

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

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

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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, γ) reaction rate without using any standard or comparator. In this study the reaction rate of (n, γ) 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 flux shape factor (α), thermal to epithermal neutron flux ratio (f), and thermal and epithermal neutron flux (ϕ_{th} and ϕ_{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 α 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 ϕ_{th} and ϕ_{epi} at 40 RR were found to be in the range from 0.87×10^{12} to $2.55 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ and from 0.41×10^{11} to $1.37 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}$, respectively. The average values of ϕ_{th} and ϕ_{epi} were $2.17 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ and $1.16 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}$ respectively, and at PTS with value of $3.89 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ for ϕ_{th} and $2.59 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}$ for ϕ_{epi} . In addition, the spectral index parameter ($r(\alpha)\sqrt{T_n/T_0}$) 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 (ϕ_{th} dan ϕ_{epi}) 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 julat 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 ϕ_{th} dan ϕ_{epi} yang diperoleh di 40 kedudukan RR masing-masing adalah dalam julat 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 ϕ_{th} dan ϕ_{epi} 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 ϕ_{th} dan 2.59×10^{11} $n\ cm^{-2}\ s^{-1}$ bagi ϕ_{epi} . Seterusnya, parameter indeks spektrum, $r(\alpha)\sqrt{T_n/T_0}$ ditentukan di 40 RR dan PTS berdasarkan formulisme Westcott. Nilai purata yang diperoleh adalah 0.0550 di RR dan 0.0493 di PTS. Kejituan dan kepersisan 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-1 dan MAG-1. Hasil kajian menunjukkan persetujuan yang baik dengan nilai kepekatan yang dilaporkan dalam sijil dengan skor-Z bernilai diantara $0 < |Z| < 2$. Selain daripada itu, sampel piawai CRMs Soil-7, NBS 1633A and MAG-1 telah dianalisis dengan menggunakan kaedah bandingan dan hasil kajian menunjukkan persetujuan 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. Sisihih bagi kedua-dua kaedah ini dalam kebanyakan kes adalah kurang daripada 10%.

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LIST OF ABBREVIATIONS

ADC	-	Analog to digit converter
Au	-	Gold
Ce	-	Cerium
CNAA	-	Cyclic Neutron Activation Analysis
Co	-	Cobalt
CRMs	-	Certified Reference Materials
Dy	-	Dysprosium
ENAA	-	Epithermal neutron activation analysis
Eu	-	Europium
eV	-	Electron volt
FEPE	-	Full energy peak efficiency
FNAA	-	Fast neutron activation analysis
FWHM	-	Full-width-at half-maximum
GCC	-	Gulf Cooperation Council
Ge(Li)	-	Germanium Lithium doped
HNO ₃	-	Nitric acid
HPGe	-	Hyper- Pure Germanium
HREE	-	Heavy rare earth element
IAEA	-	International Atomic Energy Agency
ICP-MS	-	Inductively coupled plasma-mass spectrometry
INAA	-	Instrumental neutron activation analysis
kW	-	Kilowatt
La	-	Lanthanum
LREE	-	Light rare earth element
LN ₂	-	Liquid nitrogen
Lu	-	Lutetium

MCA	-	Multichannel analyzer
MW	-	Megawatt
NAA	-	Neutron activation analysis
NaI(Tl)	-	Sodium iodide activated with thallium
NBS	-	National Bureau Standard
Nd	-	Neodymium
NM	-	Nuclear Malaysia
Np	-	Neptunium
Pa	-	Protactinium
PNAA	-	Preconcentration Neutron Activation Analysis
PGNAA	-	Prompt Gamma-ray Neutron Activation Analysis
ppb	-	Part per billion
ppm	-	Part per million
PTS	-	Pneumatic transfer system
REEs	-	Rare earth elements
RNAA	-	Radiochemical Neutron Activation Analysis
RR	-	Rotary rack
RTP	-	Reactor TRIGA PUSPATI
Sm	-	Samarium
SRM	-	Standard Reference Material
Tb	-	Terbium
Th	-	Thorium
TNAA	-	Thermal Neutron Activation Analysis
TRIGA	-	Training Research and Isotope General Atomic
UKM	-	Universiti Kebangsaan Malaysia
USGS	-	United States Geological Survey
U	-	Uranium
(U ⁺⁴)	-	Uranous ion
(UO ₂ ⁺²)	-	Uranyl ion
UZrH	-	Uranium-zirconium-hydride
Yb	-	Ytterbium

LIST OF SYMBOLS

A	-	Activity present after time (t) in (Bq)
A_0	-	Initial activity of the source (Bq)
A_{sp}	-	Specific count rate ($s^{-1}g^{-1}$)
b	-	Barn ($10^{-28} m^2$)
C	-	Counting factor ; $= (1 - e^{-\lambda t_c}) / \lambda t_c$
D	-	Decay factor; $= (e^{-\lambda t_d})$
E_{Cd}	-	Cadmium cut-off energy (= 0.55 eV)
E_r	-	Effective energy (eV)
\bar{E}_r	-	Effective resonance energy (eV)
E_0	-	0.0253 eV Maxwellian neutron energy
F	-	Thermal to epithermal flux ratio
F_{Cd}	-	Cadmium transmission factor epithermal neutrons
g	-	Statistical weight factor
$g(T_n)$	-	Westcott's g -factor
$g_{Lu}(T_n)$	-	Westcott's g -factor of Lu
G_{epi}	-	Correction factor for epithermal neutron self-shielding
G_r	-	Correction factor for resonance neutron self-shielding
G_{th}	-	Correction factor for thermal neutron self-shielding
I_0	-	Resonance integral for a $1/E$ epithermal spectrum
$I_0(\alpha)$	-	Resonance integral for a $1/E^{1+\alpha}$ epithermal spectrum
$I(E)$	-	Branching gamma ratio of energy E
k_0	-	k_0 factor
m	-	Mass of element
m_s	-	Mass of calibration standards
M	-	Atomic weight ($g.mol^{-1}$)
$n(v)$	-	Density of neutrons

N	-	Number of radioactive nuclei
N_0	-	Number of target nuclei
N_A	-	Avogadro's number ($\approx 6.23 \times 10^{23} \text{ mol}^{-1}$)
N_p	-	Number of counts in the full-energy peak
Q_0	-	Resonance integral (I/E) to 2200 m s ⁻¹ cross-section ratio
$Q_0(\alpha)$	-	Resonance integral ($I/E^{1+\alpha}$) to 2200 m s ⁻¹ cross-section ratio ($I_0(\alpha)/\sigma_0$)
$r(\alpha)\sqrt{T_n/T_0}$	-	Modified spectral index
r	-	Epithermal index
R	-	Reaction rate (s ⁻¹)
R_{Cd}	-	Cd-ratio ($= A_{sp}/(A_{sp})_{Cd}$)
R_{epi}	-	Epithermal (n, γ) reaction rate per nucleus
R_{th}	-	Thermal (n, γ) reaction rate per nucleus
s_0	-	Corresponding quantity for an ideal 1/E epithermal neutron flux distribution
$s_0(\alpha)$	-	Modified reduced resonance integral
$S_{\alpha,T}$	-	Overall uncertainty of α parameter
S	-	Saturation factor ($1 - e^{-\lambda t_i}$)
t_c	-	Counting time (s)
t_d	-	Decay time (s)
t_i	-	Irradiation time (s)
t_m	-	Measuring time (s)
$T_{1/2}$	-	Half life (s)
T_n	-	Neutron temperature (°C)
T_0	-	293.59 K (Maxwellian) neutron temperature
v	-	Neutron velocity (m s ⁻¹)
v_0	-	The Maxwellian neutron velocity (m s ⁻¹)
V_{Cd}	-	Velocity of neutron corresponding with Cd-cut off energy
W'	-	Constant value for each element
Z	-	Z-score
α	-	Epithermal neutron flux shape factor

β	-	Beta particle
γ	-	gamma abundance
ε_p	-	Full-energy peak detection efficiency
ε_{abs}	-	Absolute efficiency
ε_{int}	-	Intrinsic efficiency
ε_T	-	Total efficiency
θ	-	Isotopic abundance
λ	-	Decay constant (s^{-1}); $\lambda = \frac{\ln 2}{T_{1/2}}$
ρ	-	The absolute gamma emission probability
σ	-	Neutron capture cross section
σ_0	-	2200 m s ⁻¹ cross section
$\sigma(E)$	-	The (n, γ) cross section [in cm ²] at neutron energy E
$\sigma(v)$	-	The (n, γ) cross section [in cm ²] at neutron velocity v
τ	-	Dead time
Δm	-	Uncertainty in mass of sample
ΔN	-	Uncertainty in the number of counting
ΔR	-	Uncertainty in the reaction rate
φ_{epi}	-	Epithermal neutron flux (n cm ⁻² s ⁻¹)
φ_{th}	-	Thermal neutron flux (n cm ⁻² s ⁻¹)

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CHAPTER 1

INTRODUCTION

1.1 Introduction

The technique of nuclear activation analysis is based on activation of the elements by sub-atomic particles and the subsequent measurement of induced radioactivity. If the sub-atomic particle is a neutron then the technique is called neutron activation analysis (NAA). The technique was first discovered in 1936 by Georg von Hevesy and Hilde Levi, whereby a neutron source was used to activate a mixture of rare earth elements to determine the concentration of dysprosium (Dy) and europium (Eu). However, due to relatively weak neutron sources and inefficient γ -spectrometry available, the development of NAA was rather slow. Therefore, most data on elemental content was obtained through chemical separations and decay curve analysis. Twenty years later, the rapid progress of NAA began when the nuclear research reactor made intense sources of neutrons available.

In the early sixties, the development of neutron activation analysis accelerated when NaI (TI) gamma-ray detectors became available coupled with multichannel pulse-high analyzers. Now it is possible to obtain gamma-ray spectra data with higher efficiency and moderate energy resolution which allow for the simultaneous determination of different radionuclides without chemical separations. With the advent of the high purity Ge detector, around 1970, neutron activation

analysis became one of the major techniques in the nuclear analysis field (Abugassa, 1999; Blaauw, 1993).

The high sensitivity of NAA technique makes the technique suitable for the determination of trace elements including rare earth elements (REEs). In addition, NAA determines many elements simultaneously and non-destructively (instrumental). Furthermore, due to its simplicity and high degree of accuracy, NAA has been recommended as a method for certifying reference materials and for utilization in multidisciplinary studies (IAEA-TECDOC-1215, 2001).

Nowadays, neutron activation is among the most sensitive tools used for identification and for the quantitative elemental analysis of samples in the field of geology, agriculture, environmental science, as well as biological and biomedical studies. Most elements can now be determined at the level of ppb (part per billion). Basically in this technique, a given sample is irradiated with thermal or epithermal neutrons and becomes radioactive. The unstable isotope then decays normally by emitting beta particles and gamma quanta with specific energies into a more stable configuration. The intensity of the characteristic gamma ray lines in the spectrum, are proportional to the elemental concentration that are measured and used for quantitative identification of the element. High fluxes of neutrons are normally required for the activation process and the most popular neutron source for this specification is the nuclear reactor.

The neutron spectrum (intensity versus energy) in a reactor consists of three components: thermal, epithermal and fast neutrons. For the neutron capture processes to occur, thermal neutrons are used. However, the entire reactor neutron spectrum may also be utilized via filtering with either cadmium or boron to achieve selective activation with epithermal or fast neutrons. Activation with epithermal neutrons is known as Epithermal NAA, and activation with fast neutrons is called Fast NAA (Sood *et al.*, 2004).

There are three approaches in doing NAA: relative method, k_0 -standardization method (single comparator), and absolute method. Elemental analysis of unknown sample using the relative method is usually performed by irradiating a standard material of known mass simultaneously with the sample, then followed by comparing their gamma ray spectrum of the elements of interest and counting them under identical conditions. This method requires no detailed information of the neutron flux, ϕ at the reactor irradiation site or of the nuclear data for the isotope concerned. Here, it is assumed that, neutron flux, irradiation and counting times, geometries and other variables are the same for both the unknown sample and known chemical standard. One of the drawbacks of the relative method is the difficulty in maintaining the stability of chemical standard.

In the case of simultaneous determination of a large number of elements in one sample, the relative method requires preparation, counting and data processing of a standard for each element to be determined. There is a high possibility of instability and non homogeneity of the standard used. In addition, differences in matrix composition between the standard and sample can contribute to experimental uncertainty. It is cumbersome to irradiate a large number of standards and samples at the same time.

The k_0 -standardization method is based on simultaneous irradiation of a sample and a neutron flux monitor (normally gold), and the use of a composite nuclear constant called k_0 -factor. This technique eliminates the need of using multi-element standards to achieve better precision and accuracy of the result. Compared to the relative method, the technique of k_0 -NAA is experimentally simpler but more complex in the formulation and calculations as well as computational programming. This technique requires experts to interpret the spectrum of selected elements in the sample, which are difficult to do. For multi-element analysis, this method may not be easy as far as nuclear data and decay scheme parameters are concerned.

On the other hand, NAA based on absolute method is a more direct analysis of the irradiated samples without using any standard or comparator. 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 usual parameters that characterize the neutron flux are their fluxes, thermal and epithermal flux ratio and epithermal parameters.

1.2 Problem Statement

The reactor available at Malaysia Nuclear Agency (NM) has been utilized for INAA studies in many fields such as environmental, nuclear data studies, nutritional epidemiological studies, industrial materials analysis, geological and geochemistry studies. The main method used at NM is the relative technique, simply because of its easiness method. However, this method becomes difficult in case of multi-element analysis. The choice of standard for each element is a great challenge and is also influenced by many factors such as solubility in solvents, purity, weighing, resistance to radiation and decomposition (Kafala and MacMahon, 2007). Several approaches have been suggested to solve this problem such the use of synthetic multi-elements standard and standard reference material (SRM). In the first approach, the preparation of multi-element in one solution involves the problems of stability of individual ions during storage and adsorption of elements on container wall. Whereas the use of SRM is convenient to control the conditions of irradiation, cooling and counting of samples, it is not easy to maintain proper reference standard materials and the benefit is reduced by uncertainties in the analytical data (Kim *et al.*, 1986).

The application of the k_0 - standardization method in NM was studied by many researchers (Abugassa *et al.*, 1996; Abugassa, 1999; Abugassa *et al.*, 2004; Khoo *et al.*, 2007; Wee *et al.*, 2006; Yavar, 2012). This method had been implemented but not in a wide scales due to the lack of expert in the k_0 software management. Furthermore, the problem of choosing suitable comparator elements for multi-elements analysis may not be easy (Kafala and MacMahon, 2007)

In order to overcome the problems mentioned above, absolute method is the better choice in NAA method. In this case the direct elemental concentration calculations from few measured neutron parameters are thought to reduce the uncertainties obtained from the relative and k_0 methods. The neutron flux parameters had been measured at rotary rack irradiation positions of PUSPATI reactor but it was not utilized for absolute NAA method. With the large amount of accurate nuclear data available, the NAA absolute method became more reliable and accurate. This research will focus on the study of the capability of the absolute method at PUSPATI TRIGA Mark II research reactor and applied it to determine U, Th and rare earth elements (REEs) content in rock samples as compared with the relative method.

1.3 Research Objectives

The objectives of this research are as follows:

- a) To determine neutron spectrum parameters at PUSPATI TRIGA Mark II research reactor irradiation facilities at the forty positions of Rotary Rack (RR) and one location of Pneumatic Transfer System (PTS). These include the epithermal neutron flux shape factor α , thermal to epithermal flux ratio, f and thermal and epithermal neutron fluxes (ϕ_{th} , ϕ_{epi}).
- b) To determine the elemental concentration of certified reference materials (CRMs) of IAEA (Trace Elements in Soil-7, Lake Sediment SL-1, Stream Sediment IAEA-313 and IAEA-312), USGS (Nepheline Syenite STM-1 and Marine Sediment MAG-1) and NBS (Coal Fly Ash 1633A) by absolute method and examine their accuracy using Z-scores.
- c) To compare the results of CRM (IAEA Soil-7, Marine Sediment MAG-1 and NBS Coal Fly Ash 1633A) with the results determined by relative method.
- d) To determine the elemental concentration of U, Th and REEs in rock samples by both absolute and relative methods and compare their results based on their respective relative deviations and correlation coefficients.

1.4 Research Scope

The main aim of this work is to affirm the capability of absolute method of neutron activation analysis as one of the standardization method of NAA, which can be used by Malaysia Nuclear Agency. The concentration of elements in samples determined by absolute method is based solely on the reaction rate formulation of the neutron capture processes. The proposed method was developed based on Høgdahl convention and Westcott formalism. The Westcott formalism was used in this study to determine the elemental concentration of Eu and Lu, whereby their (n, γ) reaction showed a significant deviation from $1/v$ cross section behavior.

The absolute method requires the determination of neutron reactor parameters experimentally. In addition, the full-energy-peak efficiencies (ϵ_γ) of the gamma ray detector for counting purposes have to be determined. Practically, neutron flux parameters change according to reactor core configuration, as well as homogeneity and population of neutron flux incident in particular irradiation positions. This change may affect the accuracy of elemental concentration. Therefore, to avoid this possible effect the neutron flux parameters were determined by attaching a suitable monitor (Au and Zr) to the samples and then irradiated simultaneously.

In this study, the neutron flux parameters α , f , ϕ_{th} , and ϕ_{epi} were determined by using three monitors (Au, Zr and Co), irradiated with and without cadmium cover at forty irradiation positions of rotary rack (RR) and one location at pneumatic transfer system (PTS) facilities in PUSPATI TRIGA Mark II (thermal power capacity of 1 MW). In addition, the calibration efficiencies of two coaxial HPGe detectors, coupled with Canberra GC3018 and Ortec GEM25-76-XLB-C were determined experimentally at four different source-detector distance. A simple computer program written in MATLAB was developed to calculate the neutron flux parameters and efficiency calibration of the detectors.

In order to evaluate the accuracy and precision of the absolute method, the absolute method was applied to determine of elementals concentration in different

types of Certified Reference Materials (CRM): IAEA (Lake Sediment SL-1; Trace Elements in Soil-7; Stream Sediment IAEA-313 and IAEA-312); USGS (Nepheline Syenite STM-1 and Marine Sediment MAG-1) and NBS (Coal Fly Ash 1633A) and the results were compared with the certified values.

This study involves the application of absolute method on rock samples in order to determine the concentrations of uranium, thorium and rare earth elements (La, Ce, Nd, Sm, Eu, Tb, Dy, Yb and Lu). Rock samples were collected from six states namely: Perak, Penang, Kelantan, Negeri Sembilan, Selangor and Johor. The results were compared to NAA relative technique.

1.5 Significance of the Research

This research is for the purpose of developing standard methods in NAA, specifically the absolute method because it offers several advantages over the relative and k_0 methods such as expense, versatility and ease of automation. It also offers the possibility of multi-element analysis in one single irradiation without the use of standard or comparator which better enhances the NAA technique.

This work will introduce a simple approach to calculating nuclear parameters such as the epithermal neutron flux shape factor (α); fitting the efficiency calibration curve; determining the correction of coincidence effect; and that it is able to produce reliable results that can be effectively applied for NAA. In addition, it is hoped that this study will demonstrate the viability of the absolute NAA method in determination of elements present in different types of samples.

Furthermore, this study is contribute significant information to provide general information and baseline data for the NAA absolute method related to mathematical formulas, excel workbook format and a simple programming approach

that can be used to determine neutron flux parameters, efficiency calibration that can establish the NAA absolute method in Malaysian Nuclear Agency.

1.6 Thesis Organization

This thesis consists of six chapters. Chapter 1 presents the introduction of the NAA method; problem statement; objectives of the study; scope of the study; and significance of the study.

Chapter 2 involves a literature review of NAA including information on uranium, thorium and rare earth elements. Chapter 3 discusses the derivation of mathematical formulae based on Høgdahl Convention and Westcott Formalism for the calculation of elemental concentrations using absolute NAA. Full details on instrumentation and methodology are presented in Chapter 4, involving the information on the reactor and γ -ray spectrometry including the HPGe detector and experimental procedures. Chapter 5 covers the calibration of detectors used in this study and a discussion of results for the determination of neutron flux parameters α , f , ϕ_{th} and ϕ_{epi} , and the determination of elemental concentrations for U, Th and REEs (La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, and Lu). Conclusion and suggestions are summarized in Chapter 6.

REFERENCES

- Abugassa, I., Sarmani, S. and Samat, S. (1996). Development of k_0 -standardization method for reactor neutron activation analysis. *Sains Malaysiana*. 25(3), 47-54.
- Abugassa, I. (1999). *A study of instrumental neutron activation analysis based on k_0 -standardisation method developed for environmental materials*. Ph.D. Thesis. University Kebangsaan Malaysia.
- Abugassa, I., Sarmani, S. and El-Ghawi, U. (2004). Instrumental neutron activation analysis based on k_0 -standardization method as compared with other methods in the analysis of the IAEA inter-comparison test. *Journal of radioanalytical and nuclear chemistry*. 259(3), 381-384.
- Acharya, R., Nair, A., Reddy, A., and Manohar, S. (2002). Validation of a neutron activation analysis method using k_0 -standardization. *Applied Radiation and Isotopes*. 57(3), 391-398.
- Ahmad, A., Gray, P., Macmahon, T. and Macwani, M. (1982). Neutron activation analysis without multielement standards. *Journal of Radioanalytical Chemistry*. 72(1-2), 335-352.
- Aksoy, A. (1993). Efficiency calibration of HPGe detector in far and close geometries. *Journal of radioanalytical and nuclear chemistry*. 169(2), 463-469.
- Aksoy, A. (2011). Efficiency Inter-Comparison Measurements of Medium and Large Volume HPGe Detectors in Far and Close Geometries. *Arabian Journal for Science and Engineering*. 36(1), 131-136.
- Al-Sulaiti, H. (2011). *Determination of Natural Radioactivity Levels in the State of Qatar Using High- Resolution Gamma-ray Spectrometry*. Ph.D.Thesis. University of Surrey. UK.

- Al-Zahrany, W. (2007). *Elemental Distributions in Marine Sediments in the Straits of Melaka using Neutron Activation and Mass Spectroscopic Analyses*. Ph.D. Thesis. Universiti Putra Malaysia.
- Alnour, I., Ibrahim, N. and Liew, H. F. (2011). The accuracy of the absolute NAA method based on the analysis of standard reference materials (SRMs). *International Journal of Physical Sciences*. 6(17), 4169-4175.
- Alnour, I., Ibrahim, N. and Hossain, I. (2012a). Concentrations of ^{214}Pb , ^{214}Bi in ^{238}U series and ^{208}Tl , ^{228}Ac in ^{232}Th series in granite rock in (Kadugli) Sudan. *Indian Journal of Pure and Applied Physics*. 50(5), 285-288.
- Alnour, I., Ibrahim, N., Wagiran, H., Laili, Z., Omar, M., Hamzah, S., *et al.* (2012b). Natural radioactivity measurements in the granite rock of quarry sites, Johor, Malaysia. *Radiation Physics and Chemistry*. 81(12), 1842-1847.
- Alnour, I., Wagiran, H., Ibrahim, N., Hamzah, S., Wee, B., Elias, M., *et al.* (2013). Determination of neutron flux parameters in PUSPATI TRIGA Mark II Research Reactor, Malaysia. *J. Radioanal. Nucl. Chem*. 296(3), 1231-1237.
- Bacon, G. E. (1969). *Neutron Physics*. Wykham Publications (London) Ltd: A subsidiary of Taylor & Francis Ltd. London.
- Basri, N. A. and Ramli, A. T. (2012). Selection of Possible Candidate Area for Nuclear Power Plant In Johor, Malaysia. . *JOURNAL of NUCLEAR And Related TECHNOLOGIES*. 9 (1), 56-63.
- Bereznai, T. (1980). Methods, problems and trends of standardization in multielement reactor neutron activation analysis. *Fresenius' Zeitschrift für analytische Chemie*. 302(5), 353-363.
- Bergerioux, C., Kennedy, G. and Zikovsky, L. (1979). Use of the semi-absolute method in neutron activation analysis. *Journal of Radioanalytical Chemistry*. 50(1-2), 229-234.
- Blaauw, M., Bode, P., and De Bruin, M. (1991). An alternative convention describing the (n, γ)-reaction rate suited for use in the k_0 -method of NAA. *Journal of radioanalytical and nuclear chemistry*. 152(2), 435-445.
- Blaauw, M. (1993). *The holistic analysis of gamma-ray spectra in instrumental neutron activation analysis*. Ph.D. Thesis. Delft University of Technology.
- Bode, P., Blaauw, M. and Obrušnik, I. (1992). Variation of neutron flux and related parameters in an irradiation container, in use with k_0 -based neutron activation analysis. *Journal of radioanalytical and nuclear chemistry*. 157(2), 301-312.

- Castor, S. B. and Hedrick, J. B. (2006). Rare earth elements. *Industrial Minerals volume, 7th edition: Society for Mining, Metallurgy, and Exploration, Littleton, Colorado*, 769-792.
- Chakraborty, K., Ram, G. and Aidid, S. (1980). Rare earth element abundance patterns in alkaline basaltic lavas of Kuantan. Peninsular Malaysia. *Geological Society of Malaysia Bulletin*. 13, 103-111.
- Chaudhary, M., Ahmad, S. and Qureshi, I. (1980). Simultaneous determination of thorium and uranium in ores and SRMs by instrumental neutron activation analysis. *Journal of Radioanalytical Chemistry*. 57(1), 137-146.
- Covell, F. D. (1975) Gamma-Ray Spectrometry. In: Coomber. D. I. Ed. *Radiochemical Methods in Analysis*. New York and London: Pleunm Press.
- Currie, L. A. (1968). Limits for qualitative detection and quantitative determination. Application to radiochemistry. *Analytical Chemistry*, 40(3), 586-593.
- De Corte, F., Speecke, A. and Hoste, J. (1969). Reactor neutron activation analysis by a triple comparator method. *Journal of radioanalytical and nuclear chemistry*. 3(3), 205-215.
- De Corte, F., Moens, L., Simonits, A., Wispelaere, A. and Hoste, J. (1979). Instantaneous α -determination without Cd-cover in the $1/E^{1+\alpha}$ epithermal neutron spectrum. *Journal of Radioanalytical Chemistry*. 52(2), 295-304.
- De Corte, F., Hammami, K. S.-E., Moens, L., Simonits, A., Wispelaere, A. and Hoste, J. (1981). The accuracy and precision of the experimental α -determination in the $1/E^{1+\alpha}$ epithermal reactor-neutron spectrum. *Journal of Radioanalytical Chemistry*. 62(1-2), 209-255.
- De Corte, F., Moens, L., Simonits, A., Sordo-El Hammami, K., Wispelaere, A. and Hoste, J. (1982). The effect of the epithermal neutron flux distribution on the accuracy of absolute and comparator standardization methods in (n, γ) activation analysis. *Journal of Radioanalytical Chemistry*. 72(1-2), 275-286.
- De Corte, F. (1987). *The k_0 -standardization method—A move to the optimization of neutron activation analysis*. Ph.D. Thesis. Rijksuniversiteit Gent.
- De Corte, F., Simonits, A., Bellemans, F., Freitas, M. C., Jovanović, S., Smodiš, B., et al. (1993). Recent advances in the k_0 -standardization of neutron activation analysis: Extensions, applications, prospects. *Journal of radioanalytical and nuclear chemistry*. 169(1), 125-158.

- De Corte, F. (1994). Neutron activation analysis: an old faithful to cherish. *Acta Physica Hungarica*. 75(1-4), 189-197.
- De Corte, F., Bellemans, F., De Neve, P. and Simonits, A. (1994). The use of a modified Westcott-formalism in the k_0 -standardization of NAA: The state of affairs. *Journal of radioanalytical and nuclear chemistry*. 179(1), 93-103.
- De Corte, F. and Simonits, A. (1996). KAYZERO/SOLCOI for Neutron Activation Analysis (NAA) using the k_0 Standardization Method version 4 User's manual. DSM Research, Geleen (NL).
- De Corte, F. (2001). The standardization of standardless NAA. *Journal of radioanalytical and nuclear chemistry*. 248(1), 13-20.
- De Corte, F. and Simonits, A. (2003). Recommended nuclear data for use in the k_0 standardization of neutron activation analysis. *Atomic data and Nuclear data Tables*. 85(1), 47-67.
- Debertin, K., and Schötzig, U. (1979). Coincidence summing corrections in Ge (Li)-spectrometry at low source-to-detector distances. *Nuclear Instruments and Methods*. 158, 471-477.
- Debertin, K. and Helmer, R. G. (1988). *Gamma-and X-ray spectrometry with semiconductor detectors* (Vol. 126): North-Holland Amsterdam.
- Diaz, O., Peraza, E. H., Reyes, M. C. L., Pellon, I. A., Guevara, M. V. M. and Cabrera, M. I. (1997). Epithermal neutron flux characterization of the Triga Mark III reactor, Salazar, Mexico, for use in INAA. *Journal of radioanalytical and nuclear chemistry*. 220(1), 95-97.
- Diaz, O., Figueiredo, A., Nogueira, C., Lopez, N., Gonzalez, H., Manso, M., et al. (2005). Epithermal neutron flux characterization of the IEA-R1 research reactor, Sao Paulo, Brazil. *Journal of radioanalytical and nuclear chemistry*. 266(1), 153-157.
- Dubinskaya, N. A. Pelekis. L. L. (1971). The application of monitors in neutron activation analysis. *Journal of Radioanalytical Chemistry*. 9,(1) 61-72.
- Dung, H. M., and Sasajima, F. (2003). Determination of α and f for k_0 -NAA in irradiation sites with high thermalized neutrons. *Journal of radioanalytical and nuclear chemistry*. 257(3), 509-512.
- Dung, H., and Hien, P. (2003). The application and development of k_0 -standardization method of neutron activation analysis at Dalat research reactor. *Journal of radioanalytical and nuclear chemistry*. 257(3), 643-647.

- Ehmann, W. D. and Vance, D. E. (1991). *Radiochemistry and nuclear methods of analysis*. John Wiley & Sons, Inc. New York.
- El-Taher, A. (2007). Rare-earth elements in Egyptian granite by instrumental neutron activation analysis. *Applied Radiation and Isotopes*. 65(4), 458-464.
- El Nimr, T., De Corte, F., Moens, L., Simonits, A. and Hoste, J. (1981). Epicadmium neutron activation analysis (ENAA) based on the k_0 -comparator method. *Journal of Radioanalytical Chemistry*. 67(2), 421-435.
- Franek, M. and Krivan, V. (1993). Determination of sub-ng g⁻¹ concentrations of thorium and uranium in microelectronic materials by radiochemical neutron activation analysis. *Analytica Chimica Acta*. 274(2), 317-325.
- Friedlander, G., Kennedy, J. W., Macias, E. S. and Miller, J. M. (1981). *Nuclear and radiochemistry*: Wiley.
- Girardi, F., Guzzi, G., and Pauly, J. (1964). Activation Analysis by Absolute Gamma Ray Counting and Direct Calculation of Weights from Nuclear Constants. *Analytical Chemistry*. 36(8), 1588-1594.
- Girardi, F., Guzzi, G. and Pauly, J. (1965). Reactor Neutron Activation Analysis by the Single Comparator Method. *Analytical Chemistry*. 37(9), 1085-1092.
- Gobbett, D. J. and Hutchison, C. S. (1973). *Geology of the Malay Peninsula: West Malaysia and Singapore*: Wiley-Interscience New York.
- Greenberg, R. R., Bode, P. and De Nadai Fernandes, E. A. (2011). Neutron activation analysis: A primary method of measurement. *Spectrochimica Acta Part B: Atomic Spectroscopy*. 66(3), 193-241.
- Gultekin, A., Kaynak, G. and Gurler, O. (2006). Determination of full energy peak efficiency of HpGe detector from 59.5 to 1332.5 keV. *Indian Journal of Pure and Applied Physics*, 44(4), 281.
- Hamidatou, L. A. and Ramdhane, M. (2008). Characterization of neutron spectrum at Es-Salam Research Reactor using Høgdahl convention and Westcott formalism for the k_0 -based neutron activation analysis. *Journal of radioanalytical and nuclear chemistry*. 278(3), 627-630.
- Henderson, P. (1984). *Rare earth element geochemistry*. Elsevier Publ., Amsterdam, 510p.
- Høgdahl, O. T. (1962). *Neutron absorption in pile neutron activation analysis*: Michigan. Univ., Ann Arbor. Michigan Memorial-Phoenix Projecto.

- Høgdahl, O. (1965). Neutron absorption in pile neutron activation analysis determination of copper and gold in silver. *Radiochemical Methods of analysis, Symposium on Radiochemical Methods of analysis*: Salzburg, October 19-23, 1964, IAEA Vienna.
- Holden, N. E. (1999). Temperature dependence of the Westcott g-factor for neutron reactions in activation analysis. *Pure and Applied Chemistry*. 71(12), 2309-2315.
- Huh, C. and Bacon, M. (1985). Determination of thorium concentration in seawater by neutron activation analysis. *Analytical Chemistry*. 57(11), 2138-2142.
- Hurtado, S., Garcia-Tenorio, R. and Garcia-Leon, M. (2009). Coincidence summing corrections in gamma-ray spectrometry using GEANT4 Code. *Nuclear Science, IEEE Transactions on*. 56(3), 1531-1536.
- IAEA-295. (1989). *Measurement of Radionuclides in Food and the Environment, A Guidebook*: Tech .Rept. No. 295. IAEA, Vienna, Austria.
- IAEA-TECDOC-564. (1990). Practical aspects of operating a neutron activation analysis laboratory. *Techdoc*. 564. IAEA. Vienna.
- IAEA-TECDOC-1215. (2001). Use of research reactors for neutron activation analysis. IAEA. Vienna.
- IAEA-TECDOC-1218. (2001). *Quality aspects of research reactor operations for instrumental neutron activation analysis*. IAEA. Vienna.
- IAEA. (2010). Research Reactors: Purpose and Future. International Atomic Energy Agency, Vienna.
- Jaafar, A. (1976). *Gelogy and mineral resources of the Karak and Temerloh areas, Pahang*. Geological Survey of Malaysia. Malaysia.
- Jones, L. D. (1990). *Uranium and Thorium occurrences in British Columbia*: Mineral Resources Division, Geological Survey Branch.
- Jovanović, S., Vukotić, P., Smodiš, B., Jaćimović, R., Mihaljević, N. and Stegnar, P. (1989). Epithermal neutron flux characterization of the TRIGA MARK II reactor, Ljubljana, Yugoslavia, for use in NAA. *Journal of radioanalytical and nuclear chemistry*. 129(2), 343-349.
- Kafala, S. and MacMahon, T. (1993). Neutron activation analysis without multi-element standards. *Journal of radioanalytical and nuclear chemistry*. 169(1), 187-199.

- Kafala, S. and MacMahon, T. (2007). Comparison of neutron activation analysis methods. *Journal of radioanalytical and nuclear chemistry*. 271(2), 507-516.
- Khoo, K., Sarmani, S. and Abugassa, I. (2007). Determination of thermal to epithermal neutron flux ratio (f), epithermal neutron flux shape factor (α) and comparator factor (F_c) in the Triga Mark II reactor, Malaysia. *Journal of radioanalytical and nuclear chemistry*. 271(2), 419-424.
- Khoo, K. S., Sarmani, S., Abdul Majid, A. and Leong, T. K. (2008). Assessment of neutron flux gradients in irradiation channels at the TRIGA reactor by Au-Cr-Mo monitor set based on k_0 -INAA. *Sains Malaysiana*. 37(4), 401-404.
- Khrbish, Y. and Spyrou, N. (1991). Prompt gamma-ray neutron activation analysis by the absolute method. *Journal of radioanalytical and nuclear chemistry*. 151(1), 55-61.
- Kim, N. B., Park, K. S. and Bak, H.-i. (1986). A Single Comparator Method Using Reactor Neutron and Its Errors. *Journal of the Korean Nuclear Society*. 18(2), 85-91.
- Kim, J. I., Born, H. J. (1973). Monostandard activation analysis and its applications: Analyses of Kale powder and NBS standard glass samples. *Journal of Radioanalytical Chemistry*. 13,(2) 427-442.
- Knoll, G. F. (1989). *Radiation detection and measurement*. 2nd ed. John Wiley & Sons publisher. New York.
- Knoll, G. F. (2000). *Radiation Detection and Measurements*. 3rd ed.. John Wiley & Sons, Inc. New York.
- Küppers, G. (2001). Determination of ^{232}Th by Neutron Activation Analysis using Isotope-related k_i Factors. *Radiation protection dosimetry*. 97(2), 123-125.
- Lee, S. K. (2007). *Natural background radiation in the Kinta District, Perak, Malaysia*. MSc. Thesis. Universiti Teknologi Malaysia.
- Lenihan, J. M. and Thomson, S. J. (1969). *Advances in activation analysis*. Academic Press. London and New York.
- Liew, H. F. (2010). *The absolute method of neutron activation analysis using triga neutron reactor, nuclear agency, Malaysia*. MSc Thesis. Universiti Teknologi Malaysia.
- Lin, X., Baumgärtner, F. and Li, X. (1997). The program “MULTINAA” for various standardization methods in neutron activation analysis. *Journal of radioanalytical and nuclear chemistry*. 215(2), 179-191.

- Long, K. R., Van Gosen, B. S., Foley, N. K. and Cordier, D. (2012). *The principal rare earth elements deposits of the United States: a summary of domestic deposits and a global perspective*: Springer.
- Longoria, L., and Benitez, J. (1996). Full energy peak efficiency of a Ge detector as a function of energy and distance. *Applied Radiation and Isotopes*. 47(3), 339-343.
- Lyon, Jr. W. S. (1964). Guide to activation analysis. Robert E. Krieger Publishing Company, Huntington, New York.
- Mason, B. (1952). *Principles of geochemistry*. John Wiley & Sons, Inc. New York.
- Mason, B. and Moore, C. B. (1982). *Principles of geochemistry*. 4th ed. John Wiley & Sons, Inc. New York.
- Mizera, J. and Řanda, Z. (2010). Instrumental neutron and photon activation analyses of selected geochemical reference materials. *Journal of radioanalytical and nuclear chemistry*. 284(1), 157-163.
- MNA. (2008). *Safety Analysis Report (SAR) for PUSPATI TRIGA Reactor, NUKLEARMALAYSIA/L/2008/34 (S)*: Malaysian Nuclear Agency Bangi, Malaysia.
- Montgomery, D. and Montgomery, G. (1995). A method for assessing and correcting coincidence summing effects for germanium detector efficiency calibrations. *Journal of radioanalytical and nuclear chemistry*. 193(1), 71-79.
- Morgan, J. and Lovering, J. (1963). The determination of uranium and thorium in rocks by neutron activation analysis. *Analytica Chimica Acta*. 28, 405-417.
- Oddone, M., Meloni, S. and Genova, N. (1984). Neutron activation analysis: a powerful tool for assay of rare-earth elements in terrestrial materials. *Inorganica chimica acta*. 94(6), 283-290.
- Omar, M. and Wan Hassan, F. (1999). Naturally occurring radionuclides in Malaysian granites. *Jurnal Sains Nuklear Malaysia*. 17(2), 73-77.
- Orvini, E., Speziali, M., Salvini, A. and Herborg, C. (2000). Rare earth elements determination in environmental matrices by INAA. *Microchemical journal*. 67(1), 97-104.
- Poortmans, F., Girlea, I. and Fabry, A. (1971). Neutron cross sections for ^{152}Eu below 1 eV. *Nuclear Physics A*. 172(3), 489-498.

- Ram, G., Chakraborty, K. and Aidid, S. (1980). instrumental neutron activation analysis for rare earth elements in dolerite dykes of Kuantan area, Peninsular Malaysia. *Bulletin of the Geological Society of Malaysia*. 13, 87-92.
- Rezaee, K., Saion, E., Wood, A. K. and Reza, A. M. (2009). Rare earth elements distribution in marine sediments of Malaysia coasts. *Journal of Rare Earths*. 27(6), 1066-1071.
- Rezaee, K., Saion, E., Wood, A. K. and Abdi, M. (2010). Rare earth elements determination and distribution patterns in surface marine sediments of the South China Sea by INAA, Malaysia. *Journal of radioanalytical and nuclear chemistry*. 283(3), 823-829.
- Shawky, S., Amer, H., Nada, A., Abd El-Maksoud, T. and Ibrahim, N. (2001). Characteristics of NORM in the oil industry from Eastern and Western deserts of Egypt. *Applied Radiation and Isotopes*. 55(1), 135-139.
- Simnad, M. (1980). The U-ZrHx Alloy: Its Properties and use in TRIGA Fuel. General Atomic Division. Report. Project No 4314, E-117-833.
- Simonits, A., De Corte, F. and Hoste, J. (1976). Zirconium as a multi-isotopic flux ratio monitor and a single comparator in reactor-neutron activation analysis. *Journal of Radioanalytical Chemistry*. 31(2), 467-486.
- Sood, D., Reddy, A. and Ramamoorthy, N. (2004). *Fundamentals of Radiochemistry*: IANCAS Publications.
- St-Pierre, J. and Zikovsky, L. (1982). Use of the absolute method in neutron activation analysis: application to National Bureau of Standards coal and spinach. *Canadian Journal of Chemistry*. 60(17), 2278-2280.
- Suttle Jr, A., O'Brien, B. C. and Mueller, D. W. (1969). Neutron activation analysis of uranium in geological material by measuring tellurium-132. *Analytical Chemistry*. 41(10), 1265-1269.
- Vargas, M. J., Timón, A. F., Díaz, N. C. and Sánchez, D. P. (2002). Influence of the geometrical characteristics of an HpGe detector on its efficiency. *Journal of radioanalytical and nuclear chemistry*. 253(3), 439-443.
- Verheijke, M. (1994). Relation between the Høgdahl convention and the modified Westcott formalism for (n, γ) reactions with a pure $1/v$ n cross-section behavior. *Journal of radioanalytical and nuclear chemistry*. 183(2), 293-299.
- von Hevesy, G. and Levi, H. (1936). *The action of neutrons on the rare earth elements*: Levin & Munksgaard, Ejnar Munksgaard.

- Wan Hassan, W. F., and Hamzah, S. (1999). Rare earth element patterns in some granitic rocks of Peninsular Malaysia. Paper presented at the Ninth Regional Congress on Geology, Mineral and Energy Resource of Southeast Asia-GEOSEA'98. August 17-19. Kuala Lumpur, Malaysia, *Geol.Soc. Malaysia Bull.* 513-528.
- Wee, B. S., Dung, H. M., Wood, A. K., Salim, N. A. A. and Elias, M. S. (2006). Testing the applicability of the k_0 -NAA method at the MINT's TRIGA MARK II reactor. *Nuclear Instruments and Methods in Physics Research A.* 564, 716-720.
- Williams, J. and Gilliam, D. (2011). Thermal neutron standards. *Metrologia.* 48(6), S254-S262.
- Witkowska, E., Szczepaniak, K. and Biziuk, M. (2005). Some applications of neutron activation analysis. *Journal of radioanalytical and nuclear chemistry.* 265(1), 141-150.
- Yavar, A. (2012). *Characterisation of neutron flux parameters at TRIGA Mark II Research Reactor using the k_0 - INAA and absolute methods and comparison with MCNP-4C Code.* Ph.D. Thesis. Universiti Kebangsaan Malaysia.
- Yaziz, B.Y., Puad, B.M. & Synposis, A. (1982). The Construction, Installation and Commissioning of the PUSPATI TRIGA Reactor, *Seventh European Conference of TRIGA Reactor Users.* September 1982. Istanbul, Turkey,.
- Yücel, H. and Karadag, M. (2004). Experimental determination of the α -shape factor in the $1/E^{1+\alpha}$ epithermal-isotopic neutron source-spectrum by dual monitor method. *Annals of Nuclear Energy.* 31(6), 681-695.
- Zijp, W. L. and Baard, J. H. (1979). *Nuclear data guide for reactor neutron metrology:* Netherlands Energy Research Foundation ECN.