IMMOBILIZATION OF RADIOACTIVE WASTE IN DIFFERENT FLY ASH-ZEOLITE CEMENT BLENDS

A thesis Submitted

By

Nesreen Mohamed Sami

M.Sc. (Chemistry)

Hot Laboratories and Waste Management Center
Atomic Energy Authority

To

Chemistry Department, Faculty of Science
Ain Shams University

For

The Degree of Doctor of Philosophy in Chemistry

2013

IMMOBILIZATION OF RADIOACTIVE WASTE IN DIFFERENT FLY ASH-ZEOLITE CEMENT BLENDS

A thesis Submitted By

Nesreen Mohamed Sami
M.Sc. (Chemistry)
Hot Laboratories and Waste Management Center
Atomic Energy Authority

To

Chemistry Department, Faculty of Science
Ain Shams University

For
The Degree of Doctor of Philosophy in Chemistry

Supervised By

Prof. Dr. Sayed Sabet
Professor of Physical and Electrochemistry,
Faculty of science, Ain Shams University

Prof. Dr. Mohamed.I. El Dessouky
Professor of Nuclear Chemistry
Atomic Energy Authority

Prof. Dr. Ahmed. M. El-Kamash
Professor Nuclear Chemical Engineering
Head of Radioactive Waste Management Division
Atomic Energy Authority

Asst. Prof. Belal El-Gammal
Assistant Professor of Physical Chemistry
Atomic Energy Authority

2013

IMMOBILIZATION OF RADIOACTIVE WASTE IN DIFFERENT FLY ASH-ZEOLITE CEMENT BLENDS

A thesis Submitted By

Nesreen Mohamed Sami
M.Sc. (Chemistry)

Thesis Advisors : **Signature**

1- Prof. Dr. Sayed Thabet ……..……..
Professor of Physical Chemistry
Faculty of Science - Ain Shams University

2- Prof. Dr. Mohamed.I. El Dessouky ……..
Professor of Nuclear Chemistry
Atomic Energy Authority

3- Prof. Dr. Ahmed. M. El-Kamash ....…….
Professor Nuclear Chemical Engineering
Head of Radioactive Waste Management Division
Atomic Energy Authority

4- Asst. Prof. Belal El-Gammal .....…….
Assistant Professor of Physical Chemistry
Atomic Energy Authority

Head of the Department of Chemistry

Prof. Dr./ Maged Shafik Antonious Nakhla

Acknowledgement

*First of all and above, I thank and pray to "**ALLAH**" for explicitly known and uncountable reasons.*

*My deep appreciation and sincere thanks to **Prof. Dr./ Sayed S. Abd El-Rehim**, Professor of Physical and electrochemistry, Ain Shams University, for sponsoring this thesis and continuous encouragement and guidance.*

*I wish to express my grateful gratitude and full thanks to **Prof. Dr./ Mohamed .I. El Dessouky,** Professor of Nuclear Chemistry, Atomic Energy Authority for his supervision and valuable discussions and guidance which made this thesis in the present form.*

*Heartly, I would like to express my grateful gratitude to **Prof. Dr./ Ahmed. M. El-Kamash**, Professor of Nuclear Chemical Engineering, Head of Radioactive Waste Management Division, Atomic Energy Authority for supervising the research investigated and for suggesting the point, This research was carried out under his direction. The guidance and support given by him were indispensable to the completion of this work.*

*I am greatly indebted to **Asst. Prof./ Belal El-Gammal**, assistant professor of physical chemistry, Atomic Energy Authority for valuable discussion, continuous guidance and sincere supervision and help during the different stages of this work.*

Finally I can't forget the complete assistance and faithful encouragement of my family during the preparation of this thesis.

The author would also like to thank the staff members of the hot laboratories center, Atomic Energy Authority.

ABSTRACT

The problem of radioactive waste management has been raised from the beginning use of nuclear energy for different purposes. The radwaste streams produced were sufficient to cause dangerous effects to man and its environment.

The ordinary portland cement is the material more extensively used in the technologies of solidification and immobilization of the toxic wastes, low and medium level radioactive wastes.

The production of portland cement is one of the most energy-intensive and polluting. The use of high energy in the production causes high emission due to the nature and processes of raw materials. The cement industry is responsible for 7% of the total CO_2 emission. Thus, the cement industry has a crucial role in the global warming. The formation of alite (Ca_3SiO_5), which is the main component of the Portland cement clinker, produces a greater amount of CO_2 emission than the formation of belite (Ca_2SiO_4). The proportion of alite to belite is about 3 in ordinary Portland clinker. Therefore, by decreasing this proportion less CO_2 would be emitted. Furthermore, if industrial byproducts such as fly ash from thermal power station or from incineration of municipal solid wastes have the potential to reduce CO_2 used as raw materials and alternative hydrothermal–calcination-routes are employed for belite clinker production, CO_2 emission can be strongly reduced or even totally avoided. The availability of fly ash will help in reducing the CO_2 emissions and will also help in resolving, to a great extent, the fly ash disposal problem.

This thesis is based on focusing on the possibility of using fly ash as raw materials to prepare low cost innovation matrices for immobilization of radioactive wastes by synthesizing new kind of cement of low consuming

energy. The synthesis process is based on the hydrothermal-calcination-route of the fly ash without extra additions. The hydrothermal treatment was carried out in the presence of CaO and 1 M NaOH solution for 4 h at the temperature of 200 °C. The precursors obtained during the hydrothermal treatment were heated at temperatures of 700 °C, 800 °C, and 900 °C. Three types of new cements called fly ash-zeolite cement (FAZC) originates from dehydration by heating of the hydrated precursors formed at 700 °C, 800 °C, and 900 °C as a result of the pozzolanic reaction.

The changes of fly ash composition after different treatments and the microstructural changes produced in those precursors during heating were characterized by X-ray diffraction (XRD), FT infrared spectroscopy, surface area (BET-N_2), and thermal analyses (thermogravimetric analysis (TGA) and differential thermogravimetric analysis (DTG)). From the results obtained we concluded that the optimum temperature for obtaining the fly ash zeoilt cement clinker is 800 °C.

The physical ,chemical and mechanical properties of fly ash zeolite cement (FAZC) including the setting time, bleeding rate and compressive strength were determined. Then, these investigations are compered with those of ordinary portland cement (OPC). The effect of water immersion in different leachants for long periods on the compressive strength was studied and also, radiation resistance. Leaching behaviour of Cs^{137}, Co^{60} and Sr^{85} from both immobilized FAZC and OPC have been investigated. Based on the results obtained it can be concluded that, fly ash zeolite cement can be used for radioactive waste solidification facilities in order to satisfy toward the saftey requirements and reduce the radionuclides leach rates to the ground water in the disposal land facility.

CONTENTS

[1] Introduction

[2] Literature Review

[3] Materials And Methods

[4] Results and Discussion

LIST OF TABLES

LIST OF FIGURES