THE ABSOLUTE METHOD OF NEUTRON ACTIVATION ANALYSIS USING TRIGA NEUTRON REACTOR, NUCLEAR AGENCY, MALAYSIA

LIEW HWI FEN

UNIVERSITI TEKNOLOGI MALAYSIA

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To beloved my mom and dad.

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ABSTRACT

Neutron activation analysis (NAA) offers excellent sensitivities that are superior to other analytical techniques in performing identification and quantitative elemental analysis. The technique involves the irradiation of samples and the detection of gamma energies emitted from the isotopes formed from the process of neutron capture. Most NAA were done by comparison method, which is found to have high errors due to the differences in the matrix composition of sample as well as comparator. The purpose of this study is to demonstrate an alternative technique of activation analysis based on absolute gamma ray measurements and the direct calculation of elemental concentrations from reaction rates equation of neutron capture process. The efficiency of the gamma-ray spectrometer as well as the neutron spectrum parameters, thermal and epithermal neutron flux at four characterized irradiation position of 10, 22, 27, and 31 in the rotary rack 1-MW TRIGA reactor at Malaysia Nuclear Agency was determined. The accuracy and precision of this absolute NAA technique were verified by analyzing two certified reference materials, Soil-1 and Soil-7 provided by IAEA. The experimental results for both the materials irradiated at the four characterized irradiation positions were found to be in good agreement with the certified values. The average Z-score for the concentration values were below two signified that the concentration results were accepted for most elements. In conclusion, the proposed technique can be applied for many of future activation analyses with high accuracy without having to rely on the availability of standard samples.

ABSTRAK

Analisis pengaktifan neutron (NAA) memberikan kepekaan yang tinggi berbanding dengan teknik analisis yang lain dalam mengenalpasti elemen dan analisis kuantitatif elemen. Teknik ini melibatkan penyinaran sampel dan penentuan tenaga gama yang dipancarkan oleh isotop yang terbentuk daripada proses penangkapan neutron. Kebanyakan NAA menggunakan kaedah perbandingan yang mempunyai ralat yang tinggi disebabkan perbezaan komposisi matriks di antara sampel dan sampel perbandingan. Tujuan kajian ini adalah melaksanakan kaedah alternatif analisis pengaktifan neutron iaitu kaedah mutlak berdasarkan pengukuran mutlak sinar gama dan pengiraan terus kepekatan elemen daripada persamaan kaedah tindak balas proses penangkapan neutron. Kecekapan spektrometer sinaran gama dan parameter spektrum neutron, fluks terma dan epiterma diukur pada lokasi penyinaran 10, 22, 27, dan 31 di rak berputar 1-MW TRIGA reaktor Agensi Nuklear Malaysia. Kejituan dan ketepatan kaedah mutlak ini ditentusahkan dengan menganalisis dua sampel piawai Soil-1 dan Soil-7 yang dibekalkan oleh IAEA. Keputusan eksperimen terhadap kedua- dua sampel piawai pada keempat–empat kedudukan penyinaran didapati menunujukkan persamaan yang baik dengan nilai yang disahkan. Nilai purata Z-skor kurang daripada dua menunjukkan keputusan analisis boleh diterimapakai untuk kebanyakan elemen secara tepat. Secara keseluruhannya, teknik yang dicadangkan boleh digunapakai untuk analisis pengaktifan neutron pada masa hadapan dengan ketepatan yang tinggi tanpa bergantung kepada bahan piawai.

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LIST OF SYMBOLS

α	-	Epithermal shape parameter
f	-	Thermal to epithermal flux ratio
φ_{th}	-	Thermal neutron flux
φ_{epi}	-	Epithermal neutron flux
t_i	-	Irradiation time
t_d	-	Decay time
t_m	-	Counting time
A_0	-	Activity of irradiated sample
N_A	-	Avogadro's number
θ	-	Natural isotopic abundance
M	-	Atomic weight
R	-	HOGDAHL Reaction Rate
S	-	Correction factors for saturation during irradiation
D	-	Correction factors for decay between irradiation and γ counting
C	-	Correction factors for decay during counting
m	-	Mass of the target element
N_p	-	full energy peak net count
γ	-	Gamma abundance
$\varepsilon_\gamma(E)$	-	Detector efficiency at gamma energy
N	-	Number of interacting isotopes
σ (E)	-	Cross-section in cm^2 at neutron energy of E in eV
φ (E)	-	Neutron flux per unit of energy interval

σ_o	-	Thermal neutron capture cross section at 2200 ms^{-1}
I_o	-	Resonance integral for a 1/E spectrum
$I_o(\alpha)$	-	Resonance integral valid for $1/E^{1+\alpha}$ spectrum
E_{cd}	-	Cadmium cut-off energy
$Q_o(\alpha)$	-	α- corrected Q_o
Q_o	-	I_o/σ_o ratio of resonance integral to (n, γ) cross section
E_r	-	Effective resonance energy
R_{cd}	-	Ratio of the specific count rates of the samples irradiated without and with a cadmium cover
A_{bare}	-	Activity of bare monitor
A_{cd}	-	Activity of monitor covered with cadmium
F_{cd}	-	Ratio of the activity of a monitor with a zero cadmium cover thickness
$Z_\alpha(A_{sp},n)$	-	Specific count rate random error
$Z_\alpha(A_{sp},n)_{bare}$	-	Bare specific count rate random error
$Z_\alpha(A_{sp},n)_{cd}$	-	Cadmium specific count rate random error
$Z_\alpha(F_{cd},n)$	-	Systematic errors of Fcd
$Z_\alpha(Q_o,n)$	-	Systematic errors of Qo
$Z_\alpha(E_r,n)$	-	Systematic errors of Er
$Z_\alpha(E_{cd},n)$	-	Systematic errors of Ecd
$S_{\alpha,T}$	-	Overall uncertainty of α parameter
n	-	Number of neutron per volume
v	-	Velocity of the neutron.
P_i	-	The fitted parameters of the function
E	-	Gamma energy of the i_{th} photopeak in MeV
$t_{1/2}$	-	Half Life
Z	-	Z-score

LIST OF ABBREVIATIONS

NAA	-	Neutron Activation Analysis
HPGe	-	High-purity germanium detector
FNAA	-	Fast Neutron Activation Analysis
PGNAA	-	Prompt Gamma Ray Neutron Activation Analysis
NAA	-	Neutron Activation Analysis
MNA	-	Malaysia Nuclear Agency
CRM	-	Certified Reference Materials
MNA	-	Malaysia Nuclear Agency

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CHAPTER I

INTRODUCTION

1.0 Introduction

Neutron activation analysis (NAA) was first discovered in 1936 and offers excellent sensitivities and is superior to other analytical techniques. NAA perform both qualitative and quantitative identification for a wide variety of materials in solid, liquid, or gaseous states. The reason for the high sensitivity is that the cross section of neutron activation is high in the thermal region for majority of the elements. Because of its vast range of potential applications, neutron activation analysis is utilized extensively in field such as geological science, medicine, agriculture, soil science, and environmental studies.

Neutron activation analysis is a physical technique based on the nuclear method. This technique is a non-destructive form of analysis. It can be used without damaging the materials being tested and minimizing the risk for loss and contamination. The undisputable advantage of this analytical technique is its multi-elemental character which enables simultaneous determination of many elements without chemical separation. Moreover, NAA is capable of detecting many elements at low concentrations. Nearly 70% of elements in the Periodic Table can be analyzed

by NAA.

1.1 Background of Study

There is a wide distribution of neutron energy in a reactor. The neutron spectrum consists of three principal components (thermal, epithermal and fast neutrons) based on their kinetic energies. In most reactor, thermal neutrons component are dominant and slow neutrons are fully moderated within the reactor with kinetic energies < 0.5 eV. Activation with epithermal neutrons is known as Epithermal NAA. Cadmium or boron is used as thermal neutron filter. These neutrons have been partially moderated and consist of kinetic energies of 0.5 eV to 0.5 MeV. Activation with fast neutrons (kinetic energies > 0.5MeV) is termed of Fast NAA (FNAA).

In principal, NAA falls into two categories depending on the time of measurement. Prompt gamma ray neutron activation analysis (PGNAA) measures gamma ray emitted during the irradiation while delayed gamma ray neutron activation analysis measures gamma ray as a result of radioactive decay.

There are three standard methods of NAA: relative method, k_o-standardization method and absolute method. Element analysis of an unknown sample using the relative method is usually perform by firstly irradiating known amounts of the sample and chemical standard simultaneously followed by comparing their gamma ray spectrum of the elements interest and counting under the same configuration. k_o-standardization method is based on irradiation of a sample with a neutron flux monitor such as gold and the use of nuclear constant called k_o - factor. This technique eliminates the need for using multi-element standards, thus is simpler than the relative method in terms of experiment but involves more complex formulae

and calculation. Absolute method is a direct analysis of the irradiated samples without using standard reference. This technique consists of absolute gamma ray measurement and direct calculation of weights or concentrations from nuclear constants.

1.2 Statement of Problem

The NAA relative method has been widely used in laboratory due to its direct, simple, and accurate elemental analysis. However, in this relative method, no detailed knowledge of the neutron flux, φ in the irradiation site or the nuclear data of the isotope concerned is required. The concentration of elements determined is dependent on the flux gradient in the irradiation position. Therefore, when the samples and standard have to be irradiated at different irradiation positions, the variation of the neutron flux at different irradiation position in the reactor might influences in the accuracy and precision of the analytical result. Besides, the different radial shape of sample and standard can contribute to different spatial flux distribution.

In the case of simultaneous determination of a great number of elements in one sample, the relative method requires preparation, counting and data processing of a standard for each element to be determined. It is also difficult to irradiate a large number of standards and samples together in a vial. Therefore, this will limit the samples that can be analysis by relative method.

There are possibility of instability and not homogeneity of the standard used in NAA relative method. In addition, differences in matrix composition between the sample and standard can contribute to uncertainty. All these factors can make relative NAA method becoming cumbersome, time consuming, laborious and expensive,

despite the fact that it is a very sensitive analytical method.

1.3 Purpose of Research

The aim of this research is to develop an absolute method of Neutron Activation Analysis that can be used in the laboratory of Analytical Chemistry, Malaysia Nuclear Agency (MNA). The method is based solely on the reaction rate formulations of the neutron capture processes. This absolute method requires the input of neutron reactor parameters that have to be precisely determined by experiment before the elemental masses of the irradiated samples can be computed.

1.4 Objectives of Research

The objectives of the study are:

i. To calibrate the full energy peak efficiency of the detector for the gamma ray spectrometer used.

ii. To characterize the reactor neutron spectrum (epithermal shape parameter α, thermal to epithermal flux ratio f, thermal and epithermal neutron flux φ_{th}, $\varphi_{epi,}$) at four selected irradiation locations at rotary rack of the TRIGA MARK II reactor.

iii. To determine the elements concentration of certified reference materials (CRM) in order to examine the accuracy of the developed method.

1.5 Significant of Research

The expected outcome from this research is a viable approach of doing NAA which no longer rely on the use of multi standard materials that can be subjected to many errors. This research is hoping to overcome some of the drawbacks of conventional NAA method (Relative method) and able to produce reliable results that can be effectively applied for NAA analysis. Besides, a large number of elements can be determine simultaneously without the use of reference standards which can better enhance the NAA technique. Hopefully, this new approach can be used by many industries in the determination of elements in sample.

1.6 Research Scope

The scopes of the work have been defined to comprise:

i. To calibrate the efficiencies of two detectors name Ortec detector GEM10185 detector controlled by Gamma Vision software and Canberra GC 3018 detector at different geometry.

ii. To measure the epithermal shape parameter, α using 5 monitors (Au-Al, Zr, Zn, Co and Mo) activation method with and without cadmium cover at rotary rack facilities of 1MW TRIGA Mark II reactor at MNA.

iii. To determine the thermal and epithermal neutron flux of the irradiation site by using gold foil activation method irradiated with and without cadmium cover.

iv. To determine the element concentration of soil-1 and soil-7. This

sample is intended to be used as a standard reference material in quality control material for the assessment of a laboratory's analytical work.