



Assessment of Technologically Enhanced Pollutants Concentration in Egyptian Natural Gas

Thesis

**Submitted in Partial Fulfillment of the Requirements of the
Degree of Ph.D. in Physics**

By

Omyma Riad Megahid

Under Supervision of

Prof. Dr. Samir Ushah El-Khameesy
Department of Physics, Faculty of Science,
University of Ain Shams

Late Prof. Dr. Zainab Melegy Mohamed
Department of Physics, Faculty of Science,
University of Ain Shams

Prof. Dr. Abo-Baker Ramadan
Egyptian Nuclear and Radiological Regulatory Authority,
Cairo, Egypt

Prof. Dr. Hanan Deiab
Egyptian Nuclear and Radiological Regulatory Authority,
Cairo, Egypt

**Department of Physics, Faculty of Science,
University of Ain Shams**

Cairo, Egypt

2016

Approval Sheet

Title of the Ph. D. Thesis

**Assessment of Technologically Enhanced pollutants
concentration
in Egyptian Natural Gas**

Name of the candidate

Omyma Riad Megahid

Supervisors

Signature

Prof. Dr. Samir Usha El-Khameesy ()
Department of Physics, Faculty of Science,
University of Ain Shams

Prof. Dr. Abo-Baker Ramadan ()
Egyptian Nuclear and Radiological
Regulatory Authority,
Cairo, Egypt

Prof. Dr. Hanan M. Deiab ()
Egyptian Nuclear and Radiological
Regulatory Authority,
Cairo, Egypt

Late Prof. Dr. Zainab Melegy Mohamed
Department of Physics, Faculty of Science,
University of Ain Shams



Name : Omya Riad Megahid

Degree : Ph.D.

Department : Physics

Faculty : Science

University : Ain Shams

Graduation Date: 1996-Ain Shams University

Registration Date: 11/7/2007

Grant Date : 2017

ACKNOWLEDGEMENT

I would like to express my deepest appreciation for *Prof. Dr. Samir El-Khameesy*, Department of Physics, Ain-Shams University His encouragement, guidance and support from the initial to the final level enabled me to Develop and understanding of the subject. Without his guidance and persistent help, this thesis would not have been possible.

I would like to express my deepest appreciation for *Late Prof. Dr. Zainab Melegy Mohamed*, Department of Physics, Faculty of Science, University of Ain Shams, For her great encouragement, guidance and support at the initial work of this thesis

I would like to thank *Prof. Dr. Abo- Baker Ramadan*, Egyptian Nuclear and Radiological Regulatory Authority, Cairo, Egypt for his supervision, support, help, guidance and useful discussion during the different stages of this work.

I would like to express my utmost gratitude for *Prof. Dr. Hanan Deiab*, Egyptian Nuclear and Radiological Regulatory Authority, Cairo, Egypt for her supervision, greet assistants during the experimental work and for her support fruitful discussion and criticism.

I would like to express my gratitude for the *staff of Physics Department*, Faculty of Science, Ain-Shams University.

I offer my best regards and gratitude to my friends and colleagues at *MIDEL EAST OIL REFINERY*, who helped and encouraged me throughout this thesis.

I offer my best regards and gratitude for my friends and colleagues at *GASCO*, especially *Mr. Moustafa Abo El Makarem* for his support and guidance throughout this thesis.

Thank you Dad *Prof. Dr. Riad Megahid* for being such a great role model I always looked up to. Thank you for teaching me so much over the years.

A great Thank for my Mother for being such a positive influence in my life

Last and certainly not least, I would like to thank my husband *General. Hisham El Essawy* for his love and support.

My *Beloved Kids* for their patience, encouragement. They have kept me confident, sane and happy throughout the whole of this work.

Table of content

Acknowledgment	I
Table of Contents	II
List of Figures	VI
List of Tables	VII
List of Abbreviation	X
Abstract	XII

Chapter 1 General Introduction and Literature Review

General Introduction	1
----------------------------	---

Chapter 2 Naturally Occurring Radioactive Materials and Health Impact

2.1 Introduction.....	25
2.2 Sources of Naturally Occurring Radioactive Material.....	25
2.2.1 Thorium and Uranium Decay Series	26
2.2.2 Norm in Fossil Fuels.....	28
2.3 Natural Gas in Egypt	29
2.3.1 Natural Gas Production in Egypt	30
2.3.2 Egyptian National Gas Grid.....	31
2.4 Technically Enhanced NORM in Oil and Gas Industries	32
2.4.1 History of Discovering Te-NORM in Gas and Oil Industries	34

2.4.2 Worldwide Records for Te-NORM	35
2.4.3 Worldwide NORM Clearance Levels	38
2.5 Types and Amounts of Waste in Oil and Natural Gas	39
2.5.1 Produced Water.....	40
2.5.2 Scale	41
2.5.3 Sludge	42
2.5.4 Contaminated Equipment.....	43
2.6 Laws and Legislation for Oil and Gas Sector in Egypt	46
2.7 Management of NORM Wastes in Oil and Gas Industry	48
2.8 Exposure and Risk Assessment from NORM.....	48
2.8.1 Occupational Exposure	49
2.8.2 External Exposure.....	49
2.8.3 Internal Exposure	49
2.9 Occupational Monitoring and Protection.....	50

Chapter -3

Overview of the Main Methods and Techniques Used for Gamma Ray Activity Evaluation

3.1 Introduction.....	51
3.2 Gas Detectors	52
3.2.1 Geiger-Müller Tube	52
3.2.2 Ionization Chamber	55
3.3 Gamma Spectrometry	57
3.3.1 Gamma Spectrometry with Scintillation Detectors .	59

3.3.2 Gamma Spectroscopy with Semiconductor Detectors	61
3.3.3 Germanium Detector.....	64
3.3.4 Silicon Detector	65
3.4 Thermoluminescence Detectors	66

Chapter - 4

Experimental Procedures

4.1 Introduction.....	68
4.2 Collection of Natural Gas and Oil Samples	68
4.2.1 Collection of Natural Gas Samples	69
4.2.2 Collection of Oil Samples	70
4.3 Determination of Natural Gas Chemical Composition.....	71
4.3.1 Sampling of VOCs.....	71
4.3.2 Gas Chromatographic Analysis	72
4.3.3 Inductively Coupled Plasma Atomic Emission Spectrometer (ICP/AES).....	76
4.4 Measurements of Norm in Natural Gas and Oil Samples.....	77
4.4.1 Measurements by Gamma ray Spectroscopy	77
4.4.2 Setup of the Used Gamma Ray Spectrometer.....	78
4.4.3 Reduction of Gamma Ray Background.	79
4.4.4 Energy Calibration of Gamma Spectrometer.....	80
4.4.5 Efficiency Calibration of the HpGe Detector	80
(a) Relative Efficiency Curve of The Detector	83
(b) Absolute Efficiency Calibration Method	85

4.5 Activity calculation of Norm in Egyptian Oil & Natural Gas .86

Chapter 5

Results and Discussion

5.1 Introduction.....	88
5.2 Chemical Composition of Natural Gas	89
5.3 Activity Calculation in Egyptian Natural Gas	90
5.3.1 Norm Concentration in Waste Water.....	90
5.3.2 Norm Concentration in Sludge	92
5.3.3 Norm Concentration in Scales	94
5.3.4 Norm Concentration in Filter of Dehydration Unit .	95
5.3.5 Comparison of NORM Concentration from Different Natural Gas Samples	97
5.4 Norm Concentration in Egyptian Oil Refinery	99
5.4.1 NORM Concentration in Ash	99
5.4.2 NORM Concentration in West Water of Oil Refinery	100
5.4.3 NORM Concentration in Oil Samples	102
5-4-4 Comparison between NORM Concentrations in Different Oil Refinery Samples.....	103
5.5 Comparison Of NORM Concentration In Oil And Natural Gas	105
5.6 Radiation Risk Assessment and Risk Control	106
5.6.1 Absorbed Dose.....	106
5.6.2 Annual Effective Dose Equivalent	106
5.6.3 Radium Equivalent.....	107
5.6.4 Gamma Index	107

5.7 Natural Gas Absorbed Dose (D), Annual Effective Dose(AEDE), Radium Equivalent ($R_{a_{eq}}$) and Gamma index ($I_{\gamma r}$).....	108
5.7.1 Absorbed Dose in Natural Gas	109
5.7.2 Annual Effective Dose in Natural Gas.....	109
5.7.3 Radium Equivalent in Natural Gas	110
5.7.4 Gamma Index for Natural Gas.....	110
5.8 Radiation Hazards and Risk Control From Oil Industries	110
5.8.1 Absorbed Dose In Oil	112
5.8.2 Annual Effective Dose Equivalent	112
5.8.3 Radium Equivalent.....	112

Chapter 6

Conclusions and Recommendations

Conclusions and Recommendations	113
References.....	116

List Of Figures

2.1	Thorium-232 Decay Series	28
2.2	Uranium-238 Decay Series	28
2.3	Egyptian Natural Gas Grid	32
2.4	Types of Wastes in Oil and Gas Industries	40
2.5	Waste Water in Oil Refinery	41
2.6	Scales in Pipe from Gas Processing Plant	42
2.7	Cleaning of Sludge in a Storage Tank	43
2.8	Filters Before and After Being Used	45
3.1	Schematic of a Geiger Counter using an "End Window" Tube for Low Penetration Radiation.	55
3.2	Ionization Chambers Schematic Diagram	57
3.3	A Schematic Diagram of a Spectrometer with Scintillator Detector	60
3.4	Schematic Diagram for Electron Hole Production	63
4.1	Photographs of Sludge and Scale Samples Collected from the interior Surface of Gas Pipe Line.	69
4.2	VOCs 3M Monitors	72
4.3	A Photograph of the Gas Chromatographic Analyzer	73
4.4	Inductively Coupled Plasma Atomic Emission Spectrometer	76
4.5	Block Diagram of a Typical Gamma Ray Spectrometry System	79
4.6	HPGe Detector Relative efficiency Curve in the Range 241 -1900 keV Making Use of Ra-226 Source.	82

4.7	Absolute Efficiency- Energy Curve for Soil Samples.	84
4.8	Absolute Efficiency- Energy Curve for Water Samples	85
4.9	The Gamma Transitions for Different Radionuclides	87
5.1	Chemical Analysis of Egyptian Natural Gas	90
5.2	Activity Concentration of Measured Radionuclides in Waste Water..	92
5.3	Activity Concentration of Measured Radionuclides in Sludge	94
5.4	Concentrations of Measured Radionuclides in Scale Samples.	95
5.5	Activity Concentration of Measured Radionuclides In Filters	97
5.6	Average Activity Concentrations of Measured Radio Nuclides in Different Natural Gas Samples.	98
5.7	Activity Concentration (Bq/kg) in Ash Samples	100
5.8	Activity Concentration (Bq/kg) in Waste Water Samples	101
5.9	Activity Concentration (Bq/kg) in OIL SAMPLES	103
5.10	Average Concentration of Measured Radio Nuclides in Different Oil Samples	104

List of Tables

Table 2.1	Concentration Levels of Radium Nuclides Reported in Different Countries	36
Table 2.2	Ranges of Activity Levels in Produced Water From the Oil Fields	38
Table 2.3	The European Commission Clearance Levels.	38
Table 2.4	The Average Worldwide Activity Levels of U, Th and K	39
Table 2.5	The Exemption Activity Levels of NORM as Recommended in the IAEA Basic Safety Standards	39
Table 2.6	The Average Worldwide Levels of the Most Common Radiological Indices	39
Table 5.1	Chemical Composition of Egyptian Natural Gas	89
Table 5.2	Radioactivity Concentration Bq/l of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K from Waste Water Samples. The Uncertainties of the Results are Also Included.	91
Table 5.3	Radioactivity Concentration Bq /kg of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K from Sludge Samples Associated with Experimental Standard Deviations.	93
Table 5.4	Radioactivity Concentration of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K from Scales Samples Associated with Experimental Standard Deviations.	95
Table 5.5	Radioactivity Concentration of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K From Filter Samples Associated with Experimental Standard Deviations.	96
Table 5.6	Radioactivity concentration of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K for Natural Gas Samples Associated with Experimental Standard Deviations.	98
Table 5.7	Radioactivity Concentration of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K for Ash Associated with Experimental Standard Deviations.	99

Table 5.8	Radioactivity concentration of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K for waste water samples Associated with Experimental Standard Deviations.	101
Table 5.9	NORM concentrations in Oil Samples Associated with Experimental Standard Deviations..	102
Table 5.10	Average Concentrations of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K for the Different Sources of NORM in Oil Associated with Experimental Standard Deviations.	104
Table 5.11	Values of Absorbed Dose (D), Annual Effective Dose (AEDE), Radium Equivalent (Ra_{eq}) and Gamma Index (I_{yr}) for a Natural Gas Samples.	108
Table 5.12	Values of Absorbed dose (D), Annual Effective Dose(AEDE), Radium Equivalent (Ra_{eq}) and Gamma Index (I_{yr}) for all Oil Samples	111

Chapter -1

General Introduction and Literature Review

Naturally Occurring Radioactive Materials, NORMs are found almost everywhere. They exist in air, in soil, and even as radioactive potassium in the human body's as well as in public water supplies and some food such as nuts, peanut butter and cereal. In addition they are found in fossil fuel such as coal, oil and natural gas. It is worth to mention that the potential radiation risk that can result from the presence of NORM in all the previously mentioned forces are very low and are usually neglected. However, the use of raw material for manufacturing different sorts of chemical products such as oil, coal and natural gas could result in concentrating NORMs in the product, since the objective of the studies performed in this thesis is concerned with the effect of NORMs in oil and natural gas on people engaged in industries related to three or two fossil fuels, the studies and discussions will be confused to merely to these sources[1,2].

In general, fossil fuels are considered as very weak sources of NORM, and therefore less or no attention are given to radiation exposure that may result from them. However, the use of any one of them during manufacturing chemical products can lead to enhance the concentration of NORMs in the products or in the waste materials. In addition, the workers engaged in these industries have to be under control and kept to minimum level. Further, the concentration of NORMs like Radium – 226 and Radium – 228 may also occur in sludge that accumulates in oil field, pits and tanks. These sludge become sources of oil and gas NORM wastes.