

## CONTENTS

	<b>Page</b>
<b>Approval page</b>	ii
<b>Acknowledgements</b>	iii
<b>Abstract</b>	iv
<b>List of tables</b>	viii
<b>List of illustrations</b>	x
<b>Abbreviations and symbols</b>	xii
<b>CHAPTER 1 INTRODUCTION</b>	
1.1 Arsenic-As	1
1.1.1 General	1
1.1.2 Arsenic pollution	1
1.1.3 Toxic effects of arsenic in humans	3
1.1.4 Arsenic determination	6
1.2 Neutron Activation Analysis	8
1.2.1 NAA Method	9
1.2.2 Neutron sources	10
1.2.3 Activation with neutron	11
1.2.4 Measurement of gamma ray	17
1.2.5 Quantitative determination of activity	18
1.2.6 Instrumental and radiochemical	20
1.2.7 Sensitivity for determination of elements by NAA	21
1.3 Aim of the research	21
<b>CHAPTER 2 EXPERIMENTAL</b>	
2.1 Apparatus and Material	22
2.1.1 NAA apparatus	22
2.1.2 Chemical and material	22
2.2 Sample material	26
2.3 NA experiment	26
2.3.1 Standard and soil preparation	26

2.3.2 Preliminary study	26
2.3.2.1 Qualitative analysis	26
2.3.2.2 Quantitative analysis	27
2.3.3 Optimization conditions	27
2.3.4 Analysis validation method and flux correction	28
2.4 Analysis of soil samples	28
2.5 $\gamma$ -spectrum evaluation and calculation of results	29
<b>CHAPTER 3 RESULTS</b>	<b>30</b>
3.1 Preliminary soil analysis	32
3.1.1 Qualitative analysis	32
3.2 Quantitative analysis	36
3.2.1 Preliminary study to quantitative analysis	36
3.2.2 Optimization condition	38
3.2.3 Analysis validation method and flux correction	50
3.2.3.1 Flux correction	50
3.2.3.2 Validation method	54
3.3 Analysis of soil samples	55
<b>CHAPTER 4 CONCLUSIONS</b>	<b>57</b>
<b>CHAPTER 5 REFERENCES</b>	<b>59</b>
APPENDIX A	62
APPENDIX B	66
VITA	79

## LIST OF TABLES

Table	Page
1.1 Physical and Chemical information of arsenic	2
1.2 Arsenic toxic in different organ systems: systemic effects	5
1.3 Classification of the methods for arsenic determination	6
1.4 Examples of some techniques for arsenic determination	7
1.5 Capture cross section ( $\sigma_\gamma$ ) and resonance integral ( $I_\gamma$ )(in barns) for some typical activation targets	14
1.6 Half-life values for typical activation product	15
1.7 Estimated detection limits for INAA using decay gamma rays. Assuming irradiation in a reactor neutron flux of $1 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$	21
2.1 Irradiated and cooling times in this work for determination of arsenic	28
3.1 Determination limits for arsenic element: a comparison of NAA (assuming a thermal neutron flux of $10^{18} \text{ n m}^{-2} \text{ s}^{-1}$ ) with other analytical techniques	30
3.2 Nuclear data of $^{76}\text{As}$ radionuclide	31
3.3 Radioisotopes induced from thermal neutron activation of natural elements present in soil samples	33
3.4 Determination of arsenic concentrations in five samples from different locations where $\lambda = 4.389\text{E-}4 \text{ min}^{-1}$ .	36
3.5 Count rates and arsenic concentrations in SRM and sample soil No 5 obtained from various irradiation ( $T_i$ ) and cooling ( $T_d$ ) times	39
3.6 Arsenic average concentration in samples soil No 5 obtained with various irradiated and cooling time	42
Photopeak to Compton scattering ratio of soil samples No 5 obtained with various irradiated and cooling times	44
3.8 Counts and specific counts in a copper monitor	50
3.9 Relative count values by sample position flux correction and relative flux ratio of As/Cu; counts of Cu ratios and normalized with sample and standard position in the rabbit	51

3.10 The calculation and results of arsenic concentration in six candidate's SRM from different vial position in the rabbit	54
3.11 Arsenic concentrations in 5 candidate's sample by TNAA and ENAA compared with XRF E-T method	56
4.1 Arsenic concentrations in five candidate's sample by TNAA and ENAA	58
B1.1 Some type of detector	70
B2.1 The weights of soil sample, starch binder and $As_2O_3$ in pellets	76
B2.2 Measuring time in this experiment.	76
B3.1 Result from XRFA calculation of difference soil samples	78

## LIST OF ILLUSTRATIONS

Figure	Page
1.1 Health effect from breathing inorganic arsenic	4
1.2 Health effect from ingesting inorganic arsenic	5
1.3 The process of neutron capture by a target nucleus followed by the emission of gamma rays	9
1.4 A typical reactor neutron energy spectrum showing the various components used to describe the neutron energy regions	10
1.5 Activation curve	17
2.1 The renewed Triga Mark III nuclear research reactor (TRR-1/M1), operated by the Office of Atomic Energy for Peace (OAEP)	23
2.2 $\gamma$ Spectroscopy system manufactured by Oxford Instrument Inc., Nuclear Measurement Group, U.S.A.	24
2.3 Apparatus and material in this study	25
2.4 Scheme of Qualitative analysis of soil samples. ( <i>Nepi</i> = epithermal neutron, <i>Nth</i> = thermal neutron, <i>Ti</i> = irradiated time, <i>Td</i> = cooling time	26
2.5 The position of sample vials in rabbit ( <i>s</i> is sample vial, SRM is standard vial and $\blacksquare$ = copper wire)	29
3.1 $\gamma$ -spectra of radioisotopes induced from thermal neutron activation of natural elements present in soil samples for short-, intermediate-, and long time irradiation applied in both A4 and Ca3 core tubes	34
3.2 $\gamma$ -spectra of radioisotopes induced from thermal neutron activation of elements present in SRM 2710 (Motana soil) for short-, intermediate-, and long time irradiation applied in A4 core tube	37
3.3 Arsenic concentration in soil samples No 5 obtained with various irradiated, cooling time and position vials in the rabbit	43
3.4 $\gamma$ spectrum of soil No 1 by suitable condition of ENAA	49
3.5 $\gamma$ spectrum of soil No 1 by suitable condition of TNAA	49
3.6 Neutron flux variation along an irradiation rabbit demonstrated by the specific count in a copper monitor	51

3.7 Neutron flux variation along an irradiation tube, demonstrated by the activity induced in a cobalt monitor	53
3.8 The flux corrections by in both normalized with specific count of As / specific count of Cu ratio and normalized with sample and standard position in rabbit	53
3.9 Flux correction of SRM 2710 by sample position flux correction method	55
B.1.1 The production of characteristic X-ray	66
B.1.2 The selection rule	67
B.1.3 Decay scheme of the principle transition in $^{238}\text{Pu}$	68
B.1.4 X-ray spectrum from $^{238}\text{Pu}$ source measured with Si(Li) detector	68
B.1.5 X-ray composition	69
B.1.6 Principle of Emission-Transmission (E-T) method	72
B. 2.1 XRF apparatus system	74
B.2.2 Block diagram of detector system	74
B.3.1 X-ray fluorescence spectrum of soil sample No1	77

## ABBREVIATIONS AND SYMBOLS

AAS	atomic absorption spectrometry
AFS	atomic fluorescence spectrometry
DGNAA	delayed gamma-ray neutron activation analysis
EDXRF	energy dispersive X-ray fluorescence
ENAA	epithermal neutron activation analysis
E-T	emission transmission GFAAS
FWHM	fullwidth at half maximum
FNAA	fast gamma-ray neutron activation analysis
HG	hydride-generation
HGAAS	hydride-generation atomic absorption spectrometry
HPGe	hyperpure (or intrinsic) germanium detector
HPLC	high performance liquid chromatography
ICP-MS	inductively coupled plasma-mass spectrometry
ICP-AES	inductively coupled plasma- atomic emission spectrometry
INAA	instrumental neutron activation analysis
LOD	limit of detection
MCA	multi-channel analyzer
NIST	the Nation Institute of Standard and Technology
NAA	neutron activation analysis
PGNAA	prompt gamma-ray neutron activation analysis
SRM	standard reference material
TNAA	thermal neutron activation analysis
RSD	relative standard deviation
XRF	X-ray fluorescence
$A$	atomic weight
$A_o$	activity at the end of the irradiation time
$A_s$	the saturation activity
$A_{sam}$	activity of sample
$A_{bare}$	activity in bare tube

$A_{\text{epi}}$	activity under 1 mm of cadmium
$A_{\text{std}}$	activity of standard
$C_{\text{sam}}$	concentration of element in sample
$C_{\text{std}}$	concentration of element in standard
Ci	Curi
$N$	number of nuclei
$N_A$	Avogadro's number
(n, $\gamma$ )	(neutron, gamma) reaction
(n,p)	(neutron, proton) reaction
(n, $\alpha$ )	(neutron, alpha) reaction
nd	not detected
ppm	part per million
ppb	part per billion
$P$	branching ratio
<i>sam</i>	sample
<i>std</i>	standard
$t_c$	counting time
$t_d$	decay time
$t_m$	measuring time
$T, t$	irradiation time
$W_{\text{sam}}$	weight of sample
$W_{\text{std}}$	weight of standard
$W$	mass of element
$\epsilon$	molar absorptivity
$\phi$	neutron flux
$\phi_{\text{th}}$	thermal neutron flux
$\phi_{\text{epi}}$	epithermal neutron flux
$\theta$	isotropic abundance
$\sigma$	cross section
$\sigma_{\text{th}}$	thermal cross section
$\sigma_{\gamma}$	capture cross section



$\lambda$	decay constant
$I_{\gamma}$	capture resonance integral

## For appendix B

$\chi$	total mass absorption coefficient
$\mu_i$	individual mass absorption coefficients
$w_i$	weight fraction of interest element $i$ ,
$n$	number of components.
$\phi_1$	angle between excitation radiation from source and sample.
$\phi_2$	angle between characteristic X-ray of element $i$ in sample and detector
$E_0$	incident radiation energy.
$E_i$	energy of characteristic X-ray of element $i$ .
$I_I$	characteristic X-ray of element $i$ from sample
$I_i'$	characteristic X-ray of element $i$ from target.
$I_i^{s+}$	characteristic X-ray of element $i$ from sample and target
$\chi_i$	mass absorption coefficient of X-ray measurement of element $i$
$\rho_m D$	thickness of sample
$F_i$	absorption correction factor
$S_i$	sensitivity of element $i$