Republic of Iraq
Minister of Higher Education &
Scientific Research
AL-Nahrain University
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A study of the natural radioactivity of the petroleum waste.

A Thesis
Submitted to the College of Science Al-Nahrain University
in Partial Fulfillment of the Requirements for the Degree of
Master of Science in Physics

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Jamad al-Awal 1431
April 2010

بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

﴿رَبِّ أَوْزِعْنِي أَنْ أَشْكُرَ نِعْمَتَكَ الَّتِي أَنْعَمْتَ

عَلَيَّ وَعَلَى وَالِدَيَّ وَأَنْ أَعْمَلَ صَالِحًا تَرْضَاهُ

وَأَدْخِلْنِي بِرَحْمَتِكَ فِي عِبَادِكَ الصَّالِحِينَ﴾

صَدَقَ اللَّهُ الْعَظِيمُ

﴿سورة النمل، أيه: 19﴾

Abstract

The aim of this study is to detect the radioactivity of radionuclides and measure the specific activities and the uranium concentration in (30) samples of crude oil and oil waste were brought from three oil locations Dora refinery, Bijie refinery, and north field of al-Rumaila.

The measurements in this study were done by using two techniques:

1- Gamma rays spectroscopy system was used to measure the specific activity for radionuclides. This system is consist of high purity of germanium detector (HPGe) with personal computer as multi channel analyzer (MCA) that computerized to plot the radioactive spectrum and compute the specific activities of the radionuclides. There are seven radionuclides are detected:

(Bi-214, Ra-226, TI-208, Bi-212, Pb-212, K-40, and Cs-137), these radionuclides included; two radionuclides (Bi-214,Ra-226) belong to the U-238 series, three radionuclides (TI-208, Bi-212, Pb-212) belong to Th-232series, one natural radionuclide (K-40), and one artificial radionuclide (Cs-137).

For Dora refinery samples, the averages of specific activities of (Bi-214, Ra-226) were (7.79, 16.08) Bq/kg respectively, the averages of specific activities of (TI-208, Bi-212, Pb-212) were (7.95,4.05,5.75) Bq/kg , the average of specific activities of (Cs-137) was (0.64) Bq/kg , the average of specific activities of (K-40) was (99.3) Bq/kg.

For Bijie refinery samples, the average of specific activities of (Bi-214) was (0.39) Bq/kg, the averages specific activities of (Bi-212, Pb-212) were (0.02,0.01) Bq/kg, the average of specific activities of (K-40) was (2.28) Bq/kg , and three radionuclides (Ra-226, TI-208, and Cs-137) were not appeared in Bijie samples.

2- solid state nuclear track detectors technique (SSNTDs), the uranium concentration determined by using CR-39 track detector and fission fragment track technique, the nuclear reaction used as source of uranium fission fragment is U-235 (n-f) obtained by the bombardment of U-235 with thermal neutrons emitted from (Am-Be) neutron source with flux (5000 n/cm^2.s) for seven days, the concentration were calculated by comparison with the standard samples.

The uranium concentration in Dora refinery samples were ranged between (0.95-3.34) ppm with average (2.03) ppm for solid samples and (1.25) ppm for liquid samples.

The uranium concentration in Bijie refinery samples were ranged from (0.42-1.46) ppm with average (0.55) ppm in the solid samples and (1.24) ppm in the liquid samples.

The uranium concentration in the north field of al-Rumaila samples were ranged between (1.78 - 6.35) ppm with average (3.37) ppm.

From above results, we concluded that the maximum of uranium concentration was found in the north field of al-Rumaila in Basrah governorate, these results are agreement with the permissible limit from IAEA. The maximum of external equivalent dose rate from NORM in oil waste was found in the north field of al-Rumaila was (0.30) mSv/y less than the permissible limit from IAEA which is (1) mSv/y for the public.

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LIST OF ABBREVIATIONS & SYMBOLS

CPS	Count per second
DL	Detection limit
DRF	Dose rate conversion factor
EPA	Environmental Protection Agency
FWHM	Full Width at Half Maximum
(HPGe)	High Purity Germanium
IAEA	International Atomic Energy Agency
Nal(Tl)	Sodium Iodide with Thallium
NORM	Natural Occurring Radioactive Material
NRP	Norwegian Radiation Protection
RC	Reduce crude oil
SSNTDs	Solid State Nuclear Track Detectors
TEM	Transmission Electron Microscopy
TENORM	Technically Enhances Natural Occurring Radioactive Materials
$\mathbf{A_x}$	Specific activity of radionuclides ,(Bq/ kg) or (Bq/l)
C	Speed of light , (m/s)
ch	Channel
Ci	Curie
d	Diameter , (micrometer)
E	Energy, (Joule)
$\mathbf{E_B}$	Activation energy of bulk etching , (Joule)
$\mathbf{E_T}$	Activation energy of the track etch ,(Joule)
H (t)	The external dose rate at time (t), (nGy/y)
h	Thickness, (micrometer)
k	Boltzman constant, (J. mol/K)
L	Distance , (micrometer)
Le	Track length ,(micrometer)
M	Mass, (kg)

LIST OF ABBREVIATIONS & SYMBOLS

N	Normality
Q	Charge , (Coulomb)
Q_{eff}	Effective charge , (Coulomb)
R	Energy resolution
S	Etching sensitivity
T	Absolute temperature, (K)
V	Etching rate ratio
V_B	Bulk etching velocity, (µm/s)
V_d	Diameter growing velocity, (µm/s)
V_T	Tracks etch velocity, (µm/s)
V_o	Volume, (L)
W	Weight, (kg)
W_{eq}	Equivalent weight
Z	Atomic number
δ	Percentage of error
µm	micrometer
keV	Kilo electron volt
MeV	Million electron volt
ϕ	Dip angle
ϕ_C	Critical angle
η	Efficiency
ρ	Track density
ppm	part per million
µg	microgram

Chapter One

1.1 Introduction

The oil extraction and production industry generates several types of solid and liquid waste, scales, sludge, and water. There are typical residues can be found in such facilities and that can be contaminated with Naturally Occurring Radioactive Material (NORM). As a result of oil processing, the natural radionuclides can be concentrated in such residues, so the forming called Technologically Enhanced Naturally Occurring Radioactive Material (TENORM). Most of the radionuclides that appear in oil and gas stream belong to the U-238 and Th-232 natural series, besides K-40. Emphasis was given to the quantification of Ra-226, K-40 since these radionuclides are responsible for most of the external exposure in such facilities **[1]**.

NORM such as the ones from the U-238 and Th-232 series and K-40, are essential constituents of the crust of earth, and can be found in higher than normal concentrations in some regions of the earth, known as high natural radiation areas. The awareness about radiation doses due to natural radiation has been the cause of increased concern in recent years. The particular attention has been focused on the mining and processing industry, as these operations can be responsible for the concentration of NORM in products or residues. After the concentration of radionuclides in industrial processes the TENORM waste associated with hydrocarbon operations at the oilfields occur in the form of scale deposits, sludge, and produced water **[2]**.

Scales formed by precipitation, as well as sludge, which accumulate in the bottom of storage tanks and other equipment which contains variable amounts of the naturally occurring radionuclides from the U-238 and Th-232 series . Scales buildup may cause problems in the operation of an installation by clogging tubing and valves, thus restricting the flow. In many oil fields the produced water is re-injected into the reservoirs to maintain pressure **[2]**. Normally, the water injected in the wells has a different composition from that already in the reservoir. When these two types of water are mixed and the water-oil emulsion is carried to the surface during the pumping operation, precipitation will occur. This precipitation and the consequent scale formation is due to variations of sulphates and carbonates solubility and is associated with temperature variations, pressure changes and injection of water **[3]**.

The dissolved radium in water is brought to the surface along with the hydrocarbons. Uranium and thorium are less soluble in water and remain in the reservoir. The degree of accumulation of radioactive materials is always significant in ducts and equipment that carries produced water, for this reason TENORM concentration are found.

In general, higher in scales and sludge formed in water-handling equipment. Radiation levels measured after the water separation stage are consequently negligible or much lower than those observed in other cases where the water is still present in the mixture. As a result, a high water production rate, such as in the older oil fields can result in increased TENORM accumulation on tubing and equipment **[3]**.

1.2 Radioactivity

Radiation is the name given to the energetic particles or waves emitted by an atom at the time of radioactive decay. It is produced by radioactive decay, nuclear fission, chemical reactions, hot objects, and gases excited by electric currents, nuclei can undergo a variety of processes which result in the emission of radiation. The most common forms of radiation are alpha, beta particles and gamma-rays, our bodies contain radioactive materials such as C-14, K-40 quite naturally **[4]**.

Following the discovery of the radioactivity of uranium by Becquerel, there was a successful search for other naturally occurring radioactive elements and the recognition by Rutherford that radioactivity involved a change in mass and chemical nature of an element. The new element produced in any radioactive process was frequently radioactive as well **[5]**.

There are three radioactive series were recognized U-238,U-235, and Th-232 in which heavy elements lost mass and changed their atomic number in successive steps, the changes ending only when the element became a stable isotope of lead.

In all these spontaneous changes three types of radiation were recognized, namely (energetic helium nuclei), (electrons), and (energetic photons) radiation. Radioactivity is a number of disintegration per second, its unit for measurement is Becquerel (Bq) **[6]**.

$$1 \text{ Curie (Ci)} = 3.7 \times 10^{10} \text{ Bq} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1\text{-}1)$$

In the general sense, heat and light are also a form of radiation, but it was concerned here with other forms, which are commonly known as atomic radiation or simply as radiation, certain substances such as uranium, thorium etc., by nature is unstable. The atoms of such substances attain stability by throwing away the excess energy in the form of electromagnetic radiations or particles called nuclear radiations or ionizing radiations. The transition of radioactivity from air to human is shown in Figure (1-1) **[7]**.

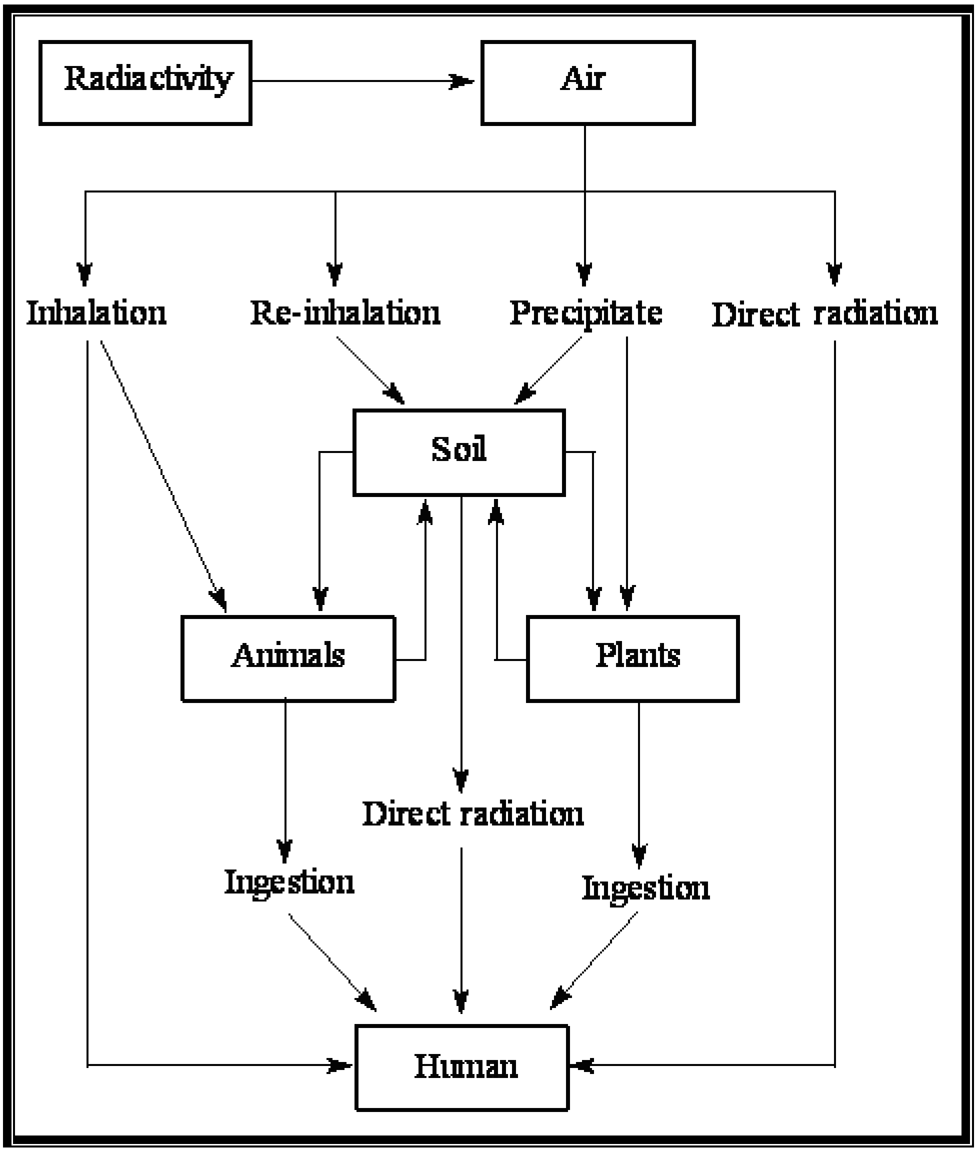

Fig (1-1): Design of transition radioactivity from air to human **[7].**

1.3 Natural radioactivity sources

All natural elements that have atomic numbers Z > 82 are radioactive; there are three natural radioactive series called uranium, thorium, and actinium series, neptunium series is included natural radioactive series but dose not occur in nature because its half-life (2.14×10^6) year shown in Table (1-1) **[8]**.

Table (1-1): The radioactive decay series **[8]**.

Series	First isotopes	Half -life (year)	Last isotopes(stable)
Uranium	U-238	4.49×10^9	Pb-206
Actinium	U-235	7.10×10^8	Pb-207
Thorium	Th-232	1.39×10^{10}	Pb-208
Neptunium	Np-237	2.14×10^6	Bi-209

1.4 Natural uranium

Uranium is considered a one of the most important primordial radionuclide in the earth crust. It was discovered in 1789 by Klaproth, who drives its name Uranus planet. It's a silver white, lustrous, dense, natural and commonly occurring weakly radioactive element. It is ubiquitous throughout the natural environment, being found in varying but small amounts in rocks, soils, water, air, plants, animals and in all human beings.

On average, about (90) μg of uranium exist in the human body comes from the normal intakes of food, water, and air, approximately 66% was found in skeleton, 16 % in the liver, 8% in the kidneys and 10% in other tissues of the body **[9]**.

The average annual intake of uranium by adults had estimated to be (460) µg from ingestion and inhalation. Uranium is widespread in nature, in a wide variety of solid, liquid and gaseous compounds.

It readily combines with other elements to form uranium oxides, silicates, carbonates and hydroxides. Natural uranium contains 99.274% of U-238, 0.720% of U-235, and 0.006% of U-234,they all have 92 protons in the nucleus, but 146,143 and 142 neutrons respectively as shown in Table (1-2). The longer half-life the less radioactive decay products appear in a given time interval and could effect human health **[10]**.

Table (1-2): Isotopic composition of natural uranium **[10]**.

Natural uranium	**U-234**	**U-235**	**U-238**	**Total**
Weight %	0.006 %	0.720 %	99.274 %	100 %
Activity %	48.9 %	2.2 %	48.9 %	100 %
Activity in 1g of U_{na}	12.356 Bq	0.568 Bq	12.356 Bq	25.280 Bq

1.4.1 Uranium -238 series

This series begins with U-238 nuclei half-life (4.5×10^{9}) year, and gradually ends with lead Pb-206, which is a stable element. The elements of this series are represented in Table (1-3) **[11]**.

Table (1-3): Uranium - 238 decay series **[11]**.

Nuclide	Particles Kind	Half Life T (1/2)	Decay's Constant $(Sec)^{-1}$	Particles Energy (MeV)
U_{92}^{238}	α	4.51×10^{9} y	4.88×10^{-18}	4.18
Th_{90}^{234}	β	24.1 d	3.33×10^{-7}	0.19
Pa_{91}^{234}	β	1.18 m	9.79×10^{-3}	2.31
Pa_{91}^{234}	β	6.66 h	2.89×10^{-5}	0.5
U_{92}^{234}	α	2.48×10^{5} y	8.8×10^{-14}	4.76
Th_{90}^{230}	α	7.6×10^{4} y	2.89×10^{-13}	4.69
Ra_{88}^{226}	α	1620 y	1.36×10^{-11}	4.78
Rn_{86}^{222}	α	3.83 d	2.1×10^{-6}	5.49
Po_{84}^{218}	β, α	3.05 m	3.79×10^{-3}	α, 6.0 β, …
At_{85}^{218}	α	1.3 sec	0.53	6.69
Pb_{82}^{214}	β	26.8 m	4.31×10^{-4}	0.65
Bi_{83}^{214}	α, β	19.7 m	5.86×10^{-4}	α, 5.5 β, 0.4 – 3.2
Po_{84}^{214}	α	1.64×10^{-4} sec	4.23×10^{3}	7.68
Tl_{81}^{210}	β	1.32 m	8.75×10^{-3}	1.99
Pb_{82}^{210}	β	22 y	10^{-9}	0.02
Bi_{83}^{210}	α, β	5.01 d	1.6×10^{-6}	α, 4.7 β, 1.16
Po_{81}^{210}	α	138.4 d	5.8×10^{-8}	5.3
Tl_{81}^{206}	β	4.3 m	2.69×10^{-3}	1.57
Pb_{82}^{206}	-	stable	-	-

1.4.2 Actinium -235 series

This series begins with U-235 nuclei half-life (7.1 × 10^8) year, which have the longest half -life compared to other elements in this series and ends with lead Pb-207, which is a stable element. The importance of U-235 is the ability of fast fission by using thermal neutrons. The elements of this series are represented in Table (1-4) **[12]**.

Table (1-4): Actinium -235 decay series **[12]**.

Nuclide	**Particles Kind**	**Half Life T (1/2)**	**Decay's Constant $(\mathrm{Sec})^{-1}$**	**Particles Energy (MeV)**
U_{92}^{235}	α	7.13×10^8 y	3.08×10^{-17}	4.18
Th_{90}^{231}	β	25.64 h	7.51×10^{-6}	0.30
Pa_{91}^{231}	α	3.25×10^4 y	6.77×10^{-13}	5.0
Ac_{89}^{227}	α, β	21.2 y	1.04×10^{-9}	α, 4.94 β, 0.04
Th_{90}^{227}	α	18.17 d	4.41×10^{-7}	5.97
Fr_{87}^{232}	β	22 m	5.25×10^{-4}	1.15
Ra_{88}^{223}	α	11.68 d	6.87×10^{-7}	5.71
Rn_{86}^{219}	α	4.0 sec	0.173	6.82
Po_{84}^{215}	α, β	1.83×10^{-3} sec	379	7.38
At_{85}^{215}	α, β	10^{-4} sec	6930	α, 8.0 β, …
Pb_{82}^{211}	β	36.1 m	3.2×10^{-4}	1.4
Bi_{83}^{211}	α, β	2.15 m	5.37×10^{-3}	α, 6.62 β, …
Po_{84}^{211}	α	0.52 sec	1.33	7.45
Tl_{81}^{207}	β	4.78 m	2.42×10^{-3}	1.44
Pb_{82}^{207}	-	stable	-	-

1.4.3 Thorium-232 series

Thorium was discovered by "J.Berzelins", which is derived from the Scandinavian "thor". This series begin with thorium Th-232 nuclei half-life (1.4×10^9) year and ends with Pb-208. The elements of this series are represented in Table (1-5) **[13]**.

Table (1-5): Thorium-232 decay series **[13]**.

Nuclide	Particles Kind	Half Life T (1/2)	Decay's Constant $(Sec)^{-1}$	Particles Energy (MeV)
Th_{90}^{232}	α	1.39×10^{10} y	1.56×10^{-18}	4.01
Ra_{88}^{288}	β	6.7 y	3.86×10^{-9}	0.05
Ac_{89}^{228}	β	6.13 h	3.14×10^{-5}	1.11
Th_{90}^{228}	α	1.91 y	1.15×10^{-8}	5.43
Ra_{88}^{224}	α	3.64 d	2.2×10^{-6}	5.68
Rn_{86}^{220}	α	56 sec	1.24×10^{-2}	6.29
Po_{84}^{216}	β, α	0.15 sec	4.62	α, 6.78 β, …
At_{85}^{216}	α	3×10^{-4} sec	2.31×10^{3}	7.79
Pb_{82}^{212}	β	10.64 h	1.81×10^{-5}	0.35
Bi_{83}^{212}	β, α	6.06 m	1.91×10^{-4}	α, 6.05 β, 2.25
Po_{84}^{212}	α	3×10^{-7}	2.31×10^{6}	8.78
Tl_{81}^{208}	β	3.1 m	3.73×10^{-3}	1.80
Pb_{82}^{208}	-	stable	-	-

1.4.4 Neptunium -237 series

Neptunium has a half-life (2.14×10^6) year, which is much shorter than the geological age of the earth. Virtually all neptunium decayed within the first 50 millions of years after the earth was found. Neptunium-237 did not find in nature but it was discovered in some stars spectrum **[14]**.

1.5 The interaction of radiation with matter

When nuclear disintegration takes place, several types of radiation may be emitted. Those most often encountered are alpha (α), beta (β), and gamma (γ) radiation. Positively charged α particles, identified as helium nuclei, are relatively large in mass and interact strongly with matter. They are absorbed by as little as a sheet of paper. Negatively charged β particles, identified as fast electrons, are smaller than alpha particle in mass. They interact less with matter than α particles and can easily penetrate paper and cardboard. However, they are absorbed by metal sheets. The third type of radiation is γ rays, which are not particles but very high energy photons. They interact with matter much less strongly than do alpha and beta particles **[15]**.

1.5.1 Interaction of α-particles with matter

Heavy charged particles, such as α-particles interact with matter primarily through Coulomb forces between their positive charge and the negative charges of the orbital electrons of the atoms in the material. If the energy is sufficient, the electron is completely removed from the atom (ionization). Energy is transferred from the α-particle to the electron, and the velocity of the α-particle decreases as result of the interaction. The energy transfer event only a small fraction of **α**-particle loses energy in many successive interactions; **α**-particle interacts with many electrons, implying that the result is a more or less continuous decrease of its velocity until the **α**-particle has stopped **[16]**.

1.5.2 Interaction of ß-particles with matter

Compared with **α-**particles, electrons lose their energy at a lower rate. Large deviations from the directions of the incoming electrons are possible because the electron mass is equal to that of the orbital electrons with which it is interacting, and a much larger fraction of its energy is lost in a single collision than for **α-**particles **[17]**. An expression similar to that for **α-**particles has loss the specific energy due to ionization and excitation (collision losses). Electrons also differ from **α-**particles because energy may be lost by radiative processes as well as by Coulomb interactions. These radiative losses may take the form of electromagnetic radiation, which might originate from any position along the electron track **[17, 18]**.

Radiative losses are most important for high electron energies but because the mean **α-**energy from decaying U-238, U-235, and Th-232 is about (1) MeV, radiative losses represent a small fraction of the energy losses due to ionization and excitation **[18]**.

1.5.3 Interaction of γ-rays with matter

When **γ** -rays pass through the matter various interactions with the atoms in the material can take place all these processes lead to the partial or complete transfer of the **γ-**rays photon energy to the electrons of the atoms in the material **[19]**. Three types of γ-rays interactions with matter are important:

1. Photoelectric absorption.
2. Compton scattering,
3. Pair production.

In the photoelectric process, a photon interacts with an absorbing atom, while the photon disappears completely. The photoelectric effect is the predominant mode of interaction of γ -rays of relatively low energies (< 0.5) MeV, as shown in Figure (1-2) **[20]**.

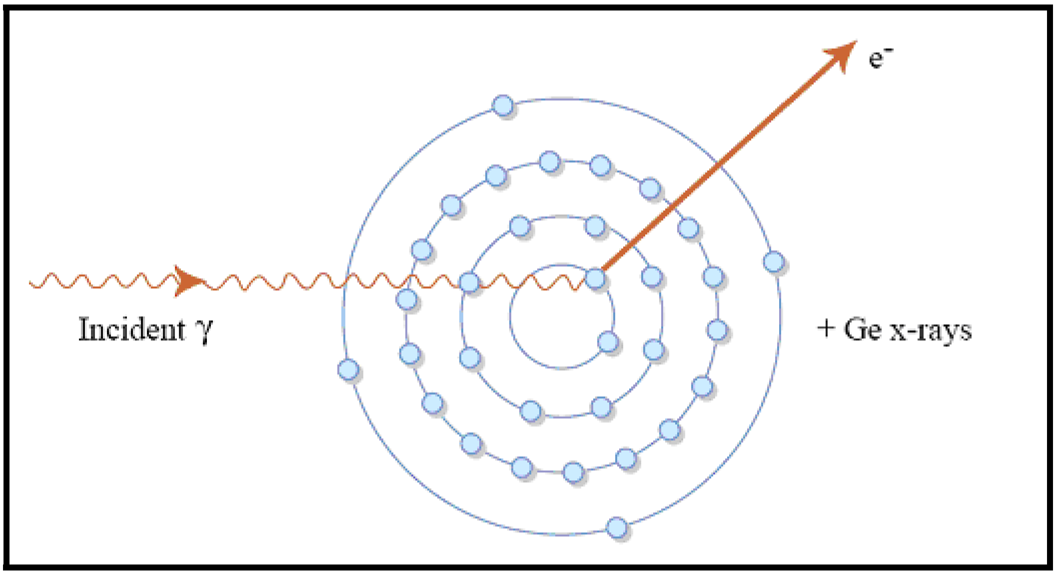

Fig (1-2): The photoelectric effect **[20]**.

The Compton scattering takes place between the incident γ -ray photon and an electron in the absorbing material. It is often the dominant interaction mechanism for γ-ray energies of radioisotope sources from (0.5–5) MeV as shown in Figure (1-3), the incoming γ-ray is scattered over an angle with respect to its original direction. A part of the photon energy is transferred to the Compton electron. The cross-section for Compton scattering (σ) per unit mass is **[20, 21]**.

$$\sigma = \text{Constant} / E_\gamma \quad \text{.........................} \quad (1\text{-}2)$$

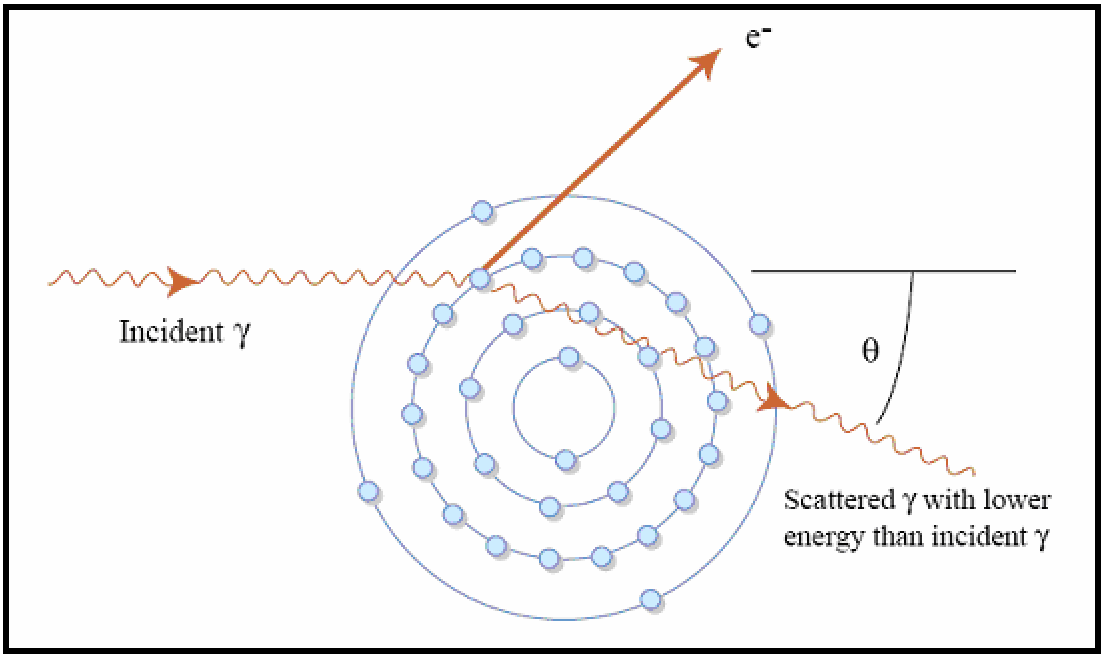

Fig (1-3): Compton Scattering **[20]**.

Pair production: If **γ**-ray quantum energy exceeds twice the rest-mass energy of an electron (1.022) MeV the process of pair production is possible. In this case, **γ**-ray quantum produces an electron-positron pair; this process shown in Figure (1-4) **[20, 22]**.

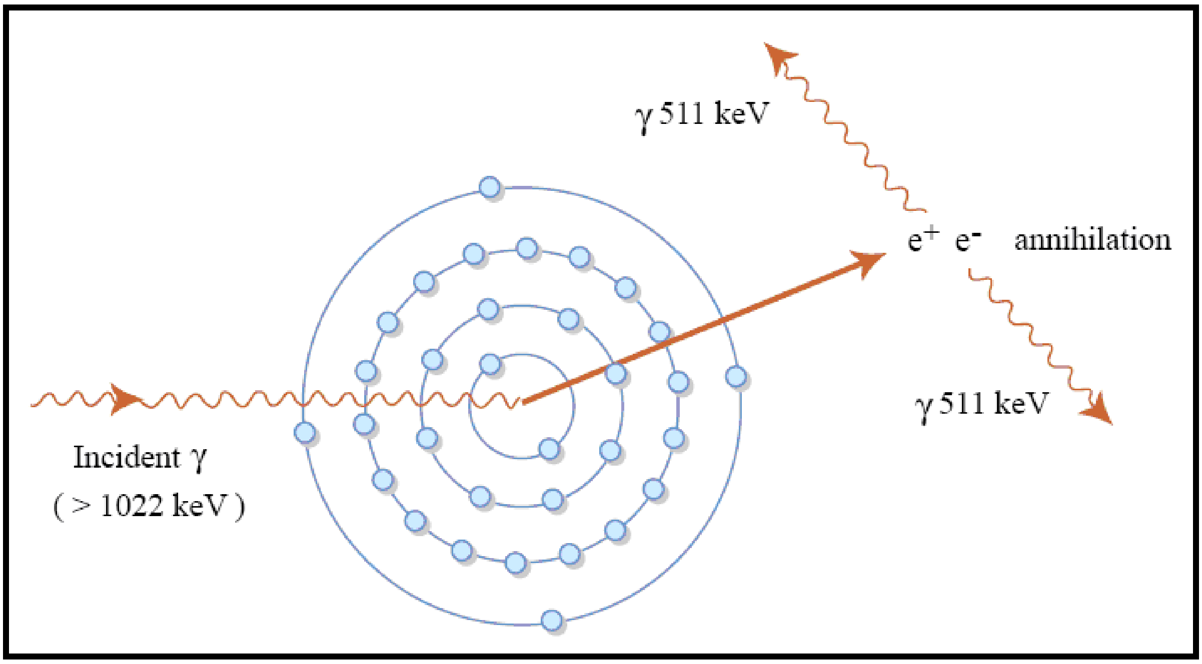

Fig (1-4): Pair Production **[20]**.

1.6 Sources of background radiation

Naturally occurring radioactive materials are common in the environment and in the human body. These materials are continuously emitting ionizing radiation from outer space (cosmic radiation) bombards the earth constantly. The ionizing radiation from these natural sources is called background radiation **[23]**.

Human activities, such as making medical X-rays, generating electricity from nuclear power, testing nuclear weapons, and producing a variety of common products that contain radioactive materials, can cause additional exposure to ionizing radiation as shown in Figure (1-5) **[24]**.

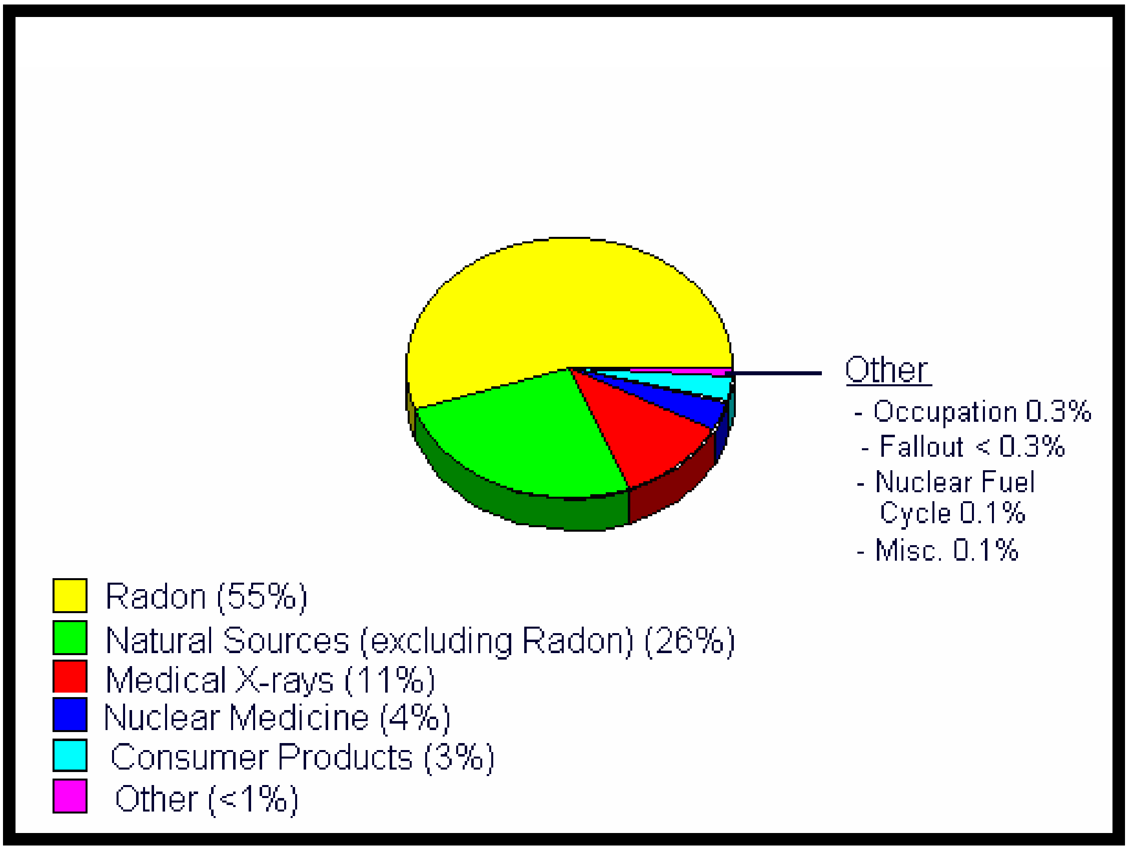

Fig (1-5): Sources of radiation exposure **[24]**.

Actual values vary, depending on where people live and how they spend their time. The worldwide average background equivalent dose for a human being is about (2.4) mSv/year as shown in Table (1-6). The equivalent dose exposure is mostly from cosmic radiation and natural isotopes in the earth **[25]**.

Table (1-6): Equivalent dose from the natural sources **[25]**.

Source	**Equivalent dose (mSv/y)**
Cosmic rays	0.4
Gamma rays	0.5
Internal	0.3
Radon	1.2
Total	2.4

All radiation detectors record some background signals because of the cosmic radiation that continuously bombards the earth atmosphere and the existence of natural radioactivity in the environment. The nature of this background varies greatly with the size and type of detector and with the extent of shielding that may be placed around it **[26]**.

The background radiations are conveniently grouped into four categories:

1. The natural radioactivity of constituent materials of detector itself.
2. The natural radioactivity of the ancillary, supports, and shielding placed in the immediate vicinity of the detector.
3. Radiations from the activity of the earth surface terrestrial radiation walls of the laboratory, or other far away structures.
4. The primary and secondary components of cosmic radiation **[27]**.

1.7 Previous works

Many studies have been performed to investigate and measure the concentrations of radioactive elements in oil waste samples and crude oil by using different techniques, some of these measurements are achieved by EPA, IAEA and NRP, also many studies for researchers were examining the radioactivity related problems and attempting NORM in oil and gas producing facilities; some of these studies are abstracted as follows:

- (EPA) found in elevated levels in the mineral scales in oil and gas production facilities, radium and thorium in British, the total specific activities was (120-520) Bq/kg ,(1999) **[28]**.
- (IAEA) investigates very high levels of radioactivity at the oil field pipe cleaning facilitates conducted at facility revealed the presence of large quantities of radioactive dust and particulates. The total specific activities were (222×10^3) Bq /kg of Ra-226 and (37×10^3) Bq /kg of Th-232 in USA, (2001) **[29]**.
- Pataki G. determined the total specific activities of NORM from oil and gas industry which was (65-500) Bq/kg in Germany, (2002) **[30]**.
- (IAEA) measured the total specific activities from oil and gas Industry was (100 - 230) Bq/kg in Holland, (2003) **[31]**.
- (NRP) measured the specific activity of Ra-226 for scales, sludge, which were taken from equipment like pumps, valves , the specific activities was (130-320) Bq/kg for scales, and (100-240) Bq/kg for sludge in Norway,(2004) **[32]**.

- (IAEA) measured the total specific activity from oil and gas industry from waste of sludge was (20.13-74.45) Bq/kg while for scales was (202-1199) Bq/kg, in Syria, (2004**)** **[33]**.
- ZayirY.M. measured the total specific activity of NORM for scales from the refinery of Dora only and it was (3.0-27.2) Bq/kg using the NaI(Tl) detector of volume ($65cm^3$) and resolution of 8.5% for Cs-137 at (662) keV and the time of measuring was (1000) sec, which may be operated in two monitoring functions. The count rate in (cps) and dose rate in selectable units and quantities, as well as accumulated dose from the time mode was enabled Iraq, (2005) **[34]**.

1.8 The aim of study

The purposes of the present work are to:

1- detect and measure the specific activities of the radionuclides in crude oil and oil waste from Dora refinery and Bijie refinery using (HPGe) detector.
2- determine the uranium concentration in crude oil and oil waste from Dora refinery, Bijie refinery, and the north field of al-Rumeala in Basrah using CR-39 track detector.
3- find the relationship between two techniques.
4- explain the effect of oil waste radioactivity on the environment.

Chapter Two

2.1 Source of petroleum

The evidence of petroleum generation and migration lies mainly in the two fields of geology and chemistry. Empirically a theory of the origin of the oil must explain two facts:

- The geological evidence.

Most hydrocarbon deposits occur in sedimentary rocks, known as source rocks, from which the petroleum migrates to the reservoirs, many hydrocarbon deposits occur in porous and permeable sandstones and carbonate rocks that totally enclosed in other impermeable seal rocks.

- The chemical evidence.

Petroleum has an organic origin, optical studies and chemical evidence refers to organic matter (algae, spores, pollen, plant cuticle, resin and pigments) accumulated in shale or fine grained carbonate rocks as being the principal source material for petroleum **[35]**.

In general, petroleum had formed as the result of layers down of organisms such plank and bacteria on the sea floor millions of years ago. Under hard conditions and migrate, the effect of heat and pressure so that the location of deposit is not the location of the initial debris accumulations. In primary production, oil flows under natural pressure into the well bottom and then is brought to the surface. In secondary production, pressure must be added to the system to move the oil into the well bottom [**35]**.

2.2 Oil waste residues

Oil field equipment can contain hard radioactive scales and softer scale bearing sludge that appears as coatings or sediments. Gas field equipment may be contaminated internally with coatings or plating of concentrated NORM. The scales and sludge containing NORM are formed primarily during the extraction and production stages, when the mixture which is contain (oil, gas, and water) is brought to the surface and forming during the separation process. Initially, NORM is brought to the surface through the down hole tubing as part of the mixture. Hard scales precipitate from the formation water onto the down hole tubing in response to changes in temperature, pressure and salinity as the water is brought to the surface **[36]**.

The scale is typically a mixture of carbonate and sulphate minerals, one of these sulphate minerals is barium $BaSO_4$, which is incorporate radium in its structure.

When compared with other NORM containing wastes in the oil and gas industry, the concentrations of Ra-226 are the largest in these scales and enhanced substantially from concentrations found in the natural environment.

The basic geological formations of rocks in Iraq that represented as sand stones formation in south and east of Iraq and limestone in north of Iraq as (U-238, Th-232) are found in most sedimentary rocks and soil in variable amounts. Igneous rocks contain higher concentrations of (U-238, Th-232) than sandstones and limestone as shown in Table (2-1) **[36]**.

Table (2-1): Average of specific activities of some rocks in Iraq **[36]**.

Type of rocks	Ra-226 Bq/ kg	U-238 Bq/ kg	Th-232 Bq/ kg	K-40 Bq/ kg
Igneous	25.21	18.15	12.52	103
Sandstone	19.27	11.82	8.05	125
Shale	21.96	12.85	10.75	114
Limestone	15.54	14.87	5.18	81

2.3 The operation system in refinery

NORM waste must comply with a set of management and disposal requirements designed to minimize the potential for human health effects. In general, scales and sludges meeting the definition for regulated NORM must be managed and disposed of by methods that provide a higher degree of containment and isolation than is required for scales and sludge.

The crude oil has different characteristics, it may be with heavier or lighter hydrocarbons, and certainly contains a complex mixture which must be separated and treated in the refinery **[37]**.

The refinery operations are divided into three categories:

1. Physical separation: distillation, solvent extraction.
2. Breaking down, cracking, vis-breaking, cooking.
3. Rebuilding processes -reforming.

The different components of the mixture are separated in distillation column by their boiling range. The crude oil has vaporized and passed into a vertical column. The vapors rise until they reach a temperature where, they condense on to trays and had taken off. The boiling range approximate oil product is shown in Table (2-2) **[38, 39]**.

Table (2-2): Boiling range of crude oil products **[38, 39]**.

Approximate boiling range °C	Number of carbon atoms	Description
20	1-4	Gas
20-60	5-6	Gasoline
40-180	6-10	Naphtha
180-260	10-14	Kerosene
260-340	14-20	Gas oil ,Diesel
≥ 340	≥ 20	Waxes

2.4 Water waste treatment

Rocks that contain oil and gas deposits often contain water as well. The water dissolves minerals and radionuclides, such as radium, that are in the rocks. Water treatment includes passing the water through various types of filters and devices that rely on physical and chemical processes to remove impurities and organisms. If the water is containing radionuclides treated by such system, it is possible to generate waste even if the treatment system not originally intended to remove radioactivity, and the water waste includes **[40,41]**:

- Filter sludge.
- Alum sludge.
- Ferric chloride residual.
- Granular activated carbon.
- Water from filter backwashes.

In petroleum refining, the primary solids waste from petroleum separator bottoms include corrosion products, carbon, catalyst fines, chemical precipitates, sand, and silt, all of which are oil coated. There are two technologies likely to produce the TENORM waste because they generate sludge and are know to remove radioactivity from water **[42]**:

- Lime softening.
- Ion -exchange.

The lower cost of lime softening that used on larger systems to soften water by the addition of quicklime (CaO), which raises the (pH) causing calcium and magnesium in water to precipitate. The precipitate along with the suspended solids was removed by sedimentation and filtration; lime is preferred in installations that use (1) ton per day **[43]**.

2.5 Oil waste treatment

Radium and other radionuclides and their radioactive decay products become concentrated in production wastes: pipe scale that tends to form inside oil, gas production pipes and equipment large volumes of waste water that accumulate in tanks or pits. The types of waste generated by the petroleum industry include pipe, scale, sludge, and equipment or components contaminated with radium. It is estimated that the industry generates waste about (150,000) cubic meters yearly **[44]**.

Field surveys have shown that petroleum pipe scale may have very high of Ra-226 concentrations. In some cases, more than (400,000) pCi/g of this waste is retained in oil and gas production equipment, while some of the scale and sludge is presently being removed and stored in drums. The industry disposes of scale and sludge wastes removed from oil and gas production equipment and also discards associated contaminated components **[45]**.

In Dora refinery, the oil wastes are collected from all units of processes in the refinery in storage tanks (AP1). At this unit, the physical treatment is used to collect the light waste from oily water mechanically using disc coil, after that the waste are separated in separation tank (AP4), at this tank, the heavy sludge are at the bottom and the water is at the top of the tank .

The water is pumped to the neutralization tank (AP7) to control the pH of water by adding NaOH or H_2SO_4, while the sludge are floated by air pressure at the tank (AP8), and the water is collected at the biological tank (AP10). In this tank H_3pO_4 and yeast are used for the growth of bacteria which are used for removing hydrocarbon pollutants from industrial water, and then the water is collected at the storage tank (AP12) and then discharged to the environment. The sludge are collected at the storage tank (AP11) after floating at tank (AP8).The sludge are treated by using quicklime (CaO) for solidification then discharged to environment, There is a huge amount of untreated sludge which is collected at pit abandonment **[46]**.

2.6 History of solid state nuclear track detectors

The science of solid-state nuclear track detectors was born in 1958 when D.A. Young discovered the first tracks in a crystal of LiF. The etch pits, later called ‘‘tracks’’, which was previously placed in contact with a uranium foil, irradiated with thermal neutrons and treated with a chemically etching solution. The thermal neutrons led to fission of the uranium nuclei and the fission fragments bombarded the LiF crystal and damaged it. The damaged regions constituted more chemically active zones than the surrounding undamaged areas, one year later; Silk and Barnes reported the finding of damaged regions in mica **[47]**.

They used the transmission electron microscope to investigate tracks of heavy charged particles in mica. Fleischer conducted extensive investigations of this method. They applied the method to mica and to other materials like minerals, plastics and glasses. There was a tremendous growth in this field of science.

The basis of solid state nuclear track detection is that charged particles damage the detector within nanometers along the track in such a way that the track can be etched many times faster than the undamaged material. Etching, typically for several hours, enlarges the damage to conical pits of micrometer dimensions that can be observed with a microscope. For a given type of particle, the length of the track gives the energy of the particle. The charge can be determined from the etch rate of the track compared to that of the bulk. If the particles enter the surface at normal incidence, the pits are circular; otherwise the elliptically and orientation of the elliptical pit mouth indicate the direction of incidence **[48]**.

2.7 Solid state nuclear track detectors (SSNTDs)

SSNTDs are insulating materials have the capabilities for measuring concentration and spatial distribution of isotopes if they emit heavy charge particles, either directly or as a result of specific nuclear reactions . The damage of these particles along their path called track (Latent track), may become visible under an ordinary optical microscope after etching with suitable chemicals. When one compares the properties of SSNTDs with the conventional detectors, it can be seen that the SSNTDs have many advantages over other detectors in general, and over nuclear emulsions in particular **[49]**.

The fact that fission fragments can be recorded and distinguished in a mixed field of unwanted light charged particles, neutrons and gamma rays, has made these detectors extremely useful in experiments where fission reaction rate is very low and a very high detection efficiency is required .

As already mentioned, the SSNTDs can perform background free detection of fission fragments in the presence of high doses of light charged particles, gamma rays and neutrons **[50]**.

They are ideal for the study of rare type of heavy charged reaction products emitted in nuclear reactions. They have been advantageously employed in the measurement of spontaneous fission decay constant of a number of heavy nuclides, and other parameters of great importance in nuclear science **[51]**.

The solid state nuclear track detectors are divided into two categories:

- Inorganic detectors such as: mica, glass, etc.
- Organic detectors (plastic) such as: CN-85, LR-115, CR-39, lexan, mikrofol, etc **[52]**.

2.7.1 Inorganic detectors

Inorganic detectors are compounds where carbon and hydrogen do not enter in structure, and created an ionic bond between its atoms. The inorganic detectors and chemical composition are shown in Table (2-3) **[53]**.

Table (2-3): Kinds of inorganic detectors **[53]**.

No.	Detector	Chemical Composition
1	Zircon	$ZrSiO_4$
2	Quartz	SiO_2
3	Mica(Biotite) Mica (Muscovite)	$K(Mg, Fe)_3\ AlSi_3O_{10}\ (OH)_2$ $Kal_3Si_3O_{10}\ (OH)_2$
4	Fluorite	$CaF_{10}\ (OH)_2$
5	Soda Lime Glass	$23SiO_2:5Na_2O:5CaO:Al_2O_3$
6	Olivine	$Mg\ Fe\ SiO_4$
7	Calcite	$CaCO_3$

2.7.2 Organic detectors

Organic detectors are the compounds that carbon and hydrogen enter in structure, and create a covalent bond between its atoms. This type of SSNTDs have a sensitivity larger than inorganic detectors because bonds of (C-C) and (C-H) which easy broken after exposing to the radiation.

The organic detectors have a high analytic power larger than inorganic detectors, while the threshold energy for organic detectors are less than inorganic detectors .There are many kinds of the organic detectors shown in Table (2-4) **[53]**.

Table (2-4): Kinds of organic detectors **[53]**.

No.	Detectors	Chemical Composition
1	Polyester (HB Pa IT)	$C_{17}H_9O_2$
2	Polyimide	$C_{11}H_4O_4N_2$
3	Cellulose, Cellulose Nitrate (CN) Cellulose Triacetate (CT)	$C_6H_8O_9N_2$ $C_3H_4O_2$
4	Polycarbonate (PC) (Lexan , Makrofol)	$C_{16}H_{14}O_3$
5	Plexi glass	$C_5H_8O_2$
6	Poly allyl diglycol Carbonate (CR-39)	$C_{12}H_{18}O_7$

2.8 Materials of SSNTDs

Particles tracks formed in bulk samples of virtually any insulating material but not in metals or other good conductor. Table (2-5) indicates the categories of track-storing and non track-storing materials and shows that there appears to be a correlation with electrical resistivity, such that materials with values above about (2000) ohm.cm generally store tracks **[54]**.

Table (2-5): Relation of track formation with the electric resistivity **[54]**.

Materials	Resistivity range (ohm.cm)
Track-Forming-Insulators	
Poor Insulators: MOS_2	$(10^{6}-10^{20})$
Semiconductors: V_2O_5	(3000- 25000)
Non Track-Forming-Semiconductors:	
(Germanium, Silicon)	(2000 - 20000)
Metals:	
(Aluminum , Copper, Gold, Platinum ,	(10 - 2000)
Tungsten	$(10^{-6} - 10^{-4})$

2.9 Threshold of SSNTDs

Track formation related to the production of dense regions of ionization by charged particle and to a first approximation, track formation are regarded as occurring when the number of ions exceeds a certain threshold value; this threshold varies from one type of material to another. The threshold for track registration can defined, as the minimum primary ionization energy required occurring track able for chemical etching **[55]**.

The track formation should related to the number of different parameters, such as total energy loss rate, primary ionization, restricted energy loss, type and energy of the ionized charge particle and depend on the chemical structure of detector material. The tracks formation criterion, that takes the form of a statement are formed in a medium when, the chosen parameter exceeds some critical value **[55]**.

These tracks formation criteria can be tested by irradiating a given solid with a number of ions at various energies and recording those cases for which etch able tracks are formed on diagram, the theoretical curves of the relative damage caused by different ions as a function of their velocities are shown in Figure (2-1) **[56]**.

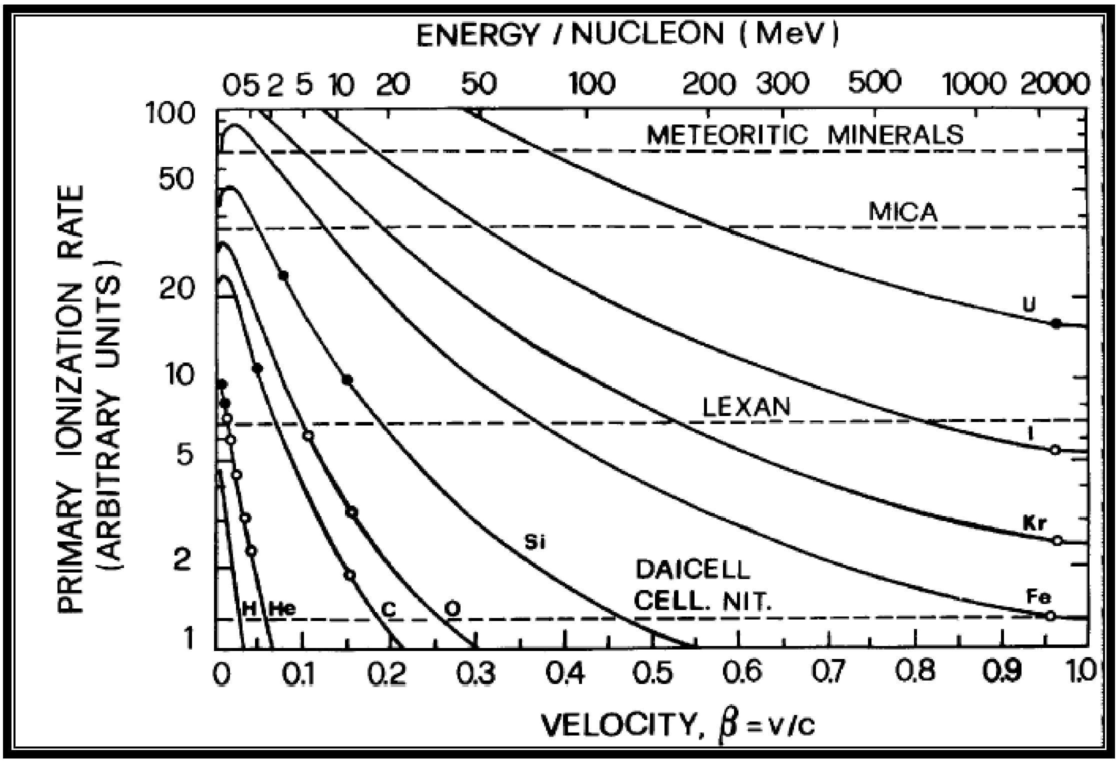

Fig (2-1): Primary ionization rate for various heavy ions versus particle velocity and track etch threshold for various materials **[56]**.

2.10 Tracks formation in the (SSNTDs)

The radiation damage in the (SSNTDs) takes two types **[57]**:

- Inorganic material such as crystal.
- Organic material such as polymer.

2.10.1 Tracks formation in inorganic materials

There are two important major factors in damage production; the primary and the secondary ionization by electrons. It obviously appears that the damage caused by interaction with the electrons. This proposal postulates that the passage of a heavily-ionizing particle leaves a narrow region containing a high concentration of positive ions (burst ionization) in its wake **[58]**. These arrays of adjacent ions are electro statistically unstable and according to coulomb interaction, following ionization, an array of interstitial ions and vacant lattice sites are produced by coulomb energy, subsequent processes include neutralization of the positive ions and elastic relation of the local stresses by spreading the strain more widely around the track path as shown in Figure (2-2a). In the final stage the long strain makes possible the direct observation of un etched tracks in crystals by (TEM) or etches tracks by optical microscope **[59]**.

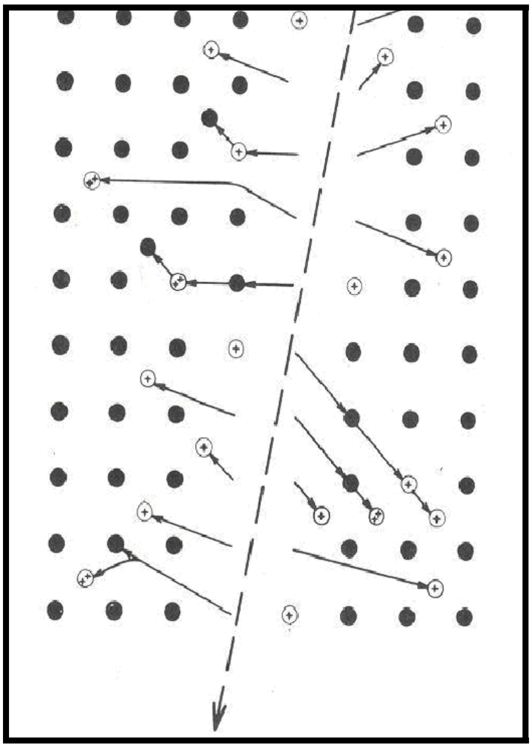

Fig (2-2a): The atomic character of a plastic track in a crystal **[59]**.

2.10.2 Tracks formation in organic materials

The favorable theory for track formation in organic materials is that based on the radiochemical damage mechanism. The etched tracks are formed by the breaking of the long polymer chains to short chains when irradiated. The result is new chain ends, i.e., chemically reactive sites are formed. The polymer is a chemical compound of large molecules which consist of small repeated units called monomers; the monomer is a collection of atoms connected by covalent bonds **[60]**.

The passage of ionizing particles e.g. (proton, deuteron, and alpha particles) through the polymer will excite and ionize the polymer molecules, some excited molecules may in their turn, de excite through the emission of radiation or through non radioactive transitions. Excitation energy can also transfer from one molecule to another.

Both ions and excited molecular chains along its path form a complex array of stable molecules, free radicals, ionized molecules and radical ions. Further reactions among these ions, free radicals and molecules will take place as shown in Figure (2-2b).However, these free ions is relatively small, only free radicals react among them or with other molecules .

The net effect on the plastic will be the production of many broken molecular chains, leading to reduction in the average molecular weight of the substance. Therefore, the rate of chemical etching attack on a plastic increases as the molecular weight decreases. It is likely that the damage region is more etch able than the bulk material which leads to the etching a long the track and is greater in rate than that of the bulk material **[61]**.

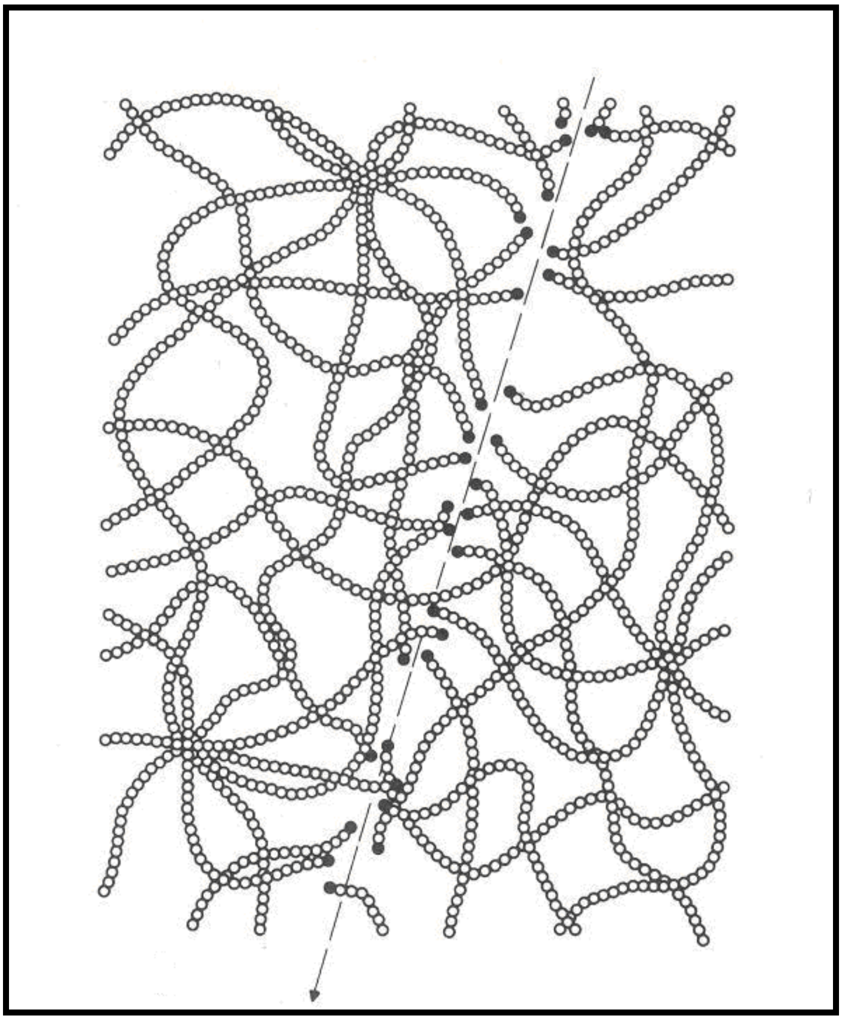

Fig (2-2b): The atomic character of a plastic track in polymer **[61]**.

2.11 The chemical etching

Ionizing particles passing through polymeric track detectors produce latent track, which are trails of radiation damage. The best means of observing the tracks is by etching the SSNTDs material with a chemical solution, which preferentially attacks, the damaged material and enlarges the original track to a size, which is visible in the optical microscope **[62]**.

In general, etchings for polymeric detectors are frequently solutions of alkali hydroxides such as KOH, NaOH with (1-12 N) at (40-60)°C. For glasses and minerals crystals such as quartz, mica and certain pyroxenes etched in aqueous solutions of acids such as (HF) with ~ 48% concentration at 20°C. Etching times can vary from few seconds to many hours. It varies according to the exact etching conditions; the temperature and the concentration of the etching solution **[62]**.

The chemical etching process depends on several parameters are **[63]**:

1. Type and composition etching solution.
2. The concentration of the etching solution.
3. Temperature of the etching solution.
4. Etching time.

The shape of the etched track depends on the energy, velocity of the incident particle, concentration and temperature of the etching solution, the stages of the chemical etching are shown in Figure (2-3).

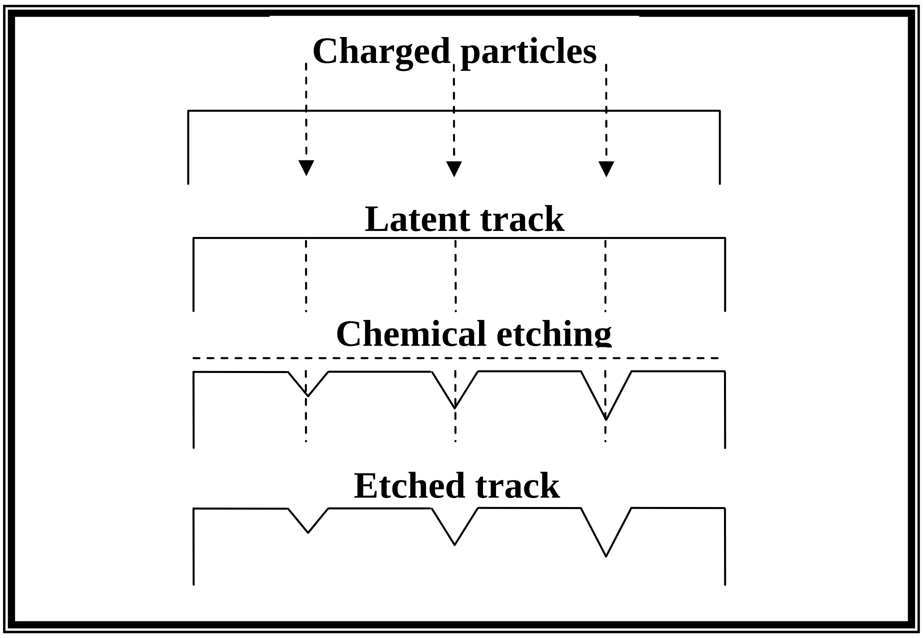

Fig (2-3): Stages of the chemical etching [63].

2.12 Track affecting parameter

There are many parameters affected on the manifestation of track **[64]**:

2.12.1 The bulk etch rate velocity

The rate of chemical etching that attacks the undamaged region surrounding the track is termed the bulk etching velocity V_B. It is defined as the thickness that is removed from one of the surfaces of the detector as per time as a result of chemical etching effect.

The bulk etching rate is a material parameter whose value depends on the structure homogeneity and isotropy of the material of SSNTDs. It is generally constant for a given material and for a given etching applied under a specific set of etching condition, but in crystals it often depends on the crystallographic orientation, and it may vary in some polymers commensurably with the depth below the original surface of the detector. It follows that the bulk etching rate of a given detector material varies with the constituents of the chemical etching, its concentration and temperature. It has been also found that for a given (isotropic & homogenous) material, the V_B varies exponentially with the etching temperature for a constant concentration and is given by the following empirical formula **[64, 65]**.

$$V_B = A \exp(-E_B / k\,T) \qquad \text{(2-1)}$$

Where:

A: constant.

E_B: activation energy of bulk etching (Joule).

k: Boltzman constant = 1.38×10^{-23} (J. mol/K).

T: absolute temperature of the etching solution.

The unit of V_B is (μm/s).

2.12.2 The track etch rate velocity

The track etch velocity V_T is the linear rate at which the detector material is etched out along the particle trajectory, i.e. it measures the amount of the removed layer of the detector along the incident particle trajectory per unit of etching time. The chemical etching dissolves the damaged regions along the track path at a linear rate V_T and attacks the bulk material at a less rate of V_B **[64, 65]**.

It depends on the material of the SSNTDs, their structural homogeneity and isotropy however, it also depends on the crystallographic orientation in the crystals, the incident particle parameters (charge Q, mass M and energy E). The etching conditions such as the concentration and temperature of the chemical etching. It has been found that in a given irradiating and etching condition of a detector, the V_T is increase exponentially with the temperature of the etching according to the following empirical formula **[64, 65]**.

$$V_T = B \exp(-E_T / k T) \quad \text{(2-2)}$$

Where

B: constant.

T: absolute temperature of the etching solution.

k: Boltzman constant = 1.38×10^{-23} (J. mol/K).

E_T: activation energy of the track etch (Joule).

The unit of V_T is (µm/s).

2.12.3 Track diameter growing velocity

The diameter growing velocity V_d is defined as the changes in the average diameter of the opening etch pits per unit time of etching and it can be obtained by calculating the slope of diameter divided time curve as **[66]**:

$$V_d = d / t = \text{the slope} \quad \text{(2-3)}$$

2.12.4 Critical angle

The critical angle ϕ_c is the angle that controlled to emerging the tracks and is represents the minimum angle below which the tracks cannot observe by etching **[67]**.

There are three types of angle incidence are shown in Figure (2-4):

(a) $\phi > \phi_c$ (normal incidence), the tracks can be observed.

(b) $\phi < \phi_c$ the tracks cannot be observed.

(c) $\phi = \phi_c$ (critical angle), the tracks can be observed.

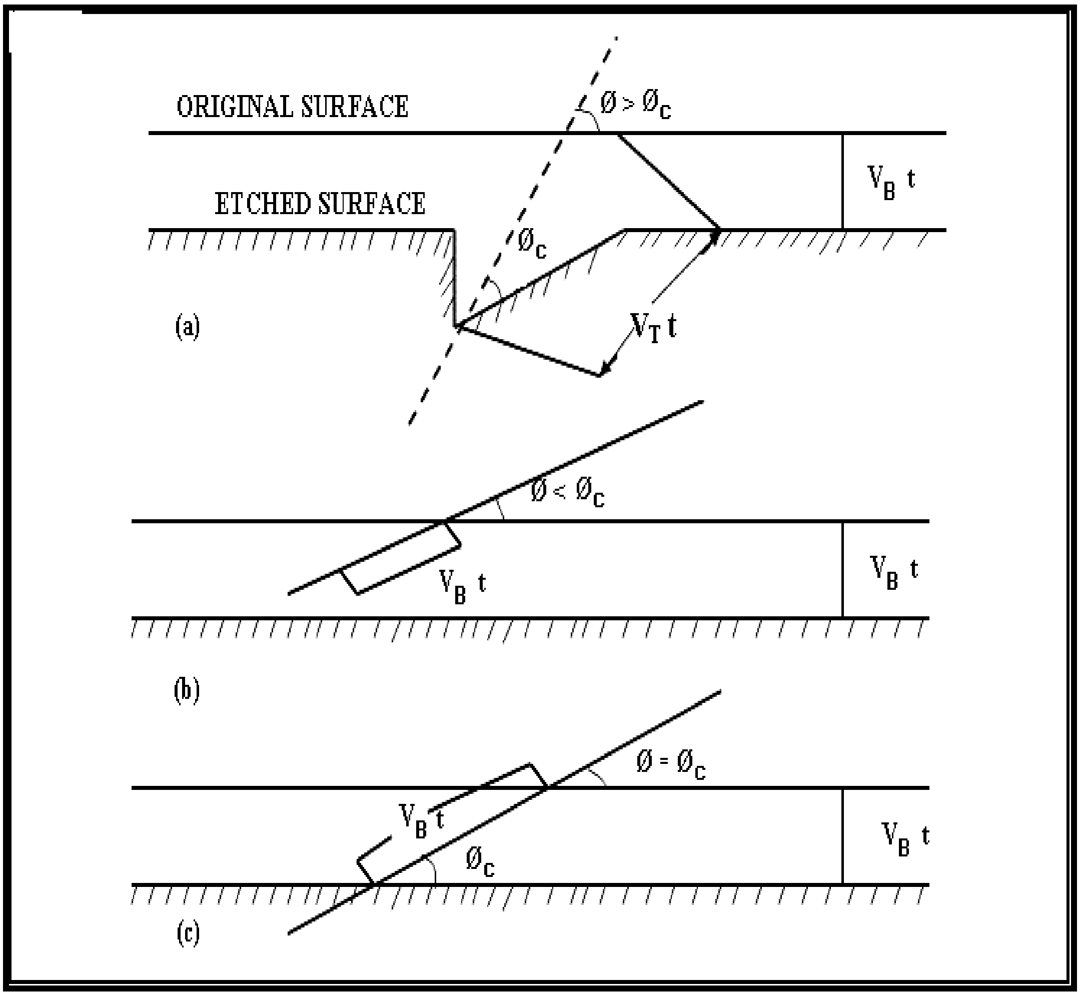

Fig (2-4): Track registration geometry with incidence angle **[67]**.

2.13 Track etching geometry

The track etching geometry depended on the critical angle for incident particle that makes with the surface detector. However, the tracks cannot recording when the angle of incident particle ϕ (placed between the particle passage and detector surface) less than critical angle. The track can be visible when (V_T) greater than (V_B), the incident particles on the track detector can be identified by charge Q, effective charge Q eff, mass M and energy E **[68]**.

The difference in these parameters of the incident particles produce changes in the track etching parameters, a total range of the particle through the detector material (track length), the track etching velocity (V_T) and the variation of (V_T) with position along the track **[68]**.

The etched track has a conical shape and the easy measurable parameters are the conical etched-out length (Le) and the diameter (d) of the etch pit opening. The etched track formation process can envisioned as the incident particle parameters (charge Q, mass M and energy E) representing the input parameters, while the etched track parameters (track length Le, axes, profile and contour) represent the output parameters.

All the processes in-between the above-mentioned parameters represent the damage formation and the etching principle. In the simple instance is a particle penetrating a detector material to its original surface shown in Figure (2-5) **[69]**.

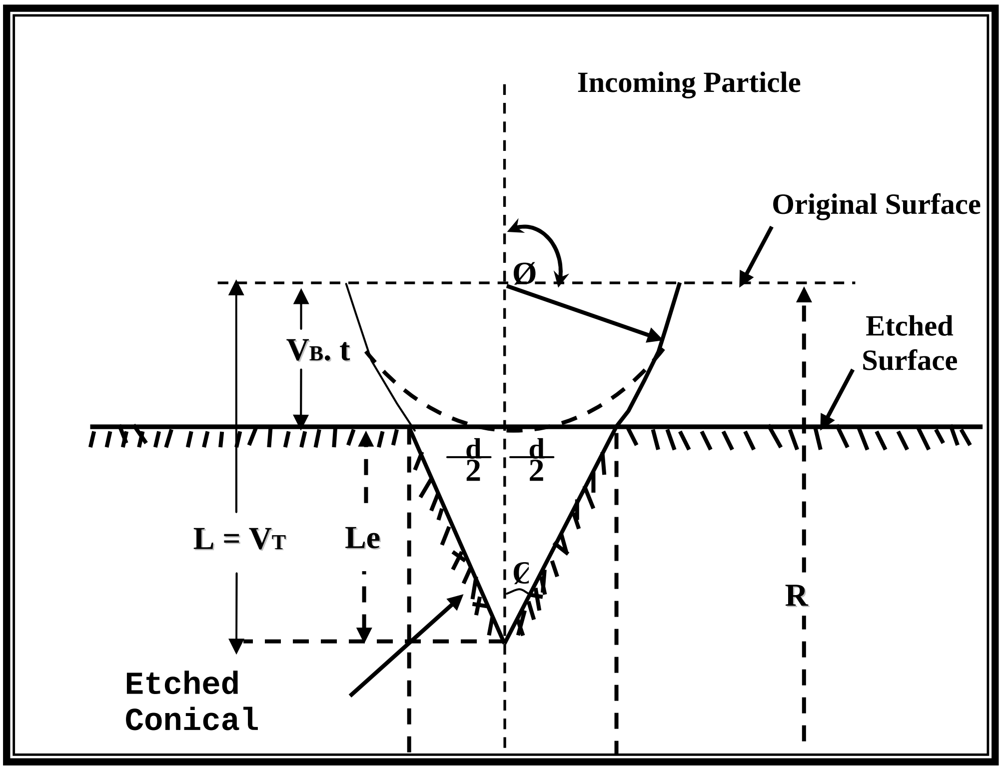

Fig (2-5): The track geometry for particle penetrates detector material normally **[69]**.

The linear of attack down the track, i.e. V_T so that in an etching time t, the etch pit will extend to a distance L from the point of origin, then **[70]**.

$$L = V_T \,.\, t \;\ldots\ldots\ldots\ldots \quad (2\text{-}4)$$

The surface is also being removed at a rate V_B, the full length of the etch pit is:

$$Le = V_T \,.\, t - V_B \,.\, t \;\ldots\ldots\ldots\ldots \quad (2\text{-}5)$$

The diameter of the etch pit is related to V_B and V_T according to this equation:

$$d = 2\, V_B \,.\, t\, [(V_T - V_B) / (V_T + V_B)]^{\frac{1}{2}} \;\ldots\ldots\ldots\ldots \quad (2\text{-}6)$$

The removed surface thickness h is:

$$h = V_B \,.\, t \;\ldots\ldots\ldots\ldots \quad (2\text{-}7)$$

The etching rate ratio is represented in this equation:

$$V = V_T / V_B \;\ldots\ldots\ldots\ldots \quad (2\text{-}8)$$

The track diameter (d) and the length of the etched track (Le) depend essentially on the competitive effects of V_T and V_B .

When $V_B = V_T$ both (Le, d) vanishes, and then no tracks produce **[70]**.

2.14 Etching efficiency and sensitivity

The etching efficiency is defined as the ratio of the counted tracks and the particle flounce impinges on the detector surface.

The efficiency η is given by **[71]**:

$$\eta = \text{No. of etched track / No. of incident particles} \;\ldots\ldots\ldots \quad (2\text{-}9)$$

$$\eta = (V_B / V_T)^{-1} \;\ldots\ldots\ldots\ldots \quad (2\text{-}10)$$

$$\eta = (1/V)^{-1} \ldots\ldots\ldots\ldots \quad (2\text{-}11)$$

Where: V is etching rate ratio (V_T / V_B).

This mean the value of the etching efficiency depends on the track etched rate velocity V_T and the bulk etched rate velocity V_B. The efficiency can define in terms of the critical angle as follows:

$$\eta = (\sin \phi_c)^{-1} \qquad (2\text{-}12)$$

Where: $\sin \phi_c = V_B / V_T$

The experiments showing that the polymers track detectors have high registration efficiency about (85-90%) while the most glass detectors in the range (40-60 %). Another etching parameter is termed etching sensitivity (S) is defined the ratio between etching velocity a long the track to the etching velocity at surface velocity **[72]**.

The sensitivity (S) can calculated using the following equations **[72]**.

$$S = (V_T / V_B)^{-1} \qquad (2\text{-}13)$$

$$S = V^{-1} \qquad (2\text{-}14)$$

$$S = (1 / \sin \phi_C)^{-1} \qquad (2\text{-}15)$$

2.15 Gamma ray spectrometer system

Gamma spectrometry system that used in this study is a fully integrated data acquisition and computation system comprising the following **[73]:**

1. High purity germanium used as a detector.
2. Preamplifier is normally an integral part of the detector unit and located very near to the detector because it is necessary for the operation of the detector with low noise.
3. Linear amplifier is usually built into the multi channel analyzer (MCA), amplifies the pulse by a factor of several hundred, it has a voltage of several volts, it also shapes the pulse so it can analyze more accurately.
4. Bias high voltage power supply is used to supply high voltage to the detector with a range from (0-5000) D.C volt, and must be increased slowly to avoid damaging the detector.
5. Personal computer is used as (MCA) stored the number of pulses detected with a given size (voltage) into a memory channel, the contents of which can later be analyzed to find information about gamma ray detected.

Each pulse which increments the number stored in a memory channel is said to add a count. The distribution of counts in the channels of (MCA) memory is called a spectrum. The block diagram of (HPGe) detector components is illustrated in Figure (2-6) **[74]**.

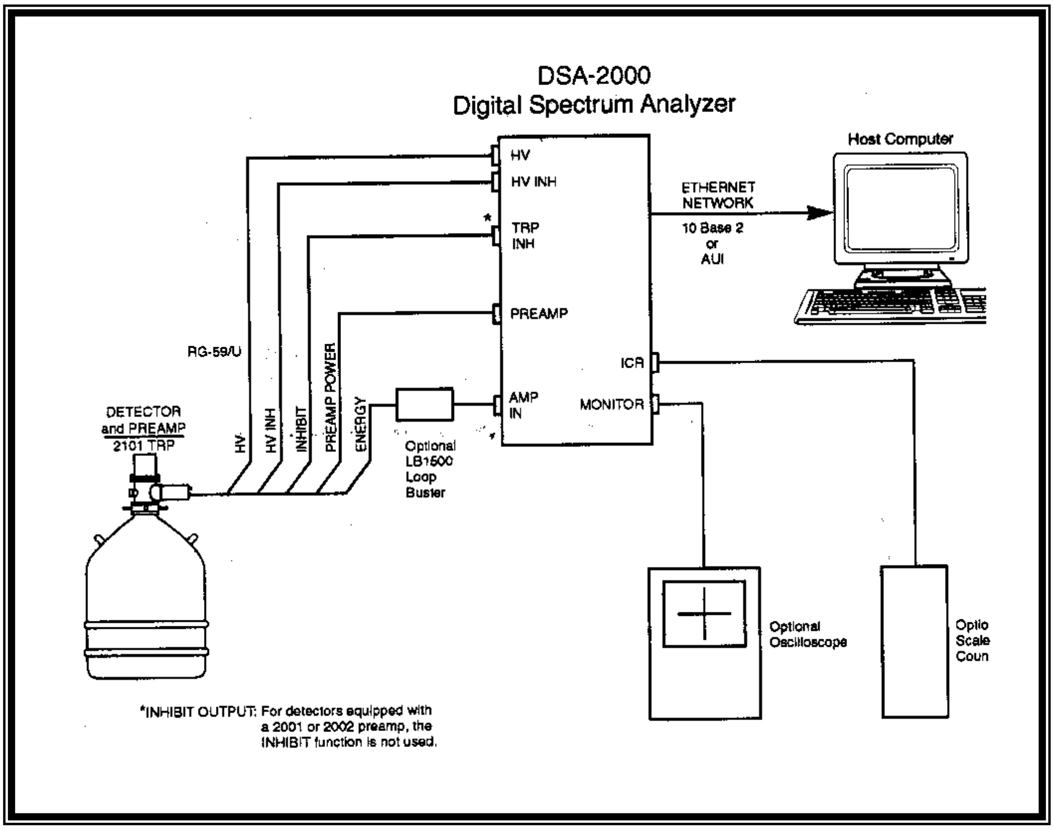

Fig (2-6): Gamma ray spectroscopy system **[74]**.

(HPGe) needs to be cooled when it used only. The cooling of semiconductors is achieved by permanently mounting the detector on a cryostat. The cryostat consists of a reservoir or Dewar containing the cooling medium and vacuum-chamber hosing the detector.

The Dewar is made of two or more concentric metal containers with the space between the two containers evacuated for thermal insulation. The cooling medium is usually liquid nitrogen temperature to (-196 °C, 77K). The cooling rod of the Dewar is made of copper as shown in Figures (2-7a) and (2-7b) [**75]**.

Fig (2-7a): photo of cryostat of the (HPGe) detector

The vacuum chamber that contains the detector is made of stainless steel. The chamber protects from dirt and, by being evacuated, prevents condensation of vapor on the detector surface or electrical discharge when high voltage is applied to the detector.

A metal envelope with thin window at its end, the incident photons surrounds the detector. The window is made of either aluminum or beryllium. Beryllium windows as thin as (25) μm have been reported. Commercial cryostats include preamplifier as a standard component **[76]**. Reduction of background in any measurement is very important.

It become necessary in cases when the sample to be counted is very weak radiation source and its activity barely exceeds the background complete elimination of the background radiation is impossible, but reduction of it is feasible by using special shields. Common shields are made of lead or steel with thickness are (0.1- 0.15) m [**77]**.

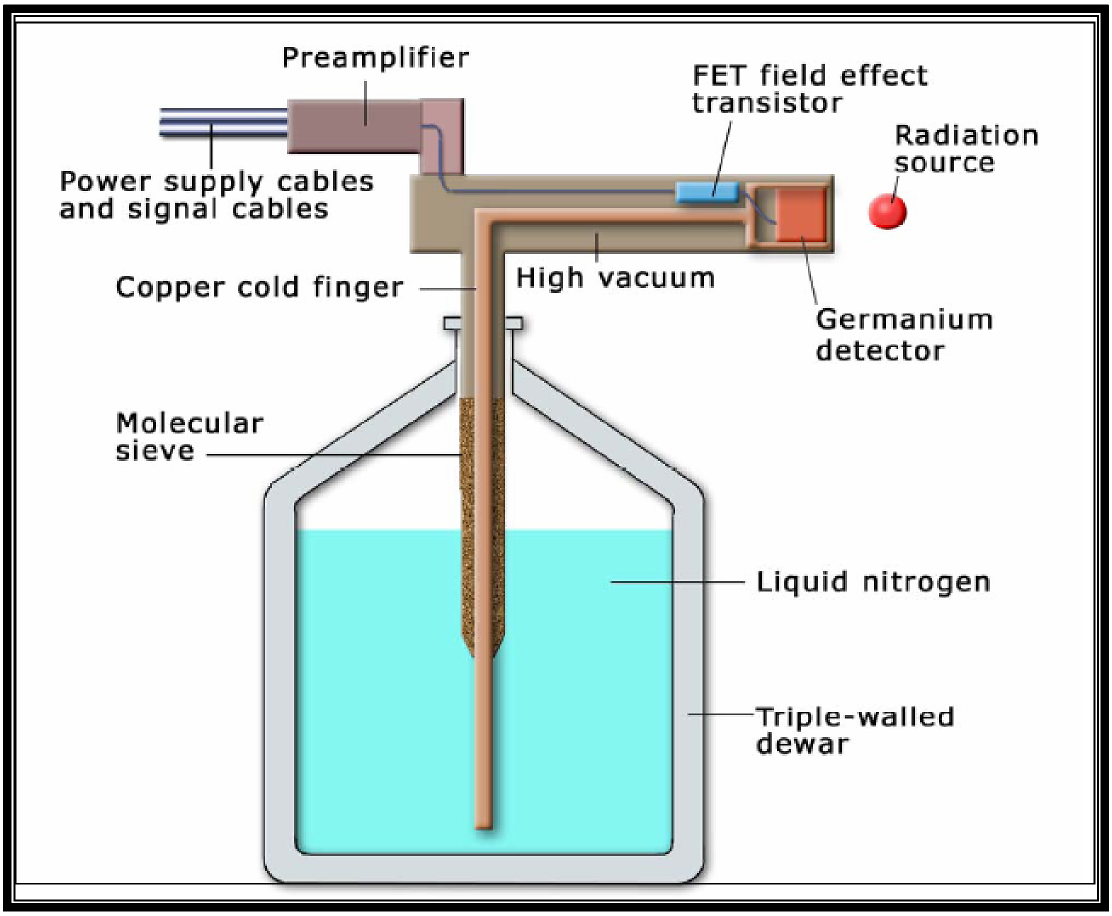

Fig (2-7b): A diagram of horizontal commercially available cryostat **[75]**.

If (HPGe) detector warms up to the room temperature, which many happen if the cryostat runs dry, it loses its ability to operate as a photon detector. It may again become a detector after the lithium drifting process is repeated the considerable. The energy resolution expressed in terms of its FWHM was (2) keV at (1332) keV for Co-60 transition. Efficiency calibration for the specified geometries and energies was performed using a reference standard solution of Europium Eu-152 as shown in Figures (2-8) and (2-9) **[77]**.

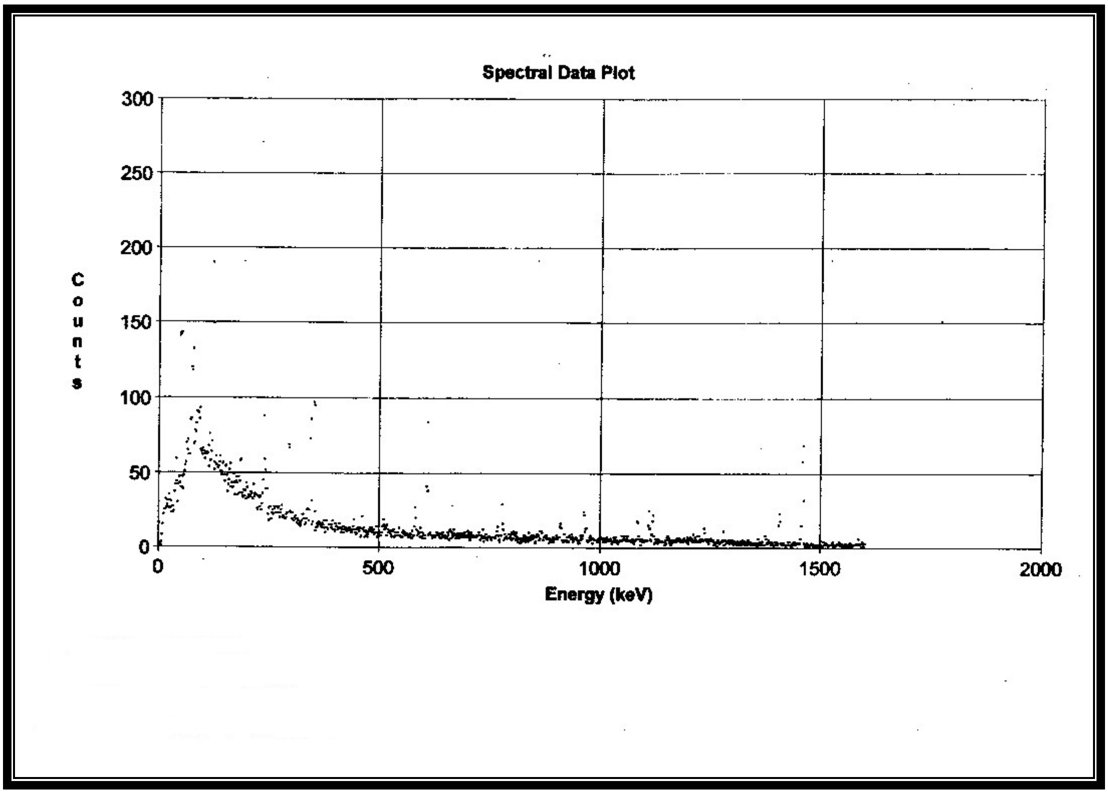

Fig (2-8): Spectrum of Eu-152 for **[77]**.

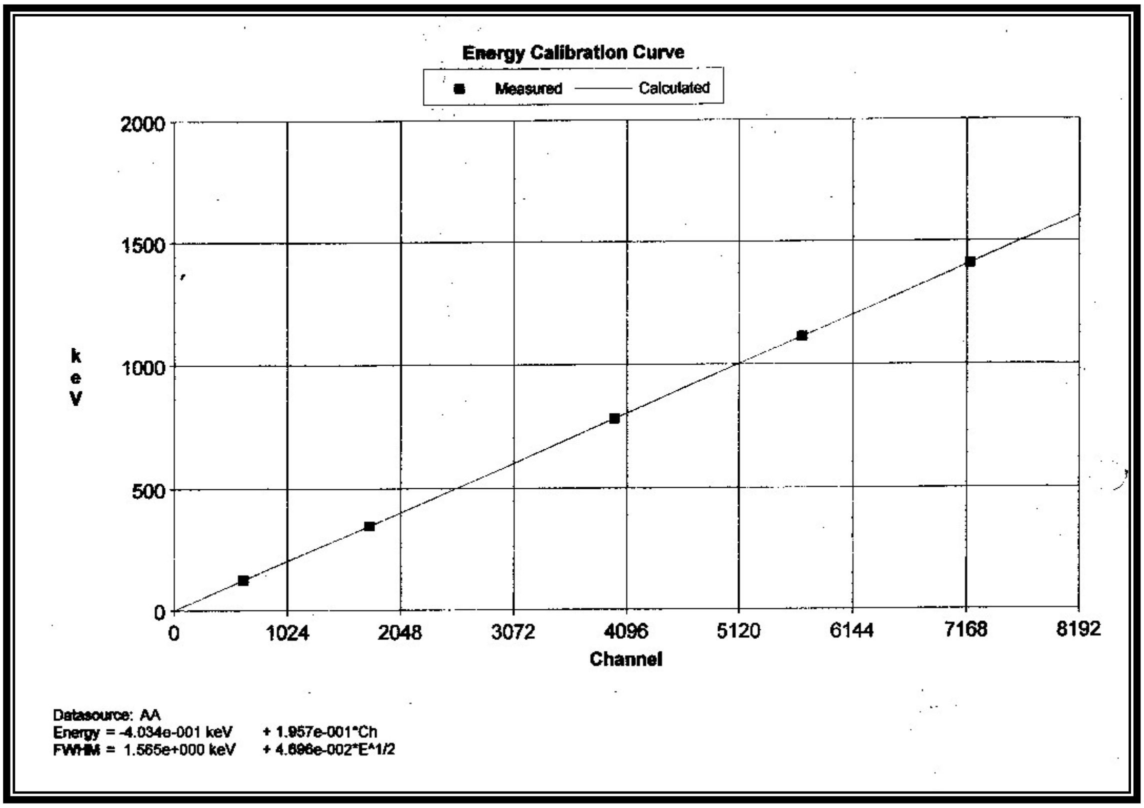

Fig (2-9): Energy calibration for the system **[77]**.

2.15.1 Detection limits of (HPGe) detector

Detection limits is a term used to express the detection capability of a measurement system under certain conditions. An estimate for lowest amount of the activity of a specific gamma-emitting radionuclide that can detect at the time of measurement can calculate from several expressions. General accepted expression for the estimate of limits, which is frequently referred to as the Detection Limit (DL) as shown in equation (2-16) **[78]**:

$$(2.77 + 3.29\sqrt{B.G}\,)\ \mathrm{x}\ \frac{\mathrm{dps}\,(\mathrm{concentration})}{\mathrm{net\ area}}\ \mathrm{DL} = \ldots\ldots\ldots\ldots\ldots\ (2\text{-}16)$$

The B.G of (HPGe) is measured by the equation (2-17):

$$\frac{(\delta)^2 - area}{2} \qquad \mathrm{B.G} = \ \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ (2\text{-}17)$$

Since (δ) is, a percentage of error measured from the final reports. The percentage of error in the measurement of the specific activity ranged from (2% - 6%). The DL provides a means of determining the operation capability of gamma measuring system for (3600) sec, as shown in Table (2-6).

Table (2-6): The detection limit for the isotopes using **(**HPGe) detector

Isotope	Energy (keV)	Detection Limit D.L (Bq/kg)	Background B.G
Ra-226	186.1	3.10	1.35
Pb-212	238.5	0.05	0.30
Bi-214	609.3	0.10	0.20
Cs-137	661.6	0.25	0.01
Bi-212	726	0.08	0.02
Ac-228	911	6.65	0.30
K-40	1460	1.31	2.28

2.15.2 Energy resolution of (HPGe) detector

The energy resolution of (HPGe) detector is an important characteristic because the resolution is a measure of the ability to distinguish between peaks that are similar in energy. Generally, more efficient (HPGe) detectors have lower energy resolution than small, less efficient detectors.

The energy resolution (R) of HPGe detector is (10-100) times better than that of NaI(Tl) scintillation detector and the energy resolution of (HPGe) detector is using the spectrum of Co-60 as shown in Figure (2-10), to measure the energy resolution using equation (2-18).

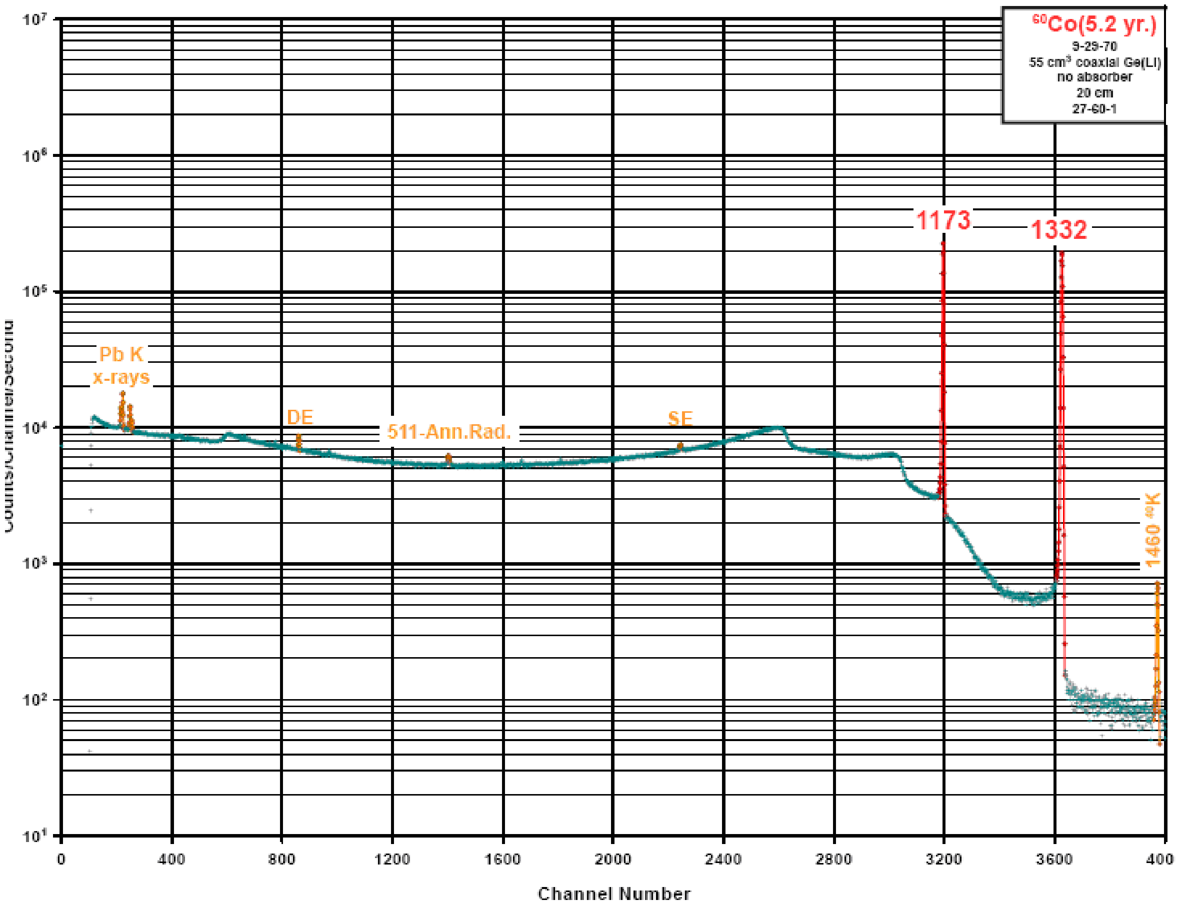

Fig (2-10): The spectrum of radioisotope Co-60 **[79]**

Figure (2-9) used to calculate the energy resolution of the detector using the flowing equation:

ch) ……………………………. (2-18) ΔE /ΔR= FWHM × (

Where:

R: The energy resolution, (keV).

FWHM: Full width at half maximum.

E: The difference in energies between two separated gamma lines. Δ

ch: The difference in channels between the photo peak. Δ

Δ E = $E_2 - E_1$ = 1332 –1173 = (159) keV

Δch = $ch_2 - ch_1$ = 3620 – 3200 = (420) ch

(Δ E / Δch) = 159 / 420 = (0.378) keV / ch

From equation (2-18), R = (5.5) ch × (0.397) keV/ch = (2.07) keV

2.15.3 Analysis of gamma spectrum with (HPGe) detector

The output of the detector is a gamma-ray spectrum, a plot of gamma ray intensity versus energy that is typically divided into (4096) channels. Multiple gamma rays are detected, often two or more from the same isotope, for example (1173) keV and (1332) keV peaks of Co-60. The area beneath these peaks is integrated; typically using computer code, for both samples and standard, and this area is proportional to isotope abundance.

The analysis of complex gamma spectrum is necessary because the spectrums entails first the assignment of energy to the peak of the spectrum and second the determination of the number of counts (the area) for each peak for example as shown in Figure (2-11). The energy assignment to the peaks of the spectrum had accomplished by calibrating the detector with the source that emits gamma of known energy and intensity **[80]**.

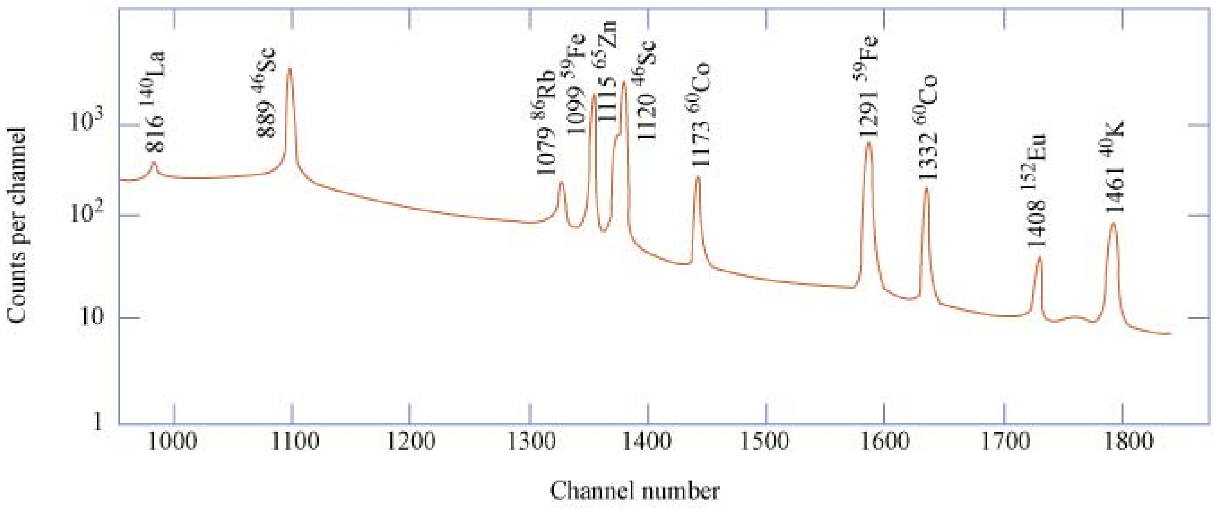

Fig (2-11): Typical gamma ray spectroscopy (1000-1800) channels **[80]**.

2.16 External Dose

The health impact from the accumulation of TENORM must be assessed. Besides the external exposure due to gamma radiation, potential exposure path ways to humans from oilfield TENORM include internal radio contamination by radon gas progeny inhalation and external exposure to the highly penetrating radiation. Internal contamination can occur mainly during equipment cleaning and decontamination activities.

Monitoring radionuclides of the U-238 series and Th-232 series besides K-40 has been done for different areas contaminated by industrial effluents including oil and gas industries **[81].**

For the wastes of the oil and gas industry, the concentrations of radionuclides from the uranium and thorium series and K-40 can be determined by high resolution gamma-ray spectrometry. Most of the radioactivity found in scales and sludge is due to enhanced levels of dissolved radium. Only minute quantities of uranium and thorium have been found in these residues.

The external dose from NORM in the oil waste from the oil locations was calculated by using the following equation **[81]**:

$$H\ (t) = A_x \times DRF \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (2\text{-}19)$$

Where

H (t): The external dose rate at time (t), nGy/h.

A_x : The specific activity of radionuclide of the sample, Bq/kg.

DRF: The dose rate conversion factor.

DRF for Th-232 series = (0.623) nGy/h per Bq/kg.

DRF for U-238 series = (0.461) nGy/h per Bq/kg.

DRF for K-40 = (0.0414) nGy/h per Bq/kg.

Chapter Three

3.1 Introduction

Nuclear spectroscopy is the analysis of radionuclide radiation by measuring the energy distribution of the source. Spectrometer is an instrument that separates the output pulses from a detector, such as (HPGe) detector according to size of crystal; the distribution is proportional to the energy of the detected radiation. The output of the spectrometer provides detailed information that is useful in identifying unknown radioisotopes and counting one isotope in the presence of another **[69].**

This chapter shows the apparatus and experimental details for gamma spectroscopic method adopted using (HPGe) detector for detection and measurement the specific activities of the radioactive elements. In addition, this chapter describes the methods for estimation NORM using CR-39 track detector to measure the uranium concentration in different samples of crude oil and oil waste from the oil locations: (Dora refinery, Bijie refinery and north field of al-Rumaila in Basrah). The materials and the apparatus that used included.

3.2 Collection of the samples

Thirty (solid, liquid) samples were collected from many places with in various dates from the oil locations; the samples were stored for one month at normal laboratory conditions. This time is necessary to get a radiological equilibrating to the samples, before counting the concentration of natural radioactive material for the samples.

3.2.1 Samples of Dora refinery

Twelve samples were taken from many places inside Dora refinery shown in Table (3-1).

Table (3 -1): The location of all samples inside Dora refinery

Sample code	Date	State-type	Location of samples
D1	2/6/2009	solid-sludge	The oil waste from operated units (Al-Sudanya).
D2	2/6/2009	solid-scale	The oil waste from (heavy products).
D3	2/6/2009	solid-sludge	The oil waste from (light products).
D4	2/6/2009	solid-sludge	The oil waste around pit abandonment.
D5	2/6/2009	solid-sludge	The oil waste from (Hi-tech).
D6	2/8/2009	solid-brines	The oil waste from beside of furnace.
D7	2/8/2009	solid-sludge	The depleting area of crude oil.
D8	9/8/2009	solid-sediment	Refining dep. down of furnace.
D9	9/8/2009	liquid-scale	The reached crude oil from Kirkuk.
D10	9/8/2009	liquid-scale	The reached crude oil from Basrah.
D11	9/8/2009	liquid-scale	The oil waste from (pit abandonment).
D12	9/8/2009	liquid-scale	Reduce crude oil (RC).

3.2.2 Samples of the north field of al-Rumaila

Eleven samples were taken from many places inside the north field of Al-Rumaila at dates (2 /9/ 2009) shown in Table (3-2).

Table (3 -2): The location of samples inside the north field of al-Rumaila

sample code	state-type	**location of samples**
B_1	solid-sludge	The fifth station (middle of station).
B_2	solid-sludge	The forth station (compressing of gas).
B_3	sediment	The fifth station (unit for drying).
B_4	solid-scale	The forth station around the pipe (skemar).
B_5	solid-sludge	The second station (separating of gas).
B_6	solid- scale	The second station (storage location of skemar).
B_7	solid- brine	The second station (right side).
B_8	sediment	The second station (down of water separating).
B_9	solid-sludge	The fifth station (down of skemar).
B_{10}	solid-sludge	The fifth station (the back side).
B_{11}	solid- scale	The fifth station (beside of skemar).

Where: Skemar is a place in the north field of al-Rumaila contains many pipes and pumps of oil.

3.2.3 Samples of Bijie refinery

Seven samples were taken from many places inside refinery of Bijie at dates 4/10/2009 shown in Table (3-3).

Table (3 -3): The location of samples inside the refinery of Bijie.

sample code	state-type	**location of samples**
J_1	solid- sludge	The vacuum residue produces from RC.
J_2	solid-scale	The extract from heavy metal from RC.
J_3	solid-scale	The foam wax heavy metal from RC.
J_4	solid -sludge	The oil waste from storage tank.
J_5	solid -scale	Asphalt.
J_6	liquid-scale	Crude oil.
J_7	liquid-scale	RC.

3.3 Materials and Apparatuses

1- Hand mill

It is made from ceramic mortar.

2- Micrometer

Using a micrometer to measure thickness of CR-39 track detectors and the pellets.

3- Sieve

Using sieve which has radius (212) µm, to get clean and pure solid samples without any strange impurities

4- Sensitive balance

Using sensitive balance to weight the samples. This balance is from type of (Mettler Garantia AE163) industrialization in Switzerland

5- Methyl cellulose powder

Using methyl cellulose powder $C_6H_{10}O_5$ as a binding material.

6- Piston

Using piston from type CARVER. INC, Wabash, Indiana, USA, which the strength of piston (1-13) ton and used to press the solid samples in to a pellet with (13) mm diameter and (3) mm thickness.

7- Water bath

The water bath is type of “Labsco” industrialization in Germany is used in this study. It includes a thermostat, which can be operated over a range of (10-100) ^{o}C. However, the water path was carried out at 60 ^{o}C, and distilled water was used as the bath liquid.

8- Irradiation of the oil waste samples

Using (Am–Be) neutron source with flux (5000 n/cm^2.s), which it emits fast neutrons from the (α, n) reaction such as: **[77]**

$$^{9}Be + {}^{4}_{2}He \longrightarrow {}^{12}C + {}^{1}_{0}n_{fast} + 5.76\ MeV$$

The neutron source consists of a rod of (Am–Be) surrounded by a paraffin wax which is used for moderating fast neutrons with energy (10^6) eV to thermal neutrons with energy (0.025) eV, as shown in Figure (3-1).

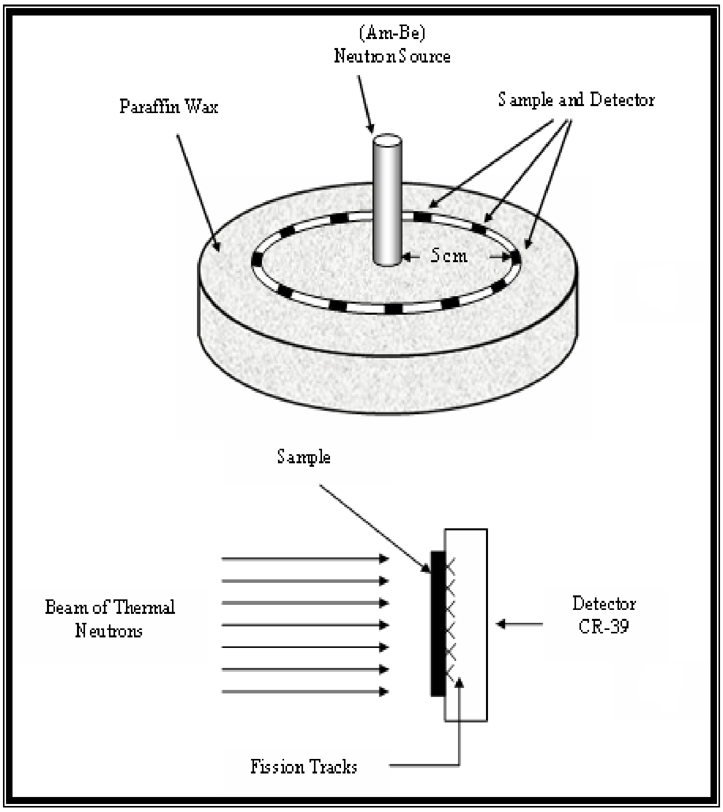

Fig (3-1): The irradiation of the detectors with samples for neutron source.

The time of irradiation was seven days with influence of thermal neutrons is shown in this equation [**77**].

$$^{235}U + {}^{1}_{0}n_{\text{thermal}} \longrightarrow {}^{236}U \longrightarrow {}^{141}Br + {}^{92}Kr + 3\,{}^{1}_{0}n$$

9- Enchant solution

Sodium hydroxide solution with (6.25N) normality has been used for the etching process, which prepared as in equation (3-1) [**72]**:

$$W = W_{eq} \times N \times V_o \qquad \ldots\ldots\ldots\ldots\ldots\ldots (3\text{-}1)$$

Where:

W: weight of NaOH needed to prepare the given normality = (62.5) g
W_{eq} : equivalent weight of NaOH = 23 + 16 + 1 = (40) g / mole

V_o: volume of distilled water = (250) ml.
N: normality = 6.25

The enchant compartment has a volume (250) ml contains the NaOH solution. This apparatus is closed assembly, except for small vent at the top of the condenser tube, which prevents any change of enchant normality concentration during the experiment due to evaporation. The etching time was (7) hr.

10- Optical microscope

The optical microscope type (Motic) industrialization in Malaysia was used. It is capable of giving magnifications of up to (400x) to measure number of tracks. The tracks density is measured from this equation **[78]**:

$$\text{Tracks density } (\rho) = \frac{\text{Average number of total pits (track)}}{\text{Area of field view}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{(3-2)}$$

The tracks in the sample that observed using CR-39 track detector is shown in Figure (3-2).

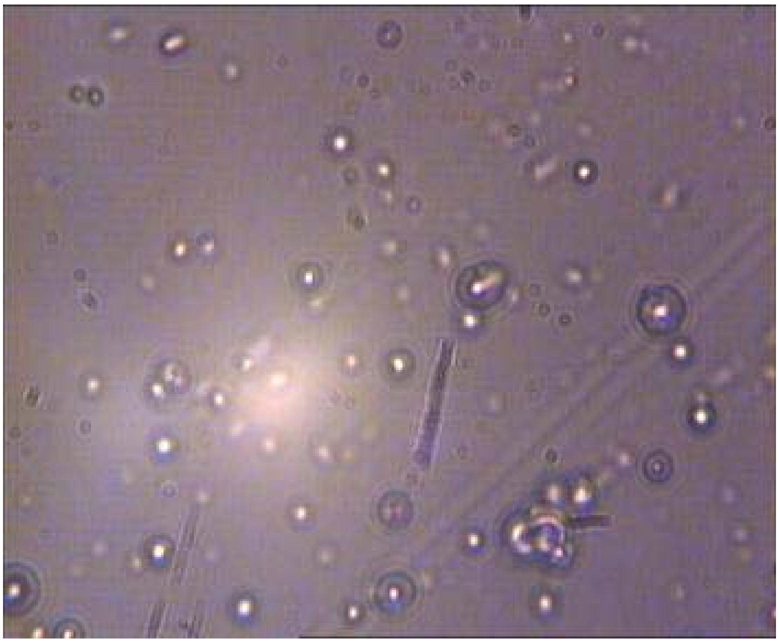

Fig (3-2): photograph of tracks in sample B_6

3.4 Types of detectors that used

In this study two techniques for measurements were used as:

1. Sheets of (250) µm thick of CR-39 plastic (supplied by Pershore Moulding LTD Co. UK) were used. These sheets were cut into small pieces each of (1 x 1) cm^2 area, and stored at normal laboratory conditions.
2. (HPGe), high purity germanium detector as gamma ray spectrometer using Canberra system.

3.4.1 CR-39 track detectors

The chemical composition for organic detector CR-39 is ($C_{12}H_{18}O_7$) and it has density (1.32) g/cm^3. The CR-39 detector is made from a liquid monomer which is made by polymerization in the form of highly cross linked in to homo-polymers, copolymers usually (methyl methacrylate and vinyl acetate) or intermediate products **[58]**.

The plastic CR-39 has a specific name called polyallydigycal carbonate. The symbol CR-39 that it given according to the "Columbia Resins". The detector CR-39 has a high efficiency to record the tracks of charge particles in company with other detectors and it has some specifications as **[59]**:

1- Optically transparent.
2- Very sensitive to radiation.
3- Highly isotropic and homogeneous.
4- Not cross-linking after radiation damage has broken the chemical bonds.
5- Having a non-solvent chemical etching.
6 -This polymer is resistant almost to all solvents and the heating.

The use of CR-39 plastic as a nuclear particle detector become generalized in the fields of dosimetry, spectroscopy and environmental science due to its high sensitivity; most of the applications of this detector are in proton, alpha and neutron dosimetry and radiography as well as for radon dosimetry and cosmic rays studies . The general structure for monomer is contain two groups of ally is ($CH_2 - CH = CH_2$) as shown in Figure (3-3) **[60]**.

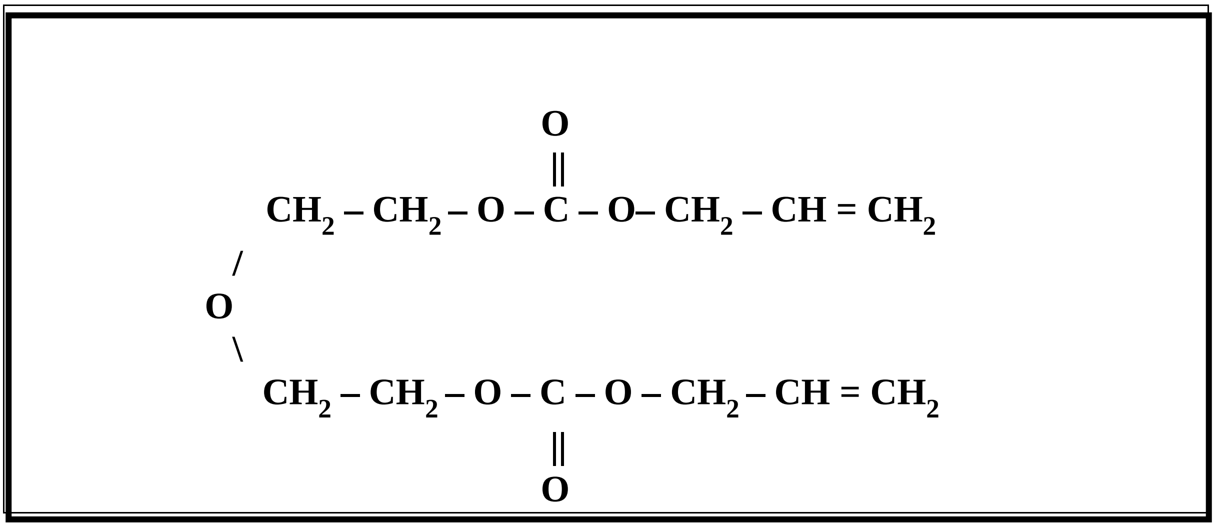

Fig (3-3): The chemical form of monomer CR-39 plastic **[60].**

The experimental details for the samples before using CR-39 track detector involved:

For solid sample:

1. (0.5) g was taken from each sample and grinds using hand mill and cleaned by using special sieve radius (212) μm, then mixed sample with (0.1)g of methylcellulose powder $C_6H_{10}O_5$ which used as a binding material. The mixture was pressed with (3) ton into a pellet. The pellet were covered with CR-39 track detectors on one side and put in a plat of paraffin wax at a distance of (5)cm from the neutron source, all samples were putting in front of the neutron source with flux (5000) n/cm^2.s , as shown in Figure (3-1).

2. After seven days to an (Am-Be) neutron source with total influence (3.024×10^9) n/cm^2, the CR-39 detectors were etched using (NaOH) solution at 60°C in the water path for (7) hr, after etching, the detectors were washed by distilling water then dried and count the number of tracks under an optical microscope.

For liquid sample:

3. (0.1) liter was taken from each sample and immersed CR-39 track detector in the liquid sample for one month by hanging it in order to measure alpha emitters as shown in Figure (3-4).
4. After one month, CR-39 detectors were etched for (7) hr by chemical etching using (NaOH) solution with (6.25 N) using the water path at 60°C, the etched detectors were washed by distilling water and dried then count the number of tracks under an optical microscope.

The background of CR-39 detector was measured then subtracted from the results.

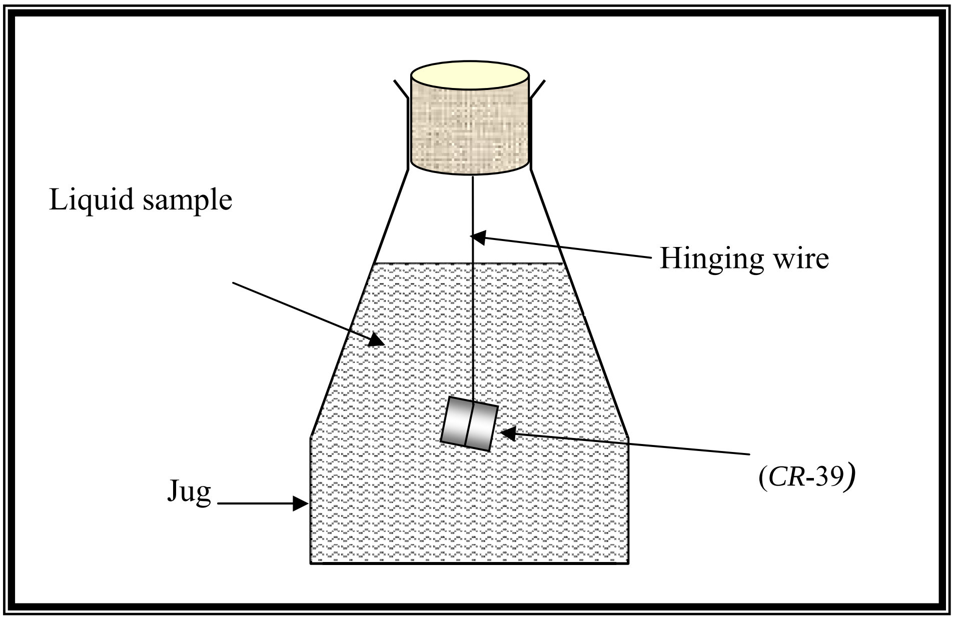

Fig (3-4): CR-39 track detector immersed in liquid sample

3.4.2 Gamma-ray spectrometry (HPGe) detector

Gamma spectrometry is a sensitive method of analysis that can yield analytical data for several different radionuclides in a single sample analysis, which need to shield the detector with suitable material shown in Figure (3-5).

Fig (3-5): photo of (HPGe) detector with shield

Gamma-ray emission from a radionuclide usually proceeded by the particles emissions from radioactive beta decay (either β^-or β^+), and sometimes by alpha decay. The decay leaves the nucleus in either a ground or excited state. If the nucleus is in an excited state, the decay to ground state typically is achieved by gamma emission; nucleus may have many different energy levels through which it must pass before attaining a ground state configuration **[78]**.

The general specification of the system is shown in Table (3-4).

Table (3-4): General specifications of Canberra system.

System	The specifications
Type of detector	Semiconductor ,Ge
Volume of crystal	(3×3) inch
Gas used	Liquid nitrogen at 77 K
Operating voltage	(-2500) V dc
No .of channel	(4096) ch
FWHM for (Co-60)	(1.33) MeV
Relative efficiency	30 %
Counting time for each sample	(3600) sec
Resolution	(2) keV
Diameter of crystal	(59.5) mm
Length of crystal	(58.5) mm
Distance from the window	(5) mm
Detector model	GC 3020
Cryostat model	7500 SL

The intensity of the gamma radiation per unit time can used to quantify the number of radionuclide atoms that have undergone decay. The sample had counted at the same geometry as the standard sample and an absolute efficiency curve obtained. Each radionuclide emits gamma rays at one or more energy characteristic to that nuclide, enabling the spectroscopic to identify radionuclides present in the sample uniquely, not all transitions have the same probability.

If a nuclide emits a gamma ray of a specified energy each time it decays, its transition probability will be 100%. Co-60 was used for calibrating the system, because it has two gamma rays (1.17) MeV and (1.33) MeV that are emitted.

The experimental details for the samples before using (HPGe) involved:

1. Oil waste and crude oil samples that had taken from the refineries are stored in the laboratory for one month to get a radiological equilibrating.
2. One kg was taken from each solid sample and one liter was taken from each liquid sample using sensitive balance.
3. Each sample is putting in the standard marinelli beaker, then tests for (3600) sec using (HPGe) detector.

For each sample that tested using (HPGe) has a report contains all data about the spectrum of the sample for example, number of peaks, energy lines, yield, FWHM, no. of channels, area, efficiency, weight.

This collected data about the sample and the spectrum is very important to the programmer to measure the specific activities of the radionuclides of the samples. The background of (HPGe) was measured for (3600) sec then subtracted from spectrums and results.

Chapter four

4.1 Introduction

In this chapter, the results that include the measurement of the average concentrations of alpha-emitters, concentration of uranium and the specific activates of the radionuclides are presented. Moreover, discussion and conclusions extracted from the present results and suggestions for future work are also presented.

4.2 Uranium concentration using CR-39

Uranium concentration in oil waste samples (solid and liquid) were measured by comparison between tracks density registered on the detectors of the samples and that of the standard samples, this relation is **[82]**:

$$\frac{\rho_x}{\rho_s} = \frac{C_x}{C_s} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots..(4\text{-}1)$$

Where

ρ_x : tracks density in the unknown sample (tracks / mm^2).

ρ_s: tracks density in the standard sample (tracks / mm^2).

C_s : concentration of uranium in the standard sample (ppm).

C_x : concentration of uranium in the unknown sample (ppm).

By plotting the concentrations of uranium with the tracks densities for the standard samples as shown in Figures (4-1) and (4-2), the slope for the straight line is equal to (ρ_s/C_s) then the equation (4-1) is become:

$$C_x = (\rho_x / \text{slope}) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots.. (4\text{-}2)$$

The equation (4-2) was used for both natural exposure and irradiation method.

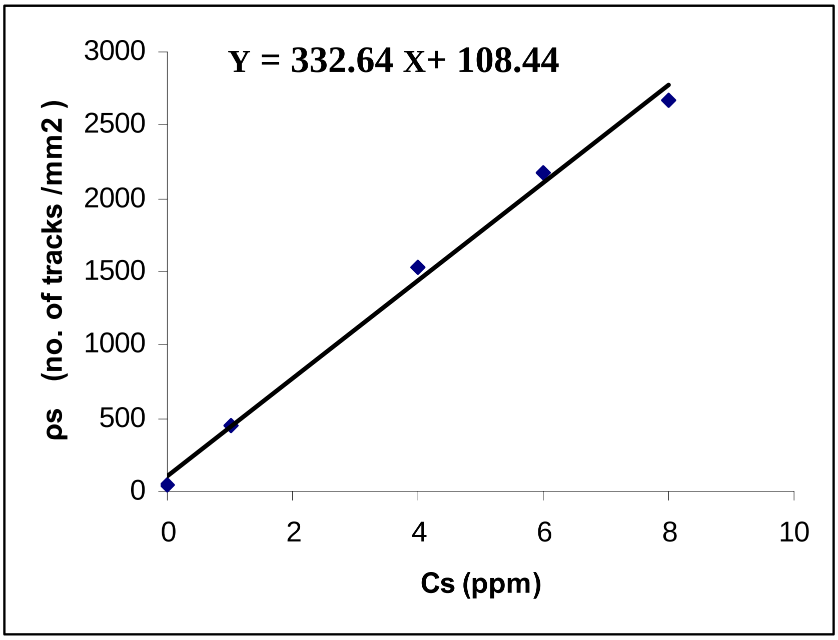

Fig (4-1): The relationship between the uranium concentrations and tracks densities in the standard solid sample **[78]**.

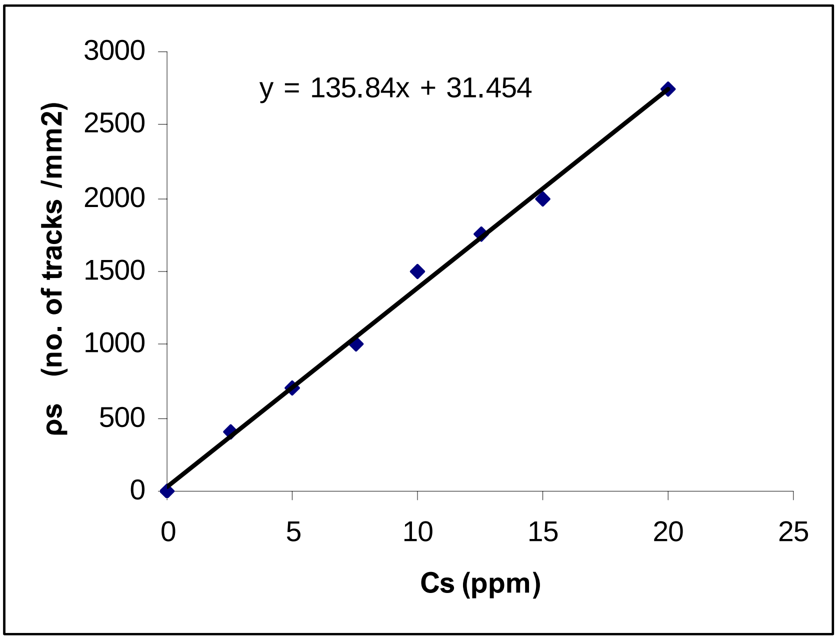

Fig (4-2): The relationship between the uranium concentrations and tracks densities in the standard liquid sample **[78]**.

4.2.1 Samples of Dora refinery

The concentrations and the specific activities of uranium in the solid and liquid samples at dates (2 /8/ 2009) and (4/10/2009) are shown in Tables (4-1), (4-2).

Table (4-1): Concentrations and specific activities of uranium in the solid samples of Dora refinery .

sample code	no. of tracks	ρ_x (tracks/mm^2)	C_x (ppm)	$\mathbf{A}_x$ (Bq/kg)
D_1	27.70 ± 1.64	409.76 ± 24.26	1.23 ± 0.07	15.16 ± 0.86
D_2	45.80 ± 2.14	677.51 ± 31.65	2.03 ± 0.09	25.03 ± 1.10
D_3	21.40 ± 1.46	316.56 ± 21.59	0.95 ± 0.06	11.71 ± 0.73
D_4	67.20 ± 2.59	994.08 ± 38.34	2.98 ± 0.11	36.74 ± 1.35
D_5	38.70 ± 1.96	572.48 ± 28.99	1.72 ± 0.08	21.21 ± 0.98
D_6	51.00 ± 2.25	754.43±33.28	2.26 ± 0.10	27.86 ± 1.23
D_7	40.00 ± 2.00	591.71 ± 29.58	1.77 ± 0.08	21.82± 0.98
D_8	75.20 ± 2.74	1112.42 ± 40.53	3.34 ± 0.12	41.18 ± 1.47
Average	45.83 ± 6.76	678.61 ± 26.05	2.03 ± 1.42	25.09 ± 5.00

From Table (4-1)

- The lowest concentration of uranium was (0.95±0.06) ppm in sample (D_3) while the highest concentration of uranium was (3.34± 0.12) ppm in sample (D_8) , and the average for the samples was (2.03 ± 1.42) ppm.
- The lowest specific activity was (11.71 ± 0.73) Bq/kg of sample (D_3) while the highest specific activity was (41.18 ± 1.47) Bq/kg of sample (D_8), and the average for the samples was (25.09 ± 5.00) Bq/kg.

Table (4-2): Concentrations and specific activities of uranium in the liquid samples of Dora refinery .

sample code	no. of tracks	ρ_x (tracks/m.m^2)	C_x (ppm)	$\mathbf{A}_x$ (Bq/l)
D_9	10.53 ± 1.02	155.76 ± 15.08	1.14 ± 0.11	14.05 ± 1.36
D_{10}	12.70 ± 1.12	187.86 ±16.56	1.38 ± 0.12	17.01 ± 1.47
D_{11}	11.65 ± 1.07	172.33 ± 15.82	1.26 ± 0.11	15.53 ± 1.43
D_{12}	11.42 ± 1.06	168.93 ±15.68	1.24 ± 0.11	15.28 ± 1.41
Average	11.57 ± 3.40	171.22 ±13.08	1.25 ± 1.11	15.46 ± 3.93

From Table (4-2)

- The lowest concentration of uranium was (1.14 ± 0.111) ppm in sample (D_9), while the highest concentration of uranium was (1.38 ± 0.12) ppm in sample (D_{10}), and the average for the samples was (1.25 ± 1.11) ppm.
- The lowest specific activity was (14.05 ± 1.36) Bq/l of sample (D_9), while the highest specific activity was (17.01 ± 1.47) Bq/l of sample (D_{10}), and the average for the samples was (15.46 ± 3.93) Bq/l.

The uranium concentrations in Dora samples are shown in Figure (4-3).

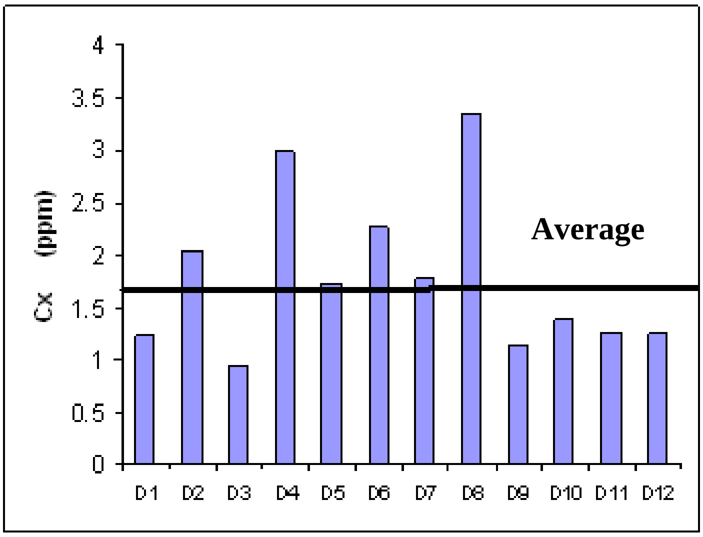

Fig (4-3): Uranium concentrations in Dora refinery samples

4.2.2 Samples of the north field of al-Rumaila

The concentrations and the specific activities of uranium in the solid samples at dates (2-5 /11/ 2009) are shown in Table (4-3).

Table (4-3): Concentrations and specific activities of uranium in the north field of al-Rumaila samples

sample code	no. of tracks	ρ_x (tracks/mm^2)	C_x (ppm)	$\mathbf{A}_x$ (Bq/kg)
B_1	40.10 ± 2.00	593.19 ± 29.58	1.78 ± 0.08	21.94 ± 0.98
B_2	66.40 ± 2.57	982.24 ± 38.01	2.95 ± 0.11	36.37 ± 1.35
B_3	60.50 ± 2.45	894.97 ± 36.24	2.69 ± 0.10	33.16 ± 1.23
B_4	92.50 ± 3.04	1368.34 ± 44.97	4.11 ± 0.13	50.67 ± 1.60
B_5	117.70± 3.43	1730.76 ± 50.73	5.20 ± 0.15	64.11 ± 1.84
B_6	143.00 ± 3.78	2115.38 ± 55.91	6.35 ± 0.16	78.29 ± 1.97
B_7	93.90 ± 3.06	1389.05 ± 45.26	4.17 ± 0.13	51.41 ± 1.60
B_8	50.40 ± 2.24	745.56 ± 33.13	2.22 ± 0.10	27.37 ± 1.23
B_9	44.30 ± 2.10	655.32 ± 31.06	1.97 ± 0.093	24.29 ± 1.14
B_{10}	45.50 ± 2.13	673.07 ± 31.50	2.02 ± 0.094	24.90 ± 1.15
B_{11}	81.40 ± 2.85	1204.14 ± 42.15	3.62 ± 0.12	44.63 ± 1.47
Average	75.97 ± 8.71	1122.91 ± 33.50	3.37 ± 1.83	41.55 ± 6.44

From Table (4-3)

- The lowest concentration of uranium was (1.78 ± 0.08) ppm in sample (B_1), while the highest concentration of uranium was (6.35 ± 0.16) ppm in sample (B_6), the average for all samples was (3.37 ± 1.83) ppm.
- The lowest specific activity was (21.94 ± 0.98) Bq/kg of sample (B_1) while the highest specific activity was (78.29 ±1.97) Bq/kg of sample (B_6), the average for all samples was (41.55 ± 6.44) Bq/kg.

The uranium concentrations in the north field of al-Rumaila samples are shown in Figure (4-4).

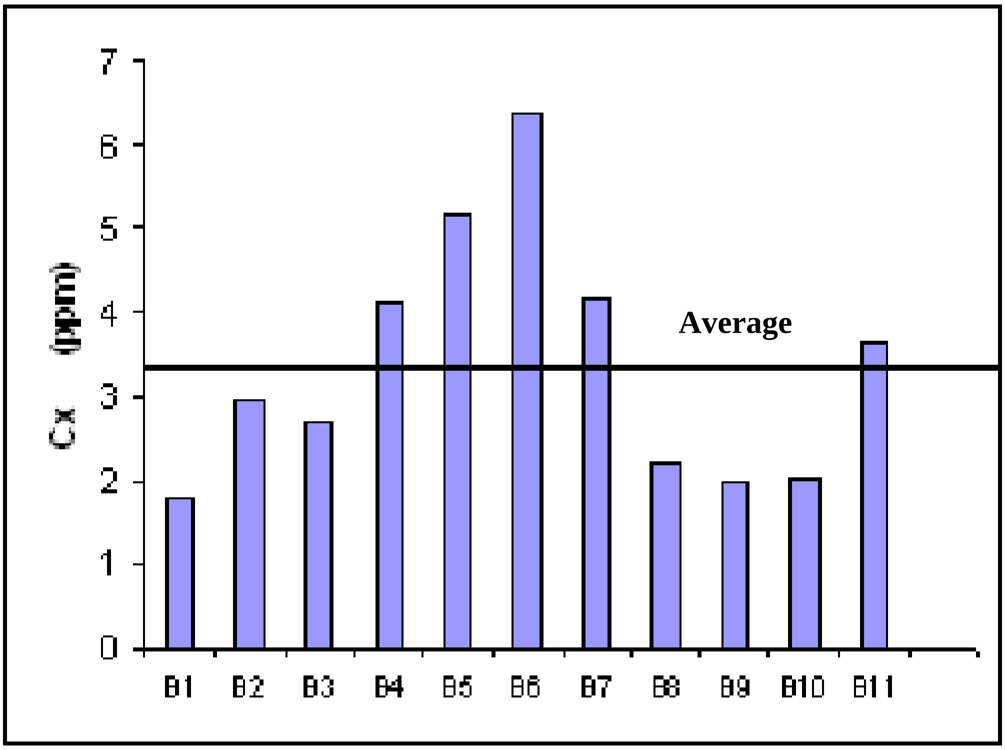

Fig (4-4): Uranium concentrations in the north field of al-Rumaila samples

4.2.3 Samples of Bijie refinery

The concentrations and the specific activities of uranium in the solid and liquid samples at dates (6 /12/ 2009) are shown in Tables (4-4), (4-5).

Table (4-4): Concentrations and specific activities of uranium in solid samples.

sample code	no. of tracks	ρ_x (tracks/mm^2)	C_x (ppm)	$\mathbf{A}_x$ (Bq/kg)
J_1	9.47 ± 0.97	140.08±14.34	0.42±0.043	5.17±0.53
J_2	10.22 ± 1.01	151.18±14.94	0.45±0.044	5.54±0.54
J_3	19.52 ± 1.39	288.75± 20.56	0.86 ± 0.06	10.60 ± 0.73
J_4	11.72 ±1.08	173.37±15.97	0.52±0.048	6.41±0.59
J_5	12.32 ±1.10	182.24±16.27	0.54±0.048	6.65±0.59
Average	12.65 ± 3.54	187.12 ± 13.67	0.55 ± 0.04	6.87± 2.62

From Table (4-4)

- The lowest concentration of uranium was (0.42±0.043) ppm in sample (J_1) while the highest concentration of uranium was (0.86± 0.06) ppm in sample (J_3), the average for the samples was (0.55 ± 0.04) ppm.
- The lowest specific activity of the samples was (5.17±0.53) Bq/kg of sample (J_1) while the highest specific activity was (10.60 ± 0.73) Bq/kg of sample (J_3), the average for the samples was (6.87± 2.62**)** Bq/kg.

Table (4-5): Concentrations of uranium and specific activities in the liquid samples.

sample code	no. of tracks	ρ_x (tracks/mm^2)	C_x (ppm)	$\mathbf{A}_x$ (Bq/l)
J_6	13.44 ± 1.15	198.81±17.01	1.46 ± 0.12	18.00 ± 1.47
J_7	9.44 ± 0.97	139.64± 14.34	1.02± 0.10	12.57 ±1.23
Average	11.44 ±3.38	169.22 ± 13.00	1.24 ± 1.11	15.28 ± 3.90

From Table (4-5)

- The lowest concentration of uranium was (1.02± 0.10) ppm in sample (J_7) while the highest concentration of uranium was (1.46± 0.12) ppm in sample (J_6), the average for the samples was (1.24 ± 1.11) ppm.
- The lowest specific activity was (12.57 ±1.23) Bq/l of sample (J_7), while the highest specific activity was (18.00 ± 1.47) Bq/l of sample (J_6), the average for the samples was (15.28 ± 3.90**)** Bq/l .

Uranium concentrations in Bijie refinery samples are shown in figure (4-5).

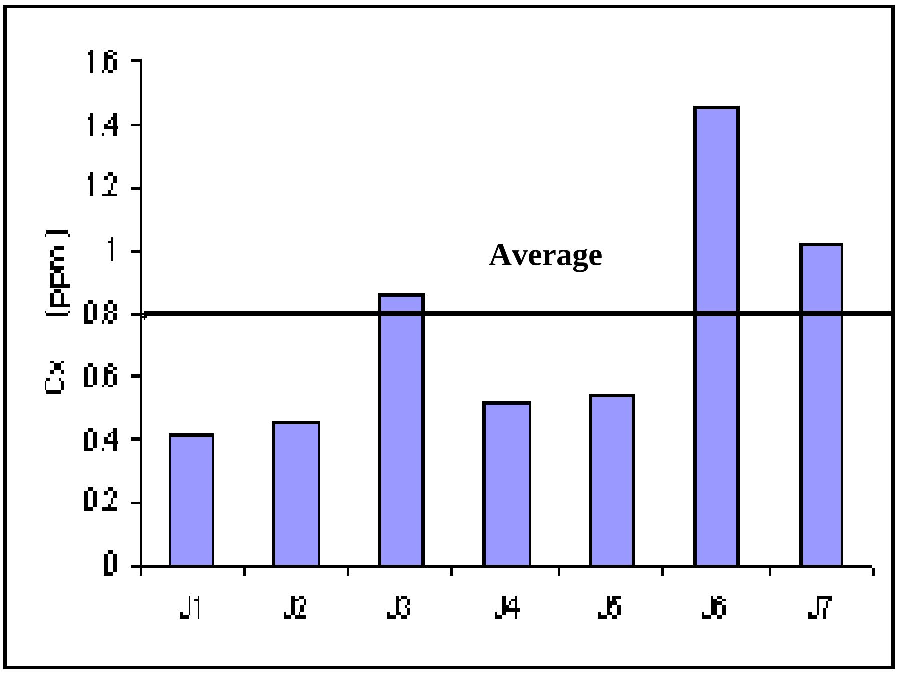

Fig (4-5): Uranium concentrations in Bijie refinery samples

The average of uranium concentrations in all samples in the oil locations are shown in Table (4-6) and Figure (4-6) below:

Table (4-6): The average of uranium concentrations in all the samples from all the oil locations.

Location of oil	C_x (ppm)
Dora refinery	1.64 ± 0.09
North field of Rumaila	3.37± 1.83
Bijie refinery	0.89 ± 0.07

From Table (4-6)

- The lowest concentration of uranium was (0.89 ± 0.07) ppm in samples of Bijie refinery while the highest concentration of uranium was (3.37± 0.11) ppm in samples of the north field of al-Rumaila.

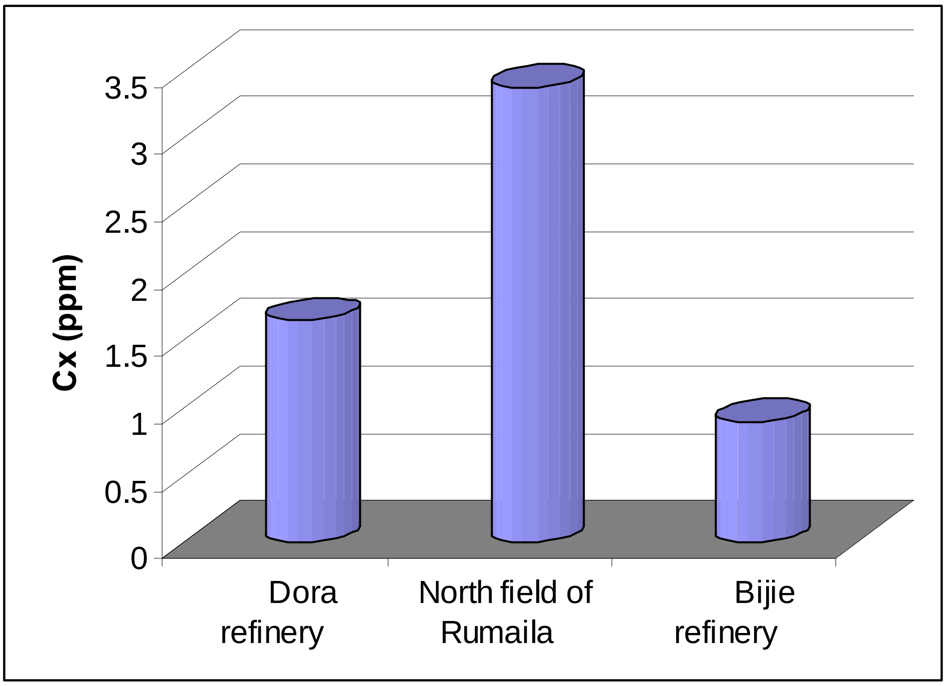

Fig (4-6): The average of uranium concentrations in all samples from the oil locations.

4.3 Specific activities of radionuclides using (HPGe) detector

Twenty samples of crude oil and oil waste were tested with counting time (3600) sec.

4.3.1 Samples of Dora refinery

Twelve samples had tested to measure the specific activities for the radionuclides as shown in Table (4-7).

Table (4-7): The specific activities of the samples of Dora refinery

Sample code	Bi-214 Bq/kg , Bq/l	Ra-226 Bq/kg, Bq/l	Tl-208 Bq/kg , Bq/l	Bi-212 Bq/kg , Bq/l	Pb-212 Bq/kg , Bq/l	K-40 Bq/kg , Bq/l	Cs-137 Bq/kg , Bq/l
D_1	6.61	12.60	7.34	3.34	5.52	117.86	B.D.L
D_2	6.92	20.31	7.12	2.22	5.81	107.56	5.20
D_3	8.41	4.11	9.91	5.61	8.32	178.9	0.70
D_4	8.52	18.6	10.00	6.92	7.93	180.66	B.D.L
D_5	6.22	14.10	7.62	4.94	5.32	140.62	0.75
D_6	8.95	29.81	11.01	6.12	6.88	184.91	0.53
D_7	8.83	14.54	10.13	6.66	6.74	189.31	0.32
D_8	39.07	79.03	32.31	18.62	22.53	76.12	0.31
D_9	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	5.24	B.D.L
D_{10}	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	6.74	B.D.L
D_{11}	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	2.23	B.D.L
D_{12}	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	1.82	B.D.L
Average	7.79±2.79	16.08±4.01	7.95±2.82	4.05±2.01	5.75±2.39	99.2±9.9	0.64±0.3

B.D.L below detection limit

Seven radionuclides had been detected (Bi-214, Ra-226, Tl-208, Bi-212, Pb-212, K-40 and, Cs-137) with energy line and yield are shown in Table (4-8).

Table (4-8): The energy lines and yield of the radionuclides

The radionuclides	Energy keV	Yield %
Ra-226	186.1	3.2
Pb-212	238.5 , 300	43.6 , 3.34
Tl-208	583.02	86
Bi-214	609.3 , 1120.2	46.1 , 15
Cs-137	661.6	85
Bi-212	726	6.6
K- 40	1460	10.6

From Table (4-7)

- The lowest specific activities of Bi-214 was (B.D.L) for the samples (D_9, D_{10}, D_{11}, D_{12}) while the highest specific activities of Bi-214 with two energy lines (609.3) keV and (1120) keV was (39.07) Bq/kg for sample (D_8) which show high existence of the U-238 radionuclide in this sample compared with the others, the average of Bi-214 for all samples was (7.79±2.79) Bq/kg.
- The lowest specific activities of Ra-226 was (B.D.L) for the samples (D_9,D_{10},D_{11},D_{12}) while the highest specific activities of Ra-226 with energy line (186.1) keV is (79.03) Bq/kg for sample (D_8), the average for all samples was (16.08±4.01) Bq/kg .
- The lowest specific activities of Tl-208 was (B.D.L) for the samples (D_9,D_{10},D_{11},D_{12}) while the highest specific activities of Ac-228 with energy line (911) keV gamma rays was (32.31) Bq/kg for sample (D_8), the average for all samples was (7.95±2.82) Bq/kg .

- The lowest specific activities of Bi-212 was (B.D.L) for the samples (D_9,D_{10},D_{11},D_{12}) while the highest specific activities of Bi-212 with energy line (762) keV was (18.62) Bq/kg for sample (D_8), the average for all samples was (4.05±2.01) Bq/kg.
- The lowest specific activities of Pb-212 was (B.D.L) for the samples (D_9,D_{10},D_{11},D_{12}) while the highest specific activities of Pb-212with two energy lines(238.5)keV and (300)keV was(22.53)Bq/kg for sample(D_8), the average for all samples was (5.75±2.39) Bq/kg.
- The lowest specific activities of K-40 was (1.82) Bq/kg for sample (D_{12}) while the highest specific activities of K-40 was (189.31) Bq/kg for sample (D_7), with the energy line (1460) keV, the average for all samples was (99.2±9.9) Bq/kg
- The lowest specific activities of artificial radionuclide Cs-137 was (B.D.L) for samples (D_1, D_4, D_9, D_{10}, D_{11},D_{12}), while the highest specific activities with energy line (661.6) keV was (5.2) Bq/kg for sample (D_2), the average of all samples was (0.64±0.3) Bq/kg.

The specific activities for Dora refinery samples are shown in Figures, (4-7), (4-8), (4-9), (4-10), (4-11), (4-12), (4-13).

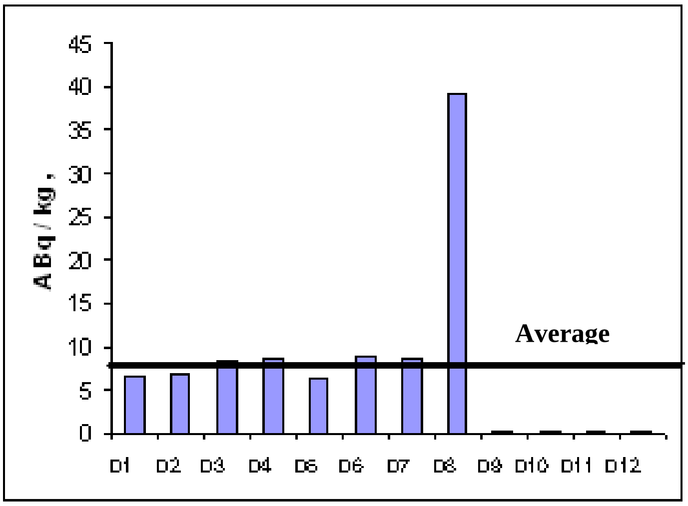

Fig (4-7): The specific activities of Bi-214 of the samples

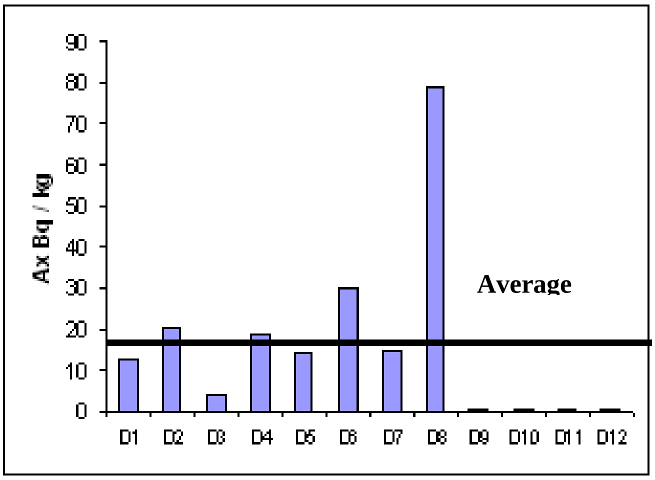

Fig (4-8): The specific activities of Ra-226 of the samples

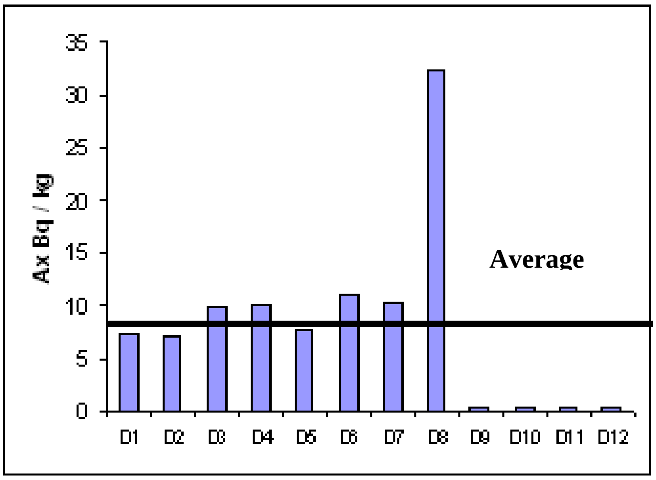

Fig (4-9): The specific activities of Tl-208 of the samples

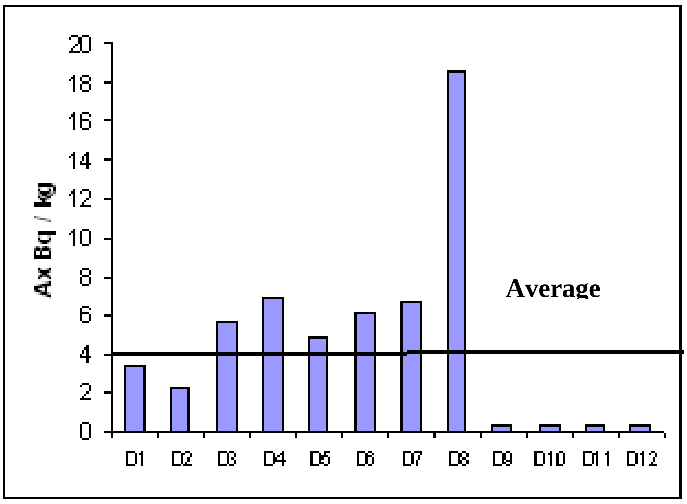

Fig (4-10): The specific activities of Bi-212 of the samples

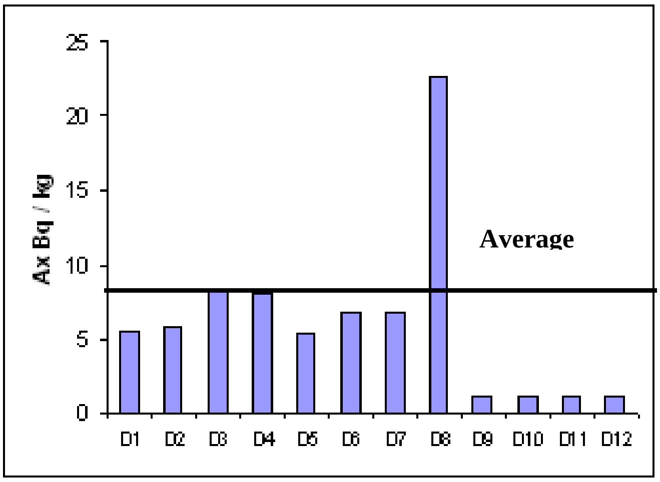

Fig (4-11): The specific activities of Pb-212 of the samples

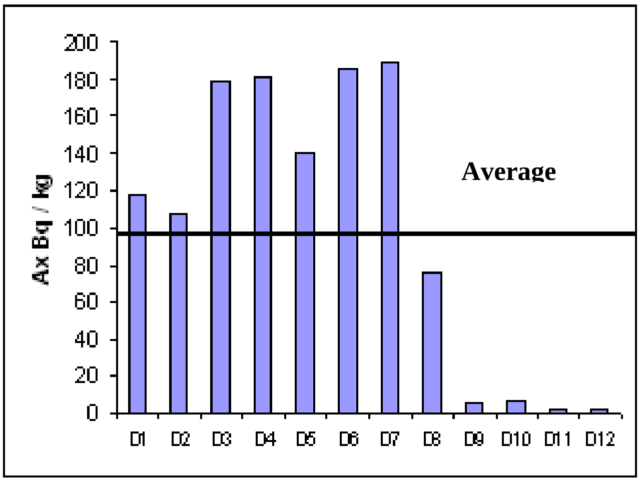

Fig (4-12): The specific activities of K-40 of the samples

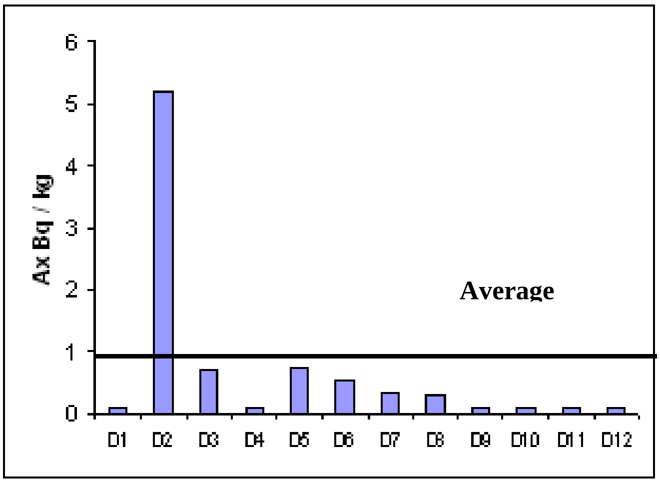

Fig (4-13): The specific activities of Cs-137 of the samples

Gamma-rays spectrum for samples of Dora refinery that observed using (HPGe) detector are shown in Figures (4-14), (4-15), (4-16), (4-17), (4-18), (4-19), (4-20), (4-21), (4-22), (4-23), (4-24), (4-25).

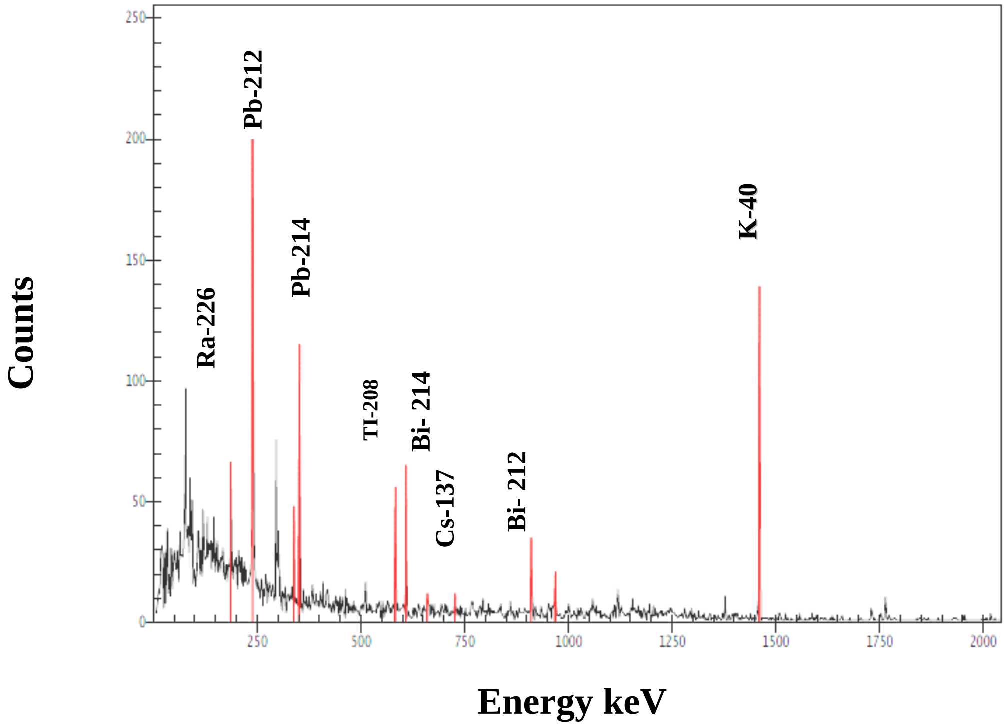

Fig (4-14): Gamma-rays spectrum for sample (D1)

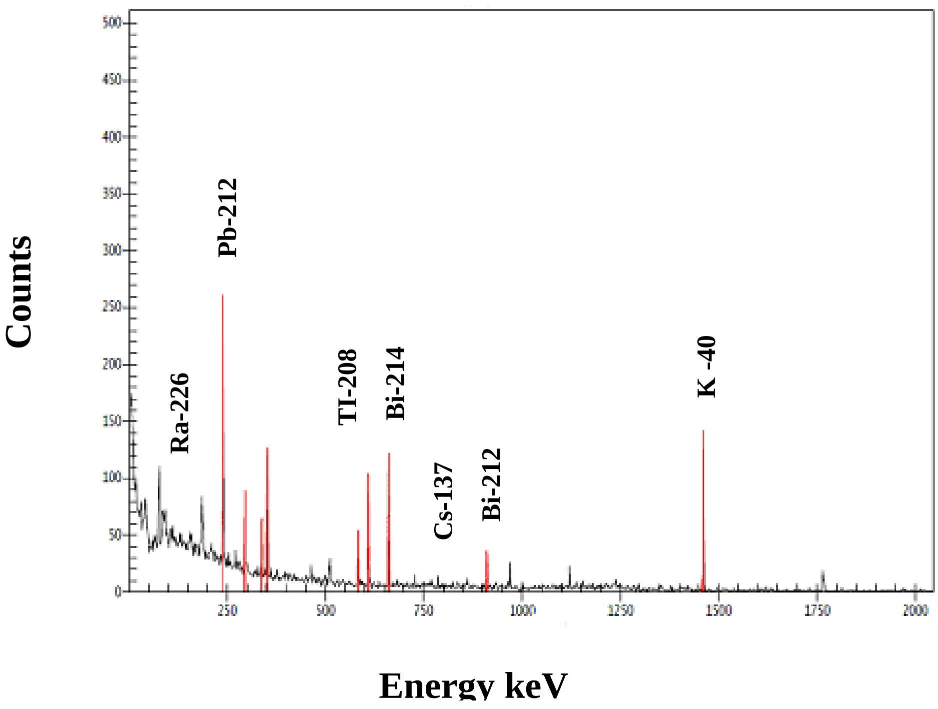

Fig (4-15): Gamma-rays spectrum for sample (D_2)

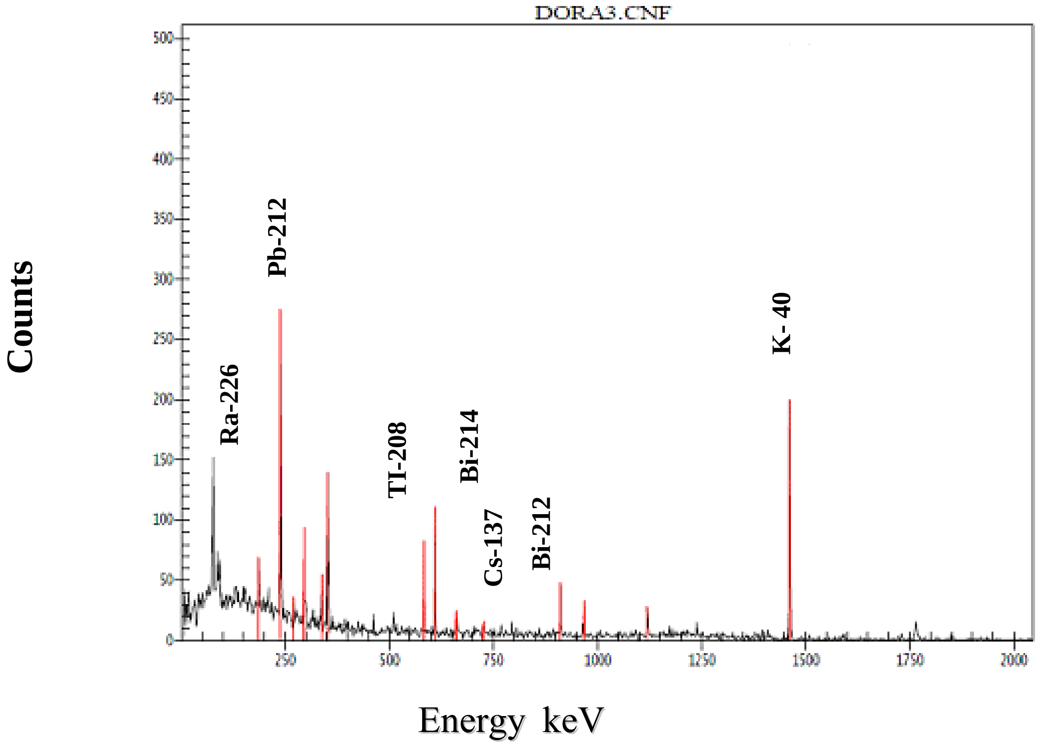

Fig (4-16): Gamma-rays spectrum for sample (D_3)

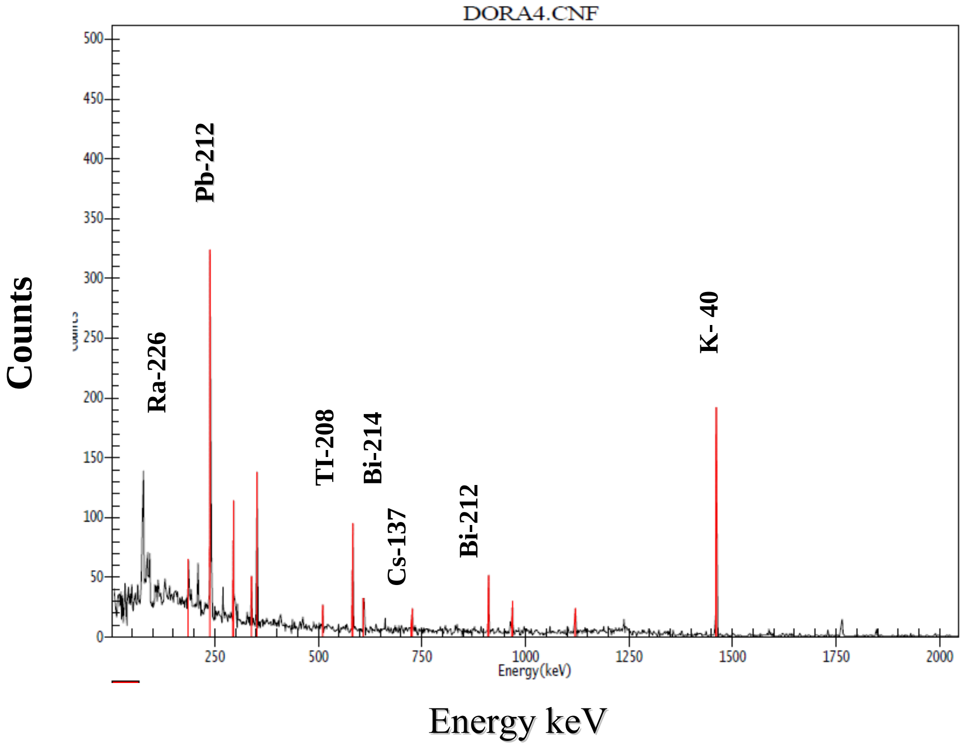

Fig (4-17): Gamma-rays spectrum for sample (D_4)

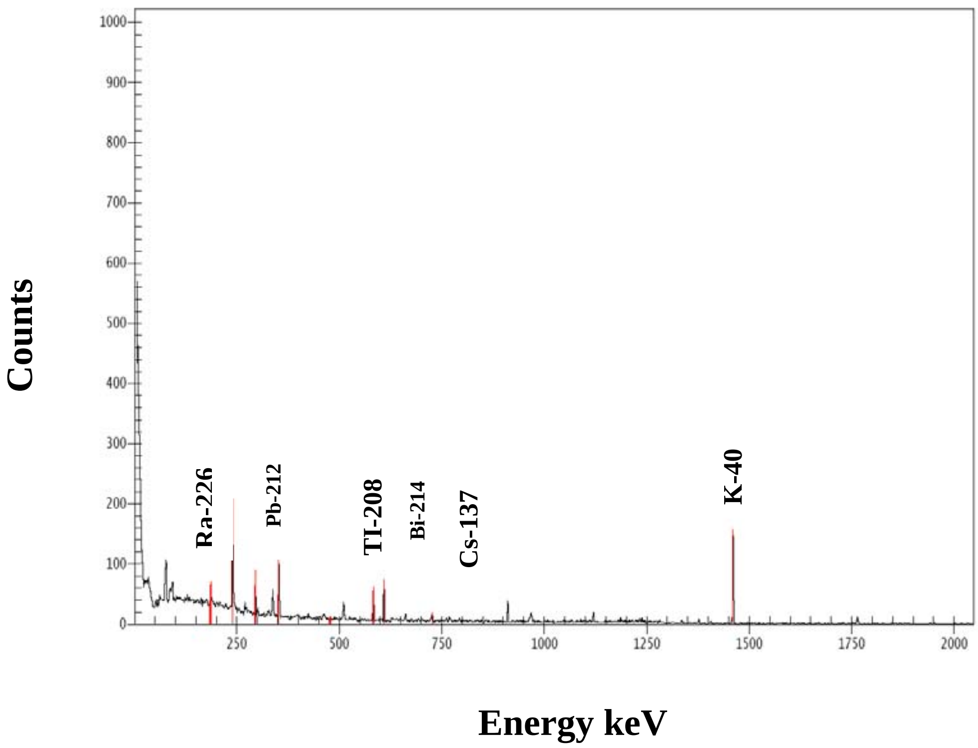

Fig (4-18): Gamma-rays spectrum for sample (D_5)

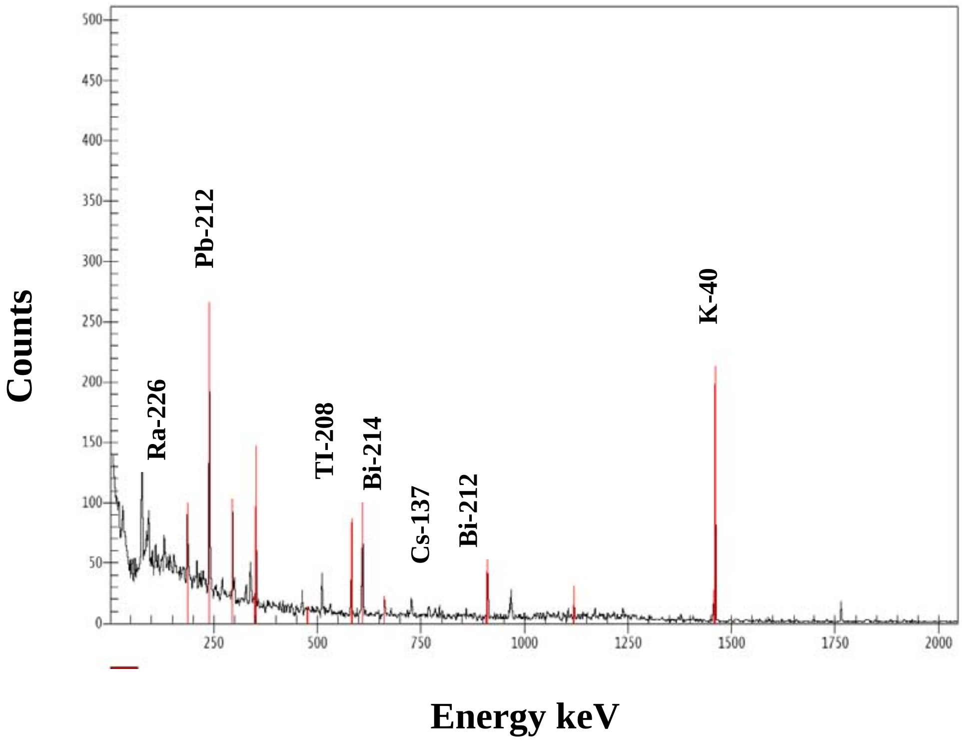

Fig (4-19): Gamma-rays spectrum for sample (D_6)

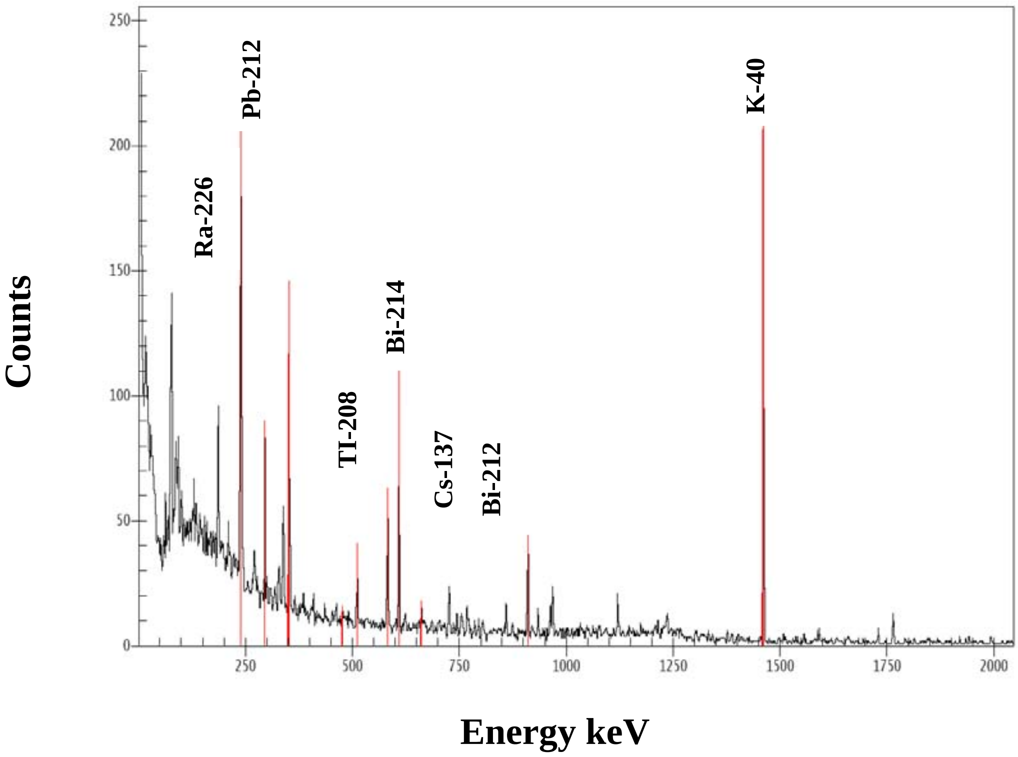

Fig (4-20): Gamma-rays spectrum for sample (D_7)

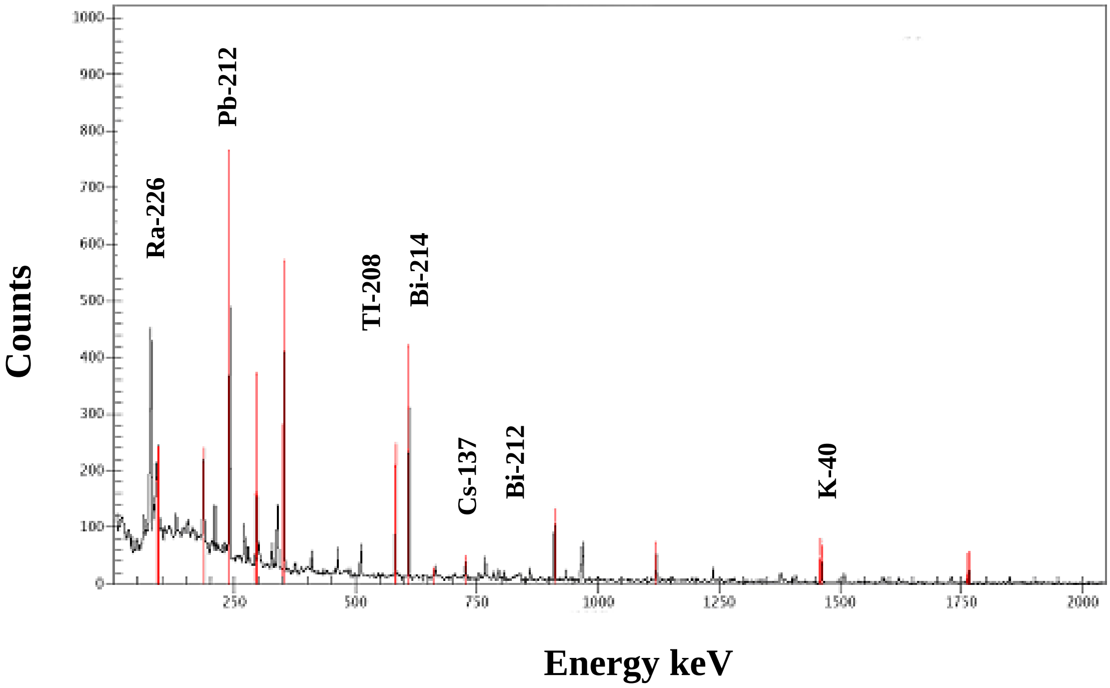

Fig (4-21): Gamma-rays spectrum for sample (D_8)

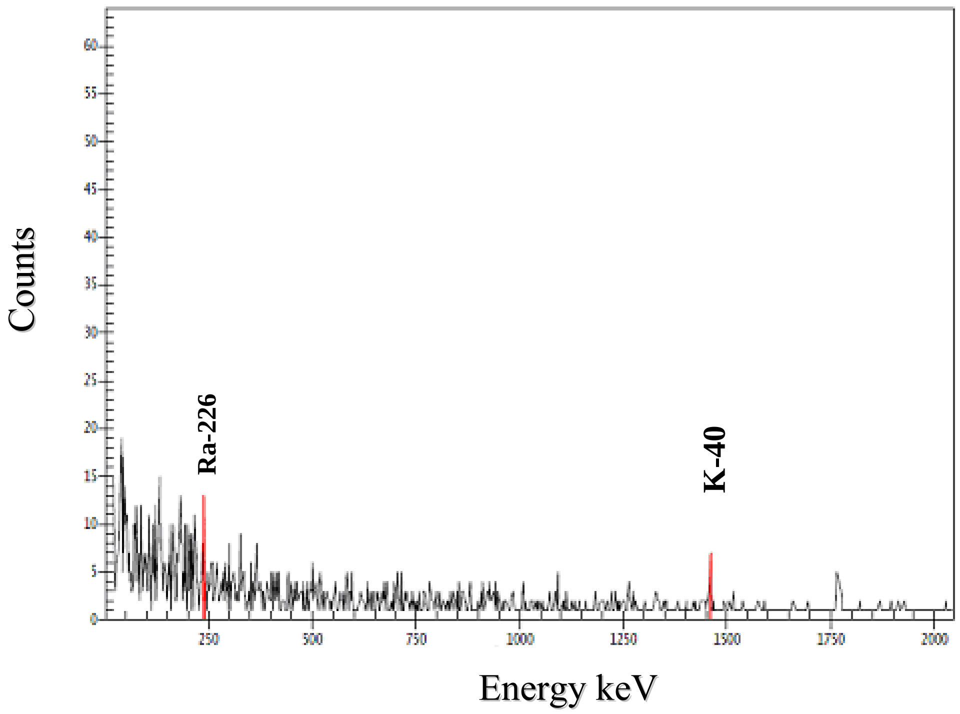

Fig (4-22): Gamma-rays spectrum for sample (D_9)

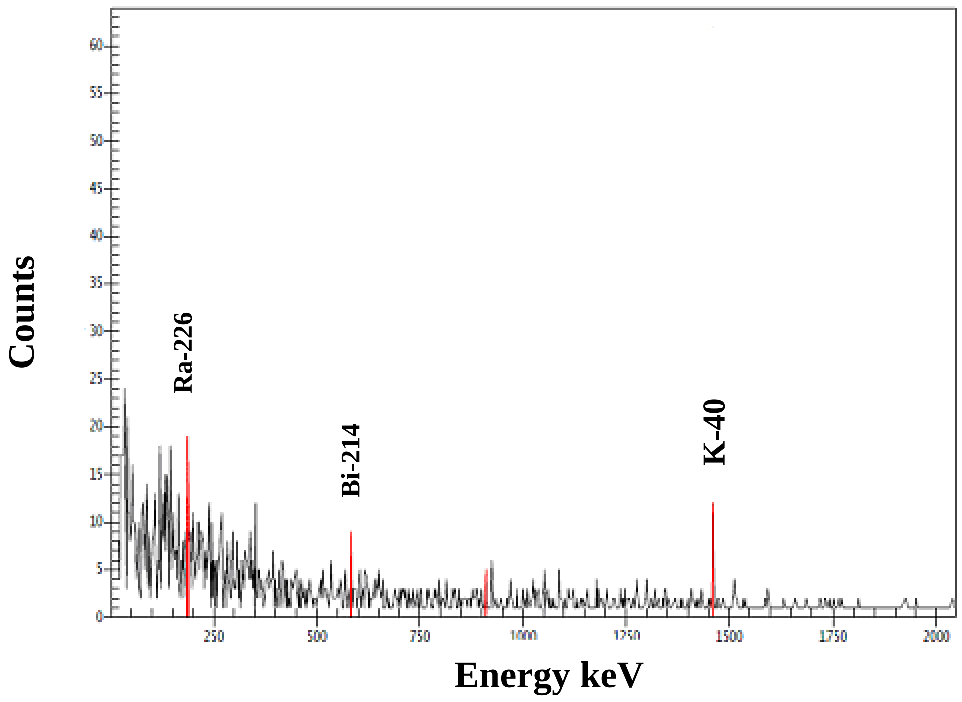

Fig (4-23): Gamma-rays spectrum for sample (D_{10})

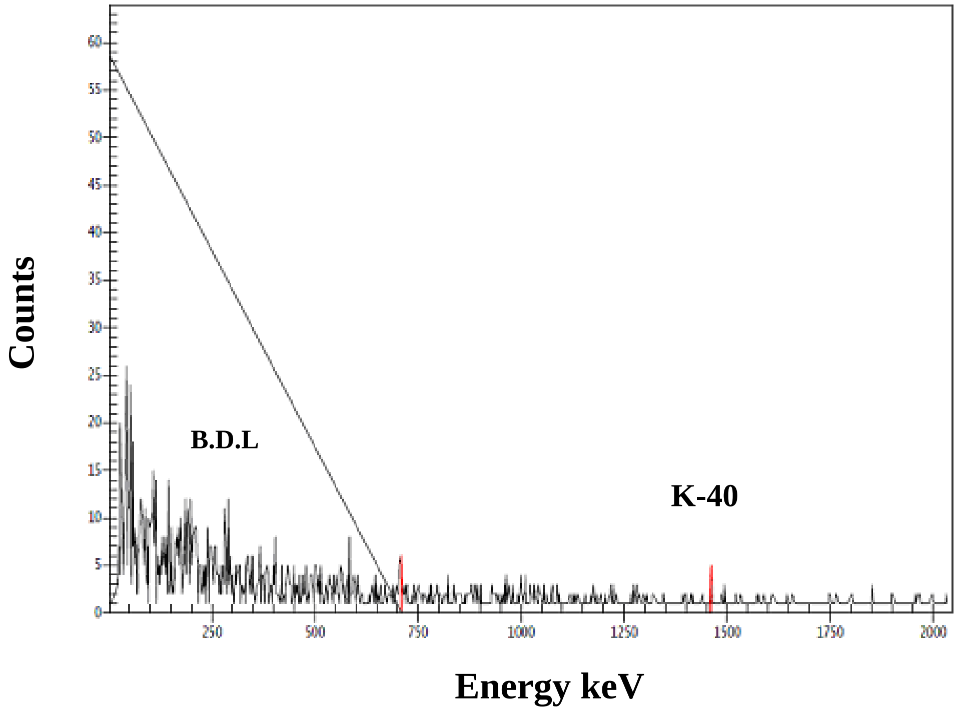

Fig (4-24): Gamma-rays spectrum for sample (D_{11})

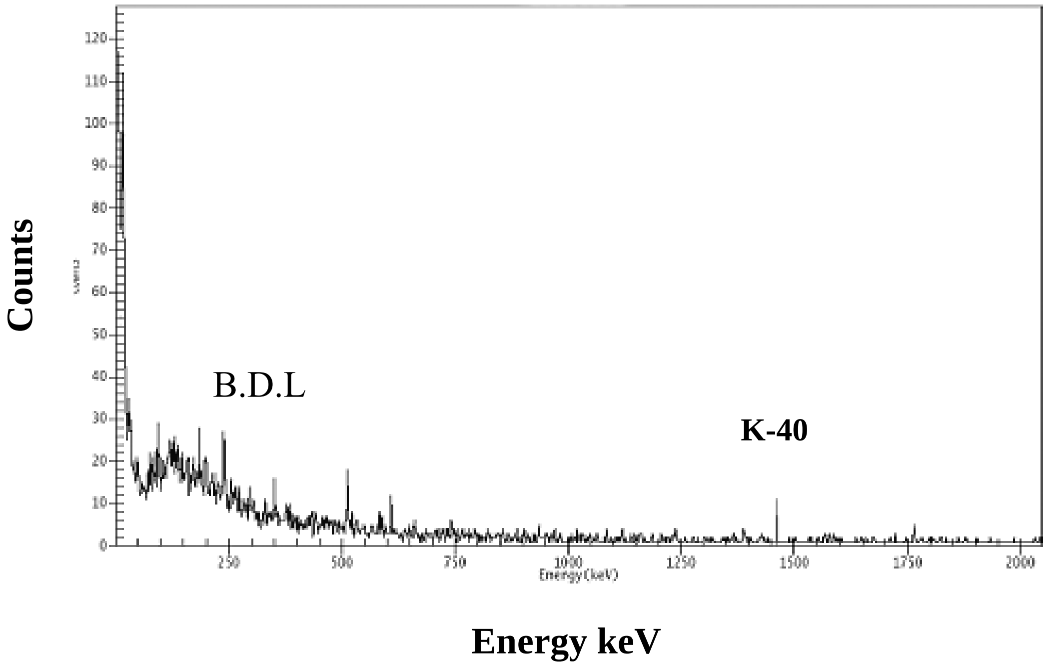

Fig (4-25): Gamma-rays spectrum for sample (D_{12})

4.3.2 Samples of Bijie refinery

Seven samples was tested to measure the specific activities for the radionuclides at dates (30 /12/ 2009), as shown in Table (4-9).

Table (4-9): The specific activities of samples of Bijie refinery

Sample code	Bi-214 Bq/kg , Bq/l	Ra-226 Bq/kg ,Bq/l	Tl-208 Bq/kg , Bq/l	Bi-212 Bq/kg , Bq/l	Pb-212 Bq/kg , Bq/l	K-40 Bq/kg , Bq/l	Cs-137 Bq/kg , Bq/l
J_1	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	1.5	B.D.L
J_2	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	1.7	B.D.L
J_3	1.90	3.2	B.D.L	B.D.L	B.D.L	5.73	B.D.L
J_4	0.15	1.5	B.D.L	0.20	0.10	2.54	B.D.L
J_5	0.21	2.0	B.D.L	B.D.L	B.D.L	3.12	B.D.L
J_6	0.53	2.4	B.D.L	B.D.L	B.D.L	4.64	B.D.L
J_7	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	1.8	B.D.L
Average	0.39±0.62	1.3± 1.14	B.D.L	0.02	0.01	3.0±1.7	B.D.L

From Table (4-9)

- Seven radionuclides had been detected in crude oil and oil waste as, (Bi-214, Ra-226, Tl-208, Bi-212, Pb-212, K-40, Cs-137).
- The lowest specific activities of Bi-214 was (B.D.L) for the samples (J1, J2, J7), while the highest specific activities was (3.2) Bq/kg for sample (J_3), and the average for all samples was (1.3± 1.14) Bq/kg.
- The lowest specific activities of Ra-226 was (B.D.L) for the samples (J1,J2,J7)), while the highest specific activities was (1.90) Bq/kg for sample (J_3), and the average for all samples was (0.39±0.62) Bq/kg.
- The specific activities and the average of Tl-208 for all samples was (B.D.L).
- The lowest specific activities of Bi-212 was (B.D.L) for all samples except sample (J_4) was (0.2) Bq/kg, and the average was (0.02) Bq/kg.

- The lowest specific activities of Pb-212 was (B.D.L) for all samples except sample (J_4) was (0.1)Bq/kg, and the average of the samples was (0.01)Bq/kg.
- The lowest specific activities of K-40 was (1.5) Bq/kg from the sample (J_1) while the highest specific activities was (5.73) Bq/kg for sample (J_3), and the average for all samples was (3.0±1.7) Bq/kg.
- The specific activities and the average of Cs-137 for all samples was (B.D.L).

The specific activities for the radionuclides in samples of Bijie refinery are shown in Figures: (4-26), (4-27), (4-28), (4-29), (4-30), (4-31) (4-32).

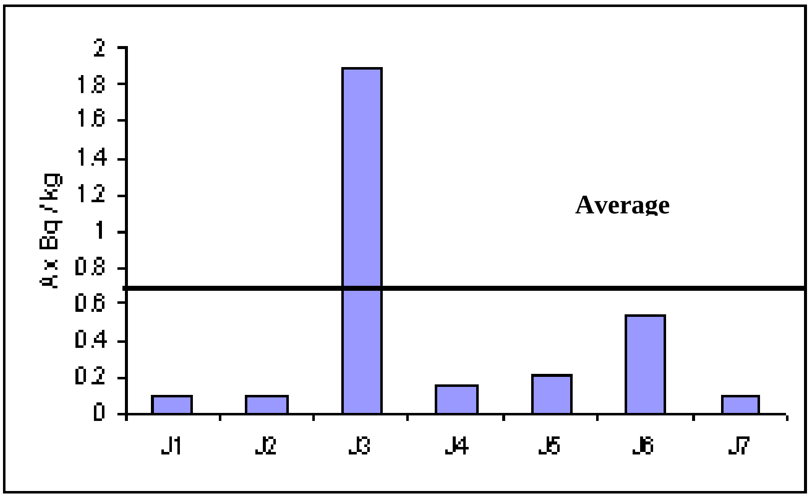

Fig (4-26): The specific activities for Bi-214 of the samples

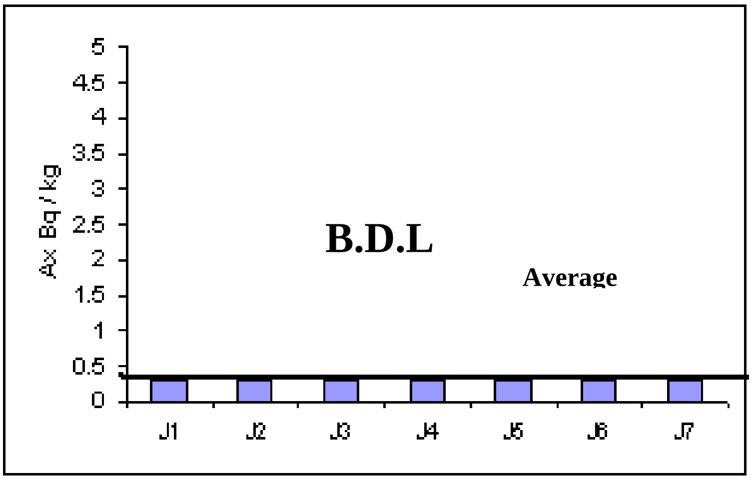

Fig (4-27): The specific activities for Ra-226 of the samples

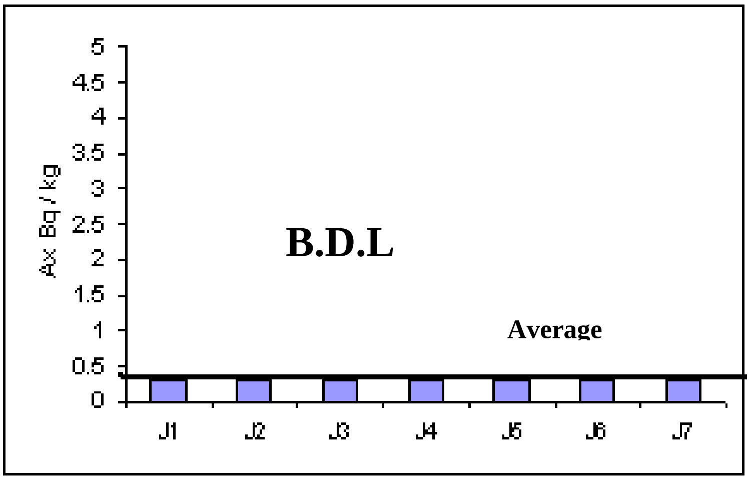

Fig (4-28): The specific activities for Tl-208 of the samples

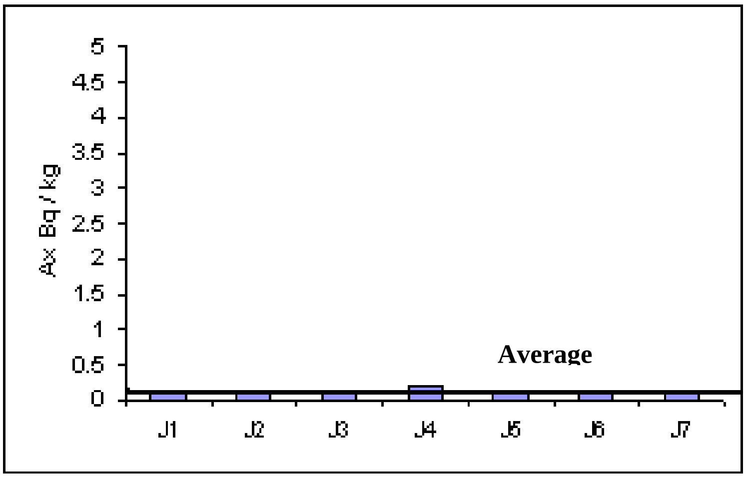

Fig (4-29): The specific activities for Bi-212 of the samples

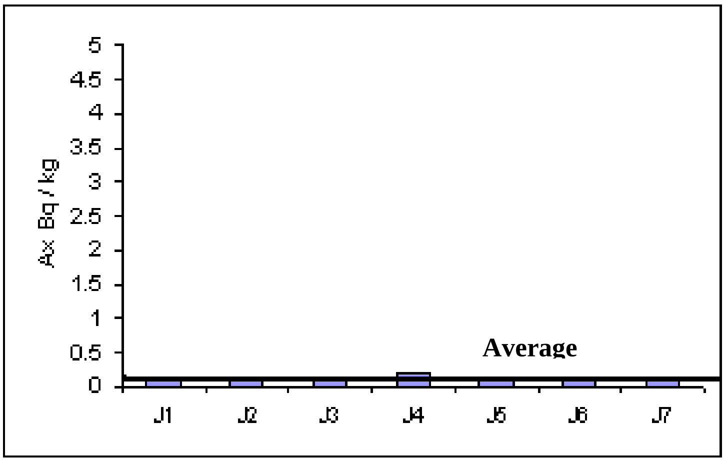

Fig (4-30): The specific activities for Pb-212 of the samples

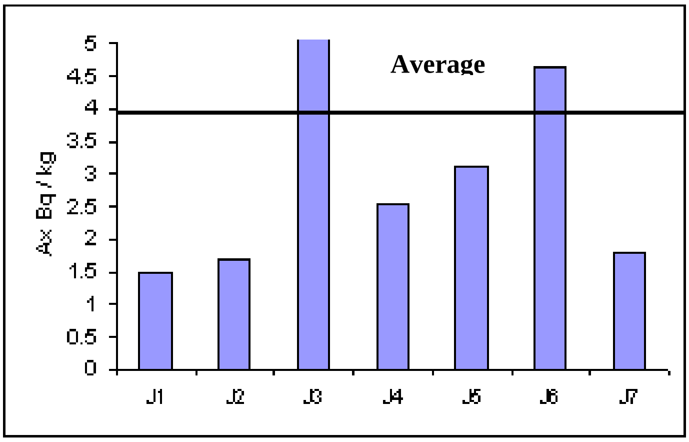

Fig (4-31): The specific activities for K-40 of the samples

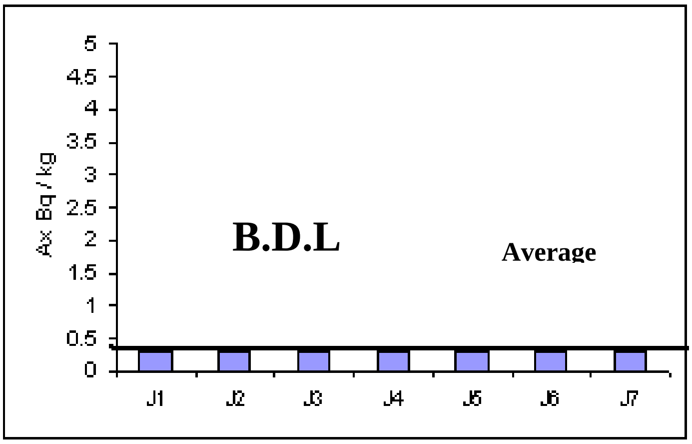

Fig (4-32): The specific activities for Cs-137 of the samples

Gamma-rays spectrum for the sample (J_4) is shown in Figure (4-33).

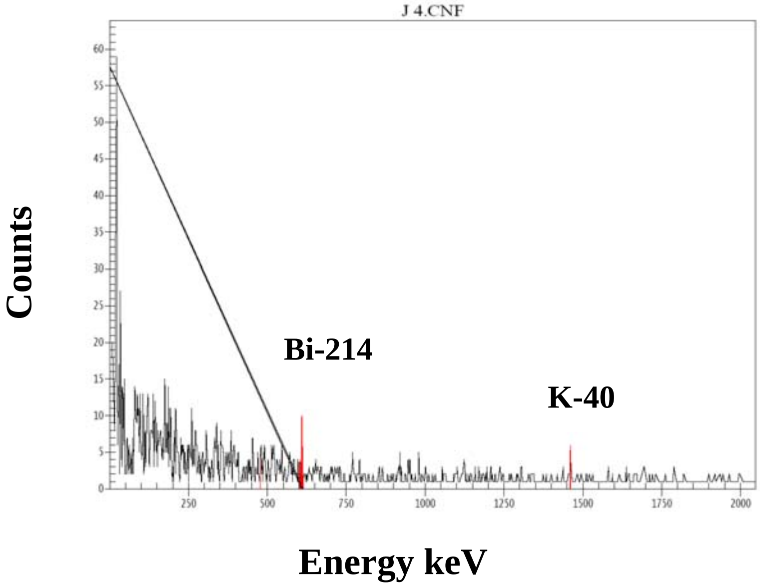

Fig (4-33): Gamma-rays spectrum for sample (J_4)

4.4 Effect of the oil waste on the environment

Scales and sludges from certain mineral processing operations exhibit specific activity of radium. The management of the oil wastes is different from country to another. If the total specific activity of the oil location is below than (1000)Bq/kg no management have done, but if the total specific activity is greater than (1000)Bq/kg then must make some management to reduce the radioactivity of NORM residues designated waste as **[83,84]**:

1- Filter bags containing the scales and sludges are sent to a dedicated waste storage facility where they are compacted by shredding and immobilized with cement before being stored in drums.

2- There is now a considerable of knowledge on appropriate methods for conditioning, storage and disposal of the various types of NORM waste, because there is huge of oil waste were through in the refinery without treatment.

3- Most of oil waste in the refinery was throughout to the pit abandonment that contains a huge of oil waste from many units in the refinery.

4- Normal landfill facilities are generally suitable, but the presence of non-radiological contaminants such as heavy metals may require the use of landfill sites specially designated for hazardous waste far away from the sources of water.

5- NORM residues from the chemical extraction of rare earths from monazite are produced in significant quantities and have characteristically high activity concentrations. It has been demonstrated that such wastes can be suitably disposed of either in earthen trenches or in engineered cells, depending on the activity concentration **[84,85]**.

The equivalent dose from NORM in the samples from all the oil location was calculated using equation (2-19), as shown in Table (4-10).

Table (4-10): The dose rate and equivalent dose from NORM in oil locations

oil location	Dose rate (nGy/h)	Equivalent dose rate (mSv/y)
Dora refinery	20.22	0.12
North field of al-Rumaila	41.55	0.30
Bijie refinery	6.83	0.05

From Table (4-10)

The lowest exposure rate was (0.05) mSv/y for Bijie refinery samples, and the highest exposure rate was (0.30) mSv/y for the north field of al-Rumaila.

All these values are below the allowed limit from IAEA that is (1) mSv/y for the public **[85]**.

4.5 Discussion

Workers and the people that live in oilfield and the refineries most likely to be exposed to the radiation from oil waste, but production sites can also pose a potential hazard to members of the public. This may be present in produced water, drilling mud, or can concentrate in pipes, storage tanks, or other extraction equipment. The contamination may be present in mineral scale, sledges, slimes, or evaporation ponds or pits. The radiation comes from (NORM) in the underground rock and sediment. When companies drill for gas or oil, the produced fluids, including water, may contain radionuclides, primarily radium-226, radium-228, and radon. The radon gas may be released to the atmosphere, while the produced water and mud containing radium are placed in ponds or pits for evaporation, re-use, or recovery.

Dora refinery samples: Table (4-1) shows the uranium concentrations of solid samples that vary from (0.95-3.34) ppm and the average of the uranium concentrations was (2.03) ppm, these results are quite low compared with the allowed limit (11.7) ppm **[86]**. For the liquid samples, Table (4-2) shows uranium concentrations are varies from (1.14-1.38) ppm with average (1.25) ppm, in these results are quite low compared with the allowed limit **[87**].

Table (4-7) shows seven radionuclides had been detected using (HPGe) detector as: (Bi-214, Ra-226, Tl-208, Bi-212, Pb-212, K-40, and Cs-137).

The appearance of radionuclides (Bi-214,Ra-226) appeared in (D_1,D_2,D_3,D_4,D_5,D_6,D_7,D_8) with average specific activities (7.79,16.08) Bq/kg respectively which show that the existence of U-238, the radionuclides (Tl-208, Bi-212, Pb-212) appeared in the samples (D_1,D_2,D_3,D_4,D_5,D_6,D_7,D_8) with average specific activities (7.95,4.05,5.75) Bq/kg which belong to the Th-232 series, these radionuclides belong to the natural radioactivity decay series with allowed limit **[88]**.

The natural radionuclide (K-40) appeared in all samples with range (1.82-189.91) Bq/kg with average specific activity (99.3)Bq/kg, the natural potassium is one of the element existences in the nature. The artificial radionuclide (Cs-137) half-life (30) year that appeared in samples (D_2,D_3,D_5,D_6,D_7,D_8) with average specific activities (0.64)Bq/kg was below than the devised contamination limit (600)Bq/kg **[86]**.

North field of al-Rumaila samples, Table (4-3) shows the uranium concentrations in varies from (1.78 - 6.35) ppm with average (3.37) ppm , this concentration was quite low expect in samples (B_4,B_5,B_6,$B_{7,}$) which indicate that sites are higher than other regions because these regions were a fields of military operations. The uranium concentrations in the oil waste were less than the allowed limit **[88].**

Bijie refinery samples, Tables (4-4) and (4-5) shows the uranium concentration in varies from (0.42-1.46) ppm with average (0.55) ppm for the solid samples and (1.24) ppm for the liquid samples. The uranium concentration in the oil waste from Bijie refinery was less than the allowed limit **[86]**. Table (4-9) shows seven radionuclides had been detected as:
(Bi-214, Ra-226, Tl-208, Bi-212, Pb-212, K-40 and Cs-137).

The appearance of the radionuclide (Bi-214, Ra-226) which belongs to uranium-238 series appeared in samples (j_3, j_4, j_5, j_6), and the average of specific activities was (0.39, 1.3) Bq/kg respectively.

The appearance of the radionuclides (Bi-212, Pb-212), which belongs to Th-232 series appeared in the samples (j_4) with specific activities (0.02,0.01) Bq/kg respectively. The natural radionuclide (K-40) appeared all samples with average specific activities (2.28) Bq/kg. The artificial radionuclide (Cs-137) is not appeared in all samples which indicate that Bijie refinery was not contaminate with (Cs-137).

The average of uranium concentrations in Dora refinery, Bijie refinery and the north field of al-Rumaila are shown in the Table (4-6) and Figure (4-6), which show that the maximum concentration of uranium were in the north field of al-Rumaila and the minimum concentration of uranium were in Bijie refinery. The reason of highest concentration of uranium in the oil waste in north field of al-Rumaila because Basrah governorate was a field of a military operation during the Iraq war 2003 and contaminate with depleted uranium which used in the war and there are huge of oil waste were throughout to the nature without good treatment. Uranium concentration in Dora refinery is higher than Bijie refinery because the most crude oil of Dora refinery comes from Basrah governorate which contains some activities of uranium concentration.

4.6 Conclusions

In this study we concluded the following knowledge:

1- The minimum of uranium concentration in oil waste using CR-39 track detector was (0.42) ppm from Bijie refinery.

2- The maximum of uranium concentration in oil waste using CR-39 track detector was (6.35) ppm from north field of al-Rumaila.

3- The average of uranium concentration in north field of al-Rumaila was (3.37) ppm is higher than average of uranium concentration in Dora and Bijie refineries and all values were less than the allowed limit.

4- The average specific activities of (Bi-214, Ra-226) radionuclides which belong to U-238 series in Dora refinery were higher than the average of specific activities in Bijie refinery.

5- The average of specific activities of (Tl-208, Bi-212, Pb-212) radionuclides in Dora refinery were higher than the average activities in Bijie refinery.

6- The average specific activities of the natural radionuclide (K-40) in Dora refinery was higher than the average specific activities in Bijie refinery.

7- The artificial radionuclide (Cs-137) was not found in Bijie refinery and the average of specific activities (0.97) Bq /kg in Dora refinery less than allowed limit.

8- The results of uranium concentrations using CR-39 track detector were in a good agreement with the results of specific activities by using (HPGe).

9- The maximum of equivalent dose rate was (0.30) mSv/y from NORM in oil waste that found in the north field of al-Rumaila.

4.7 Future work

1- Measurement of specific activities of NORM and TENORM from a all oil and gas fields and oil industry in Iraq.

2 - Measurement of specific activities of NORM and TENORM from the produce water from oil industry in Iraq.

3 - Measurement the concentration of radon gas inside all oil refineries in Iraq.

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الخلاصة

الهدف من هذه الدراسة هو الكشف عن النويدات المشعة وحساب الفعالية الإشعاعية النوعية وقياس تراكيز اليورانيوم لثلاثين نموذج من المخلفات النفطية الصلبة والسائلة تم أخذها من ثلاثة مواقع نفطية هي : مصفى الدورة و مصفى بيجي و حقل الرميلة الشمالي .

تم القياس في هذه الدراسة بأستخدام تقنيتين :

1- منظومة مطيافية أشعة كاما لحساب الفعالية الإشعاعية النوعية للنويدات المشعة . وهذه المنظومة تتكون من كاشف الجرمانيوم عالي النقاوة (HPGe) يتصل مع حاسوب يحتوي محلل طيفي متعدد القنوات(MCA) مبرمج لرسم الطيف الإشعاعي وحساب الفعالية النوعية الإشعاعية للنويدات المشعة. حيث تم الكشف عن سبع نويدات مشعة وهي :
هذه النويدات تشمل: (Bi-214, Ra-226, TI-208, Bi-212, Pb-212, K-40,Cs-137) النويدتين (Bi-214, Ra-226) واللتان تعودان إلى سلسلة تحلل U-238 ، أما النويدات الثلاثة (Ac-228, Bi-212, Pb-212) فتعود الى سلسلة تحلل Th-232 ، ونويدة طبيعية واحدة هي (K-40) ، ونويدة صناعية واحدة هي (Cs-137) .

بالنسبة الى نماذج مصفى الدورة تم حساب الفعالية الإشعاعية النوعية وقد كان معدل الفعالية الإشعاعية النوعية للنويدتين (Bi-214, Ra-226) وعلى التوالي Bq/kg (16.08, 7.79)، أما معدل الفعالية الإشعاعية النوعية للنويدات الثلاثة (TI-208, Bi-212, Pb-212) فكانت على التوالي Bq/kg(7.95, 4.05,5.75) أن معدل الفعالية الإشعاعية النوعية النويدة الطبيعية (K-40) كان Bq/kg (99.3) . أما معدل الفعالية الإشعاعية النوعية للنويدة الصناعية (Cs-137) كان Bq/kg (0.97) . بالنسبة إلى نماذج مصفى بيجي فقد تم حساب معدل الفعالية الإشعاعية للنويدتين (Bi-214, Ra-226) وكان Bq/kg(0.39,1.3)، أما بالنسبة للنويدتين (Bi-212,Pb-212) فقد كان معدل الفعالية الإشعاعية النوعية لهما Bq/kg (0.02,0.01) على التوالي ، أما النويدة الطبيعية (K-40) فقد كان معدل الفعالية الإشعاعية النوعية Bq/kg (3.0) . أما النويدات (TI-208,Cs-137) فلم تسجل أي فعالية أشعاعية.

2- تقنية كواشف الأثر النووي في الحالة الصلبة (SSNTDs) . أن قياس تركيز اليورانيوم تم باستخدام كاشف الأثر (CR-39) للكشف عن جسيمات ألفا وشظايا الانشطار، حيث أستخدمت تقنية عد أثار شظايا الأنشطار النووي الناتج من أنشطار نواة اليورانيوم (U -235) بقصفها بالنيوترونات الحرارية المنبعثة من المصدر النيوتروني (Am–Be) بفيض نيوتروني حراري ($5\times10^{3}n.cm^{-2}s^{-1}$) لمدة سبعة أيام. حيث تم حساب تركيز اليورانيوم بالمقارنة مع تركيز اليورانيوم في النماذج القياسية .

بالنسبة الى نماذج مصفى الدورة فقد تراوحت تراكيز اليورانيوم بين ppm (0.95 - 3.34) وبمعدل ppm (2.03) للنماذج الصلبة و ppm (1.25) للنماذج السائلة.

بالنسبة الى نماذج مصفى بيجي فقد تراوحت تراكيز اليورانيوم بين ppm (0.42 -1.46) وبمعدل ppm (0.55) للنماذج الصلبة و ppm (1.24) للنماذج السائلة .

أما نماذج حقل الرميلة الشمالي فقد تراوحت تراكيز اليورانيوم بين ppm (1.78- 6.35) وبمعدل ppm (3.37) .

تشير النتائج العملية أن أعلى تراكيز لليورانيوم كان في نماذج حقل الرميلة الشمالي في محافظة البصرة . وهذه النتائج هي ضمن الحدود المسموح بها من قبل الوكالة الدولية للطاقة الذرية.

وتم حساب معدل الجرعة المكافئة والناتجة عن وجود (NORM) في المخلفات النفطية حيث بلغت أعلى جرعة مكافئة mSv/y (0.30) في حقل الرميلة الشمالي وهو أقل من الحد المسموح به من قبل الوكالة الدولية للطاقة الذرية لعموم الناس mSv/y (1) .

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من قبل

عصام محمد رشيد درويش

بكالوريوس/ 1988

أشراف

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جماد الأول 1431
نيسان 2010