentary rocks [NCRP, 1975]. Oil is formed by thermal cracking of organic matter (kerogen) trapped in sedimentary rock. As oil migrates, naturally occurring radionuclides (NORs) may be taken up in the up flowing fluid stream and may be transferred into connate waters, which may be produced with oil as production water. During thermal cracking of kerogen, uranium or thorium remain with the residual organic matter and they will not be leached in a reducing environment into passing fluids. Radium will not leach into a hydrocarbon phase, but it may leach into the aqueous phase [Bloch and Key, 1981]. In this work, NORs in scale/sludge originating from oil fields in the western desert and the Red Sea region have been characterized and compared with the exempt concentration levels given by the IAEA [IAEA, 1988, 1994].

### 2. Material and Methods

Fifteen petroleum scales samples were collected at different Oil fields in the Red Sea Refineries (Company) for Petroleum Services in the Eastern Desert of Egypt, Egyptian General Authority for Petroleum Resources, Ministry of Petroleum. The Scale and Sludge samples put into clean containers. Sample were stored for 30 days before its counting radioactivity to  $^{224,226}\text{Ra}$  period to achieve the secular equilibrium  $\lambda_B \gg \lambda_A$  between radium and its products and then measure the samples 3600 sec. The energy and intensity of various gamma-ray lines have been measured using a system consist of Canberra coaxial High-Purity Germanium detector (HPGe) which has a photo peak efficiency of 70%. The energy resolution of 2 keV Full-Width at Half Maximum (FWHM) for the 1332 keV gamma-ray line of  $^{60}\text{Co}$ . A cylindrical lead shield of 5 cm thickness, which contains inner concentric cylinder of Cu with thickness of 10 mm, was used to shield the detector and to reduce the effect of background. The detector was cooled to liquid nitrogen temperatures and coupled to a PC-based 8K multichannel analyzer and an ADC with Genie 2000 for data acquisition and analysis. The calibration of the detector was carried out by using standard point sources  $^{60}\text{Co}$  (1173.2 and 1332.5 keV),  $^{133}\text{Ba}$  (356.1 keV),  $^{137}\text{Cs}$  (661.9 keV) and  $^{22}\text{Na}$  (1368.6keV) besides  $^{226}\text{Ra}$  (186.2keV). Absolute efficiency calibration curves are calculated for activity determination of the sample by using standard  $^{226}\text{Ra}$ , contained in the same cylindrical bottles as the samples. The samples were prepared with a uniform geometry. An empty bottle with the same geometry

was measured for subtracting the background. The gamma-ray transitions of energies 1120.3 keV ( $^{214}\text{Bi}$ ) and 1764 keV ( $^{214}\text{Bi}$ ) were used to determine the concentration of the  $^{238}\text{U}$  series. The gamma-ray transitions of energies 911.1 keV ( $^{228}\text{Ac}$ ) and 2614 keV ( $^{208}\text{Tl}$ ) were used to determine the concentration of the ( $^{232}\text{Th}$ ) series. The 1460 keV gamma-ray transition of  $^{40}\text{K}$  was used to determine the concentration of  $^{40}\text{K}$  in the samples as shown in Table (1) and their intensities. The spectra of the samples were perfectly analyzed

using a special PC Genie 2000 software to calculate the concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and their decay products

### 2.1. Gamma-Ray Spectrometer System

The instrumentation used to measure  $\gamma$ -rays from radioactive samples consists of a HPGe semiconductor detector, associated electronics, and a computer-based multichannel analyzer [ Verma, 2007] as shown in Fig. ( 1 )

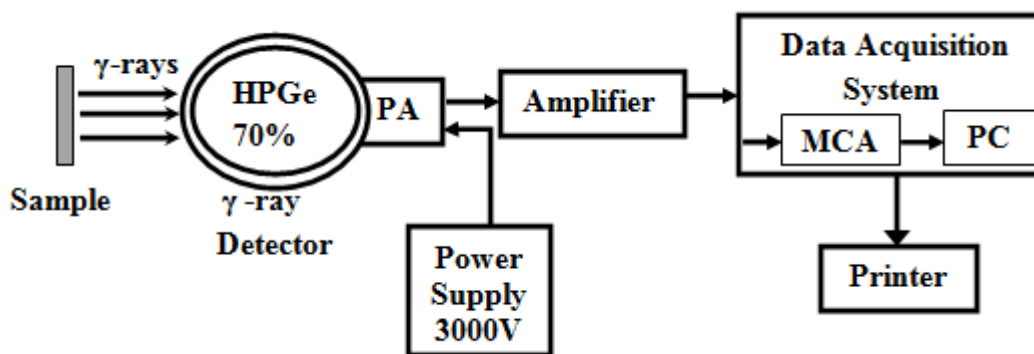


Fig.1: Blocked diagram of HPGe  $\gamma$ -ray spectrometer system.

### 2.2. Sample Collection and Preparation

Samples were collected from different places in one of the work sites of Egyptian Petroleum location. Each sample, 50- 400g, was packed in a plastic container, sealed well and stored for 30

days before analysis this allow the in-growth of uranium and thorium decay products and prevent the escape of radiogenic gases  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  and allow secular equilibrium of  $^{238}\text{U}$  and it's decay products see Table (1).

Table (1): The natural radionuclides, their gamma lines used and their intensities [VIENNA, 1990].

Parent Nuclide	Max. Activity According to (UNSCEAR)	Daughter nuclide	$\gamma$ -ray energy (keV)	Abundance (%)
$^{238}\text{U}$	130 Bq/Kg	$^{226}\text{Ra}$	186.2	3.29
		$^{214}\text{Pb}$	295.2	18.7
		$^{214}\text{Pb}$	351.9	35.8
		$^{214}\text{Bi}$	609.3	45.0
		$^{214}\text{Bi}$	1120.3	14.9
		$^{214}\text{Bi}$	1764.5	16.0
		$^{214}\text{Bi}$	2204.1	5.0
$^{232}\text{Th}$	85 Bq/Kg	$^{212}\text{Pb}$	238.6	45.0
		$^{208}\text{Tl}$	583.1	30.0
		$^{228}\text{Ac}$	911.1	29.0
		$^{228}\text{Ac}$	968.6	17.5
		$^{208}\text{Tl}$	2614.7	36.0
$^{40}\text{K}$	1600 Bq/Kg		1460	10.67

## 3. Results and Discussion

### 3.1. Natural specific activity measurement

The activity levels for radionuclides in the measured samples are computed using the following equation [ Amrani 2001 ]

$$A = C_R / \epsilon(E) I_Y W \quad (1)$$

Where:

$A$  = The activity level of a certain radionuclide (Bq/ kg)

$C_R$  = The net count rate of the sample (counts / seconds)

$\mathcal{E}(E)$  = The detector efficiency for the specific gamma ray energy

$I_\gamma$  = The intensity of gamma-line in a radionuclide

$W$  = The dried sample weight in kg.

Activities due to the presence of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides have been determined in the samples. The minimum, maximum and mean activity values of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  found in these samples are listed in Table 2. As may be seen in this table the measured values of activity in the samples due to  $^{232}\text{Th}$  vary from  $34339\text{ Bqkg}^{-1}$  to  $427\text{ Bqkg}^{-1}$ ,  $^{226}\text{Ra}$  activities vary from  $285823\text{ Bqkg}^{-1}$  to  $66\text{ Bqkg}^{-1}$  and variation in  $^{40}\text{K}$  activities ranges from  $1031\text{ Bqkg}^{-1}$  to  $51\text{ Bqkg}^{-1}$ .

The activity concentration is shown in Table 2 where all activity is very higher than world average except value.

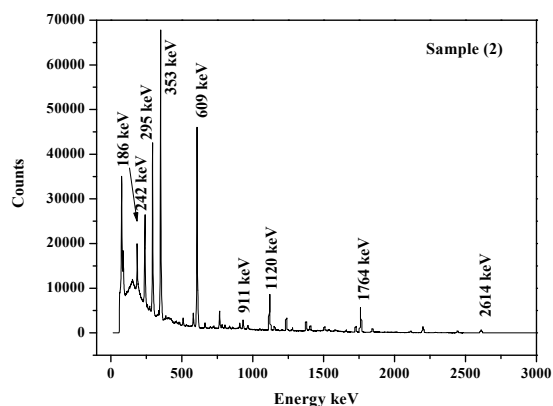
**Table 2:** Minimum, maximum and mean activity concentration values for scales and sludge samples.

Radionuclide	Minimum (Bq / kg)	Maximum (Bq / kg)	Medium (Bq / kg)
<b>Scales</b>			
$^{238}\text{U}$	9140	285823	147481.5
$^{232}\text{Th}$	427	34339	17383
$^{40}\text{K}$	51	1031	541
<b>Sludge</b>			
$^{238}\text{U}$	66	1567	816.5
$^{232}\text{Th}$	0	0	0
$^{40}\text{K}$	787	1544	1165.5

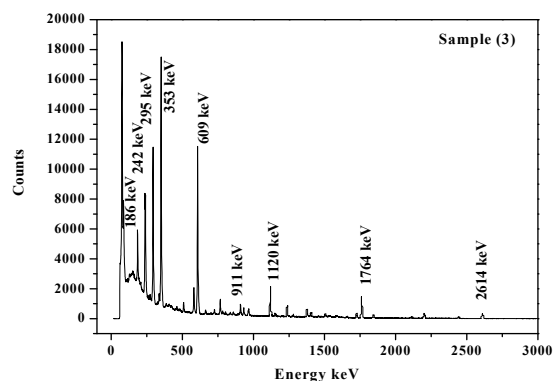
For  $^{238}\text{U}$  activity concentration was determined by measuring the 295.2 keV (18.7%) and 351.9 keV (35.8.1 %) gamma-rays from  $^{214}\text{Pb}$  and the 609.3 keV (45 %) and 1120.3 keV (14.9%) gamma-rays from  $^{214}\text{Bi}$ .  $^{232}\text{Th}$  activity was determined from the gamma-rays of 238.6 keV (45 %) from  $^{212}\text{Pb}$  and 338.4 keV (12 %), 911.1 keV (29 %) and 968.6 keV (17.5 %) from  $^{208}\text{Tl}$  and 583.1 keV (30 %) gamma-rays from  $^{208}\text{Tl}$ .  $^{40}\text{K}$  concentration was measured from its 1460 keV (10.67%) gamma-ray line.

The obtained spectrum of the background gamma radiation was subtracted from the measured gamma ray spectra of the samples. The characteristic gamma-ray emitters are marked above the corresponding peaks.

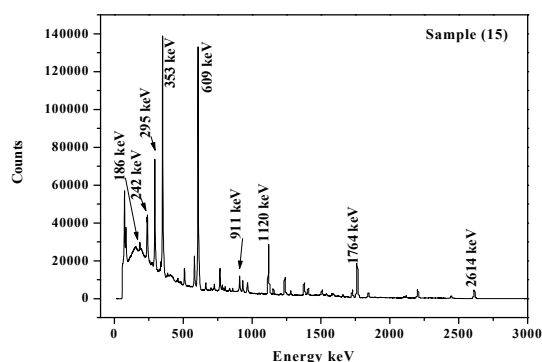
A selected one of the obtained spectrum for sample is shown in Figs. 2, 3 and 4



**Figure (2) :** The gamma ray spectrum of NORM Petroleum sample code (2).



**Figure (3) :** The gamma ray spectrum of NORM Petroleum sample code (3).



**Figure (4) :** The gamma ray spectrum of NORM Petroleum sample code (15).

To assess the radiological hazard of the scales sample, it is useful to calculate an index called the radium equivalent activity,  $Ra_{eq}$ , defined according to the estimation that 1 Bq / kg of  $^{226}\text{Ra}$ , 1.43 Bq / kg of  $^{232}\text{Th}$  and 0.077 Bq / kg of  $^{40}\text{K}$  produce the same  $\gamma$ -ray dose [Amrani, 2001]. This index  $Ra_{eq}$  is given as:

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_K \quad (2)$$

Where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentration in  $\text{Bqkg}^{-1}$  of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively. The maximum value of  $Ra_{eq}$  in petroleum scales samples must be less than 370 Bq  $\text{kg}^{-1}$  for safe use [UNSCEAR, 1993], i.e., to keep the external dose below 1.5 mSv  $\text{y}^{-1}$ . The values of  $Ra_{eq}$  are higher than this criterion limit. In petroleum scales samples, the  $Ra_{eq}$  activity are not within the recommended safety limit when used

in industry. The calculated values of the radium equivalent  $Ra_{eq}$  for the studied petroleum scales samples are given in Table 3.

Another radiation hazard index, the representative level index,  $I_{\gamma r}$ , used to estimate the levels of  $\gamma$ -radiation hazard associated with the natural radionuclides in specific samples, is defined as [KAFALA, 2007].

$$I_{\gamma r} = (A_{Ra} / 150) + (A_{Th} / 100) + (A_K / 1500) \quad (3)$$

Where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentrations in  $\text{Bq/kg}$  of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively. The values of  $I_{\gamma r}$  for the studied samples are given in table 3. It is clear that the studied samples exceed the upper limit for the representative level which is unity.

**Table ( 3):** Activity Concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , Radium equivalent activity ( $\text{Bqkg}^{-1}$ ), External Annual dose ( $\text{mSv/y}$ ), Gamma-Radiation external and internal hazard ( $I_{\gamma r}$ ,  $H_{in}$ ), Absorbed dose ( $\text{nGy/h}$ ) and the Annual Effective Dose ( $\mu\text{Sv}$ ).

Sample code	$^{238}\text{U}$ (Bq/kg)	$^{232}\text{Th}$ (Bq/kg)	$^{40}\text{K}$ (Bq/kg)	Dose Rates nGy/h)	Ra equiv.	EAD ( $\mu\text{Sv}$ )	$H_{in}$	$I_{\gamma r}$
1	15152	-	-	6471	15152	0.0684	95.899	40.951
2	172563	6769	1031	78209	181712.847	0.8157	1118.520	492.736
3	86060	6216	657	40892	94440.029	0.4222	568.820	256.731
4	66	-	1103	75	150.931	0.0007	0.647	0.408
5	240	-	787	136	300.599	0.0014	1.683	0.812
6	9140	-	-	3915	9140	0.0413	57.848	24.703
7	595	-	1544	320	713.888	0.0033	4.087	1.929
8	10209	427	51	4644	10785.107	0.0484	66.273	29.251
9	316	-	-	135	316	0.0014	2.000	0.854
10	1567	-	-	669	1567	0.0071	9.918	4.235
11	77	-	-	32	77	0.0003	0.487	0.208
12	-	-	-	13	0	0.0000	0.000	0.000
13	101024	12954	-	51714	118382.36	0.5258	689.408	323.053
14	9452	9687	-	46774	22432.58	0.0949	97.224	62.947
15	285823	34339	-	144778	331837.26	1.4753	1941.589	905.078

### 3.2. Radiation Hazard Index

This factor is used to estimate the level of gamma radiation hazard associated with natural radionuclides in specific petroleum scales samples. The external hazard index is obtained from  $Ra_{eq}$  expression through the assumption that its maximum value allowed (equal to unity) corresponds to the upper limit of  $Ra_{eq}$  ( $370 \text{ Bqkg}^{-1}$ )

according to UNSCEAR, 1993. This index value must be less than unity in order to keep the radiation hazard insignificant; then, the internal hazard index ( $H_{in}$ ) can be defined also as the potential radiological hazard posed by the different samples was calculated using the following equation [Ngachina, 2007],

$$H_{in} = (A_{Ra} / 185) + (A_{Th} / 259) + (A_K / 4810) \quad (4)$$

Where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (in  $\text{Bq}\cdot\text{kg}^{-1}$ ) were calculated for the investigated samples to indicate different levels of external  $\gamma$ -radiation due to different combinations of specific natural activities in specific petroleum scales samples.

### 3.3. External Annual Dose

The external annual effective dose (EAD) is calculated for a room with dimensions of  $4\text{ m} \times 5\text{ m} \times 2.8\text{ m}$ , estimated that the samples is put. The equation used to calculate the annual effective dose may be defined as [Ngachina, 2007 and Hussain et al., 2010] The external annual effective dose (EAD) is calculated,

$$\text{EAD} = (0.92 A_{Ra} + 1.1 A_{Th} + 0.08 A_K) \times 10^{-9} (\text{Gy/h}) \times (0.7 \text{ Sv/Gy}) (24 \times 365 \times 0.8 / \text{y}) \quad (5)$$

Where, 0.92, 1.1 and 0.08 are the specific dose rates of Ra, Th and K, respectively; with an estimated indoor occupancy factor of 0.8.

The variation of the activity concentration ( $\text{Bq/kg}$ ) of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  radionuclides in the collected samples in location of Petroleum Company is represented in Table (3).

The absorbed dose in tissues are calculate using the conversion factor in ( $\text{nGy/h}$ ), which are Known to be  $C_K = 0.0437$  for K,  $C_{Th} = 0.662$  for  $^{232}\text{Th}$  and  $C_U = 0.427$  for  $^{238}\text{U}$

The equivalent dose rate is calculated according to the equation:-

$$D = R_U C_U + R_{Th} C_{Th} + R_K C_K \quad (6)$$

Where  $R_U$ ,  $R_{Th}$ , and  $R_K$  are the conversion factors expressed in activity of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{Sv/h}$

$$R_U = 0.427, R_{Th} = 0.662 \text{ and } R_K = 0.0437$$

$C_U$ ,  $C_{Th}$  and  $C_K$  are the specific activity of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively expressed in  $\text{Bq/kg}$ .

Figure (2- 4) for sample code 2, 3 and 15 show the spectrum of the energy lines for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  from their decay products  $\gamma$ -lines energy.

The U-283, Th-232 and K-40 in NORM samples activity concentration values reported in this study are higher than the international recommended limits.

### 3.5. Radon Emanation Coefficient and Radon Exhalation Rate of the Waste Samples (Active Method)

The active method for measuring radon exhalation rate is carried out in Zagazig University, Faculty of Science, Physics Department by using the pulse type ionization chamber (Alpha Guard, Genitor Instruments, Frankfurt, Germany). Alpha Guard was calibrated in Egyptian National Institute for Standard. Each sample and Alpha Guard monitor were placed together at the same time in the Alpha Guard chamber as shown Fig. 6. Alpha Guard chamber is a container consisting of a firm corrosion-resisting stainless steel container with a removable gas-tight lid. The container dimensions were 45.0 cm diameter and 31.7 cm height. Its volume was 50.4 liters. The lid was equipped with three gas-tight electric ducts. One duct server, together with a special charger, was used as a power supply for the radon analyzer or monitor. The second duct was used to connect the fan in the middle of the inner side of the lid to the power supply, by means of a power adapter. The fan was used to ensure an even distribution of the radon exhalation from the sample in the interior of the container. The third duct provided communication between the Alpha Guard in the interior of the container and an external PC. The concentration of radon emanated from each sample inside the exhalation container was allowed to build up with time and, was measured in every one hour in diffusion mode of Alpha Guard monitor system to avoid thoron gas concentration effect for an average time of 3 days.

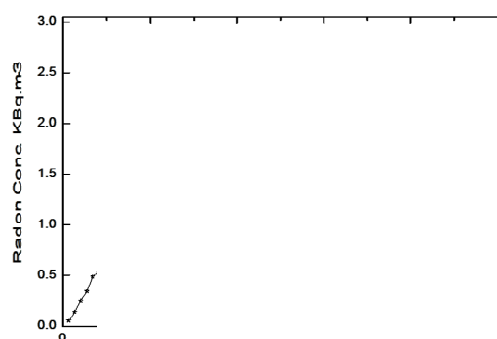


Fig. ( 5 ): Radon growth curve for sample code number 2 as an example.

For the Alpha Guard (diffusion mode), the sensitivity for thoron is about 10% of radon sensitivity [ISHIKAWA, 2004]. Moreover, the typical petroleum samples show the radium concentration is higher than thorium. Even if we assume the radon and thoron exhalation rate are the same, the effects of thoron on measured concentrations can be neglected. A

direct measurement of radon concentration for each sample is obtained. Alpha Guard was used to measure the radon concentration at a time  $t$  during the growth of radon inside the chamber. From the radon concentration, the equilibrium concentration of radon for each sample (the saturated constant radon

concentration in the sealed space which is the expected concentration at  $t > 30$  days) can be estimated. The radon exhalation rate of any sample, is defined as the flux of radon released from the surface of material, was also calculated.

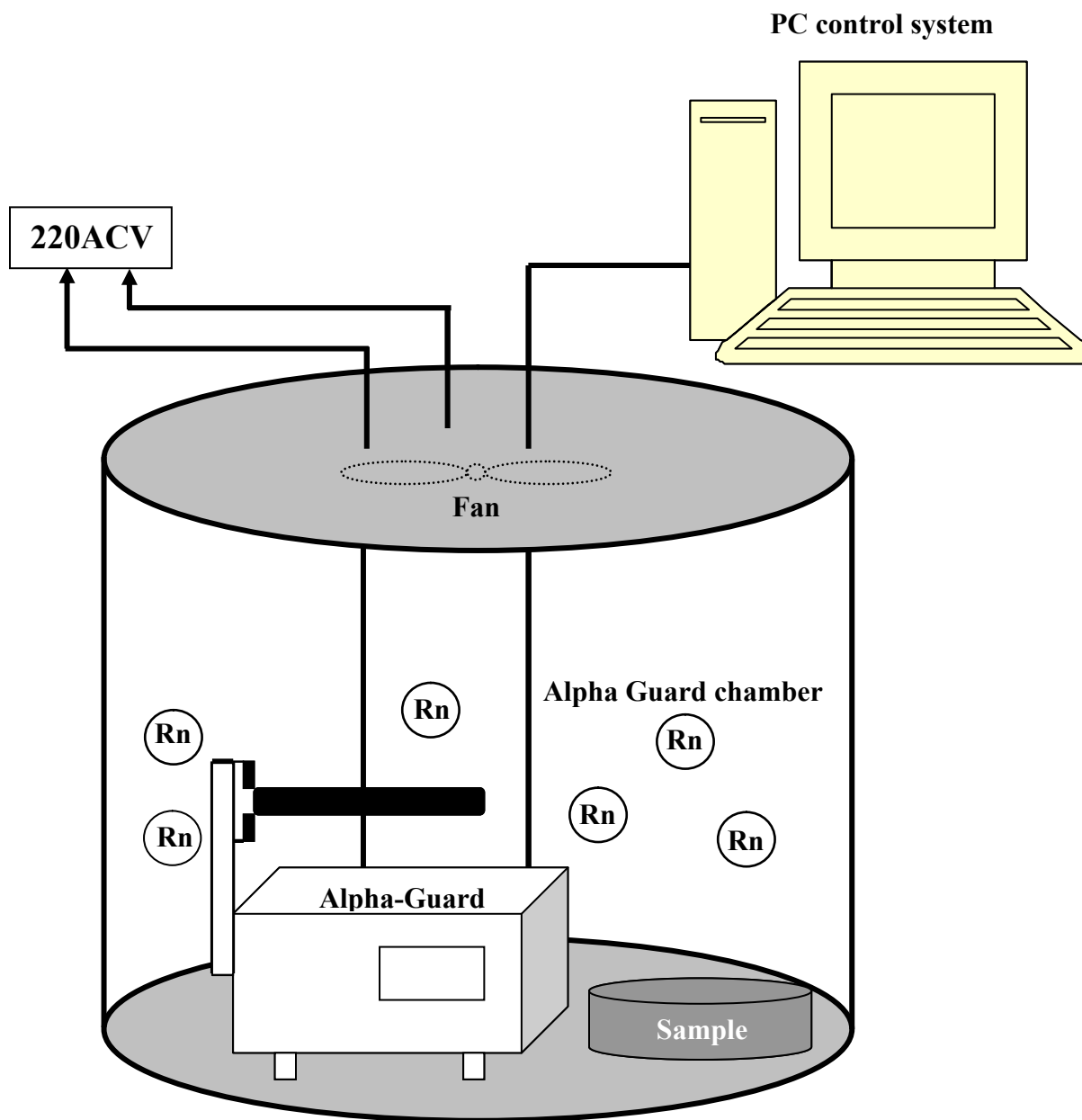


Fig. ( 6 ): Active technique set up for measuring radon exhalation rate.

To determine the radon ( $^{222}\text{Rn}$ ) emanation coefficient (EC), the samples code number 2 were initially counted for 3 days, and counted again after reaching the radioactive equilibrium between  $^{222}\text{Rn}$  decayed from  $^{226}\text{Ra}$  and its respective short-life daughters. The  $^{222}\text{Rn}(\text{EC})$  was determined using the formula described by [White and Rood, 2001; El Afifi and Awwad, 2005].

$$^{222}\text{Rn}(\text{EC}) = N_{\text{Rn}} \cdot m / (N_{\text{Ra}} \cdot V) \quad (7)$$

$$E = N_{\text{Rn}} \cdot V \cdot \lambda / \text{sample surface area} \quad (8)$$

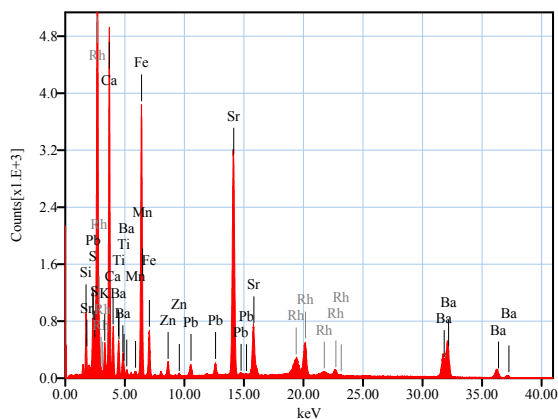
where  $^{222}\text{Rn}(\text{EC})$  is the radon emanation coefficient,  $N_{\text{Rn}}$  is the equilibrium radon concentration ( $6940 \pm 92$  Bq),  $N_{\text{Ra}}$  is the Radium concentration, (172563 Bq),  $\lambda$  is radon decay constant ( $2.1 \times 10^{-6} \text{ s}^{-1}$ ),  $V$  is the Sample volume ( $0.011775 \text{ m}^3$ ),  $m$  is the mass sample (85gm) and  $E$  is the radon exhalation rate. The value of  $\text{Rn}(\text{EC})$  and  $E$  from equation 7, 8 is equal to  $0.341547312 \text{ gm m}^{-3}$  and  $111.5 \times 10^{-4} \text{ Bq m}^{-2} \text{ s}^{-1}$  respectively.

### 3.6. XRF analysis

Table (4) and Fig. (7), represents the analysis of the waste petroleum samples using the XRF technique. The data showed major elements (Sr, Ba, Si, Pb, K, Zn, S, and Ti) and alkaline earth elements (Mn and Ca) as well as trace amounts of Fe in all the samples.

**Table (4):** Results of analysis of the waste petroleum samples using XRF-technique.

Element	ms %	Sigma	Intensity
Si	9.2928	0.9662	9284
S	1.4606	0.2263	3686
K	3.6806	0.6623	5239
Ca	42.7984	0.5323	70639
Ti	1.4246	0.4729	2358
Mn	0.5073	0.2631	1453
Fe	21.1092	0.2374	67435
Zn	0.7863	0.1878	4049
Sr	6.7239	0.1279	84388
Rh	-	-	-
Ba	10.8879	1.2263	12739
Pb	1.3285	0.3149	7841
	~100		



**Fig. (7):** Illustrates the analysis in one of the petroleum samples using XRF technique

## 4. Conclusions

An investigation was carried out to find out the concentration of Naturally Occurring Radioactive Materials (NORMs) in petroleum scales samples which collected at different Oil fields in the Red Sea Refineries (Company) for Petroleum Services in the Eastern Desert of Egypt. Activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in scales and sludge generated during oil extraction and production operations were determined using an HPGe gamma spectrometric system. Concentrations ranged from  $66$  to  $285823 \text{ Bq kg}^{-1}$  for  $^{238}\text{U}$ ,  $427$  to  $34339 \text{ Bq kg}^{-1}$  for  $^{232}\text{Th}$  and  $51$  to  $1031 \text{ Bq kg}^{-1}$  for  $^{40}\text{K}$ . The magnitude of these results demonstrates the need of screening oil residues for their radionuclide content in order to decide about their final disposal. While NORM-contaminated equipment has been a concern in the Red Sea Refineries (Company) for Petroleum Services in the Eastern Desert of Egypt well drilling, the results of this investigation show that NORM contamination of Egypt equipment is significant. Egypt well drilling equipment and wastes constitute a health risk for the country residents a potential degradation of the country environment. From the present results, it may be concluded that, for different samples NORM around location of petroleum companies, the level of natural radioactivity and hazard parameters are higher than the international recommended limits.

A recommendation that risks should be reduced to account for lower-dose-rate exposures:

- The workers at oil fields are the most threatened. Therefore, they should be classified as occupationally radiation workers.
- Such workers should follow approved radiation protection regulations and receive regular medical surveillance.
- The sites of NORM activities require qualified radiation protection experts to safeguard workers and environment from exposure and contaminations during all stages of oil production, separation and maintenance.
- Restricted regulations should be applied for the workers that performing cleaning of the contaminated equipment.

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