NEUTRON ACTIVATION ANALYSIS ABSOLUTE METHOD AT PUSPATI TRIGA MARK II RESEARCH REACTOR FOR ELEMENTAL ANALYSIS

IBRAHIM ALFADIL ALNOUR ISHAG

A thesis submitted in fulfilment of the requirements for the award of the degree of Doctor of Philosophy (Physics)

Faculty of Science
Universiti Teknologi Malaysia

SEPTEMBER 2013
To the soul of my father and to my mother, brother, sisters and friends
ACKNOWLEDGEMENT

First of all, I would like to express my deepest appreciation to all those who provided me the possibility to complete this work. A special gratitude is given to my supervisors Prof. Dr. Husin Wagiran, Prof. Dr. Nooriddin Ibrahim, Dr. Suhaimi Hamzah and Dr. Wee Boon Siong for their encouragement, fruitful suggestions, and generous support throughout the course of this research work. I also like to express my sincere gratitude to Department of Physics, Faculty of Science, Universiti Teknologi Malaysia.

The work reported in this thesis has been carried out in the Malaysian Nuclear Agency; nuklear Malaysia (NM). Furthermore I would also like to acknowledge with much appreciation the crucial role of NM, to allow me to use all required equipment and the necessary materials to complete this work. I am indebted most of all to Mr. Md Suhaimi Elias for his generosity and unlimited assistance by taking me by his car during my work which extend to one year and for discussion and ideas. My sincerely grateful also to Mr. Ariffin Talib for his taking care of me in many ways. I would also like to thank Mr. Lim Ching for providing very useful references and assistance. I am also grateful to other staff in NM especially Mr. Hj. Halim Baharuddin, Mr. Mohd Khushairi Awang, Ms. Jamaliah Mat Yatim, Ms. Pirmala Devi, Mr. Azhar Harun, Ms. N. Ashifa Salim and Ms. Ezwiza Bt Sanuri for their cooperation and assistance during the experimental work. A special thanks goes to the staff of Reactor Engineering Section for their helps and providing the information especially, to Dr. Zarina, Mr. Suhaimi Kassim, Ms. Juli, Mr. Fauzi and Mr. Gaphor. My sincere thanks also go to Dr. Muhamat Omar, Dr. Abdul Kadir, Mr. Yii and Ms. Zalina for the discussions, opinions, suggestions and ideas. My deepest and appreciation also extend to Prof. Dr. Wan Fuad Wan Hassan and Quarry sites (Johor) for providing samples.

I deeply gives appreciation to International University of Africa for financial support and the opportunity to complete my study in Malaysia.

I would specially like to express my deepest gratitudes to my mother, brothers and sisters for their continuous encouragement and undying support. Finally, I extend many thanks to my friends and colleagues for their encouragement and support.
ABSTRACT

This research aimed to study the capability of the neutron activation analysis absolute method by using PUSPATI TRIGA Mark II research reactor, at the Malaysia Nuclear Agency (NM). Most of the quantitative measurements of neutron activation analysis (NAA) were done through comparative method, which was found to have high degree of errors due to the differences in the matrix. NAA based on absolute method is a more direct analysis of the irradiated samples based on \((n, \gamma)\) reaction rate without using any standard or comparator. In this study the reaction rate of \((n, \gamma)\) was adopted based on the Høgdahl conventions as well as Westcott formalism. This technique requires absolute gamma ray measurements and neutron spectrum parameters for the calculation of weights or concentrations of elements present in the sample. The neutron spectrum parameters such as epithermal neutron shape factor \((\alpha)\), thermal to epithermal neutron flux ratio \((f)\), and thermal and epithermal neutron flux \((\phi_{th} \text{ and } \phi_{epi})\) were determined at 40 irradiation positions of the rotary rack (RR) as well as at one location of pneumatic transfer system (PTS). The value of \(\alpha\) in 40 RR was found to be in the range of 0.0060 to 0.1170 with an average of 0.0172 and 0.0028 at PTS. The \(f\) parameter ranged from 14.74 to 30.26 with an average value of 19.00 at 40 RR, while at PTS the value was 15.00. The results of \(\phi_{th}\) and \(\phi_{epi}\) at 40 RR were found to be in the range from 0.87 \times 10^{12} \text{ to } 2.55 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1} \text{ and from } 0.41 \times 10^{11} \text{ to } 1.37 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}, \text{ respectively. The average values of } \phi_{th} \text{ and } \phi_{epi} \text{ were } 2.17 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1} \text{ and } 1.16 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}, \text{ respectively, and at PTS with value of } 3.89 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1} \text{ for } \phi_{th} \text{ and } 2.59 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1} \text{ for } \phi_{epi}. \text{ In addition, the spectral index parameter } (r(\alpha)\sqrt{\mathcal{F}_n/\mathcal{F}_0}) \text{ was determined at 40 RR and PTS based on Westcott formalism. The average value was found as 0.0550 at RR and 0.0493 at PTS. The accuracy and precision of the proposed method were investigated by analyzing CRMs Soil-7, SL-1, IAEA-313, IAEA-312, NBS 1633A, USGS STM-1 and MAG-1 standard samples. The results showed a good agreement with the values reported in certificate with Z-score within 0< |Z|<2. Moreover, CRMs Soil-7, NBS 1633A and MAG-1 were analysed for relative method, and the results were found to be in good agreement with certified values. However, the results were slightly less than the results obtained by absolute method. Finally, the absolute NAA method was applied to determine the elemental concentration of U, Th and rare earth elements in rock samples based on Høgdahl convention except for Eu and Lu which were determined based on Westcott formalism. All samples were also analysed using relative method for comparison with the results obtained by absolute method. The deviations of both methods in most cases were found to be less than 10%.
ABSTRAK

Penyelidikan ini bertujuan untuk mengkaji keupayaan kaedah mutlak analisis pengaktifan neutron menggunakan reaktor penyelidikan PUSPATI TRIGA Mark II di Agensi Nuklear Malaysia (NM). Kebanyakan analisis kuantitatif pengaktifan neutron (NAA) dilakukan menggunakan kaedah perbandingan yang didapati mempunyai darjah ralat yang tinggi disebabkan oleh perbezaan matrik. NAA berdasarkan kaedah mutlak merupakan analisis penyinaran sampel yang lebih langsung berdasarkan kadar tindak balas (n, γ) tanpa menggunakan sampel piawai atau sampel bandingan. Dalam kajian ini kadar tindak balas (n, γ) berdasarkan konvensyen Høgdahl dan juga formulisme Westcott telah digunakan. Teknik ini memerlukan pengukuran sinar gamma mutlak dan parameter spektrum neutron bagi pengiraan berat atau kepekatan unsur yang terdapat dalam sampel. Parameter spektrum neutron seperti faktor bentuk neutron epiterma (α), nisbah neutron terma dan epiterma (f), dan fluks neutron terma dan epiterma (φth dan φepi) telah ditentukan di 40 kedudukan rak berputar (RR) dan juga di satu kedudukan pada sistem pemindah pneumatik (PTS). Nilai α di 40 kedudukan RR telah diperoleh dalam jual 0.0060 hingga 0.1170 dengan nilai purata 0.0172 dan 0.0028 di PTS. Parameter f yang diperoleh bernilai 14.74 hingga 30.26 dengan nilai purata 19.00 di 40 kedudukan RR, manakala di PTS bernilai 15.00. Nilai φth dan φepi yang diperoleh di 40 kedudukan RR masing-masing adalah dalam jual 0.87 × 10^{12} hingga 2.55 × 10^{12} n cm^{-2} s^{-1} dan 0.41 × 10^{11} hingga 1.37 × 10^{11} n cm^{-2} s^{-1}. Nilai purata φth dan φepi masing-masing adalah 2.17 × 10^{12} n cm^{-2} s^{-1} dan 1.16 × 10^{11} n cm^{-2} s^{-1}, dan di PTS dengan nilai 3.89 × 10^{12} n cm^{-2} s^{-1} bagi φth dan 2.59 × 10^{11} n cm^{-2} s^{-1} bagi φepi. Seterusnya, parameter indeks spektrum, $r(\alpha) \sqrt{T/\hat{T}}$ ditentukan di 40 RR dan PTS berdasarkan formulisme Westcott. Nilai purata yang diperoleh adalah 0.0550 di RR dan 0.0493 di PTS. Kejadian dan kepersisian kaedah yang dicadangkan ini diselidiki dengan menganalisis sampel piawai yang terdiri daripada CRMs Soil-7, SL-1, IAEA-313, IAEA-312, NBS 1633A, USGS STM dan MAG-1. Hasil kajian menunjukkan kesesuaian yang baik dengan nilai kepekatan yang dilaporkan dalam sijil dengan skor-Z bernilai di bawah 0 < |Z| < 2. Selain daripada itu, sampel piawai CRMs Soil-7, NBS 1633A dan MAG-1 telah dianalisis dengan menggunakan kaedah bandingan dan hasil kajian menunjukkan kesesuaian yang baik dengan nilai yang dilaporkan dalam sijil, tetapi kurang jitu sedikit berbanding dengan hasil yang diperoleh menggunakan kaedah mutlak. Akhir sekali, analisis pengaktifan neutron telah digunakan bagi menentukan kepekatan unsur U, Th dan unsur nadir bumi dalam sampel batuan berdasarkan konvensyen Høgdahl kecuali bagi unsur nadir bumi Eu dan Lu ditentukan berdasarkan formulisme Westcott. Seterusnya, kesemua sampel dianalisis menggunakan kaedah perbandingan untuk dibuat perbandingan dengan hasil yang diperoleh dengan kaedah mutlak. Sisihan bagi kedua-dua kaedah ini dalam kebanyakan kes adalah kurang daripada 10%.