

شبكة المعلومات الجامعية التوثيق الإلكتروني والميكروفيلو

بسم الله الرحمن الرحيم





MONA MAGHRABY



شبكة المعلومات الجامعية التوثيق الإلكتروني والميكروفيلو



شبكة المعلومات الجامعية التوثيق الالكتروني والميكروفيلم



MONA MAGHRABY



شبكة المعلومات الجامعية التوثيق الإلكترونى والميكروفيلم

جامعة عين شمس التوثيق الإلكتروني والميكروفيلم قسم

نقسم بالله العظيم أن المادة التي تم توثيقها وتسجيلها علي هذه الأقراص المدمجة قد أعدت دون أية تغيرات



يجب أن

تحفظ هذه الأقراص المدمجة بعيدا عن الغبار



MONA MAGHRABY



Experimental Assessment and Modeling of Radio-contaminants Transport in the Soil of a Radioactive Waste Disposal Site

BY

Mahmoud Mahfouz Gouda Ragab

(MSc. in Geology, 2016) Assistant lecturer - Waste Management Department Atomic Energy Authority

A Thesis Submitted for Doctor Degree of Philosophy in Science "Geology"

> To Geology department - Faculty of Science Ain Shams University

Under the Supervision of:

Prof. Dr. Yehia Hassan Dawood

Prof. of Mineralogy and Geochemistry Geology Department - Faculty of Science Ain Shams University

Dr. Mohamed Ragheb Mohamed

Ass. Prof. of Inorganic Chemistry Radioactive Waste Management Department Atomic Energy Authority

Prof. Dr. Ahmed Ahmed Zaki

Prof. of Chemical Nuclear Engineering Head of Radioactive Waste Management Department - Atomic Energy Authority

Dr. Ahmed Gad Abdel Wahid

Ass. Prof. of Environmental Geochemistry Geology Department - Faculty of Science Ain Shams University

Acknowledgement

Before all and above I am deeply thankful to Allah, by the grace of progress and success of this work was possible.

First, I would like to express my gratitude to Prof. Dr. Yehia Hassan Dawood, Prof. of Mineralogy and Geochemistry, Ain Shams University, for his supervising and useful comments during this work.

I would like to express my deep sincere thanks and appreciation to Prof. Dr. Ahmed Ahmed Zaki, Prof. of Chemical Nuclear Engineering, Atomic Energy Authority, for his supervision and helpful advice to complete this work.

Also, I would like to express my deep sincere thanks and appreciation to Dr. Mohamed Ragheb El-Naggar, Ass. Prof. of Inorganic Chemistry, Atomic Energy Authority, for his supervision, guidance, motivation and endless supports the work.

I would like to express my gratitude and sincere appreciation to Dr. Ahmed Gad Abdel Wahid, Ass. Prof. of Environmental Geochemistry, Ain Shams University, for effective supervision as well as fruitful discussion throughout the progress of this work.

Also, I would like to express my gratitude and sincere appreciation to, Dr. Haneen Abdel Salam and Dr. Mohammed Shaban, Atomic Energy Authority, for their internal supervision and helpful advice to complete this work. I would like to express my gratitude and appreciation to Dr. Hamida Sayed Mekhemar for her support along this work.

Finally, I would like to express my thanks to all the staff members of waste management department for their help and support.

Dedicated to

My mother and my father

To my wife, my brothers and sister

To my children
(Anas, Jana and Noor)

And all of my family



ABSTRACT

Low and intermediate level radioactive wastes are produced during research activities of the radiochemical laboratories, research reactors, radioisotope and metallurgical laboratories, activation analysis units, nuclear medicine divisions in hospitals, universities and research institutes as well as industrial activities. These wastes should be isolated to protect human and environment under controlled conditions which is one of the most important factors to be considered for the long-term safety assessment of a radioactive waste disposal facility, particularly in wet geological formations.

Representative sediment samples were collected from a radioactive waste disposal site at Inshas locality. The mineralogy and geochemistry of sediment samples are studied to characterize the sediments and to study the possible roles of grain size, mineral and chemical composition. The clay minerals are considered as important factors in the adsorption of metal ions and as backfill in the radioactive waste disposal sites. The sand and sandstone from the study area have high contents of Fe₂O₃, CaO and MgO reflecting a possible contribution from the nearby basaltic extrusive rocks. The chemical index of alteration (CIA) values reflect a low degree of chemical weathering.

Batch, diffusion, column experiments, adsorption and desorption kinetics were performed to evaluate the radionuclide

behavior in the subsurface environment. The maximum adsorption capacity of Cs^+ , Co^{2+} and Ni^{2+} onto clay sample was found to be greater than that adsorbed onto different sand samples. The kinetic study showed that the adsorption is followed the pseudo-second-order model with a good correlation coefficient ($R^2 = 0.999$). The equilibrium investigations of adsorption data were fitted using Freundlich, Langmuir, Dubinin–Radushkevich and temkin isotherm models. The adsorption process of Cs^+ is exothermic and favored at low temperatures, on the other hand the adsorption of Co^{2+} and Ni^{2+} are endothermic and favored at higher temperatures.

The total adsorbed amount of Cs⁺, Co²⁺ and Ni²⁺ ($Q_{tot,S}$) and bed capacity (mg/g) in sand column increase with increasing the initial metal ion concentrations. Column performance decreases with increasing the initial concentrations onto the saturated brown sand column. The analysis of the breakthrough curve was done using Thomas, Yoon-Nelson and Adams–Bohart models. The simulation program (GOLDSIM) was used to simulate the radionuclide transport through soil of Inshas disposal site and calculate the maximum effective dose. The air and soil activity of are very low concentration of 60 Co, 137 Cs and 63 Ni radionuclides. All values for resident farmer and transient in air and all pathways do not exceed the dose constraint of 0.3 mSv/yr.

CONTENTS

Abstract	
List of Tables	I
List of Figures	III
Abbreviation	XI
CHAPTER 1	
INTRODUCTION	
1.1. Waste classification	1
1.1.1. Exempt waste (EW)	2
1.1.2. Very short-lived waste (VSLW)	2
1.1.3. Very low-level waste (VLLW)	3
1.1.4. Low level waste (LLW)	3
1.1.5. Intermediate level waste (ILW)	4
1.1.6. High level waste (HLW)	4
1.2. Radioactive waste	6
1.3. Waste disposal	7
1.3.1. Marine disposal	8
1.3.2. Land and shallow land disposal	9
1.3.4. Radon system	10
1.3.5. Underground disposal	11
1.4. Transport of the radionuclides	11
1.5. Safety assessment of radioactive waste disposal	16

1.6. Previous works	18
1.7. Aim of the present study	28
CHAPTER 2	
GENERAL GEOLOGY	
2.1. Location of study area	29
2.2. Stratigraphy	29
2.3. The source and direction of the groundwater flow	24
in Inshas area	34
CHAPTER 3	
MATERIALS AND METHODS	
3.1. Sample collection	36
3.2. Characterization of sediment samples	37
3.2.1. Granulation test	37
3.2.2. Mineralogical identification	38
3.2.3. Fourier transform infra-red spectra	39
3.2.4. Thermal analysis	40
3.2.5. Surface area analysis	40
3.3. Geochemical investigations	40
3.4. Adsorption investigations	41
3.4.1. Batch techniques	43
3.4.2. Fixed bed column technique	48
3.5. Safety assessment methodology of radioactive waste disposal site	49

CHAPTER 4

MINERALOGY AND GEOCHEMISTRY

4.1. Characterization of the sediments	52
4.1.1. Granulation test	52
4.1.2. Mineral composition of the studied sediments	54
4.1.3. Fourier transform infra-red analysis	60
4.1.4. Thermal analysis	62
4.1.5. Specific surface areas	66
4.2. Major elements geochemistry	68
4.2.1. Geochemical classification	70
4.2.2. Provenance	72
4.2.3. Weathering in the source area	74
4.2.4. Maturity and climatic conditions during	77
sedimentation	11
4.2.5. Type of the source rocks	78
CHAPTER 5	
ADSORPTION INVESTIGATION AND	
MODELING	
5.1. Batch adsorption investigations	79
5.1.1. Preliminary studies	79
5.1.2. Kinetic modeling	93
5.1.3. Adsorption at chemical equilibrium	108
5.1.4. Thermodynamic studies	131
5.2. Fixed-bed column investigations	135

5.2.1. Effect of flow rate	136
5.2.2. Effect of initial metal ion concentration	137
5.2.3. Effect of aluminum oxide as colloid	137
5.2.4. Analysis of mass transfer zone (MTZ)	138
5.2.5. Application of dynamic models	144
5.2.6. Desorption of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto/from sand	153
5.3. Modeling the transport of Cs ¹³⁷ , Co ⁶⁰ and Ni ⁶³	
radio-contaminants and safety assessment using	156
GOLDSIM package	
5.3.1. Soil and air activity concentrations	158
5.3.2. The total effective dose equivalent (TEDE)	162
CHAPTER 6	
SUMMARY AND CONCLUSIONS	166
REFERANCES	178
APPENDICES	208
ARABIC SUMMARY	(1-6)

LIST OF TABLES

Table (3.1): The collected sediment samples from Inshas site.	37
Table (3.2): The used chemicals and reagents.	41
Table (4.1): Percentages of the granule gravel, sand, silt and clay collected sediment samples.	52
Table (4.2): Relative abundances of heavy minerals in the sand and sandstone collected sediment samples.	56
Table (4.3): Major oxides of the collected samples.	68
Table (4.4): Calculated weathering, alteration indices and oxides ratios in the sediment samples.	76
Table (5.1): Temperature dependent pseudo 1st- and 2nd- order kinetic models parameters for the adsorption of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto clay, brown (BS2) and	102
yellow sand samples. Table (5.2): Elovich kinetic model parameters of Cs ⁺ , Co ²⁺ and Ni ²⁺ adsorbed onto clay, brown (BS2) and yellow sand samples at different temperatures.	107
Table (5.3): Temperature dependent Langmuir and Freundlich isotherm models parameters for the adsorbed of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto clay, brown (BS2) and yellow sand sediment samples.	120
Table (5.4): Temperature dependent D-R and Temkin isotherm models parameters for the adsorbed of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto clay, brown (BS2) and yellow sand samples.	130
Table (5.5): Thermodynamic parameters of Cs ⁺ , Co ²⁺ and Ni ²⁺ adsorbed at different temperatures onto clay	134

brown (BS2) and yellow sand samples.	
Table (5.6): The bed capacity, column performance and mass	1.40
transfer zone for adsorption of Cs ⁺ , Co ²⁺ and Ni ²⁺	143
onto brown sand (BS2) saturated column.	
Table (5.7): Thomas model parameters to describe the	
adsorption of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto brown sand	147
(BS2) column, at different of initial metal	
concentration and flow rate.	
Table (5.8): Yoon-Nelson model parameters to describe the	
adsorption of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto brown sand	150
(BS2) column at different of initial metal ion	
concentration and flow rate.	
Table (5.9): Adams–Bohart model parameters to describe the	
adsorption of Cs ⁺ , Co ²⁺ and Ni ²⁺ onto brown sand	153
(BS2) column at different of initial metal ion	
concentration and flow rate.	
Table (5.10): Total adsorbed and desorbed amounts of Cs ⁺ ,	
Co ²⁺ and Ni ²⁺ onto/from onto brown sand (BS2)	154
column at different metal ion concentrations and	
flow rate of 1.0 ml/min.	
Table (5.11): Input data was used for the radioactive waste	157
disposal site in Inshas locality.	
Table (5.12): Solubility's, distribution coefficients and	158
inventories of ⁶⁰ Co, ¹³⁷ Cs and ⁶³ Ni.	
Table (5.13): The activity concentration of surface soil, air	
concentration, water well and the total effective	1.00
dose equivalent (TEDE) of air and all pathway for	163
farmer and transient by radionuclides of ⁶⁰ Co,	

 137 Cs and 63 Ni.

LIST OF FIGURES

Fig. (1.1): Conceptual illustration of the waste classification scheme (IAEA, 2009a).	5
Fig. (1.2): Concept of the final cover cap in low and intermediate level radioactive wastes at the El Cabril Disposal Facility (Spain).	10
Fig. (1.3): Improvement of Safety Assessment Methodologies (ISAM) project methodology.	17
Fig. (1.4): Example conceptual model of radionuclide release, transport and exposure.	18
Fig. (2.1): Location map of the study Inshas area.	30
Fig. (2.2): Simplified geological map of the study area (after EGSMA, 1998).	32
Fig. (2.3): Litho-stratigraphic units of the borehole in the study area (after DRC, 1993).	33
Fig. (3.1): The location of the sediment samples in Inshas site.	36
Fig. (3.2): Experimental setup of transport in saturated sand column.	49
Fig. (4.1): Grain size analysis of the collected eight sediment samples. [BS1 to BS5: brown sand, YS: yellow sand, SS: sandstone and CS: clay sample].	53
Fig. (4.2): Photomicrographs for mounted grains of (a) brown sand (BS2), (b) yellow sand under crossed polar (XP).	54