

**DESIGN OF A PROTOTYPE APPARATUS FOR ON-LINE
ELEMENTAL ANALYSIS USING NUCLEAR TECHNIQUES**



Ph.D. THESIS

Başak UNTUÇ

Physics Engineering Department

Physics Engineering Program

FEBRUARY 2018

**DESIGN OF A PROTOTYPE APPARATUS FOR ON-LINE
ELEMENTAL ANALYSIS USING NUCLEAR TECHNIQUES**

Ph.D. THESIS

**Başak UNTUÇ
(509082107)**

Physics Engineering Department

Physics Engineering Program

**Thesis Advisor: Prof. Dr. İskender A. REYHANCAN
Eş Danışman: Prof. Dr. M. Nizamettin ERDURAN**

FEBRUARY 2018

**NÜKLEER TEKNİKLER KULLANARAK ÇEVİRİM-İÇİ
ELEMENT ANALİZİ YAPAN PROTOTİP CİHAZIN TASARIMI**

DOKTORA TEZİ

**Başak UNTUÇ
(509082107)**

Fizik Mühendisliği Anabilim Dalı

Fizik Mühendisliği Programı

**Tez Danışmanı: Prof. Dr. İskender A. REYHANCAN
Eş Danışman: Prof. Dr. M. Nizamettin ERDURAN**

ŞUBAT 2018

Başak UNTUÇ, a Ph.D. student of ITU Graduate School of ScienceEngineering and Technology 509082107 successfully defended the thesis entitled “DESIGN OF A PRO-TOTYPE APPARATUS FOR ON-LINE ELEMENTAL ANALYSIS USING NUCLEAR TECHNIQUES”, which he/she prepared after fulfilling the requirements specified in the associated legislations, before the jury whose signatures are below.

Thesis Advisor : **Prof. Dr. İskender A. REYHANCAN**
Istanbul Technical University

Co-advisor : **Prof. Dr. M. Nizamettin ERDURAN**
Sabahattin Zaim University

Jury Members : **Prof. Dr. Cenap ÖZBEN**
Istanbul Technical University

Doç. Dr. Ayşe DURUSOY
Yıldız Technical University

Yrd. Dr. Zuhale ER
Istanbul Technical University

Doç. Dr. Baki ALTUNCEVAHİR
Istanbul Technical University

Doç. Dr. Erol KAM
Yıldız Technical University

Date of Submission : **05 January 2018**
Date of Defense : **02 February 2018**





To my family...



FOREWORD

During my PhD, I would like to express my gratitude and thanks to my supervisor Prof. Dr. İskender Atilla Reyhancan, for providing me motivation, sharing his scientific knowledge and experiences, and giving me opportunity to work in TUBITAK Project.

I would like to thank my co-supervisor Prof. Dr. M. Nizamettin Erduran who helps me for the project with his invaluable experiences, endless understanding and guides through the study.

I wish to acknowledge TUBITAK (The Scientific and Technological Research Council of Turkey) for their financial support. (Project Number: 113M034).

I wish to acknowledge I.T.U Scientific Research Projects Unit for their financial support (Grant Number: 38201).

I would like to thank to Dr. Mehmet Türkmen for his valuable helps in MCNP5 software.

I would like to thank to Prof. Dr. Altuğ Şişman for giving me opportunity to work in Energy Institute at I.T.U.

I would like to thank to Dr. Muhittin Okka for his moral support and presenting a safety working environment.

I would like to thank to Dr. Murat Sertkol for sharing his experiences throughout this study.

I would like to thank to Filiz Öztekin for her unique friendship and moral support during my Ph.D education.

I am also grateful to my family for their support through my all life. Without their encouragement, it is impossible to complete the duration of my PhD.

February 2018

Başak UNTUÇ
(M.Sc. Physics Engineer)

TABLE OF CONTENTS

	<u>Page</u>
FOREWORD.....	ix
TABLE OF CONTENTS.....	xi
ABBREVIATIONS	xiii
SYMBOLS.....	xv
LIST OF TABLES	xvii
LIST OF FIGURES	xix
SUMMARY	xxi
ÖZET	xxiii
1. INTRODUCTION	1
1.1 Purpose of Thesis	2
1.2 Literature Review	3
1.3 Hypothesis	5
2. THEORY	9
2.1 Neutron Transport.....	9
2.1.1 Neutron cross section	11
2.1.2 Neutron shielding	13
2.1.3 Neutron generators	14
2.1.3.1 Structure of neutron generators	15
2.1.3.2 Pulse neutron sources and system components	16
2.1.3.3 The method of neutron generators and their applications	17
2.2 Photon transport.....	20
2.2.1 Photoelectric effect.....	20
2.2.2 Pair production	21
2.2.3 Compton scattering.....	22
2.2.4 Photonuclear interactions	24
2.2.5 Rayleigh (Coherent) scattering.....	24
2.2.6 Photon cross-sections	24
2.2.7 Gamma-ray detectors	27
2.2.7.1 Gas-filled detectors.....	28
2.2.7.2 Solid-state detectors.....	28
2.2.7.3 Scintillation detectors	30
2.2.8 Detection System.....	32
2.2.8.1 High-voltage power (bias) supply.....	33
2.2.8.2 Preamplifier.....	33
2.2.8.3 Amplifier.....	34
2.2.8.4 Multi-channel analyzer (MCA)	34
2.3 Structure of Coal.....	35

2.3.1 The methods of analysis for coal contents	36
2.3.1.1 Non- Destructive Analyzing of Coal Contents	40
2.3.1.2 The MCLLS Method for Element Analysis	44
3. EXPERIMENTAL SET-UP AND TEST STUDIES	47
3.1 BGO Test Studies	47
3.1.1 Energy calibration.....	48
3.1.2 Pulse-shape calibration.....	50
3.1.3 Linearity	51
3.1.4 Energy resolution.....	55
3.1.5 Response functions of BGO	57
3.1.6 Efficiency calibration.....	58
3.1.6.1 Intrinsic Efficiency Measurement of BGO.....	60
3.2 Coal Spectrum Obtained By Pu-Be Source.....	66
3.3 PFTNA Calibration Process	68
4. MONTE CARLO SIMULATION METHODS	77
4.1 The Structure of MCNP5.....	78
4.2 Design of prototype apparatus.....	81
4.2.1 MCNP5 simulation of prototype apparatus.....	82
4.2.2 Generating of MCNP5 element library	89
4.3 The Structure of GEANT4	94
4.3.1 Geometry construction	95
4.3.2 Particle Description	96
4.3.3 Physics processes.....	96
4.3.4 Tracking and particle transportation.....	97
4.3.5 Stack management.....	97
4.3.6 Scoring techniques.....	97
4.3.7 User-interface and visulation.....	98
4.4 GEANT4 Simulation of Prototype Apparatus	98
4.4.1 Generating of GEANT4 element library	101
5. RESULTS	107
5.1 Manufactured Prototype Apparatus.....	107
5.1.1 The MCLLS method for the MCNP5 simulated coal samples.....	109
5.1.2 The MCLLS method for the experimental spectra.....	111
6. CONCLUSION	117
REFERENCES.....	121
APPENDICES	129
APPENDIX A	130
APPENDIX B.....	134
CURRICULUM VITAE.....	136

ABBREVIATIONS

NDA	: Non-Destructive Analysis
BGO	: Bismuth Germanium Oxide
MCNP5	: Monte Carlo N-Particle 5
GEANT4	: GEometry And Tracking 4
MCLLS	: Monte Carlo Library Least Square
MCNP5 LLS	: Monte Carlo N-Particle 5 Library Least Square
GEANT4 LLS	: GEometry And Tracking 4 Library Least Square
NAA	: Neutron Activation Analysis
BNL	: Brookhaven National Laboratory
PGNAA	: Prompt Gamma Neutron Activation Analysis
DGNAA	: Delayed Gamma Neutron Activation Analysis
CSIRO	: Commonwealth Scientific and Industrial Research Organisation
R&D	: Research and Development
DUET	: Dual Energy Gamma-ray Transmission
PP	: Pair Production
NISTA	: Neutron Inelastic Scattering and Thermal-capture Analysis
NIS	: Neutron Inelastic Scattering
TNC	: Thermal Neutron Capture
LLS	: Library Least Square
PFNA	: Pulsed Fast Neutron Analysis
PFNTS	: Pulsed Fast Neutron Transmission Spectroscopy
TOF	: Time Of Flight
API	: Associated Particle Imaging
DAQ	: Data Acquisition
EURITRACK	: EURopean Illicit TRAfficking
UNCOSS	: Underwater Coastal Sea Surveyor
PFTNA	: Pulsed Fast Thermal Neutron Analysis
XRF	: X-Ray Fluorescence
PMT	: Photo-multiplier Tube
HVPS	: High Voltage Power Supply
NIM	: Nuclear Instrumentation Module
AC	: Alternate Current
DC	: Direct Current
MCA	: Multi-Channel Analyzer
PHA	: Pulse Height Analyzer
ADC	: Analog to Digital Converter
NISTA	: Nuclear Inelastic and Thermal Capture Analysis
SPA	: Single Peak Analysis



SYMBOLS

σ	: Cross-section
Σ	: Macroscopic cross-section
Σ_R	: Removal cross-section
μ	: Total attenuation
γ	: Gamma-ray
n	: Neutron
p	: Proton
α	: Alpha particle
β^-	: Beta-minus particle
β^+	: Beta-plus particle
A	: Atomic mass number
b	: Barn
Ci	: Curie
Bq	: Becquerel
N_A	: Avogadro number
h	: Planck constant
c	: Speed of light
D	: Deuterium
T	: Tritium
$D-D$	: Deuterium-Deuterium
$T-T$	: Tritium-Tritium
$D-T$	: Deuterium-Tritium
Q	: Amount of released energy for the reaction
x	: Thickness of the target material
I_0	: Number of incident neutrons
I	: Number of transmitted neutrons
$\dot{D}$	: Dose rate of fast neutrons with shielding material
$\dot{D}_0$	: Dose rate of fast neutrons without shielding material
B	: Buildup factor
ns	: Nano second
μs	: Micro second
eV	: Electron Volt
MeV	: Mega Electron Volt
keV	: Kilo electron volt
MJ	: Mega joules
Z	: Atomic number
E_γ	: Gamma-ray energy
E_k	: Kinetic energy
E_n	: Neutron energy
E_b	: Binding energy
n/s	: Neutron number per second



LIST OF TABLES

	<u>Page</u>
Table 1.1 : The comparison of the experimental precision of the nuclear techniques [19], [20], [17].	4
Table 2.1 : The shielding calculations of fast neutrons [25].	14
Table 2.2 : Properties of Scintillation Detectors.	32
Table 2.3 : The HVPS values for different type of detectors [74].	33
Table 2.4 : The classification of coal ranks.	36
Table 2.5 : Gamma energies used in NAA of some coal elements [83].	42
Table 3.1 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV.	49
Table 3.2 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV.	53
Table 3.3 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV and experimental peaks of H (2.22 MeV), C (4.43 MeV) and O (6.13 MeV).	54
Table 3.4 : The response functions of BGO detector by using Cs-137, Co-60, Eu-152 sources and C (4.43 MeV).	58
Table 3.5 : The features of ^{137}Cs , ^{60}Co , and ^{152}Eu sources.	61
Table 3.6 : The total errors and uncertainties of efficiency measurements at distances between 2 cm and 10 cm.	64
Table 3.7 : The total errors and uncertainties of efficiency measurements at distances between 15 cm and 25 cm.	65
Table 3.8 : The total errors and uncertainties of efficiency measurements at distances between 28 cm and 35 cm.	66
Table 3.9 : The comparison of alpha- neutron sources.	66
Table 3.10 : The weight percentages of compounds in synthetic coal sample (%).	71
Table 3.11 : Amount of carbon element and oxide compounds inside of the sets (g).	71
Table 3.12 : The study on the lignite coal, collected from different regions of Turkey [99].	72
Table 3.13 : The dose rate of neutron and gamma accumulated on the concrete walls.	75
Table 4.1 : The tally cards and their specifications [98].	80
Table 4.2 : The cross sections of gamma ray energies induced by thermal and fast neutrons for interested elements of coal.	86
Table 4.3 : The concentration of the compounds in each coal set.	88
Table 4.4 : Effective dose rate of neutrons on polyethylene and lead surfaces.	89
Table 5.1 : The regression variables of compounds and regression coefficients of each coal set in MLR.	110

Table 5.2	: The comparison of the concentration of elemental contents between prepared and calculated values for the each coal set.	111
Table 5.3	: The element contents of analyzed coal samples.....	112
Table 5.4	: The calculation results of element contents of coal samples by MCNP5 LLS method.....	113
Table 5.5	: The calculation results of element contents of coal samples by GEANT4 LLS method.....	113
Table 5.6	: The deviation values of C, H, O, N, S ash and moisture obtained by MCNP5 LLS and GEANT4 LLS respectively.....	114
Table 1	: The data used for efficiency calibration at distance of 2 cm.	130
Table 2	: The data used for efficiency calibration at distance of 4 cm.	130
Table 3	: The data used for efficiency calibration at distance of 6 cm.	130
Table 4	: The data used for efficiency calibration at distance of 10 cm.	131
Table 5	: The data used for efficiency calibration at distance of 15 cm.	131
Table 6	: The data used for efficiency calibration at distance of 20 cm.	131
Table 7	: The data used for efficiency calibration at distance of 25 cm.	132
Table 8	: The data used for efficiency calibration at distance of 28 cm.	132
Table 9	: The data used for efficiency calibration at distance of 30 cm.	132
Table 10	: The data used for efficiency calibration at distance of 35 cm.	133

LIST OF FIGURES

	<u>Page</u>
Figure 1.1 : The sources of electricity production in (a) world [1] (b) Turkey [2].	1
Figure 2.1 : Neutron cross-section of some elements depending on their energies [31].	12
Figure 2.2 : The cross-sections of C and H elements according to their interactions types [32].	12
Figure 2.3 : The structure of a neutron generator [49].	15
Figure 2.4 : The diagram of API technique [48].	19
Figure 2.5 : The diagram of PFTNA pulses [48].	19
Figure 2.6 : The dominant region of photon interactions [62].	20
Figure 2.7 : Representantative view of Compton scattering [63].	23
Figure 2.8 : Cross section of Cu for photons between 10 keV and 100 GeV [64].	26
Figure 2.9 : The comparison of photon interaction cross sections for low and high atomic number material for (a) carbon and (b) tungsten [65].	27
Figure 2.10 : Illustration of a gas-filled detector [66].	28
Figure 2.11 : Typical arrangement of components in a solid-state detector [69].	29
Figure 2.12 : Illustration of various solid-state detector crystal configurations: (a) open-ended cylindrical or true coaxial, (b) closed-ended cylindrical, and (c) planar [67].	30
Figure 2.13 : Simplified picture of a scintillation detector [70].	31
Figure 2.14 : Block diagram of a detection system.	32
Figure 2.15 : The diagram of charge collection [73].	34
Figure 2.16 : The diagram of pulse height spectrum [73].	35
Figure 2.17 : The proximate analyzer [77].	37
Figure 2.18 : The ultimate analyzer [78].	39
Figure 2.19 : PGNAA and DGNAA processes via Co irradiation.	41
Figure 2.20 : Thermo Gamma and Sabia Installatios in U.S.A [22].	42
Figure 3.1 : BGO detector of 3"x3" crystal size used in prototype apparatus [84].	47
Figure 3.2 : BGO detector of 3"x3" crystal size used in prototype apparatus.	48
Figure 3.3 : Pulse-shape calibration of BGO detector between 0-3000 keV.....	50
Figure 3.4 : Resolution calibration of BGO detector between 0-3000 keV.	50
Figure 3.5 : Different MCA calibration plots for three values of amplifier gain [26].	51
Figure 3.6 : The comparative illustration of integral and differential linearities of MCA [26].	52
Figure 3.7 : The relation between energy and channel number up to about 6.5 MeV.....	53
Figure 3.8 : The relation between Δ_E and gamma energy up to about 3 MeV.....	54
Figure 3.9 : The relation between Δ_E and gamma energy up to about 6.5 MeV...	55

Figure 3.10:	The determination of FWHM for a scintillation detector.	55
Figure 3.11:	Energy resolution of BGO detector between 0-3000 keV.	57
Figure 3.12:	Response functions of BGO detector between 0 and 4.5 MeV.	58
Figure 3.13:	The relation between efficiency and energy of BGO detector at 2 cm, 4 cm, 6 cm, and 10 cm distances.	62
Figure 3.14:	The relation between efficiency and energy of BGO detector at 15 cm, 20 cm, 25 cm, and 28 cm distances.	63
Figure 3.15:	The relation between efficiency and energy of BGO detector at 30 cm and 35 cm distances.	63
Figure 3.16:	The experimental set-up of Pu-Be neutron.	67
Figure 3.17:	Gamma-ray spectrum of the coal sample induced by Pu-Be neutron source.	67
Figure 3.18:	The neutron generator used for prototype apparatus.	69
Figure 3.19:	The interface of MP320 neutron generator by Thermo Fisher Scientific.	69
Figure 3.20:	Experimental set-up used for the measurement and the neutron generator.	70
Figure 3.21:	NIM electronics and data acquisition system with the industrial PC.	71
Figure 3.22:	Synthetic coal samples prepared and used for calibration and test purposes.	73
Figure 3.23:	The total gamma-ray spectrum of synthetic coal set-1.	74
Figure 3.24:	The comparison spectrum of synthetic coal set-1 induced by fast and thermal neutrons.	74
Figure 3.25:	The comparison spectrum of 8 synthetic coal sets induced by fast and thermal neutrons.	75
Figure 4.1 :	Random history of a neutron incident on a slab of material [87].	77
Figure 4.2 :	Schematic view of the prototype apparatus.	82
Figure 4.3 :	Front view of the prototype apparatus geometry obtained by MCNP5.	83
Figure 4.4 :	Top view of the prototype apparatus geometry obtained by MCNP5.	84
Figure 4.5 :	The gamma-ray histogram of the Coal Set-1 obtained by MCNP5. .	85
Figure 4.6 :	The gamma-ray spectrum of the Coal Set-1 obtained by MCNP5.	86
Figure 4.7 :	The comparison of back-scattering and transmission gamma rays spectra of the Coal Set-1 obtained by MCNP5.	87
Figure 4.8 :	The comparison of gamma-ray spectra of five synthetic coal sets obtained by MCNP5.	88
Figure 4.9 :	The spherical fixed-geometry for elemental library spectra.	89
Figure 4.10:	The gamma-ray histograms and spectra of C and CaO.	90
Figure 4.11:	The gamma-ray histograms and spectra of Al ₂ O ₃ and Fe ₂ O ₃	91
Figure 4.12:	The gamma-ray histograms and spectra of MgO and SiO ₂	92
Figure 4.13:	The gamma-ray histograms and spectra of S and N.	93
Figure 4.14:	The schematic view of GEANT4 classes [100].	94
Figure 4.15:	The illustration of the prototype apparatus obtained by GEANT4. ..	99
Figure 4.16:	The illustration of production of neutrons obtained by GEANT4.	100
Figure 4.17:	The gamma-ray histogram of coal sample obtained by GEANT4.	100
Figure 4.18:	The gamma-ray spectrum of coal sample obtained by GEANT4.	101

Figure 4.19:	The gamma-ray histograms and spectra of C and CaO.	102
Figure 4.20:	The gamma-ray histograms and spectra of Al ₂ O ₃ and Fe ₂ O ₃	103
Figure 4.21:	The gamma-ray histogram and spectra of MgO and SiO ₂	104
Figure 4.22:	The gamma-ray histograms and spectra of S and N.	105
Figure 5.1 :	The illustration of manufactured prototype apparatus with 6m conveyor belt.....	108
Figure 5.2 :	The comparison of gamma-ray spectra of coal sample obtained by MCNP5, GEANT4 and experimental.	108
Figure 5.3 :	Experimental gamma-ray spectrum of coal sample obtained by manufactured prototype apparatus.	109
Figure 5.4 :	Comparison the gamma-ray spectrum of Coal Set-1 and MLR method obtained by MCNP5.	110
Figure 5.5 :	The MCNP5 LLS fit for gamma-ray spectrum of coal Set-1 obtained by MCNP5.	111
Figure 5.6 :	The fitting gamma-ray spectrum on Coal Set-1 by using MCNP5 LLS.	112
Figure 5.7 :	The fitting gamma-ray spectrum on Coal Set-1 by using GEANT4 LLS.	113
Figure 5.8 :	The comparison of laboratory results of C and ash contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.	114
Figure 5.9 :	The comparison of laboratory results of S and N contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.	115
Figure 5.10:	The comparison of laboratory results of moisture contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.	115
Figure 5.11:	The comparison of laboratory results of H and O contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.	116
Figure 1 :	Class diagram of MCNP5 Simulation For Prototype Design.....	134
Figure 2 :	Class diagram of GEANT4 Simulation For Prototype Design.....	134



DESIGN OF A PROTOTYPE APPARATUS FOR ON-LINE ELEMENTAL ANALYSIS USING NUCLEAR TECHNIQUES

SUMMARY

Coal has a significant role as an energy source of the future in electricity generation worldwide. In Turkey, coal is essential as an energy source, since coal-fired power plants currently fuel about 31% of global electricity. For our country, the processes of improving the quality and preparation of the coal is important to produce the energy efficiently from the coal power plant. For that purpose, an instrument using some nuclear techniques was developed to be able to determine the content of the ash, moisture and calorific value of the coal moving on the conveyor belt as on-line.

Due to the fact that being non-destructive and sensitive for some elements, neutron activation analysis (NAA) is an important technique for identifying the specific elements in materials. The technique of most devices is based on using a radioisotope such as ^{252}Cf . The irradiation of samples depend on neutron interactions which are neutron inelastic scattering ($n, n'\gamma$), and the neutron radiative capture (n, γ). Because of the average neutron energy of ^{252}Cf is about 2.5 MeV, this neutron source is convenient to analyze the samples including only the process of thermal neutron capture (n, γ). The carbon content of coal which is important parameter to determine the quality of the coal, can not be detected by using ^{252}Cf source. Therefore, a neutron generator is used to produce fast neutrons of 14 MeV releasing from deuterium – tritium (D-T) fusion reactions for the prototype device.

The prototype device is composed of a neutron generator, a scintillation detector, an iron block for shadow shielding and lead shield of BGO detector. The neutron generator is a device which can be operated with pulsed mode. Therefore, the obtained gamma rays can be classified according to their interactions induced with thermal neutrons or fast neutrons. The choice of a detector is pretty significant issue for designing the prototype device. The scintillation type of detector is preferred for the reason that has high efficiency and more endurance to neutron radiation damage. As a scintillation detector, Bismuth Germinate Oxide (BGO) is chosen. The characteristic features of the BGO is studied in detailed. The energy resolution, efficiency, shape calibration are calculated by using radioactive point sources such as ^{60}Co , ^{137}Cs , ^{152}Eu . Additionally, The response functions of BGO detector are experimentally found to apply on the spectrum obtained from simulation programs.

A prototype apparatus can be modelled to provide nuclear techniques by using Monte Carlo simulation programs. Monte Carlo N-Particle 5 (MCNP5) and Geometry And Tracking 4 (GEANT4) simulation programs are used to design the geometry and physical interactions of the prototype apparatus. The synthetic coal samples including different element contents are simulated and analyzed.

There are plenty of ways to analyze a bulk system including gamma spectrum. Some of them are peak area determination, neural network algorithm and Monte Carlo

Library Least Square (MCLLS) fitting. Analyzing the obtained spectra from Pulsed Fast and Thermal Neutron Analysis (PFTNA) experiment is very difficult due to its non-linear nature. Library Least Square is a common mathematical approach for treating non-linear radiation effects in cases such as PFTNA. In this study, MCLLS technique is applied by using MCNP5 and GEANT4 simulation programs. With the aid of fixed spherical geometry, the elements inside of the coal sample are simulated individually.

The obtained spectrum from MCNP5 and GEANT4 are used to build up an element library of coal sample. The method is applied on experimental spectrum which have analysis reports including element contents. So the experimental spectrum are provided to calibrate according to element library. Then, the calibrated element library is used to calculate the contents of unknown spectrum obtained experimentally. By using this mathematical approach, the element contents of experimental coal spectrum are aimed to calculate in the range of measurement precision defined commercially before.



NÜKLEER TEKNİKLER KULLANARAK ÇEVİRİM-İÇİ ELEMENT ANALİZİ YAPAN PROTOTİP CİHAZIN TASARIMI

ÖZET

Dünya genelinde, enerji üretiminin yaklaşık %25 i kömürden sağlanmaktadır. Türkiye de ise bu oran %31 i bulmaktadır. Enerji üretiminde bu kadar büyük pay sahibi olan kömürün içeriğinin tespiti önem arz etmektedir. Kömür, organik ve inorganik içeriğe sahip, serbest karbon, mineraller (kül yapıcı maddeler), uçucular, kükürt ve içerisinde nem barındıran enerji üretiminde kullanılan bir hammaddedir. Yanma aşamasında kömür bir dizi dönüşüme uğrayarak çeşitli ürünlere dönüşür. Kükürt yanarak kömürden ilk ayrılan bileşendir. Nem yüksek sıcaklıkta ısı alarak kömür bünyesinden ayrılır. Kükürt yanarak baca gazı salınımına katkı yapar ve atmosferik kirlenmeye yol açabilir. Gerek maden ocaklarında gerekse termik santrallarda kömürün işlenmesi sürecinde kömürün daha verimli yanması ondan elde edilecek enerjiyi etkileyen en önemli faktördür. Bu sebeple, kömürün kalitesini belirleyen ana kriterlerden olan kalorifik değer, kül, nem ve kükürt değerlerinin önceden bilinmesi gerekmektedir.

Türkiye'deki maden sahalarından çıkarılan kömür çeşidi linyit olup, yanma kalitesi açısından düşük aynı zamanda içerisinde bulunan yüksek miktardaki kükürt sebebiyle çevreyi olumsuz etkilemektedir. Buna ilaveten kömür işletmesinin kurulu olduğu maden sahasından çıkarılan kömür de nem, kül ve kalori değerleri açısından farklı değerlere sahip olabilir. Bu değerler önceden tespit edilerek; düşük karbon ve yüksek kül değerlerine sahip olan yığının, yüksek karbon ve düşük kül değerlerine sahip olan yığınla karıştırılmasıyla yanma verimi artırılabilir.

Kömürün içeriğinin tespitinde çeşitli yöntemler kullanılmaktadır. Bunlar kimyasal ve nükleer analiz teknikleri olarak sınıflandırılabilir. Laboratuvar ölçeğinde yapılan kimyasal analizlerin gerçekleştirilmesi, hem zaman alması hem de analizi yapılan numuneye zarar verdiğinden günümüzde işlevselliğini yitirmiştir. Ticari olarak en çok kullanılan teknik, ^{252}Cf gibi izotropik nötron kaynağının konveyör bant veya oluk üzerindeki numunenin nötronla ışınlanmasına dayanmaktadır. Bu methodla, gamma (γ) ışınlarıyla sonuçlanan elastik olmayan nötron saçılması içeren ($n, n'\gamma$) ve ($n, p\gamma$) yöntemi ile radyoaktif nötron yakalama (n, γ) yöntemi, içeren nükleer-nötron etkileşimlerinden faydalanılır.

^{252}Cf gibi radyoaktif kaynak yöntemleri ortalama 2.5 MeV civarında nötron üretirler. Bu sebeple, bu sistemler yoğunlukla yavaş nötronlar üzerinden gerçekleşen, nötron yakalama (n, γ) işlemlerine dayanır. Özellikle kömür analiz cihazlarında, element analizinde önemli rol oynayan elementlerden birisi karbondur. ^{252}Cf kaynak kullanarak karbon elementinin tespiti, C (n, γ) reaksiyonundaki gama ışınının (4935.301 keV) ölçülmesiyle gerçekleştirilir. Bu reaksiyon kanalı oldukça zayıf olup dahası Si (4933.889 keV) gibi başka elementlerle gamma enerjileri üstüste binmiştir. Öte yandan izotropik nötron kaynakları daha düşük enerjide ve yoğunlukta nötron yayımlar. Karbon elementinin en ideal tespiti, tesir kesit değerinin en yüksek olduğu 4.439 MeV

ile mümkündür. Ancak karbonun bu gamma enerjisindeki tespiti hızlı nötronların malzemeye etkileşimi sonucu açığa çıkmaktadır. Bu yüzden reaksiyon eşik enerjileri yüksek olan C, N ve O elementlerinin anında belirlenebilmesi, bu tür kaynaklarla pek olanaklı değildir. Bu sorunu aşmak için, 14 MeV (hızlı) nötronların üretildiği döteryum – trityum (D-T) füzyon nötron üreteçleri kullanılır. Bu cihazlar izotropik nötron kaynaklarına göre çok pahalı olmasına ramen, istenildiğinde daha yüksek enerjide nötron akısı sağlar. Dahası, aynı nötron kaynak gücü için, izotropik kaynağın maliyeti, darbeli nötron üreticiyle kıyaslanabilir. Üstelik, izotropik nötron kaynağın sürekli radyasyon yayması sebebiyle, radyasyon güvenlik mevzusu da oldukça kritiktir.

Bu çalışmada, kömür işletme sahasındaki konveyör üzerinde hareket kömürün içindeki kalori (kcal/kg), nem (%), kül (%) ve kükürt (%) miktarının nükleer teknikler kullanılarak belirleyecek bir prototip cihazın tasarlanıp, modellenmesi amaçlanmıştır. Bu amaçla yığmsal malzeme analizinde kullanılan “Nötron nelastik Saçılma” ve “Termal Nötron Kapma” tekniklerinin beraberce kullanıldığı "Nötron nelastik Saçılma - Termal Nötron Kapma" (NISTA) nükleer tekniğinden kullanılmıştır.

Prototip cihaz, darbeli modda çalışabilen nötron üretici, gama ışını dedektörü, dedektörü yüksek enerjili nötronlardan koruyacak gölge zırhlaması olarak kullanılan demir blok ve radyasyon zırhlaması özelliğine sahip kurşun bloklardan oluşmaktadır. Darbeli olarak çalışabilen nötron üretici, elektronik olup döteron-trityum reaksiyonu sonucunda ortaya çıkan 14 MeV enerjili nötronlar üretmektedir. Bu çalışmada nötronların indüklemesi sonucunda kömür içindeki elementlerin kendine has karakteristik gamma ışınları bir sintilasyon tipi gama ışını dedektörü ile sayılmaktadır. Sintilasyon dedektörleri arasında nispeten daha düşük çözünürlüğe sahip olsa da, yüksek verimliliği sebebiyle BGO sintilasyon dedektörü tercih edilmiştir.

Prototip cihazın imalatından önce çeşitli performans ve test çalışmaları yapılmıştır. BGO dedektörü üzerinde enerji, şekil, lineerlik, çözünürlük ve verim kalibrasyonu çalışmaları yapılmıştır. Türkiye'nin Ege Bölgesindeki linyit kömürünün içerdiği nem, karbon, kükürt ve küle sebep olan mineraller üzerinde yapılmış çalışmalardan yola çıkarak sentetik kömür numune setleri hazırlanmıştır. Bu numune setleri kullanarak, nötron üreticinin darbeli modda çalışması test edilmiştir. Nötron üretici açık konumdayken, hızlı nötronların malzemeye etkileşmesi sonucu elde edilen gama spektrumları; nötron üretici kapalı konumdayken yavaş nötronların malzemeye etkileşmesi sonucu elde edilen gama spektrumları elde edilmiştir. Hazırlanan 2 kg'lık kömür numunelerine ait toplam gama spektrumları arasında dikkate değer bir fark elde edilememiştir. Bu spektrum sonuçlarına, deney düzeneğini çevreleyen beton duvarların sebep olduğu yüksek miktarda çevresel radyasyonun sebep olduğu düşünülmüş ve ilerleyen aşamalarda daha yüksek miktarlarda kömür numunesiyle çalışma yoluna gidilmiştir.

Prototip cihazın modellenmesinde MCNP5 ve GEANT4 olmak üzere iki ayrı simülasyon programı kullanılmıştır. Simülasyon kapsamında prototip cihazı oluşturan parçalar; gerçek ölçüsüne, geometrisine ve konumlanmasına uygun olacak şekilde oluşturulmuştur. Ege Bölgesindeki linyit kömürünün içerdiği nem, karbon, kükürt ve küle sebep olan minerallerin oranları dikkate alınarak, simülasyon programında kullanılan kömür malzemesi oluşturulmuştur. Bu kapsamda kömür malzemesi olarak, nem miktarı için H_2O ; kül için SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , MgO bileşikleri; hava kirliliğine sebep olan kükürt ve enerjinin kalorifik değerinin elde edileceği karbon elementinin kütlece yüzdeliği uygun aralıkta girilerek, simülasyonda kullanılan

sentetik kömür örnek setleri oluşturulmuştur. Elde edilen simulasyon kömürlerin gama spektrumları, imalatı tamamlanan prototip cihazın kullanılması sonucu elde edilen gerçek kömür numunesine ait gama spektrumlarıyla karşılaştırılmıştır.

Prototip cihazın lineer olmayan yapısından dolayı, deneysel olarak elde edilmiş darbeli modda Hızlı ve Yavaş Nötron Aktivasyon Analizi (PFTNAA) spektrumun analizi oldukça zordur. PFTNAA gibi deneysel analiz yöntemlerinin kullanıldığı durumlarda, spektrumlarda gözlemlenen lineer olmayan radyasyon etkileri düzelten en yaygın matematiksel yaklaşım “En Küçük Kareler Kütüphanesi” (LLS) metodudur. Bu teknik hem deneysel olarak elde edilen element içerikleri bilinen kömür örneklerini kullanarak yapılabileceği gibi hem de modellemede kullanılan MCNP5 ve GEANT4 kodlarının kullanılmasıyla da yapılabilmektedir. Böylece, cihazın sebep olduğu lineer olmayan deneysel etkiler, iterasyon metodu ve simulasyon kütüphanesine dayalı olarak hesaplanan sonuçların, daha önceden bilinen sonuçlarla karşılaştırılmasıyla en aza indirgenebilir.

Bu yaklaşımdan yola çıkarak, Monte-Carlo En Küçük Kareler Kütüphanesi (MCLLS) metodu kömür numunelerinin element içeriğini analiz etmek için kullanıldı. Bu çalışmada deneysel olarak elde edilen kömür örneklerine ait spektrumların hem MCNP5 hem de GEANT4 kodlarıyla elde edilen element kütüphaneleri kullanılarak spektrum analizleri gerçekleştirilmiştir. Element kütüphanesi oluşturulurken sabit küresel bir hacim, sadelik teşkil etmesi açısından her iki simulasyon programı için de tercih sebebi olmuştur. Deneysel spektrumlar için kullanmadan önce LLS metodu, MCNP5 ten elde edilen kömür spektrumu üzerinde test edilmiştir. Sonuçların tutarlı bulunması üzerine metodun deneysel spektrum üzerinde uygulanmasına geçilmiştir.

LLS metodu için, öncelikle tek tek bütün elementlere ait spektrum kütüphanesi bilinmelidir. Bu sebeple H, C, O, Mg, Al, Si, S, Ca, Fe, N elementleri için ayrı ayrı MCNP5 ve GEANT4 simulasyon programı kullanılarak; MCNP5 LLS ve GEANT4 LLS ye ait spektrumlar elde edilmiştir. 10 elemente ait içeriğin bulunabilmesi için en az 11 adet element içeriği önceden bilinen deneysel spektrum gerekmektedir. Farklı kimyasal içeriğe sahip ve analizi önceden yapılmış olan yaklaşık 40 kg lık 11 adet kömür numunesi hazırlanmıştır. Her bir kömür numunesi 1 er dakika prototip cihaz bünyesinde bulunan nötron üretici ile ışınlanmış, elde edilen gama ışınları BGO detektörü ile kaydedilmiştir. Elde edilen spektrumlar LLS nin matris hesaplamaları yardımıyla matematiksel bir işleme tabi tutulmuştur. İçeriği bilinen kömür spektrumları ile element kütüphanelerinin matris çarpımlarının tersinden yola çıkarak önce bu element kütüphaneleri için uygun katsayılar elde edilerek kalibrasyon sağlanmıştır. Daha sonra, elde edilen bu katsayılarla ilişkilendirilmiş olan element kütüphaneleri, aynı deneysel kömür numuneleri üzerine içerikleri bilinmiyormuş gibi uygulanarak fit edilmesi sağlanmıştır. Böylece element içeriklerinin yüzdelikleri belirlenmiş olur. Hem MCNP5 hemde GEANT4 LLS metodunun kullanılmasıyla elde edilen element içerik değerlerinin laboratuvar analiz sonuçlarıyla karşılaştırılması yapılmıştır. Karbon, kül, nem, sülfür ve azot için; her iki simulasyon programıyla elde edilen sonuçların analiz sonuçlarından sapma miktarı bu cihazlar için belirlenen yüzde 0,5 lik güvenilirlik değerinin altında bulunmuştur.

Üretilen bu prototip cihazla, herhangi bir yığılmal malzeme içindeki temel elementlerin yüzdelik olarak oranları belirlenebildiği için oldukça geniş kapsamlı bir özelliğe de sahiptir. Cihaza farklı konumlarda dedektör takviyesi yapılarak, daha farklı endüstriyel ürünlerin de analizleri yapılabilir. Hali hazırda ülkemizde henüz üretilmemiş olan

bu tip bir prototip cihazın, yurt dışında bulunan benzer cihazlarla ölçüm hassasiyeti konusunda kıyaslaması yapıldığında, gayet tatmin edici sonuçlara sahip olduğu bulunmuştur.



1. INTRODUCTION

Coal has an important role as an energy source for electricity generation in worldwide [1]. The consumption of the various energy forms in all over the world and Turkey is comparatively shown in Figure 1.1. In Turkey, coal is essential as an energy source, since coal-fired power plants are used to supply about 31% of global electricity of the country [2]. The accomplishment of this action depends significantly upon the determining of the coal quality and an understanding of its requirements.

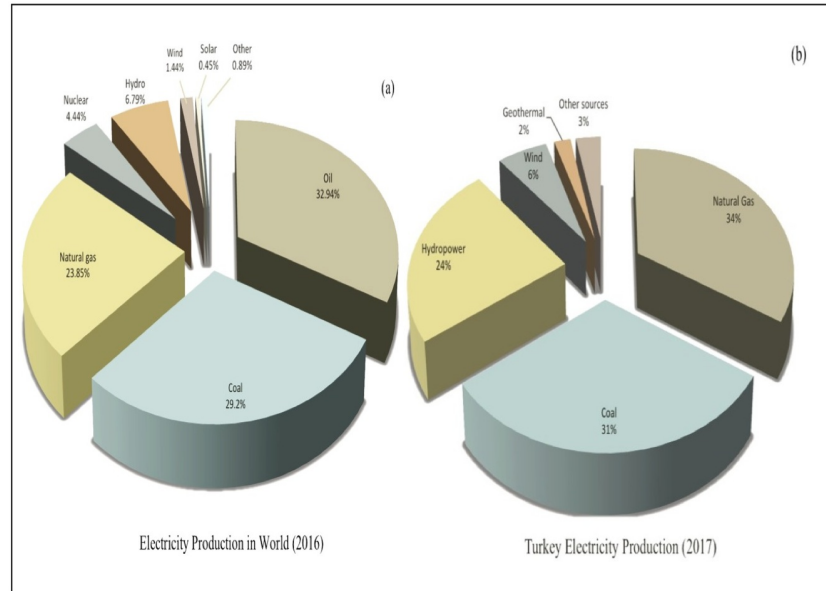


Figure 1.1 : The sources of electricity production in (a) world [1] (b) Turkey [2].

High quality coal is expected to have more content value of carbon and less content value of ash and moisture inside of itself. In the commercial sense, chemical analysis methods or instrumental techniques can be used to determine the coal quality in the mining field. Instrumental techniques are more preferred because of providing on-line measurement during coal flow on the conveyor belt. Moreover some of these techniques are based on methods of Non-Destructive Analysis (NDA). At this point, analysis may be performed with nuclear techniques. Thus, coal streams are classified and necessary blending operation is carried out before entering into the boiler and homogeneous feed streams are achieved for better combustion. On the purpose of

using some nuclear techniques [3], [4], the moisture, ash and sulfur content of coal and the calorific energy value depending of carbon content can be analyzed and classified by using this kind of apparatus.

Although the most crucial quality parameters from the commercial viewpoint are moisture, ash and fixed carbon, and calorific value; a wide knowledge of other parameters is also needed. Furthermore, high ash and sulfur content may adversely affect on the environment owing to the release of toxic elements in combustion gas and fly ash streams. Low quality coal may be blended with high quality coal and better combustion may be achieved. Despite there being some uncertainty about the relative magnitude of this problem, it is necessary to be able to measure the distribution of such elements in coal and its combustion products.

1.1 Purpose of Thesis

The aim of this thesis is to investigate the performance of a prototype apparatus which is used to analysis the content of coal material by using simulation techniques.

This study includes simulation and test study of a prototype apparatus which is supported by a TUBITAK Project with a number of 113M034 and I.T.U Scientific Research Projects Unit with a grant number of 38201. The prototype apparatus is equipped with a scintillation detector, a neutron generator, and a shielding material. As a fully electronics device, the neutron generator produces neutrons of 14 MeV energy which occur in consequence of deuterium-tritium reaction. The sample is induced by using of these neutrons of 14 MeV energy. The neutron generator is a device which can be operated with pulsed mode and continuous mode. Because of obtaining coal spectrum precisely in a short time, the operation with pulsed mode is preferred. Therefore, the obtained gamma rays can be classified according to their interactions induced by thermal neutrons or fast neutrons. The frequency of pulse cycle and duration of neutron generation can be altered optionally . For obtaining the most suitable condition, the different spectra are studied by altering the settings of the neutron generator.

As a scintillation detector, Bismuth Germanium Oxide (BGO) is chosen because of having high efficiency. The characteristic features of the BGO is studied in the course

of this work. The energy resolution, efficiency, shape calibration are determined by using radioactive point sources such as ^{60}Co , ^{137}Cs , ^{152}Eu . Additionally, the experimental response functions of BGO detector are applied to gamma ray histograms obtained simulation programs. After interaction between sample and neutrons, characteristic gamma rays of elements in coal are obtained. These characteristic gamma rays which can be either prompt gamma rays or delayed gamma rays, are counted by BGO. As a result of collected gamma rays, the spectrum is investigated for identifying content of the coal sample.

This scenario is provided to happen Monte Carlo simulation programs. Monte Carlo N-Particle 5 (MCNP5) and Geometry And Tracking 4 (GEANT4) simulation programs are used to design the geometry and physical interactions of the prototype apparatus. The synthetic coal samples including different element contents are simulated and analyzed. The consistency and reliability of the simulation results are put forward by taking advantage of the spectrum of experimental measurement results.

There are plenty of ways to analyze the gamma spectrum of a bulk system. Some of them are peak area determination, neural network algorithm and Monte Carlo Library Least Square (MCLLS) fitting [5], [6], [7]. In this study, MCLLS technique is applied by using MCNP5 and GEANT4 simulation programs. With the aid of fixed geometry, the elements inside of the coal sample are simulated individually. The obtained spectrum from MCNP5 and GEANT4 are used to build up an element library of coal sample. By using this mathematical approach, the element contents of experimental coal spectrum is aimed to calculate in the range of measurement precision defined commercially before.

1.2 Literature Review

Due to the fact that being non-destructive and sensitive for some elements, neutron activation analysis (NAA) is an important technique for identifying the specific elements in materials. The method of NAA was discovered by Hevesy and Levi in 1936. Neutrons were used to excite the sample of earth elements by emitting particles from its atom [8]. The first application of NAA technique was done by scientists of Brookhaven National Laboratory for solving problems of archaeological in the mid-1950s [9].

In conjunction with increasing the research in archaeological field, the usage of NAA has been widely known between the years in 1960 and in 1980 by Perlman and Asaro in 1969 [10], Bishop in 1975 [11], Bieber et al. in 1976 [12], Harbottle in 1982 [13].

As compared with analytic methods, NAA is a more preferred method. Since NAA presents high precision measurements, a process containing the sample preparation, immediate detection of numerous elements at same time [33].

NAA technique is based typically on measurement combination of two nuclear reactions as prompt gamma neutron activation analysis (PGNAA) and delayed gamma neutron activation analysis (DGNAA) for elemental analysis [14]. For the reason that providing the best performance requirements and its low cost, the PGNAA analyzers have been used in coal industry for 25 years.

The PGNAA systems were started to use commercially for the industry of coal mining in the 1980's. They were manufactured by Commonwealth Scientific and Industrial Research Organization (CSIRO) in Australia [15]. PGNAA systems are preferred as being most reliable techniques among coal analyzers upto now. In the middle of the 1980's, these kind of on-line analysis instruments have began to improve and R&D studies in the coalmine developed country such as U.S.A, Australia, and China [3], [4], [16], [17].

At the present time, Dual Energy Gamma-ray Transmission (DUET), Pair Production (PP), PGNAA [16], Neutron Inelastic Scattering and Thermal-capture Analysis (NISTA) [17], are the nuclear techniques which are used by the on-line analysis instrument [16]. The comparison of the measurement precision of the nuclear techniques is given in Table 1.1.

Table 1.1 : The comparison of the experimental precision of the nuclear techniques [19], [20], [17].

Content	DUET	PGNAA	NISTA
Carbon	%5	% 1.4-3.5	% 0.3
Ash	% 2	% 0.3-1	% 0.4
Sulfur	% 0.2	% 0.03-0.08	% 0.08
Moisture	measured by hygrometer	%0.4-0.8	% 0.5

By using PGNAA technique, the elements of coal on the conveyor belt are provided to be activated with neutrons. The quantitative analysis of activated elements is

performed to determine the ash, moisture, sulfur and carbon content as a percentage. In addition to coal-mining sector, the on-line sorting out with PGNA technique is used for many areas such as cement, iron, and oil industry in U.S.A, China and Australia.

The improvement of on-line analysis instruments using various nuclear techniques is allocated the considerable budget by big companies [22]. In this study, the accepted measurement precision and accuracy values of these instruments were also tried to obtain for the prototype apparatus.

By using different simulation programs and nuclear analysis techniques, the element library was obtained to determine the element contents of coal sample [3], [6].

For our country, the processes of improving the quality and preparation of the coal carries great importance to produce the energy efficiently from the coal mines. For that purpose, an instrument using some nuclear techniques ([3], [4]) was developed to be able to determine the content of the ash, moisture and calorific value of the coal moving on the conveyor belt as on-line.

1.3 Hypothesis

In this study, the content of bulk coal is intended to define as a combination of ash (%), moisture (%), carbon (%), sulfur (%) during flow on conveyor belt on-line in operation field. Therefore, the bulk coal in motion can be sorted according to their calorific values and element content without interruption. From the proprietor's point of view, the categorized coal supplies both energy saving and control of environmental pollution with low emission of sulfur. A prototype device which uses nuclear analysis techniques is designed and modeled.

The technique of most devices is based on using a radioisotope source such as ^{252}Cf , ^{241}Am - ^9Be , and ^{239}Pu - ^9Be . The most common isotropic neutron source is ^{252}Cf . The irradiation of samples depends on neutron interactions which are neutron inelastic scattering($n, n' \gamma$), and the neutron radiative capture (n, γ). Due to the fact that the average neutron energy of ^{252}Cf is about 2.5 MeV, this neutron source is convenient to analyze the samples including only the process of thermal neutron capture (n, γ). The carbon content of coal which is important parameter to determine the quality of the coal, can not be detected by using ^{252}Cf source.

The cross section of carbon gamma ray (4.43 MeV) which occurs from inelastic scattering of neutron, is larger than carbon gamma ray (4.95 MeV). Moreover, the energy of neutron source has to be larger than 5 MeV to detect the carbon gamma ray (4.43 MeV) [23]. Therefore, a neutron generator is used to produce fast neutrons of 14 MeV releasing from deuterium – tritium (D-T) fusion reaction for the prototype device. In this project, As a combination of Neutron Inelastic Scattering (NIS) and Thermal Neutron Capture (TNC) commonly referred as PGNA, NISTA technique is applied to evaluate the results of analysis. Therefore, the more precisely measurement of carbon element is possible by using the NISTA technique which is not used in current devices at present. With the aid of this precise measurement, calorific value of coal sample can be calculated accurately [17], [19].

The choice of a detector is pretty significant issue for designing the prototype device. The scintillation type of detector is preferred for the reason that has high efficiency and more endurance to neutron radiation damage. Due to the fact that BGO detector has more efficiency than NaI(Tl) detector for gamma energies larger than 4 MeV, BGO detector with a crystal of 3"x3" inches is chosen for prototype device.

Before manufacturing of the device, some simulation programs are utilized during R&D studies. By using MCNP5 and GEANT4 software packages, the on-line NISTA system including detector, neutron source, bulk sample and shielding material is simulated for test study of prototype apparatus. The real neutron generator of the prototype device is represented as a point neutron source of 14 MeV for simulation of system by MCNP5 and GEANT4. The shadow shielding material, shielding of BGO detector and collimator material of neutron source are located in conformity with real dimensions of prototype apparatus.

The coal samples with changing concentrations are simulated by including required physical interaction. The characteristic gamma rays of coal elements are defined well for comparing with the prototype device. By combining the information of interaction and simple spherical geometry, gamma ray spectrum of each element induced with neutrons are obtained individually. The each spectrum of H, C, O, Mg, Al, Si, S, Ca, Fe, N in coal represents the information of their element library.

The integration of simulation spectrum to experimental spectrum is provided by using a mathematical method which is called Library Least Square (LLS). The MCNP5 and GEANT4 element libraries are used to analyze the element contents of experimental coal spectrum [6]. The calculation results are expected to be compatible with chemical analysis done before.





2. THEORY

2.1 Neutron Transport

Neutrons as uncharged particles can begin a nuclear reaction and penetrate the material with low neutron energies [24]. The neutrons being unstable particles can survive about 15 min. out of the nucleus. They can interact with protons, electrons and electron neutrinos via Beta decays with the condition of the conservation of hadron and lepton numbers. By carrying no charge, they cannot undergo electromagnetic interactions and thus the coherent and incoherent processes can be performed without any energy loss for the ionization [24], [25]. According to their energies, neutrons can be classified into five main groups as cold neutrons, thermal neutrons, epithermal neutrons, fast neutrons and high energy neutrons.

- Cold Neutrons: Their energy range is typically between 0 and 0.025 eV just below the range of thermal energies. Cold neutrons can join into absorption reactions and scattering reactions which are dominant for the most of biological materials.
- Thermal Neutrons: The energy of thermal neutrons are found about 0.025 eV at room temperature. By using moderator materials mostly including hydrogenous element, the kinetic energy of neutrons are decreased. At this energy region, reaction of neutron absorption is more dominant which plays important role for PGNA.
- Epithermal Neutrons: The energy range of epithermal neutrons which is intermediate zone between thermal and slow neutrons is between 0.025 eV and a few eV. In this region, the absorption of the neutron can create a series of oscillations called resonance via an excited states of a nucleus [28].
- The energy range of fast neutrons is between 0.5 and 20 MeV which are produced by fission processes or (α ,n) reactions. Neutrons having kinetic energy greater than 1 MeV are usually called fission neutrons which have a mean energy of 2 MeV

(for ^{235}U fission) [27]. The fast neutrons produced by a nuclear reactor are lost their energies until it reaches the thermal energies, so that the process of neutron thermalization occurs. In order to obtain fast neutrons, typical reactions are listed below:

- The D-D fusion reaction: $^2\text{H} + ^2\text{H} \rightarrow ^3\text{H} + \text{n}$ ($Q=3.27$ MeV, $E_{\text{n}} = 2.45$ MeV, $E_{\text{He}} = 0.82$ MeV)
- The D-T fusion reaction: $^2\text{H} + ^3\text{H} \rightarrow ^4\text{He} + \text{n}$ ($Q=17.59$ MeV, $E_{\text{n}} = 14.1$ MeV, $E_{\text{He}} = 3.49$ MeV)
- The T-T fusion reaction: $^3\text{H} + ^3\text{H} \rightarrow ^4\text{He} + \text{n} + \text{n}$ ($Q=11.3$ MeV)
- High energy neutrons are known as ultra fast neutrons too. They have an energy of greater than 20 MeV. An important source of effective dose coming from neutrons is the high energy neutrons in space.

The types of neutrons classified above can undergo interactions of scattering and absorption that can happen at any energy of neutrons. By dominating the region of energy of under a hundred keV, the neutron scattering process occurs the way of elastic or inelastic scattering [29].

- Elastic scattering interaction: After a neutron strikes to a particle of alike mass in nucleus, it becomes free again and gives small amount of its energy to the nucleus by conserving the total energy, momentum and kinetic energy. The lost energy amount of the neutron can be calculated in equation 2.1.

$$E_{\text{loss}} = \frac{2 \cdot E}{(A + 1)^2} \quad (2.1)$$

where A is atomic mass number of the interacted nucleus with incident neutron, and E is the kinetic energy of the incident neutron. The reduction of neutron kinetic energy can be used for shielding mechanism by using a proper material of low mass such as hydrogen as seen in equation 2.1. After energy transferring to the nucleus, it results by producing some kinds of particles.

- Inelastic scattering interaction: It is a reaction, which the kinetic energy and momentum of incident neutron are not conserved. During inelastic scattering, the collided nucleus can be excited and splinted into some pieces of recoil particles by means of transferring energy of the incident neutron. The reaction is dominant with the material of high atomic numbers especially for fast neutrons at 1 MeV [29].

The reaction is called absorption interaction, which the nucleus absorbs the incident neutrons by involving reasonably large in number of energy. This compound nucleus system combined with the incident neutron exists in the excited state. The daughter nucleus can be de-excited by emission of radiation with charged or uncharged secondary particles such as β^- , β^+ , α , γ , p. The most important de-excitation mechanism is called prompt gamma emission, which occurs almost promptly as the neutron absorption by the nuclides [30]. The absorption interaction can be dominated at low energy range of neutrons such as thermal or slow neutron energies.

2.1.1 Neutron cross section

The probability of a specific event of neutrons interaction with a given material is defined by its cross section in units of the barn ($10^{-24}cm^2$). The cross section changes depending on the energy of neutrons and type of the nuclei and for each nuclear event. For different elements, the relation between energy of neutrons and their cross section is shown in Figure 2.1. The contribution of the cross sections of the reactions such as elastic scattering, inelastic scattering, absorption reaction and fission reaction is given as total cross section which is related with microscopic cross section per atom for a given nuclei as seen in equation 2.2.

$$\sigma_{total} = \sigma_{scatter} + \sigma_{capture} + \sigma_{fission} \quad (2.2)$$

The macroscopic cross section Σ total in units of cm^{-1} is obtained from the product of the microscopic cross section and the number of atoms N per unit volume in cm^3 as given in equation 2.3.

$$\Sigma_{total} = N \cdot \sigma_{total} = \frac{(6.02 \times 10^{23} \cdot \sigma_{total}) \cdot d}{A} \quad (2.3)$$

where d is density g/cm^3 , A is the isotopic mass number (g/mole). According to their reaction types, The comparison of cross sections of Carbon and Hydrogen is shown in Figure 2.2.

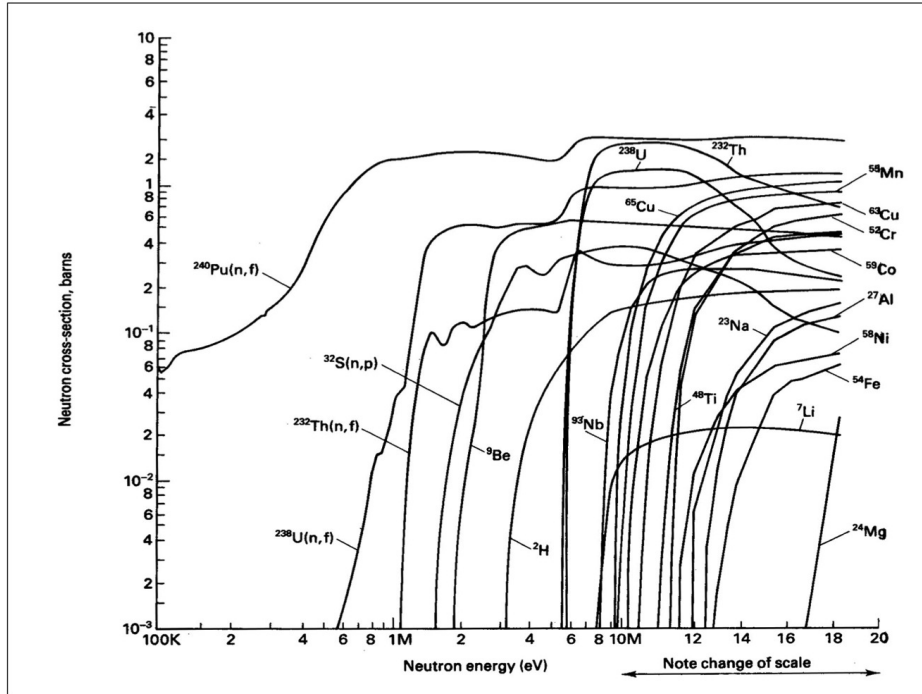


Figure 2.1 : Neutron cross-section of some elements depending on their energies [31].

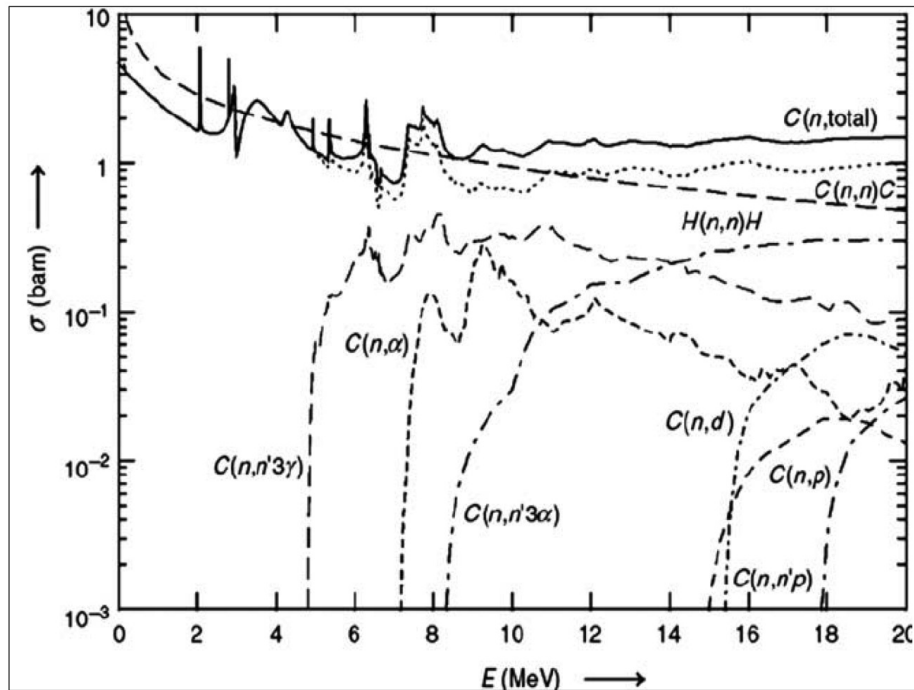


Figure 2.2 : The cross-sections of C and H elements according to their interactions types [32].

The macroscopic cross section is used to define the linear or mass absorption coefficients of neutrons just as photons. The absorption coefficient of neutron is given in equation 2.4 [26].

$$I/I_0 = e^{(-N \cdot \sigma_{total} \cdot x)} \quad (2.4)$$

Where I_0 is the number of incident neutrons on the absorber material, I is the number of transmitted neutrons passing through the absorber material, N is the number of atoms per cm^3 , σ_{total} is the microscopic cross section and x is the thickness of absorber material. It is obviously seen that the number neutrons transmitted the material I , is inversely proportional to the thickness of absorber material x .

2.1.2 Neutron shielding

The shielding of neutrons is achieved in three main steps. The first step is slowing the neutrons to thermal energies (0.025eV) by using materials containing hydrogen such as paraffin, plastic or water. Each material has disadvantages due to its characteristic. For instance, the disadvantages of water are evaporation and leakage. Being costly of plastic is disadvantage of its. For very fast neutrons, heavy metals such as iron or lead can be placed front side of materials containing hydrogen.

Second step is absorbing neutrons by using hydrogenous materials again. For 1H , the cross section of neutron absorption is quite high as 0.33 in term of b by emitting gamma ray energies of 2.22 MeV. As a second option Boron can be used to absorb the neutrons by resulting of gamma emission of low energy. The absorption of gamma rays is the third step for the neutron shielding. The processes of neutron slowing down and neutron absorption are resulted by emission of gamma rays. These gamma rays are emitted from neutron capture, scattering and activation reactions. Absorption and slowing process can be achieved at same time by using mixed type materials such as concrete mixed with barium or plastic mixed with boron. Because of neutrons are uncharged particles, they can penetrate through the shielding material excessively without interaction. Therefore, the terms of total dose and effective dose count in designing the thickness of the shielding material [34]. For a given shielding material, the dose rate of fast neutrons can be calculated by using the following equation 2.5.

$$\dot{D} = \dot{D}_0 \cdot e^{(-\Sigma_R \cdot x)} \quad (2.5)$$

Where $\dot{D}$ is the dose rate with shielding material, $\dot{D}_0$ is without shielding material, x is the thickness of the shielding material and $\Sigma \cdot R$ is removal cross section. The equation 2.6 was modified from equation 2.5 by adding the contribution of the effect of radioactive neutron source [36].

$$\dot{D} = B \cdot \dot{D}_0 \cdot e^{(-\Sigma_R \cdot x)} \quad (2.6)$$

Where the B term is the buildup factor of the related neutron source. Equation 2.6 is useful, on the condition that there is an environment of 50 cm thickness including water or hydrogenous material equivalent to water. The removal cross sections value that is 2/3 to 3/4 of total cross section, are shown in Table 2.1 for some materials [36].

Table 2.1 : The shielding calculations of fast neutrons [25].

Material	Removal Cross Section $\Sigma_R(cm^{-1})$
Water	0.103
Iron	0.1576
Ordinary concrete	0.0942
Barytes concrete	0.0945
Graphite	0.0785

2.1.3 Neutron generators

The NDA techniques based on neutron induced cover wide range for identification of bulk material in the industry. Especially, the mining industry that requires in situ and rapidly implementation takes the advantage of the usage.

In these techniques, the bulk material is irradiated by a neutron source that causes the nuclear reactions such as neutron scatterings, neutron absorption. Due to irradiation of the bulk material, emission of characteristic gamma energies are detected by the gamma spectroscopy. In order to identify the elemental contents of bulk material, these gamma energies are effectively utilized. The total quantity information of the related element in bulk sample is reached with the aid of the gamma energy detection.

The source of neutrons can be a pulsed neutron system, a radioactive point source or an accelerator based systems depending on the intended use [48]. The area of usage

of pulsed neutron system is broadly extended such as coal analysis [37], [39], cement industry [38], metal alloys [40], geological and soil analysis [41], oil well logging [42], cancer treatment [43], and security applications [44], [45], [46], [47], [48] .

2.1.3.1 Structure of neutron generators

The structure of a neutron generator is basically consisted of an ion source, an accelerator and a metal hydride target. The cathode side of ion source is grounded on the side of anode cylinder, which is applied 1-2 kV potential. Because of the potential difference between these two sides, the d-t gas molecules inside the anode cylinder are ionized by producing cold plasma. The electric field surrounding the anode cylinder causes magnetic field to accelerate the positive ions through the cathode side. The neutrons from D-D or D-T or T-T fusion reaction are obtained in consequence of the ions hitting to the target. The optimum neutron yield from the D-T reactions is about 10^8 - 10^9 n/s whereas the neutron yield from the D-D reaction is about 10^6 n/s. Trying to obtain higher neutron yield causes to reduce the operating life time of the neutron generator. The representative structure of a neutron generator is illustrated in Figure 2.3.

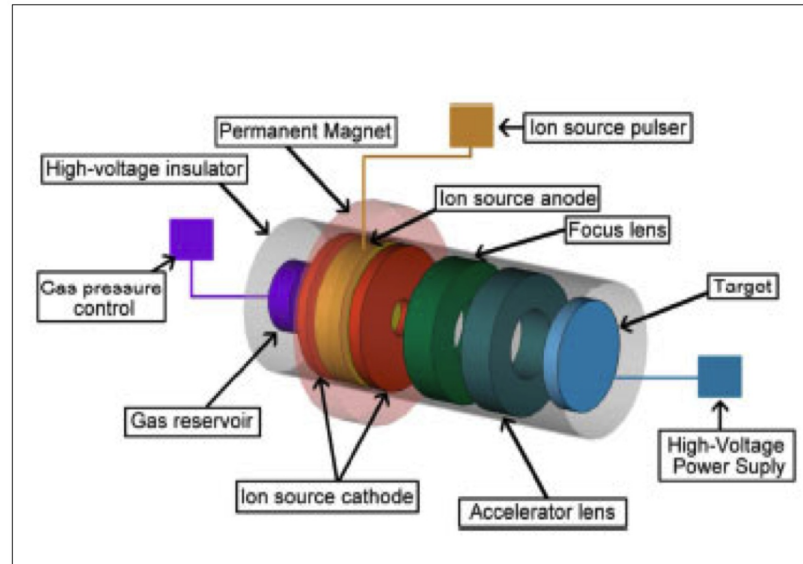


Figure 2.3 : The structure of a neutron generator [49].

^{252}Cf , ^{239}Pu , and ^{241}Am are the most common radioisotopes neutron sources. ^{252}Cf provides an average neutron energy of 2.5 MeV by spontaneous fission reactions. The average energy of the emitted neutrons from ^{239}Pu , and ^{241}Am radioisotopes is 4.5 MeV while light nuclides like Be or Li can be occurred via (α , n) reactions.

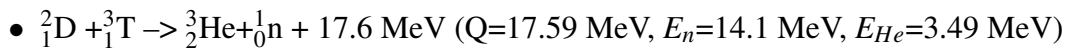
Even though the neutron generators used the radioactive sources are turned off, this technique needs significant shielding material for radiation protection.

2.1.3.2 Pulse neutron sources and system components

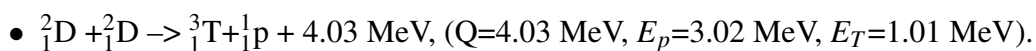
A pulsed neutron analysis system basically includes a neutron source, one or more gamma detectors, shielding material, and data acquisition system. The gamma energies emitted from the sample are induced by fast and thermal neutrons, can be recorded separately by using list mode data processing in the pulsed neutron systems. The pulsed neutron source based on nuclear reactor can be operated with a periodic mode. The energy of neutron beams bound up with the kind of moderator inside of reactor, disintegration of the nuclear fission spectrum. Due to the fact that the reactors are unportable and costly systems, the usage of nuclear reactors as a neutron generator does not gain advantage from bulk material analysis.

The D-D and D-T fusion neutron generators are known as accelerator based system which emits mono energetic neutrons. In these systems, fast neutrons are struck on a target by using charged particles. The other reactions of neutron source used in neutron generators are exemplified as $^9\text{Be} (d,n) ^{10}\text{Be}$, $^7\text{Li} (d,n) ^8\text{Be}$, $^7\text{Li} (p,n) ^7\text{Be}$, $^7\text{Be} (p,n) ^7\text{B}$ [48]. The pulsed neutron generator is preferred to be enduring, lower treatment cost, supervise the duration of neutron emission and radiation safety by using remote controller electronic equipment. The most important fusion reactions are D-D and D-T reactions [50].

The neutron generator based on D-T reactions as a neutron source is preferably used to analyze some of elements such as carbon and oxygen from inelastic scattering interactions of neutrons. In D-T reactions, emission of the neutrons is isotropically with having an energy of 14.1 MeV. The reaction occurs according to the conservation of momentum as shown in below [51].



In D-D reactions, neutrons are produced at lower cross sections than D-T reactions. Following both of reactions in D-D fusion occur with the 50% probability. D-D fusion reaction can be applicable for elements analysis based on absorption reactions.



- ${}^2_1\text{D} + {}^2_1\text{D} \rightarrow {}^3_2\text{He} + {}^1_0\text{n} + 3.27 \text{ MeV}$, ($Q=3.27 \text{ MeV}$, $E_n=2.45 \text{ MeV}$, $E_{He}=0.82 \text{ MeV}$).

The other neutron source of the neutron generators is T-T fusion. The cross section of the T-T fusion just as D-D reaction is a hundred times lower than the D-T reaction. The energy of produced neutrons in T-T reaction can take the value up to about 11 MeV which may be used as a source of slow and fast neutrons.

- The D-D fusion reactions: ${}^2_1\text{H} + {}^2_1\text{H} \rightarrow {}^3_1\text{H} + {}^1_0\text{n}$ ($Q=3.27 \text{ MeV}$, $E_n = 2.45 \text{ MeV}$, $E_{He} = 0.82 \text{ MeV}$)
- The D-T fusion reactions: ${}^2_1\text{H} + {}^3_1\text{H} \rightarrow {}^4_2\text{He} + {}^1_0\text{n}$ ($Q=17.59 \text{ MeV}$, $E_n = 14.1 \text{ MeV}$, $E_{He} = 3.49 \text{ MeV}$)
- The T-T fusion reactions: ${}^3_1\text{H} + {}^3_1\text{H} \rightarrow {}^4_2\text{He} + {}^1_0\text{n} + {}^1_0\text{n}$ ($Q=11.3 \text{ MeV}$)

2.1.3.3 The method of neutron generators and their applications

In order to accomplish the analysis of bulk material, there are several methods based on detection the gamma rays of neutrons induced. The most common techniques and their specifications are listed below:

- PGNAA: The bulk sample is continuously irradiated with fast neutrons by using a radioisotope (e.g. ${}^{252}\text{Cf}$), a reactor or a neutron generator. After absorption of neutrons, the sample emits gamma rays called prompt gamma rays which are fingerprints of the elements identified by a gamma detector. This non destructive technique is widely used in coal and cement industry.
- Pulsed Fast Neutron Analysis (PFNA): PFNA was developed an idea by Tsahi Gozani, Peter Sawa, and Peter Ryge at SAIC in 1987 [52]. Electric field surrounding anode cylinder periodically clears away the beam from the metal hydride target. This operation causes neutron pulses which last a few nanoseconds depending on the users choice. The transferred kinetic energy from the accelerator and the Q value of the D(d,n) reaction are well known. A neutron with an kinetic energy of 2.5 MeV has the velocity of 6 cm/ns. Hence, the creation time and the position of neutron can be exactly calculated. By using time of flight, the information of pulse duration helps to find signal-to-noise ratio that is necessary

to analyze the elements in bulk material. With the aid of the neutron beam can evaluate the object up to 9 cm x 12 cm of its center, The PFNA method can be an effective technique to scan the cargo containers [53].

- Pulsed Fast Neutron Transmission Spectroscopy (PFNTS): The PFNTS technique was aim to use for scanning systems at airports where have to check a great number of luggage contents in a minute in 1999 [55]. The detection rate of the scanning was suggested as 93% [54]. Only with PFTNS, a neutron beam is channeled to the neutron detector rather than a gamma detector. By using time of flight (TOF) measurements from 4 to 10 m, the energy of neutron spectrum is resolved. Especially for light elements with high cross sections, the detection of element contents is possible. However the systems with PFTNS were not sufficiently become widespread in security screening. One of the reasons is that the system based on TOF measurements is needed to voluminous area.
- Associated Particle Imaging (API): In this technique, TOF measurement is executed by using a gamma detector and an alpha detector which detects the alpha particles emitted from D-T reactions. The ZnO(Ga) alpha detector that has efficiency about 90% for 3.49 MeV alpha particles, is placed inside of the tube of neutron generator [56], [57]. The detection of alpha and gamma particles is recorded in ns by Data Acquisition (DAQ) system. The time interval of this logic signal is called coincidence window. The illustration of the API technique is shown in Figure 2.4. In order to prevent the jitters of the signals, the choice of detectors must be done carefully. Also, Increasing the neutron flux of the generator leads to raise the rate of random coincidences [?]. The European Illicit Trafficking (EURITRACK) Countermeasures Kit [58], and Underwater Coastal Sea Surveyor (UNCOSS) [59] are some of the systems being used the API technique.
- Pulsed Fast Thermal Neutron Analysis (PFTNA): The method was enhanced by George Vourvopoulos, Phillip Womble, and Frederick Schultz [61]. In PFTNA system, the gamma ray spectrum obtained from inelastic scattering (fast neutrons) and absorption reactions (thermal neutrons) are collected in different memory addresses for each neutron pulse by DAQ systems. When the neutron generator is turned on, data from inelastic reaction ($n, n'\gamma$) is acquired. As the neutron generator

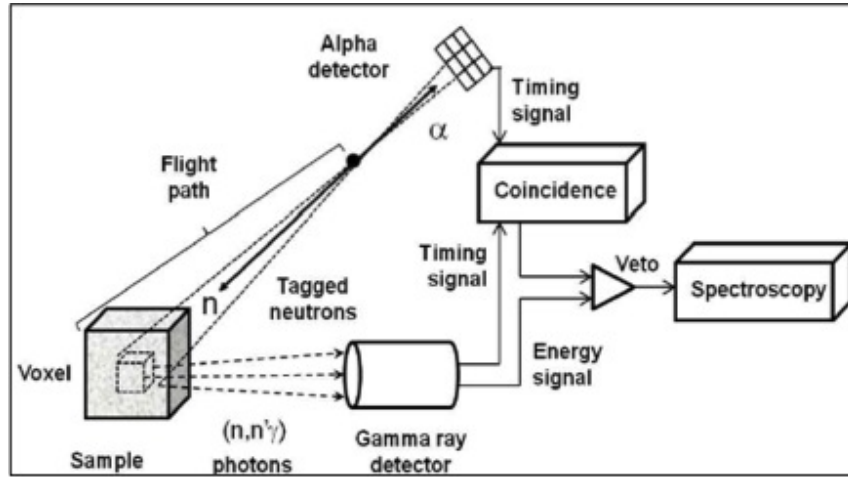


Figure 2.4 : The diagram of API technique [48].

is turned off, data from absorption reaction (n,γ) is acquired. The products from the activation reactions such as (n,p) and (n,α) are collected for a few minutes.

In order to maximize thermal neutron flux, the minimum pulse duration is upgraded to 5 ms whereas it is about 2 ns for PFTNA technique. In the neutron generator of the PFTNA, clamping circuit as a source voltage is used for the pulsing. Because of this circuit works at constant duty cycle, lower pulse frequency causes higher pulse timing.

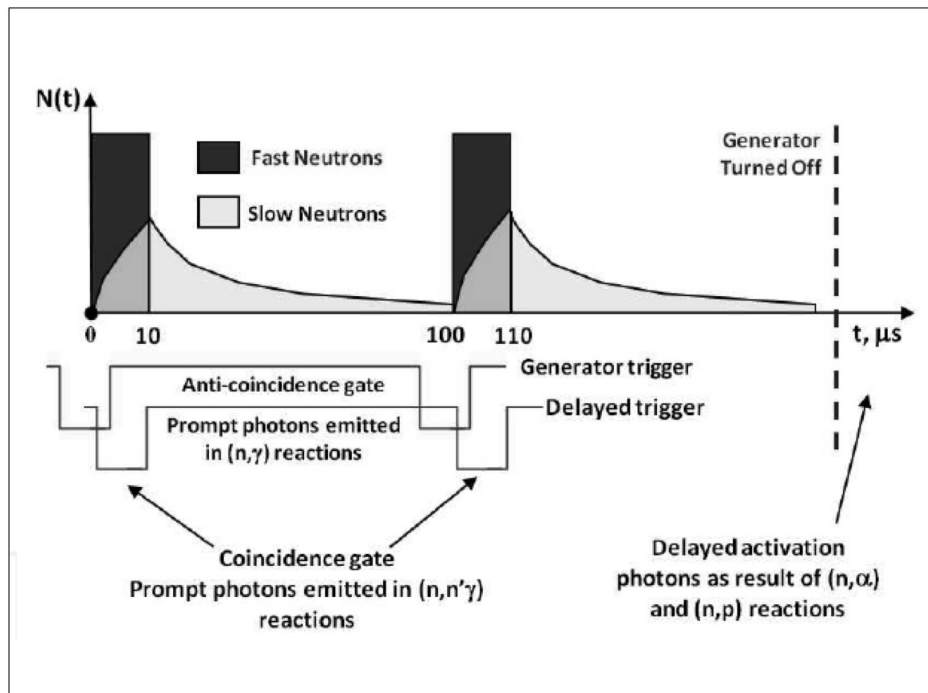


Figure 2.5 : The diagram of PFTNA pulses [48].

The characteristic of thermal neutron flux is assigned by adjusting the frequency of neutron pulse. If the frequency of the pulse is chosen more than 5 kHz, the thermal neutron flux is almost constant [60]. In order to obtain constant thermal neutron flux, the frequency of the pulse is usually adjusted nearly as 10 kHz. The diagram of PFTNA pulses is shown in Figure 2.5. For the biologic radiation protection, the user should operate the PFTNA systems 8 meters away in the case of no shielding.

2.2 Photon transport

The interaction of photons with matter is basically inclusive of pair-production, photoelectric effect, Compton scattering, photo-nuclear interactions, and Rayleigh scattering [62]. By reason of transferring the energy to secondary electrons, pair-production, photoelectric effect, Compton scattering are more important than the others. The atomic cross-sections of the related photon interaction are illustrated by delineating the dominant regions of them in Figure 2.6.

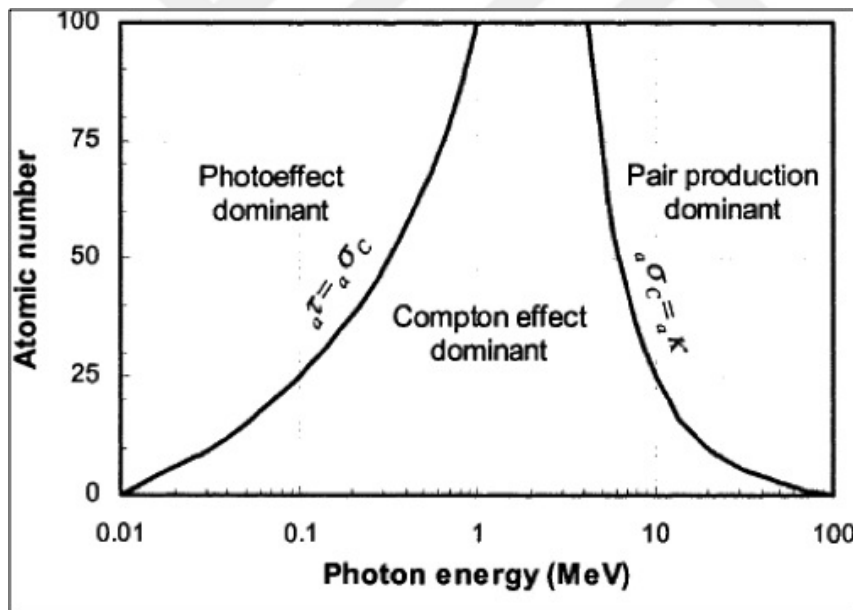


Figure 2.6 : The dominant region of photon interactions [62].

2.2.1 Photoelectric effect

The photoelectric interaction is explained by transferring the whole energy of photon to the electron of the atom. As the photon completely vanishes, either the electron is excited to the higher energy level of the atom or the electron is released with a kinetic energy rest of its binding energy. The formulation of the photoelectric effect is shown in equation 2.7.

$$E_K = h\nu - E_B \quad (2.7)$$

where $h\nu$ is the energy of incident photon, E_B is the binding energy of electron, and E_K is the maximum kinetic energy of released electron.

If the binding energy of electron is less than the energy of the incident photon, the electron is unbinded by making a hole of itself. This free electron is called as photo-electron. The vacancy of being created of photo-electron is filled by an electron cascade which is occurred towards to inner shells from outer shell of the atom. The process calling de-excitation results with different process such as Auger emission, characteristic x-rays emission and Coster-Kronig emission. The sum of the process is found both experimentally and theoretically as shown in equation 2.8.

$$w + a + f = 1 \quad (2.8)$$

where the w is for x-ray emission, a is for auger emission and f is Coster Kronig. The cross-section of photoelectric interaction changes depending on atomic number and incident photon energy as shown in equation 2.9.

$$\sigma \approx \frac{Z^5}{E_\gamma^{3.5}} \quad (2.9)$$

2.2.2 Pair production

If the energy of incoming photon is at least 1.022 MeV, an electron- positron pair can be created via the aid of strong electromagnetic interaction in the surrounding area of the nucleus. This process is called as pair production. In a reaction $M_1 + M_2 \rightarrow M_3 + M_4 + M_5 + Q$ in laboratory frame, threshold energy of the interaction can be found as shown in equation 2.10.

$$E_{th} = \frac{Q}{2M_2c^2} (Q - 2(M_1c^2 + M_1c^2)), \quad (2.10)$$

where M_2 is for a particle at rest. For a pair production process: $\gamma + M \rightarrow M + m_e + m_e + Q$, M_1 equals to 0 for γ , M_2 and M_3 is M for same particle, M_4 and M_5 are rest mass

energy for electron and positron as m_e . Hence, the released energy (Q) and threshold energy (E_{th}) of pair production process are found in equation 2.11 and equation 2.12 respectively.

$$Q = 2m_e c^2 (E_{th}) \quad (2.11)$$

$$E_{th} = \frac{2m_e c^2 (m_2 c^2 + M c^2)}{M c^2} \quad (2.12)$$

For the greater energy than 1.022 MeV, the remaining energy transform the kinetic energy of positron and electron. Although the nucleus remains unchanged, it gains extremely tiny amount of recoil energy to keep the conservation of the momentum. The electron- positron pair loses its kinetic energy and disappears in the absorber material so that annihilation process occurs. As a result of annihilation process, two photons are created with energies of 0.511 MeV. By absorbing both annihilation photons, their energies are located in the full energy peak in the related spectrum. If one of the photons can not be detected, the single escape peak with energies of 0.511 MeV is observed in the related spectrum. If none of annihilation photons can be detected, the double escape peak with energies of 1.022 MeV is observed in the related spectrum. The photon having energies less than 1.022 MeV, can not create the pair production process.

Above 1.022 MeV, the probability of occurring of the pair production in nucleus is proportional to square of atomic number Z , especially for heavy elements as shown in equation 2.13.

$$\sigma_n \sim Z^2 \quad (2.13)$$

2.2.3 Compton scattering

Compton interaction is simply explained as interaction between a photon and an electron assuming free. This process occurs as an inelastic scattering owing to the photon leaves the medium with less energy by transferring some of it to the electron. The schematic view of Compton scattering mechanism is shown in Figure 2.7.

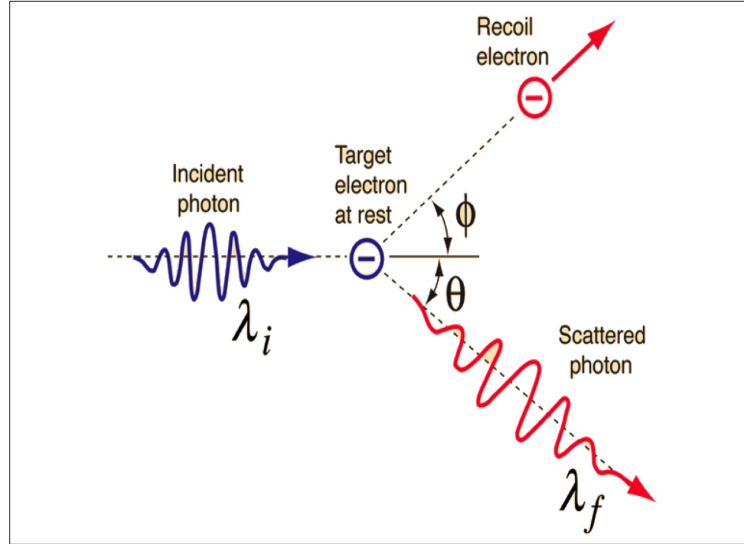


Figure 2.7 : Representative view of Compton scattering [63].

In due course of the interaction, the quantity of transferring energy to the electron determines the directions of the scattered particles. The energy of scattered photon is given by the following equation 2.14.

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}(1 - \cos\theta)} \quad (2.14)$$

where m_0c^2 is rest mass energy of electron of 511 keV, $h\nu$ is the energy of incoming photon, θ is the angle between target and scattered photon, and $h\nu'$ is the energy of scattered photon. For the negligible value of the ϕ angle, the energy of scattered photon takes close value to the incident photon. Because of the minimum value of cosine function is -1 at 180° , the energy of scattered photon is minimum. From equation 2.14, the kinetic energy of recoil electron is found as $E_e = h\nu - h\nu'$. By occurring the process of Compton scattering in the detector, the energy of recoil electron is absorbed by the medium of the detector. Depending on the energy lost of incident photon, the process is resulted as an pulse in the Compton edge region of the related spectrum.

By reason of the Compton scattering needs for a electron loosely bounded to nucleus, the probability of occurrence of the process is almost independent of the atomic mass Z . The probability of interaction is directly proportional to electron density of

the material and inversely proportional to increasing of the photon energy unlike the processes of pair production and photoelectric effect.

2.2.4 Photonuclear interactions

A photon with energy of exceeding a few MeV, excites a nucleus, a proton or a neutron is emitted from the nucleus. The proton emission (γ, p) is the reaction which most of energy is used by interaction of pair production, the rest is released as kinetic energy in matter. For that reason, the process has negligible radiation dose for medical practice. The neutron emission (γ, n) is more important process which can cause issue for radiation protection. In order to protect the hardware of medical accelerator, this process has to be considered for choosing the ideal shielding material [62].

2.2.5 Rayleigh (Coherent) scattering

Due to the fact that the atom is not excited and the energy loss of the photon is disregarded, Rayleigh scattering is also known as elastic interaction. This interaction therefore emits small amount energy of radiation. Only, scattering direction of the photon changes. The possibility of the occurrence of coherent scattering increases by higher atomic numbers of material and lower energy of incident photon.

2.2.6 Photon cross-sections

The probability of an occurring of an interaction is related with sum of the cross sections of the nuclear reactions. Especially for the simulation of the nuclear reactions, expression of cross section is needed to describe the range of particle's travel. Different types of attenuation coefficients can express the term of cross section. These are basically linear attenuation coefficient and mass attenuation coefficient. The relation between mono energetic photons entering a material and interacted with material is given by the equation 2.15.

$$dN = -\mu N dl \quad (2.15)$$

where N is number of photons hitting the material, dl is the immeasurably small thickness layer of the material, μ is the probability of number of interacting photons

with material in $1/m$ (m^{-1}). By solving the equation 2.15, an exponential relation is obtained in equation 2.16.

$$N = N_o e^{-\mu l} \quad (2.16)$$

where N is the number of transmitted photons, l is the thickness of material, and N_o is the photon number entering the slab of material. In equation 2.16, number of transmitted photons are presumed as no include the secondary photons. The term μ corresponds to linear attenuation coefficient is used to refer the expression of the probability of photon interaction. Total attenuation μ is given by the sum of the linear attenuation of photoelectric absorption, pair production, Compton scattering and Rayleigh scattering in equation 2.17. The photo-nuclear interaction term is usually neglected for the equation 2.17.

$$\mu = \mu_{photo} + \mu_{pair} + \mu_{Compt} + \mu_{coh} \quad (2.17)$$

The mass absorption coefficient μ in units of cm^2/g , is obtained by dividing both side of equation 2.17 by density of material ρ as shown in equation 2.18.

$$\mu_m = \frac{\mu}{\rho} = \frac{\mu_{photo}}{\rho} + \frac{\mu_{pair}}{\rho} + \frac{\mu_{Comt}}{\rho} + \frac{\mu_{coh}}{\rho} \quad (2.18)$$

The μ_m is bound up with the atomic cross section term σ_{tot} ($cm^2/atom$ number)

$$\frac{\mu}{\rho} = \sigma_{tot} \frac{N_A}{A}, (\sigma_{tot} = \sigma_{photo} + \sigma_{pair} + \sigma_{Compt} + \sigma_{coh}) \quad (2.19)$$

where N_A is Avogadro's number, A is the atomic mass number of attenuator material in equation 2.19. The cross sections of photon interactions consist of the energy of incident photon and the atomic mass number of the attenuator material. For copper element, the cross sections of photon interactions are shown in Figure 2.8.

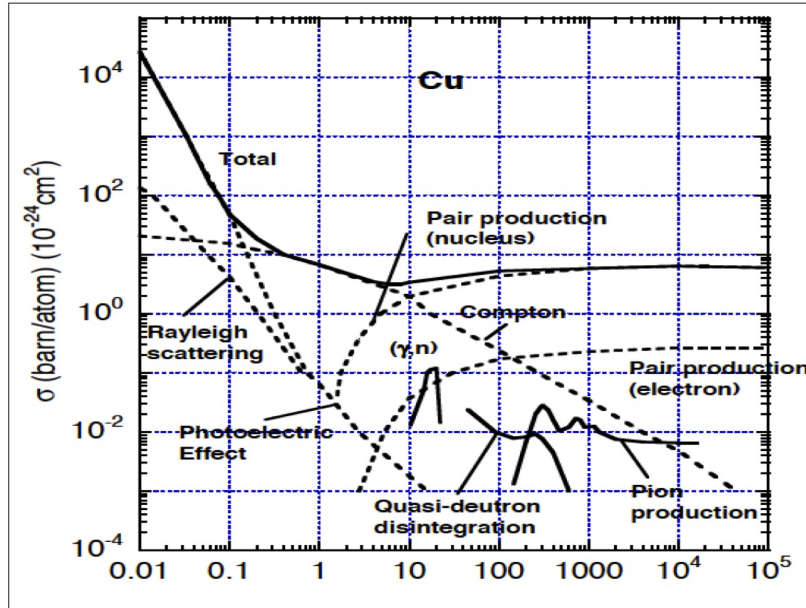


Figure 2.8 : Cross section of Cu for photons between 10 keV and 100 GeV [64].

For high atomic number Z and low photon energies, photoelectric absorption is dominant. The cross section of photoelectric absorption sharply drops in with increasing the incident photon energy.

Because of having more density of electron, The cross section of Compton scattering is higher for the low atomic number material such as containing materials with hydrogen. Also, in the range of photon energies between the predominance of pair production and photoelectric absorption, Compton scattering takes a limited place to show its precedence for the contribution of photon interactions to total cross sections. The cross section of pair production increases with the square of the atomic number of attenuator material (Z^2) and higher energies above 1.022 MeV.

The cross section of Rayleigh scattering is proportional to atomic number Z and lower photon energies. Due to the fact that it is coherent scattering, the cross section of Rayleigh scattering is usually negligible for photon transportation in Monte Carlo simulation. The contribution of photo-nuclear interactions to total cross section is about %10 in the photon energies region between 5 and 40 MeV. The cross section does not display an orderly behavior due to the atomic number of attenuator material and incident photon energies. By comparing the cross sections of photon interactions of low and high atomic number, the representation for carbon and tungsten materials is shown in Figure 2.9.

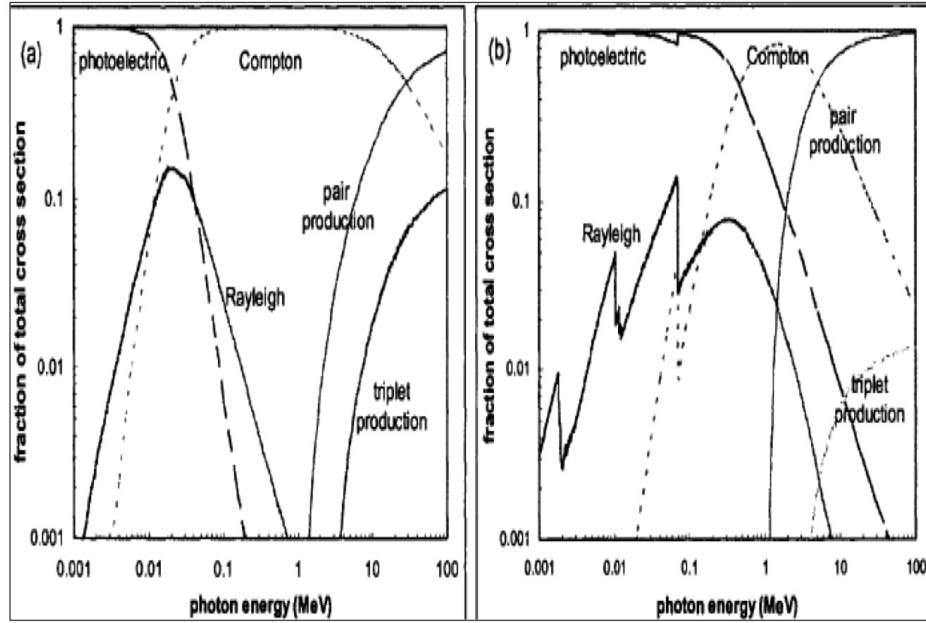


Figure 2.9 : The comparison of photon interaction cross sections for low and high atomic number material for (a) carbon and (b) tungsten [65].

2.2.7 Gamma-ray detectors

As an uncharged particle, a photon does not ionize or excite the material directly. Hence, it can be detected by following the resultant interactions with an electron. After the photon transfers all or some of its energy to an electron, the electron interacts with other electrons of the atom and causes them to be free. The collected free charges either directly via a solid-state semiconductor detector or indirectly via scintillation detector create an electrical pulse proportional to the energy of incident gamma ray inside of the material of the detector [69].

For a gamma-ray spectroscopy, the significant interactions are photoelectric effect, Compton scattering and pair production, which were discussed in section 2.2. Each interaction dominates different energy range of the gamma-ray spectrum. The gamma-ray detectors can be classified according to the material used in it. There are several types of detectors but the most used for nondestructive applications are gas-filled detectors, solid-state detectors, and scintillation detectors. The features of these detectors are detailed in below.

2.2.7.1 Gas-filled detectors

The working principle of a gas filled detector is based on ionization radiation phenomena. The gas-filled detector usually consists of a cylindrical volume full of gas and two electrodes applying electric field between them. The typical gas filled detector is shown in Figure 2.10.

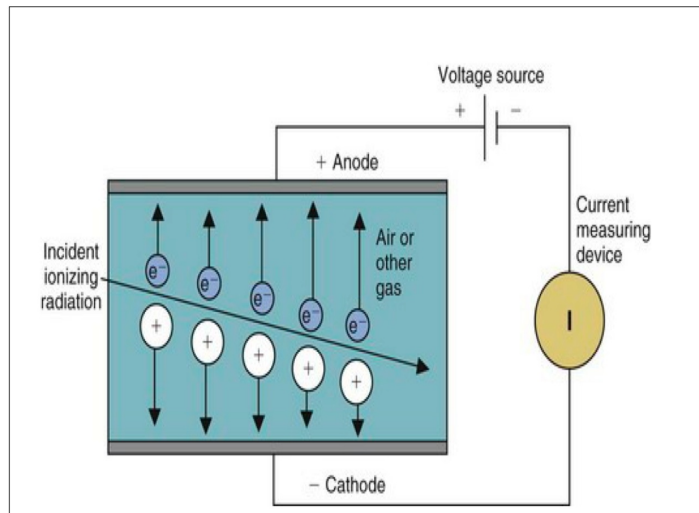


Figure 2.10 : Illustration of a gas-filled detector [66].

As a result of ionization radiation, the particles transfer some or all of their energies to create ionized electrons. With the aid of electric field, these ionized electrons carry the charges which cause measurably a current as an output signal. The magnitude of the output signal is related to the amount of the energy deposition caused by particles inside of the gas filled volume. As the electric field between the electrodes increases, the energy deposition of the detector builds up.

The common example of gas filled detector is Geiger Mueller counters which are used in dosimeters. The aim of Geiger Muller counters is to measure the number of particles not to find the type or energy of the incident particles. Gas filled detectors are commonly preferred for neutron detection, while they are not usually used for NDA of nuclear. material [69].

2.2.7.2 Solid-state detectors

The operation principle of the solid-state detectors is similar to the gas- filled detectors. Unlike ionized electrons to carry the charges, electron and hole in the structure of a

semiconductor are used for solid-state detectors. A semiconductor is a solid material which conductivity property varies with a change in temperature. Typical illustration of a solid-state detector is shown in Figure 2.11 [69]. The solid state detector is consist of depleted region applied electric field inside which is surrounded two electrodes adjacent to semiconductor crystals called p-n junction. While the amount of holes is greater than electrons in p-type semiconductor; n-type semiconductor is just the opposite of p-type junction.

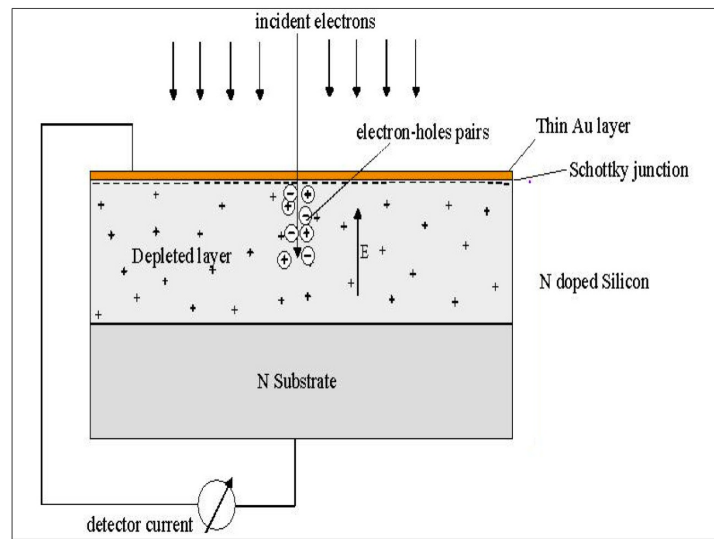


Figure 2.11 : Typical arrangement of components in a solid-state detector [69].

A semiconductor has a narrow energy gap which places between a valence band full of electrons and an empty conduction band. When an electron of the valence band is excited in some way like an increasing of the temperature or radiation exposure, the electron moves toward the conduction band by leaving a hole behind it. In the structure of crystal in solid-state detector, germanium and silicon are commonly used by being doped with lithium known as Ge(Li) and Si(Li) detectors. Compared to others, the solid-state detectors provide grate energy resolution for spectral analysis. The shape of solid-state detectors can be either coaxial or planar which is illustrated in Figure 2.12.

Their intended usage depends on the measured gamma energy range. For higher energies, the coaxial shaped detectors provide better efficiency, whereas planar shaped (Si(Li)) detectors provide better detection efficiency for low energy measurements such as x-ray fluorescence (XRF) [69]. Due to the fact that the crystal materials of solid state detectors are sensitive to radiation damage, they are not preferred to use for the NDA measurements based on a source of large neutron flux.

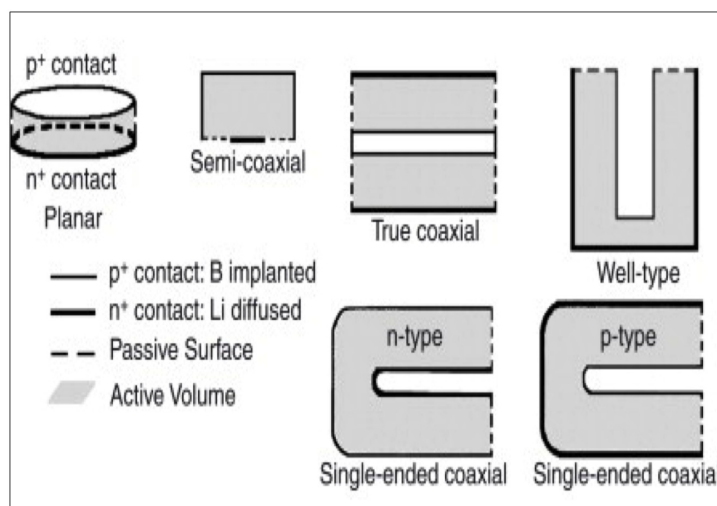


Figure 2.12 : Illustration of various solid-state detector crystal configurations: (a) open-ended cylindrical or true coaxial, (b) closed-ended cylindrical, and (c) planar [67].

These types of detectors are suitable for detection of low energy x-rays and gamma rays. Silicon detectors are mostly used in charged-particle spectroscopy and for spectroscopy of high-energy gamma rays [69].

2.2.7.3 Scintillation detectors

Scintillation is described as a process of a photon emission during the return to the ground state of an excited electron. The emitted photon is detected by a photomultiplier tube (PMT). The scintillation yield is quite small to detect. The electrodes inside of the PMT, called as dynodes, amplify the amount of photons causing photo-electrons by a multiplication factor of more than 10^4 . With the aid of applied high voltage to the PMT, the photo-electrons are accelerated from cathode to anode of the tube. The amplified photo-electrons being reached to the anode of the tube are converted to a measurable signal proportionally to the energy of the incident photon by using electronic equipment. A scintillation detector is mainly composed of a scintillating crystal, a light pipe, a PMT, a preamplifier and an electronic module. The schematic diagram of the scintillation detector is shown in Figure 2.13. The structure of scintillator composition can be any of the gas, liquid and solid states. There are three types of scintillation detectors such as organic, inorganic (crystal) and gaseous scintillators.

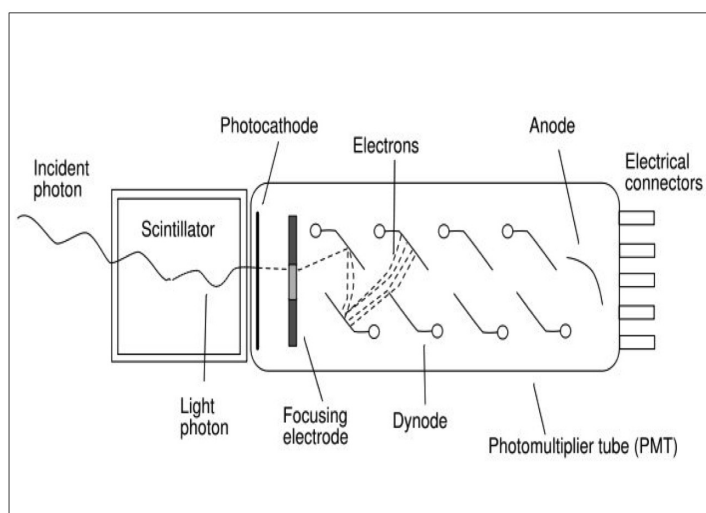


Figure 2.13 : Simplified picture of a scintillation detector [70].

- The working principle of organic scintillators is based on molecular transition of unbound valence electrons to excited state by means of ionization radiation. Liquids, plastics, hydrocarbons such as naphthalene ($C_{10}H_8$), and anthracene ($C_{14}H_{10}$) can be given as the most known example of the structure for organic scintillators.
- The operating principle of an inorganic scintillator detector is based on electron transition from allowed energy band to forbidden energy band. After this transition, a photon emission is occurred which is known as luminescence of the crystal. The favorite elements of the inorganic scintillator detectors are NaI(Tl), CsI(Tl), LiI(Eu), CaI(Na), ZnS(Ag) and BGO. These types of detectors frequently use a material doped impurity in different concentration, which is in charge of luminescence of the scintillator crystal.
- Gaseous scintillators contain a gas mixture of including noble gas. The scintillation mechanism of gaseous scintillator is achieved by atomic transition. For heavy and charged particles, the energy measurement is usually performed by using gaseous scintillators. The reason of this, it can provide a light output independently of the charge and mass for the energy deposition measurement of a particle.

Some common inorganic scintillation materials are sodium iodide (NaI(Tl)), cesium iodide (CsI(Tl)), zinc sulfide (ZnS(Ag)), and lithium iodide (LiI(Eu)). The most common scintillation detectors are solid, and the most popular are the inorganic crystals NaI(Tl) and CsI(Tl).

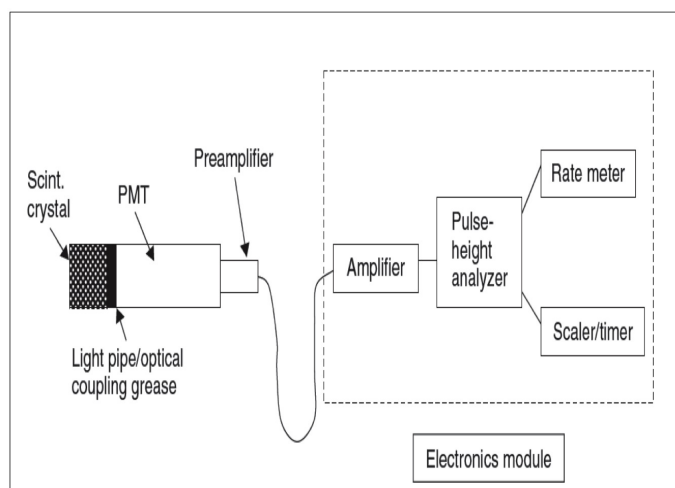
Table 2.2 : Properties of Scintillation Detectors.

Property	NaI(Tl)	BGO	LSO(Ca)	GSO(Ca)	CaI(Tl)	LuAP(Ca)	Plastics
Density (g/cm ³)	3.67	7.13	7.40	6.71	4.51	8.34	1.03
Effective atomic number	50	73	66	59	54	65	12
Decay time (nsec)	230	300	40	60	1000	18	2
Photon yield	38	8	20-30	12-15	52	12	10
Index of refraction	1.85	2.15	1.82	1.85	1.80	1.97	1.58
Hygroscopic	Yes	No	No	No	Slightly	No	No
Peak Emission	415	480	420	430	540	365	Various

Interaction of photon with scintillation material causes excitation of atoms of scintillators. During de-excitation of these atoms, photons are emitted. Moreover, the emitted photons are quite energetic. PMT is sensitive to this energy range of photons. The amount of charge flow is effected from amount of charge released of photo-cathode. Because of the photoelectric effect, there is a relation between photo-electron numbers and photon number on photo-tube. This relation is resulted as energy deposition in the scintillator.

2.2.8 Detection System

The function of a detector is to generate a pulse for each particle entering into the system. The working principle of the detector is based on the interaction of particles with matter. A detection system is basically consists of a high voltage power supply (HVPS), preamplifier, amplifier, a multi-channel analyzer (MCA). The configuration of the detection system is shown in Figure 2.14. The features of detection system components are explained in detailed.

**Figure 2.14** : Block diagram of a detection system.

2.2.8.1 High-voltage power (bias) supply

In order to collect the charges obtained by a reaction in the gamma-ray detector, it is needed to an electric field which is provided by the HVPS. The supplied voltage to the detector can be positive or negative voltage. The HVPS of the detector is usually implemented to the preamplifier via a connection apparatus [74]. For some detector types, the values of the high voltage are listed in Table 2.3. The bias supply is the part of handling the gamma-ray detector and it is reliable and easiest unit to operate in the detection system.

Table 2.3 : The HVPS values for different type of detectors [74].

Detector	High Voltage (V)
Ionization Counters	Up to 1000
Wire Chamber	0,5 – 10000
Proportional Counters	500 to 1500
Geiger Mueller	500 to 1500
Germanium Detector	Up to 4500
Silicon Detector	50 to 500
PMT	1000 to 3000

In addition to bias supply, A system of electronic modules is commonly used to convert easily the alternate current (AC) voltage direct current (DC) voltage which is called Nuclear Instrumentation Module (NIM) developed in 1960's. The NIM system adjusts the desired voltage value by using switching equipment.

2.2.8.2 Preamplifier

The preamplifier has a function as a bridge to make the processing between the product of the detector and detection system optimum. Other function of the preamplifier is to try eliminating the noise that has a negative effect on a signal. The signal at the output of the detector is generally weak in the range of mV and a short duration current pulse of 200 ns. The current information of the pulse is converted to voltage information that is related with gamma energy of an incident particle inside of the detector. Because the full information of detector output coming from gamma-ray interactions is required to transfer safely, the placement of the preamplifier is preferred to be the closest location to the detector. Also this placement of the preamplifier supplies to reduce the

possible noise level. The structure of preamplifiers usually is not available to external intervention, which is often placed as shielded with detector.

2.2.8.3 Amplifier

The main amplifier conducts the actual amplifying process of the pulse, which is mostly an integrated system to MCA. The output pulse of preamplifier reaches to an ultimate pulse shape by the amplifier. By comparing with the previous ones, the multiplication factor increases the pulses as many as 10^3 times or more. For all of the pulses, the amplifier uses same multiplication factor. The acceptable value of the output voltage of the main amplifier usually does not exceed the value of 12 V.

The amplifiers have electronic filtering systems to obtain the intended shape of pulses such as differentiation and integration filtering. By choosing the frequency of the signal as low or high, a time constant parameter of the detector is affected depending on intended use. The time constant parameter is a value of vital importance for defining the characteristic features of a detector, which are noise, dead time, and energy resolution [68].

2.2.8.4 Multi-channel analyzer (MCA)

The purpose of MCA is to record and sort the pulses, output of the amplifier, according to their heights. Therefore MCA is called as the pulse height analyzer (PHA). The height of the pulses is proportional to the energy of the incident particle to the detector. According to pulse height, the diagram of charge collection is shown in Figure 2.15.

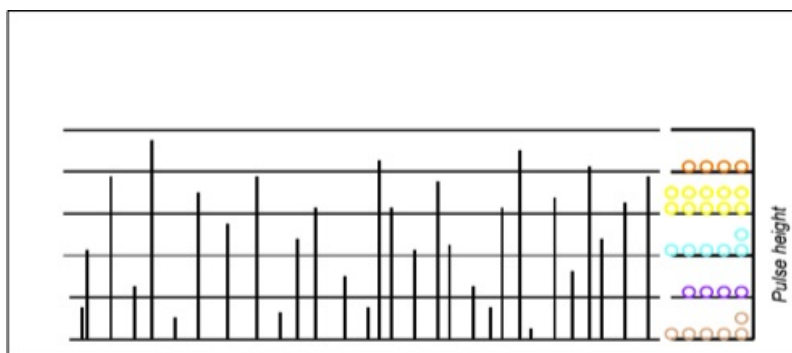


Figure 2.15 : The diagram of charge collection [73].

Each pulse is sorted individually in a channel according to its exact energy value. At the end of the sorting process, the illustration of the distribution of the particles

energies' can be drawn on the screen of the MCA as a spectrum by means of software of a computer [26] as shown in Figure 2.16.

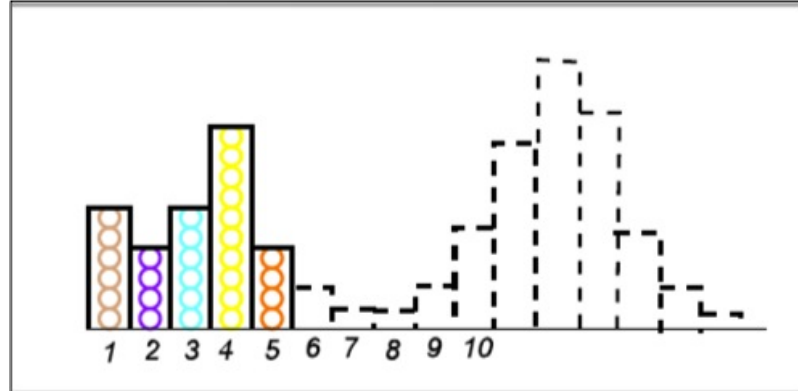


Figure 2.16 : The diagram of pulse height spectrum [73].

This illustration represents mostly the information of channel number or energy versus to number of particles per channel. Each of the channels is labeled to a certain voltage value by means of an analog to digital converter (ADC). There is an acceptable voltage value for the ADC. The ADC found at the entrance of MCA takes to process the pulses as an input coming from the amplifier and converts to a binary number. The addressing of the channel is performed dividing by a certain voltage value to the total channel number of the MCA. Since the voltage value is proportional to the gamma-ray energy of the particle, a linear relationship between a channel and energy is obtained as shown in equation 2.20.

$$E = ax + b \quad (2.20)$$

where E is the energy value, a is the slope value corresponding to amplifier gain, x is the channel number and b is the zero intercept value.

2.3 Structure of Coal

The coal is a material which was formed from peats including remains of plants and animals by fossilizing under high temperature and pressure [71]. The creation of coal arises from peatification, compaction and coalification processes respectively. The sediments inside of the peats are compressed by elimination of the water compound from the mixture. The decreasing of peats volume occurs the compaction process. Over time, buried peats began to sink into deep of the ground. The sediments are

subject to temperature increasing of 1 °C by each of 33 m deeper. This temperature increasing also causes increasing the pressure on the material. This mechanism enables to connect the chemical bonds among C, H, and O elements which are the constitution of the hydrocarbon molecules. By subtracting of water molecules and some gases from hydrocarbon structure, the coalification process happens.

The duration of coalification process and the geological structure of the region are the important factor to determine the quality of the coal material. According to the coalification process, the quality of the coal material is categorized in four groups which are lignite, sub-bituminous, bituminous, and anthracite. Lignite coal is the lowest quality whereas anthracite coal is the highest quality [75]. According to their contents, the classification of coal ranks are shown in Table 2.4.

Table 2.4 : The classification of coal ranks.

Content	Anthracite	Bituminous	Subbituminous	Lignite
Moisture (%)	3–6	2–15	10–25	25–45
Volatile matter (%)	2–12	15–45	28–45	24–32
Fixed carbon (%)	75–85	50–70	30–57	25–30
Ash (%)	4–15	4–15	3–10	3–15
Sulfur (%)	0.5–2.5	0.5–6	0.3–1.5	0.3–2.5
Hydrogen (%)	1.5–3.5	4.5–6	5.5–6.5	6–7.5
Carbon (%)	75–85	65–80	55–70	35–45
Nitrogen (%)	0.5–1	0.5–2.5	0.8–1.5	0.6–1.0
Oxygen (%)	5.5–9	4.5–10	15–30	38–48
Density (g/mL)	1.35–1.70	1.28–1.35	1.35–1.40	1.40–1.45

The classification of the coal quality is eventually based on the content of the coal that is caused by coalification. Depending on combined structure of organic and inorganic contents, coal is very complicated formation to analyze. The coal is an organic material due to fact that including C, H, N, O, S elements, where as the coal is an inorganic material by combining of nearly 60 elements inside of it [76]. Among the organic and inorganic elements, the coal material is commonly analyzed to classify according to concentration of the element contents which are moisture, ash (Si, Fe, Al, Ca, Mg, etc.), volatile matter, C, H, N, O, S, N.

2.3.1 The methods of analysis for coal contents

Predetermination of coal content is quite significant issue because of both minimizing the environmental pollution and effectivity of energy usage. The coal analysis can be

achieved in the scope of proximate and ultimate analysis. The parameters of these analysis can be classified as independent of each other. The proximate and ultimate analysis can be achieved by using destructive or non-destructive techniques. The destructive techniques are generally based on chemical analysis methods performed at laboratory conditions. The non-destructive techniques are based on nuclear activation analysis (NAA). The chemical analysis methods are not explained to provide a basis for measurements obtained a PFTNA system.

- Proximate analysis: The proximate analysis is commonly applied for preparing and mining process of the coal. The parameters used for proximate analysis are ash content, moisture content, fixed-carbon content, and volatile matter content and calorific value which are expressed in detailed. The typical proximate analyzer is shown in Figure 2.17.



Figure 2.17 : The proximate analyzer [77].

- i. Ash content: Ash is the noncombustible structure of the coal material. Ash content is an inorganic (mineral) composition which is basically consist of combination SiO_2 , Fe_2O_3 , Al_2O_3 , MgO , CaO . The concentration of ash varies according to the mining region of the coal. At high temperatures, the ash structure transforms into cinders or clinkers and adheres inside of flue pipes. This formation affects negatively the combustion efficiency of the coal plant. The rate of ash content is measured by heating the sample in an industrial oven at defined circumstances before. Also the temperature which forms the ash structure, acts as a guide for possible clinkers formation of the coal.

- ii. Moisture content: After coal is dehumidified at temperatures of 105–110 °C, the residual structure is found as moisture free coal. The weight difference can be defined as ratio of moisture content. The decreasing of the moisture content indicates often higher combustion quality of the coal as calorific value. In addition to this, the high moisture content affects the storing conditions of the coal, shipping and processing cost due to the increasing in weight.
 - iii. Volatile matter content: The volatile matter is able to turn into gaseous material which consist of CO_2 , NO_x , and SO_x . At the temperature of 950 °C and with proper conditions, The coal sample is released from the volatile material. The content of volatile matter is determined by measuring the weight loss of coal sample. Having high value of volatile matter causes easily to burst into flames of the coal. This situation is not desirable for high combustion quality.
 - iv. Fixed-carbon content: The fixed carbon plays an significant role to determine the combustibility criteria of the coal. As the gaseous form of carbon within volatile matter is eliminated from the coal sample, residual combustible part is called as the fixed carbon. The content of fixed carbon can be calculated as remaining part after the summation of other contents are subtracted from the total amount of coal sample.
 - v. Calorific value: Calorific value is the fundamental criteria to determine the combustibility of the coal. It is commonly given in terms of MJ/kg which is unit of the quantity of chemical energy to transform to thermal energy of elements within the coal. The classification of coal quality is executed according to their calorific value.
- Ultimate analysis: This analysis method is used to determine the contents of organic molecules within coal. The proximate analysis is conducted by burning of the coal in analyzer which is shown in Figure 2.18. The proximate analyzer can determine the parameters as weight percent. The parameters used for ultimate analysis are total carbon, total hydrogen, total sulfur, total oxygen, total nitrogen which are expressed in detailed.

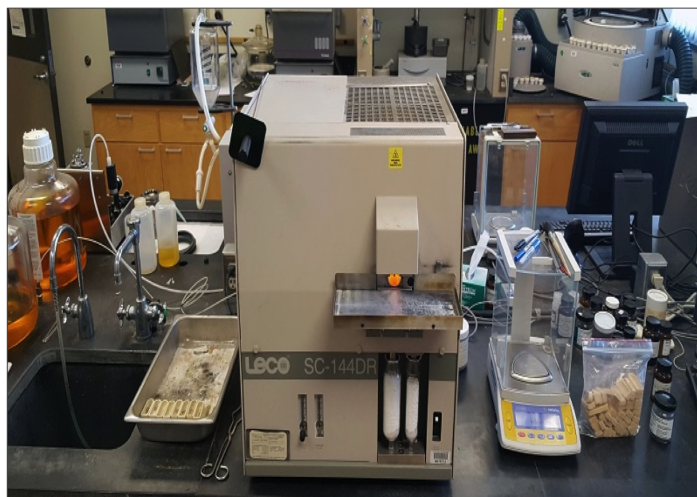


Figure 2.18 : The ultimate analyzer [78].

- i. **Total carbon:** Unlike the proximate analysis, the carbon content is calculated as a sum of fixed carbon and carbon coming from volatile content for ultimate analysis. The measurements of total carbon is conducted by combusting of coal sample in the analyzer at 1350°C . The products of CO_2 and CH_4 are produced at the end of reaction. As a result of this, amount of total carbon content is determined.
- ii. **Total sulfur:** The amount of total sulfur is based on the sum of sulfur contributions from element organic, inorganic and structure. Total sulfur is measured from the amount of SO_2 product coming from a chemical combustion reaction in the sulfur analyzer. At 1370°C , The reaction of sulfur with oxygen is concluded by an emission of SO_2 gas via endothermic process. By using a detector inside of the ultimate analyzer, the weight percent of total sulfur is calculated. The predetermination of the total sulfur is essential to prevent the emission of SO_2 gas which is main reason of acid rain (H_2SO_4) due to oxidization reaction of sulfur with oxygen. In addition to the issue of environmental pollution, the excess of sulfur content is the reason of the flue sludge and boiler abrasion.
- iii. **Total nitrogen:** The concentration of total nitrogen is calculated with the aid of ultimate analysis. During combustion of the coal, the reaction of nitrogen with oxygen results in NO_x product. Afterwards, NO_x reacts with the steam in the air. As a result of this, other source of acid rain namely HNO_3 is formed. The

predetermination of total nitrogen is connected with reason of environmental pollution.

- iv. Total hydrogen: The amount of total hydrogen content is based on the combination of the moisture content and structure of hydrocarbon. By combustion of the coal at proper conditions in the ultimate analyzer, data of water vapor and hydrous silicate material are obtained. By subtracting the hydrogen in moisture content from this data, combustible hydrogen content is reached as following relation.

$$\text{Hydrogen (fuel-based content \%)} = \text{Hydrogen (analyzer result (\%))} - ((0.1119 * \text{moisture (result of ultimate analyzer (\%))}))$$

The moisture content can be used for determining the oxygen content coming from the moisture part. The predetermination of hydrogen content is significant criteria because of contributing to the structure of moisture, and formation of H_2SO_4 and HNO_3 .

- v. The total oxygen: The amount of total oxygen can be calculated after other four parameters of ultimate analyzing are determined. The contribution of ash components and moisture content to oxygen element makes stand alone measurement difficult. Firstly, total oxygen content (%) is determined by subtracting the sum of other elements excepting the oxygen element from the total coal weight as following relation.

$$\text{Total oxygen content (\%)} = 100 - (\text{total carbon content (\%)} + \text{total hydrogen content (\%)} + \text{total nitrogen content (\%)} + \text{total sulfur weight (\%)} + \text{total ash weight (\%)})$$

Then, the total oxygen content is eliminated from moisture contribution to find correct oxygen concentration for a fuel-based value as following relation.

$$\text{Total oxygen (fuel-based content (\%))} = \text{Total oxygen (analyzer result (\%))} - ((0.8881 * \text{moisture (analyzer result (\%))}))$$

2.3.1.1 Non- Destructive Analyzing of Coal Contents

NDA is conducted by using nuclear techniques. Among the nuclear techniques, NAA is the most common techniques especially for mining industry. NAA consists of PGNA and DGNA processes [79]. These processes depend on identification time of

obtaining gamma rays from materials induced with neutrons. When the matter interacts with neutrons via inelastic scattering or absorption mechanism, the gamma-ray is emitted promptly. The observation of gamma rays from neutron absorption reaction is possible at low energy ranges of neutrons which are called as thermal neutrons. As the result of gamma-rays emission, excited the matter returns to its stable energy level in a duration of 10-14 seconds.

So, the occurrence process is PGNAA. The emitted gamma rays are specific for each of the elements. If the process of neutron irradiation results with a radio-activated particle, the duration of energy decreasing takes more time than period of half-life. This process is called DGNAA. The illustration of PGNAA and DGNAA processes are shown in Figure 2.19.

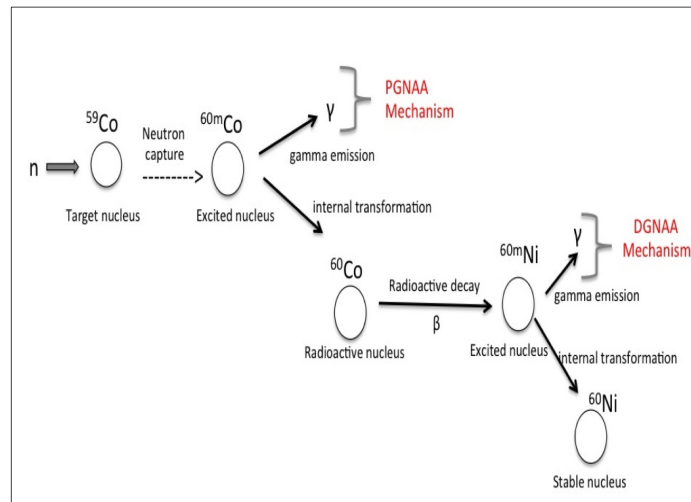


Figure 2.19 : PGNAA and DGNAA processes via Co irradiation.

Until today, the usage of radioactive sources such as ^{252}Cf has been at the center of the analyzing coal contents [17]. Due to the neutron energy of radioactive sources, the inorganic materials especially ash content, could be measured for determining of coal classification. PGNAA analyzers have been commercially available for many years, and are presently manufactured by a number of companies. The on-line elemental analysis devices in mine site which are produced in U.S.A, is shown in Figure 2.20.

The online elemental analysis devices uses the PGNAA mechanism including information of TNC and NIS reactions separately. For occurrence of TNC (n, γ), nucleus induced with thermal neutrons should exist in the excited state of nearly 8 MeV. Detection of some elements can be more probable by using TNC process. Some of these elements are Si, Ca, H, S, Fe, Cl. In order to utilize NIS ($n, n'\gamma$) mechanism,



Figure 2.20 : Thermo Gamma and Sabia Installatios in U.S.A [22].

energy of fast neutrons requires from fusion reactions which can be obtained from $^{241}\text{Am} - ^9\text{Be}$, or a neutron generator. NIS reactions can be used to determine the C, N, and O elements of bulk material. Unlike TNC process, spectrum of NIS is easy to understand because of being free of reactions including thermalization of neutrons [79]. The characteristic gamma energies of most important elements relating to coal coming from the process of TNC and NIS is shown in Table 2.5. The cross-sections of elements coming from NIS interactions are obtained with neutrons of 14 MeV.

Table 2.5 : Gamma energies used in NAA of some coal elements [83].

Nucleus	Kind of neutrons	Process	Cross section (barn)	Gamma energy (MeV)
^1H	Thermal	TNC	0.33	2.22
^{12}C	Fast	NIS	0.42	4.43
^{14}N	Thermal	TNC	0.08	10.32, 10.82
^{16}O	Fast	NIS	0.31	6.13
^{24}Mg	Fast	NIS	0.645	1.369, 2.754, 3.866, 4.641
^{27}Al	Fast	NIS	0.17	0.843, 1.014
^{28}Si	Fast	NIS	0.52	1.78
^{32}S	Thermal	TNC	0.534	0.841, 2.38, 3.22, 5.42
^{40}Ca	Thermal	TNC	0.431	1.94, 6.40
^{56}Fe	Thermal	TNC	0.256	7.63, 7.64

As different from previous devices, in this study, Nuclear Inelastic and Termal Capture Analysis (NISTA) technique is utilized for analyzing the coal sample as a combination of TNC and NIS [17]. On-line analysis of H, S, Fe, C, O, Si, Al and other elements of bulk material are realized with such a method. In order to use this technique, a 14 MeV pulsed neutron source, a BGO detector, electronic equipment for data recording and processing are put inside the prototype apparatus with neutron shadow shielding.

The neutron emission from pulsed- generator occurs isotropically in the energy range of fast neutrons. The emitted neutrons collide with elements of coal sample. During the operation mode of neutron generator, specific gamma-rays induced by fast neutrons are collected in a file. Especially, detection of gamma rays for C, N, and O elements is critical issue to use PFTNA systems. By turning off the generator, slowing down of neutrons in coal sample occurs via thermal neutron interactions [80]. The characteristic gamma-rays induced by thermal neutron are sort out in another file. So, the separation of spectrum is handled according to energy range of neutrons. The identification of ash content and sulfur element is made by using gamma-rays induced thermal neutron. For this prototype device, BGO detector is decided to detect the gamma-rays rather than NaI(Tl) detector as a conventional choice of scintillation detector. BGO is superior to NaI(Tl), because of having higher efficiency and lower background contribution to main spectrum [80].

The PFTNA technique is chosen for design the prototype apparatus, since it has some advantages by comparing the other techniques. It can be thought as requirements of analysis for total carbon and O element in moisture of coal sample, exposing to lower radiation due to switching off the generator, analyzing the multi-seams as the criteria of great performance in choosing PFTNA.

In order to analyze the spectrum of PFTNA , the most common methods are LLS and Single Peak Analysis (SPA) upto now. For SPA, matrix of material content and some correction techniques are needed to solve the spectrum in addition to weight factor of elements. But for a sample including too many elements, applying of correction is not easy to perform. Since density of gamma flux is significantly altered indistinguishably linked with entirely contents of sample [81]. For these reasons, it is noticed that LLS produce a solution about double time better than SPA method [82].

By using MCLLS, the issue of material content matrix can be solved spontaneously. The Monte Carlo technique is designed to produce elemental libraries for interested sample by taking into account the prompt gamma ray contributions from shielding material and mixture of elements. In addition to this, non-linear effects of detection system can be fixed with MCLLS method. The obtained algorithm from elemental library is set to on-line coal analyzer device via neural network programs. The results of the analysis are displayed on a screen for an operator to visualize easily. During

a movement of coal sample on conveyor belt, the operator can perform classification process without having an extensive knowledge.

2.3.1.2 The MCLLS Method for Element Analysis

MLR is a very useful mathematical method to represent any unknown coal spectrum as the sum of relative elemental contents. Mathematical formulation of the MLR was given below in equation 2.21.

$$y_i = \sum_{j=1}^n a_{ij}x_j \quad (i = 1, 2, 3 \dots 1024) \quad (2.21)$$

where the parameter a_{ij} is the count rate of spectrum at the i th channel for the relative element library in the spherical geometry for j th element, x_j is the relative content of j th element, y_i is coal spectrum at the i th channel. From equation 2.21, the least square criterion is used to find the most probable value (M) as shown in equation 2.22.

$$M = \sum_{i=1}^{1024} (y_i - \sum_{k=1}^n a_{ik}x_k)^2 \quad (i = 1, 2, 3 \dots 1024) \quad (2.22)$$

where M is the most probable value for elemental contents of the given related net coal spectrum. By assuming the a_{ij} is constant, the partial derivative of the term of M with respect to the elemental contents shown by x_j is set to zero given by equation 2.23

$$\frac{\delta M}{\delta x_j} = \sum_{i=1}^{1024} (y_i - \sum_{k=1}^n a_{ik}x_k) a_{ij} \quad (2.23)$$

The matrix form of the equation 2.23 was shown in equation 2.24. In order to calculate the concentrations of elemental contents, the equation 2.25 was solved for X.

$$A^T Y - (A^T A) X = 0 \quad (2.24)$$

$$X = (A^T A)^{-1} (A^T Y) \quad (2.25)$$

where X is matrix the elemental contents of the coal spectrum, A is the matrix of the combination of the single element library obtained from the simulation, Y is the

matrix of the related net coal spectrum, A^T is the transpose matrix of A, $(A^T A)^{-1}$ is the inverse matrix of the multiplication of A^T and A. By using equation 2.25 , the X matrix is calculated for gamma-ray spectrum of each coal set. So, the element contents of coal sample can be found.





3. EXPERIMENTAL SET-UP AND TEST STUDIES

3.1 BGO Test Studies

BGO is a scintillator detector with a chemical composition $Bi_4Ge_3O_{12}$. BGO is the crystalline form of an inorganic oxide with cubic eulytine structure, colorless, transparent, and insoluble in water. Due to its high density, BGO is a very efficient gamma-ray absorber. BGO has a high effective atomic number (Z) which increases the probability of a photoelectric event at the first interaction site. This helps to reduce inter-crystal scatter, which becomes significant as the individual crystal size is reduced to achieve higher spatial resolution. Because of its high detection efficiency, BGO scintillation crystals are used in applications where a high photofraction is required such as neutron activation analysis. The photopeak efficiency which is the ratio of the number of counts in the total absorption photopeak to the total number of counts as a function of the γ -ray energy was shown in Figure for NaI(Tl) and BGO crystals of 1.5" x 1.5" [84].

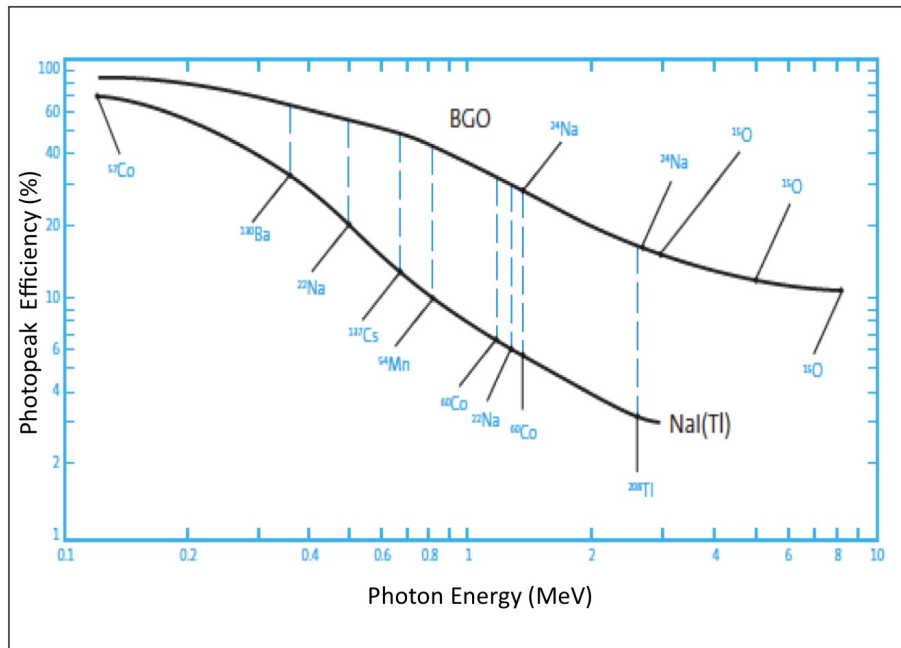


Figure 3.1 : BGO detector of 3"x3" crystal size used in prototype apparatus [84].

With its elevated stopping power, high efficiency scintillation, first-rate energy resolution and non-hygrosopes, BGO is an excellent scintillation material and is ideal for a wide range of applications in high energy physics, nuclear physics, space physics, nuclear medicine, geological prospecting and other industries [85]

In order to obtain the optimum results from the NDA, the gamma-ray spectroscopy system should be known well. BGO detector used in the prototype apparatus was tested to examine the characteristic features. The calibration study of BGO detector includes the measurements of energy calibration, pulse shape analysis, energy resolution, efficiency calibration and linearity and of the detector. These measurements were achieved by using gamma peak energies of ^{137}Cs , ^{60}Co , ^{152}Eu radioactive point sources and delay gamma energy of aluminum source at 2754 keV. The experimental response functions of BGO detector were used to convert the histograms to spectra for MCNP5 and GEANT4 simulations.



Figure 3.2 : BGO detector of 3"x3" crystal size used in prototype apparatus.

3.1.1 Energy calibration

In order to identify unknown samples correctly, the measurements are necessary to be performed accurately. The process of energy calibration has to be done before measurement of the sample. The aim of energy calibration is to present the information which is related to channel number of peak and energy of the interested peak. By using the well known energies of radioisotope sources, the energy calibration of detector is

concluded. It is desired to provide the gamma energies used for calibration process span as a whole of the spectrum.

The energy scale of BGO was adjusted to 10 MeV. The radioactive point sources used for the calibration are ^{137}Cs , ^{60}Co , ^{152}Eu , and delay gamma energy of aluminum source at 2754 keV which is obtained by irradiation from the source of Pu-Be. By using these sources, the energy calibration of BGO was able to achieve up to 3 MeV. The gamma energy, channel number, standard deviation of Gaussian distribution in terms of σ , and FWHM values were shown in Table 3.1.

Table 3.1 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV.

Energy (keV)	Channel #	$\sqrt{E}$ (keV)	σ (keV)	FWHM (keV)
122	12	11.03	13.82	32.477
245	22	15.64	17.37	40.82
344	34	18.55	20.34	47.80
662	64	25.7	28.04	65.90
779	72	27.9	23.94	56.26
964	90	31.04	26.4	62.04
1173	114	34.24	28.88	67.86
1332	130	36.49	30.43	71.51
1408	132	37.53	32.97	77.48
2504	244	50.03	61.53	144.60
2754	268	52.47	93.83	220.50

By using the information of Table 3.1, the relation between channel versus to energy is shown in Figure 3.3. This curve can be used to determine the energy of a photon that is responsible for an unknown peak in the spectrum by using the equation 3.3.

$$Ch\# = a(E) \pm b = 0.0976(E) - 1.5528 \quad (3.1)$$

where a is the slope of the fitting line, E is the energy value in keV, b is the intercept and $Ch\#$ is the channel value. The values of a and b are usually determined with fitting a linear function to all the available points by the software. The relation between energy and channel of BGO detector was found in linear as shown in Figure 3.3. The obtained values were recorded by the software of the detector. Before each measurement, they were recalled to determine an unknown spectrum.

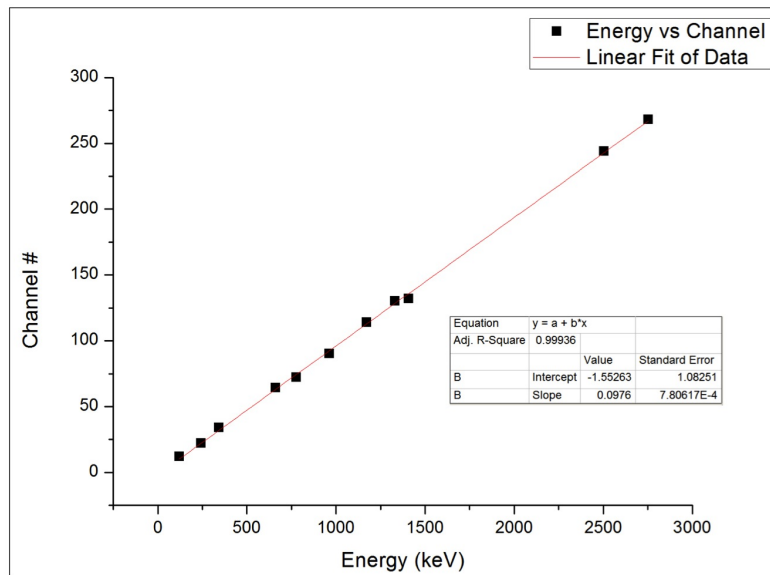


Figure 3.3 : Pulse-shape calibration of BGO detector between 0-3000 keV.

3.1.2 Pulse-shape calibration

For NDA analysis, the pulse shape behavior of the detector should be well known. To accomplish the calibration of pulse shape, the peak width information and related gamma energy are obtained by using known energies of radioisotope sources. The pulse-shape calibration was shown in Figure 3.4.

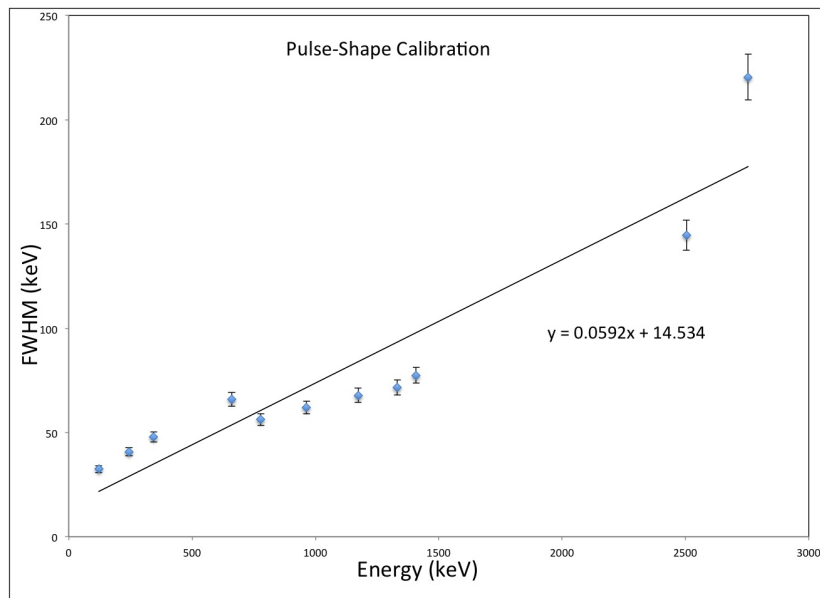


Figure 3.4 : Resolution calibration of BGO detector between 0-3000 keV.

The correlation between FWHM value and energy seems like a product of 3rd degree polynomial function. However, getting infinite FWHM value related gamma energy is

not reasonable. So linear fitting process was applied to experimental data between 0 and 3000 keV.

As seen in Figure 3.4, some scattered point was obtained from the fitting line. There was not obtained a smooth line between FWHM and gamma energy. Because, measuring the peak width experimentally includes some uncertainties related to nature of the spectrum.

3.1.3 Linearity

In theory, the relation between channel number and energy is expected to be linear. But electronic equipment such as MCA and amplifier, connected to detector may not allow to set this relationship. The pulse of an event passing through the amplifier, the channel number of gamma energy can take a different value due to changeable gain value of amplifier. The diagram of pulse height to channel number with different gain values are shown in Figure 3.5. In a similar manner, the gain of ADC in MCA can effect the relation between channel number and value of gamma energy.

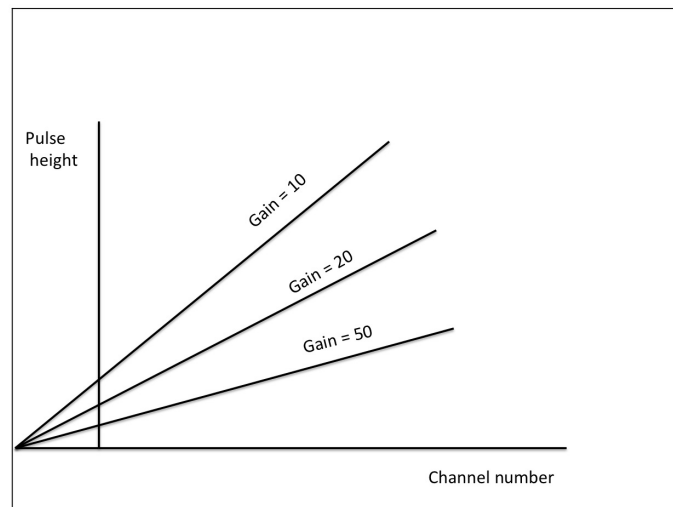
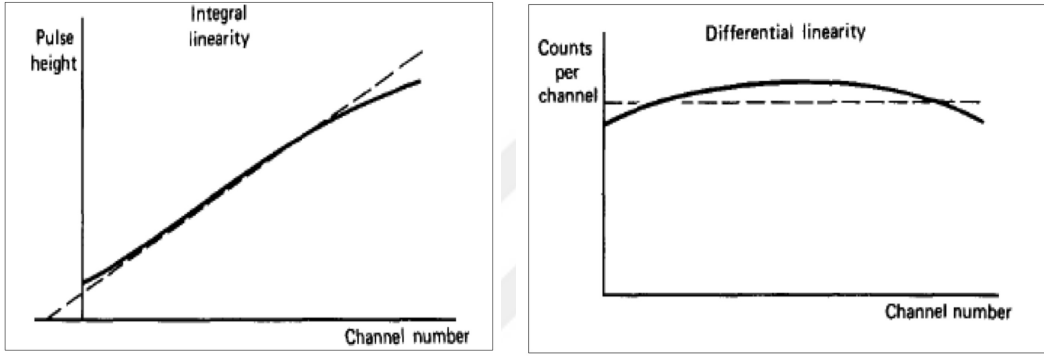


Figure 3.5 : Different MCA calibration plots for three values of amplifier gain [26].

However, after setting the gain value of electronic instruments, the analysis of spectrum is achieved by using known gamma-ray energies of radioisotope source. A relationship is built between their gamma energy values and channel numbers. The relationship usually includes a linear fitting on them. This calibration equation gives significant information about linearity of the detection system. The level of non-linearity has an acceptable value depending on the source of the problem. While non-linearity of ADC system at level of 0.02%, this value can increase up to level of 0.05 for amplifier%

[86]. The increasing of non-linearity causes more faulty measurements. The most common linearity measurements are integral linearity and differential linearity [26]. The integral linearity is obtained by calibration fit from energy versus channel number. The amount of deviation from this calibration fit line is shown as a percent quantity of non-linearity of the device. The differential linearity is related with statistically enough event collection in the channel. The comparison of integral and differential linearity was shown in Figure 3.6. The deviation amount is acceptable up to the value of a few percent [26].



(a) Illustration of integral linearity of MCA.

(b) Illustration of differential linearity of MCA.

Figure 3.6 : The comparative illustration of integral and differential linearities of MCA [26].

In order to obtain the linearity of BGO in experimental, two data sets were used. In first data set, gamma energy values were used between 0 and 3 MeV by using of ^{137}Cs , ^{60}Co , ^{152}Eu sources. The calibration function of first data set was shown in equation 3.2.

$$Ch\# = 0.0976(E) - 1.5528 \quad (3.2)$$

In second data set, gamma energy values were extended up to about 6.5 MeV by adding experimental peaks of H (2.22 MeV), C (4.43 MeV) and O (6.13 MeV). The calibration function of second data set was shown in equation 3.3.

$$Ch\# = 0.1124(E) - 14.728 \quad (3.3)$$

The relation between energy and channel number up to about 6.5 MeV was shown in Figure 3.7.

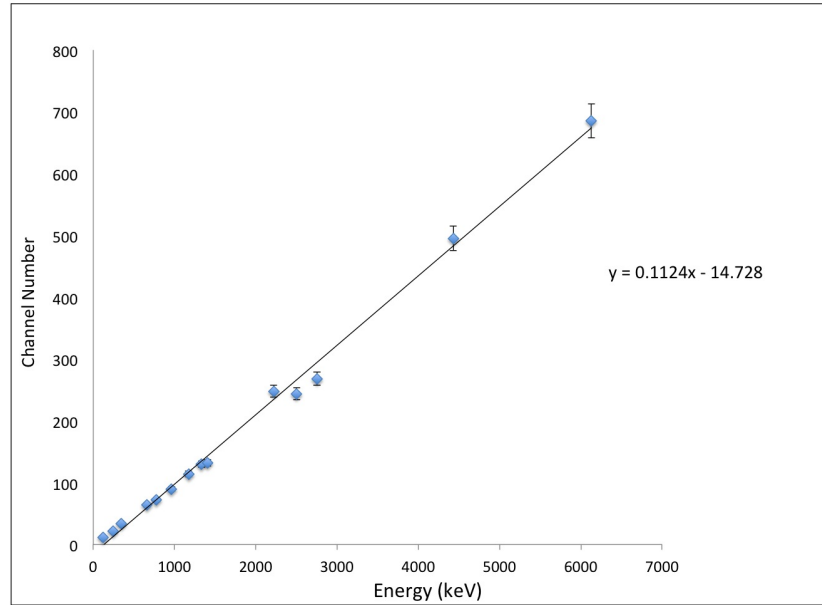


Figure 3.7 : The relation between energy and channel number up to about 6.5 MeV.

The equation 3.2 and 3.3 were used for linearity calculation which was based on determination the difference between fitted and reference energy value. These gamma energy values obtained from equation 3.2 and 3.3 were shown in Table 3.2 and 3.3 for two data sets respectively. For linearity calculation, equation 3.4 was used by using values of Table 3.2 and 3.3.

$$\Delta E = \frac{E_{calibration} - E_{reference}}{E_{reference}} \quad (3.4)$$

Table 3.2 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV.

$E_{reference}(keV)$	Channel No.	$E_{calibration}(keV)$	ΔE
122	12	138.86	0.14
245	22	241.31	-0.013
344	34	364.27	0.058
662	64	671.64	0.016
779	72	753.61	-0.032
964	90	938.04	-0.027
1173	114	1158.31	-0.012
1332	130	1347.88	0.012
1408	132	1368.37	-0.028
2504	244	2515.91	0.004
2754	268	2761.81	0.002

Table 3.3 : Calibration data for Cs-137, Co-60, Eu-152 sources and delay gamma energy of aluminum source at 2754 keV and experimental peaks of H (2.22 MeV), C (4.43 MeV) and O (6.13 MeV).

$E_{reference}(keV)$	Channel No.	$E_{calibration}(keV)$	Δ_E
122	12	233.807	0.92
245	22	322.775	0.32
344	34	429.537	0.247
662	64	696.44	0.053
779	72	767.615	-0.014
964	90	927.758	-0.037
1173	114	1141.281	-0.027
1332	130	1283.63	-0.036
1408	132	1301.423	-0.075
2220	248	2333.451	0.051
2504	244	2297.864	-0.082
2754	268	2511.38	-0.088
4430	495	4530.96	0.022
6130	685	6221.35	0.015

For two data sets, the relations between Δ_E and energy value were shown in Figure 3.8 and Figure 3.9 respectively.

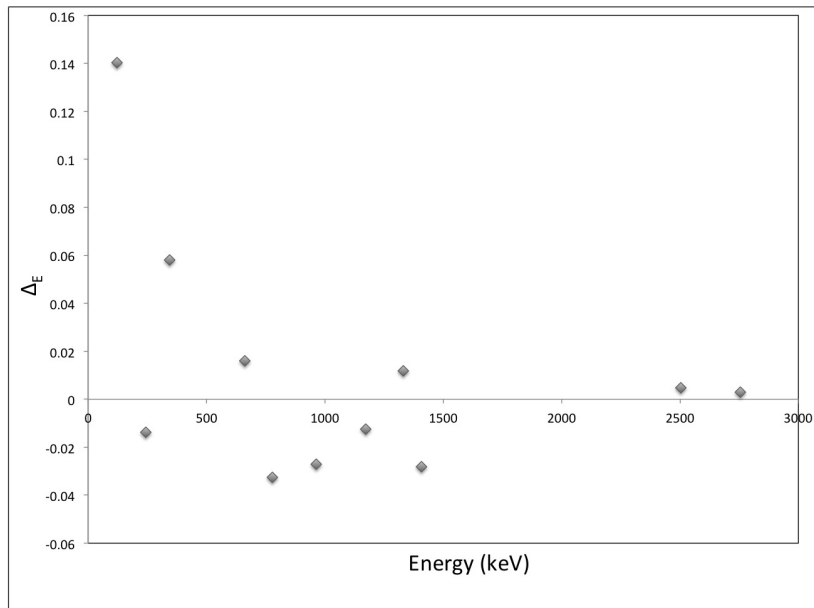


Figure 3.8 : The relation between Δ_E and gamma energy up to about 3 MeV.

As seen in Figure 3.8, there was a non-linearity between 600 keV and 1500 keV by ignoring below of 500 keV. After adding experimental peaks to the calibration, the non-linearity was significantly decreased expect for below of 250 keV as shown in Figure 3.9.

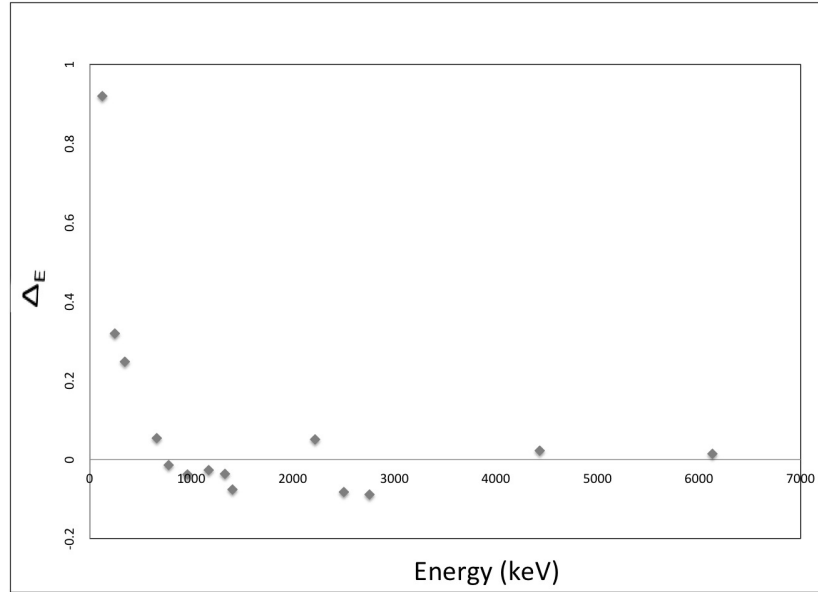


Figure 3.9 : The relation between ΔE and gamma energy up to about 6.5 MeV.

3.1.4 Energy resolution

The pulse height of events coming from a known source (e.g. Co-60 at 1172 keV) should be same in theory. However, the same height value experimentally occupies a few of channels depending on the detector choice. In a spectrum, The Full Width at Half the Maximum height (FWHM) term explains the occupation of channels at specific peak energy. FWHM means a quantity to separate two neighbor peaks. So, this term is a measure for resolution of the detector. The shape of peak shows a Gaussian distribution as shown in Figure 3.10.

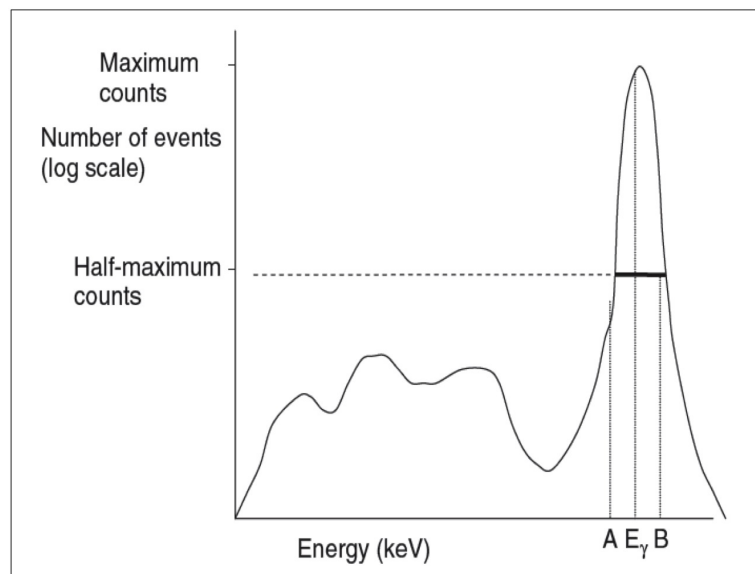


Figure 3.10 : The determination of FWHM for a scintillation detector.

By using the notations of Figure 3.10, The value of FWHM can be found as shown in equation 3.5. The entire calculation of FWHM has to be consistency in units by using either channel number or energy value of keV.

$$FWHM = \frac{B - A}{E_\gamma} \quad (3.5)$$

Resolution of a detector is described in two ways which are relative resolution (%) and absolute resolution in keV. The relative resolution is measured by comparing with resolution of NaI detector at a specific gamma energy. The resolution parameter is mathematically expressed in equation 3.6.

$$R = \frac{FWHM}{H_o} = \frac{2.35K\sqrt{N}}{KN} = \frac{2.35}{\sqrt{N}} = 2.35\sigma \quad (3.6)$$

Where H_o is energy of the centroid peak and R is the resolution of the peak. As the R value approaches to zero, The resolution of the detector becomes higher. The FWHM value increases with the gamma-ray energy. In addition to value of gamma energy, this value is effected by count number of the event, specifications of detector and noise of electronic instruments. The resolution of BGO was calculated by using the equation 3.7

$$R = \frac{FWHM}{E} \times 100 \quad (3.7)$$

where E is the energy of the centroid peak and R is the resolution (%). ^{137}Cs , ^{60}Co , ^{152}Eu were used to measure the energy resolution. between 0-3000 keV. The Table 3.1 includes the FWHM values obtained from PeakFits analysis program applied on Maestro software of Ortec. The relation between FWHM and energy was shown in Figure 3.11.

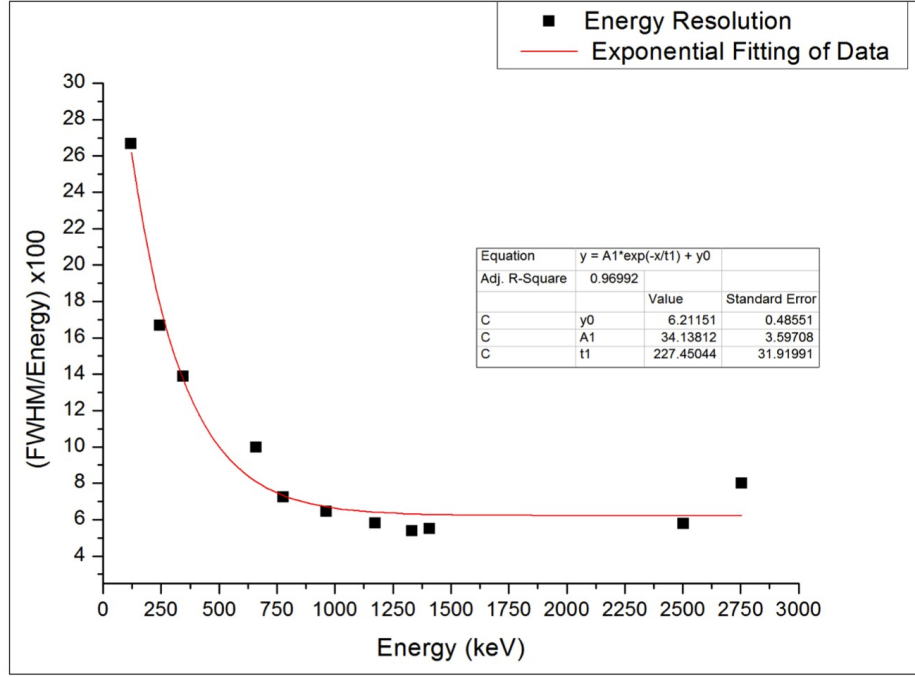


Figure 3.11 : Energy resolution of BGO detector between 0-3000 keV.

3.1.5 Response functions of BGO

The other significant thing is to determine the Gaussian distributions of peaks obtained from the experiment. There is a relation between energy and Gaussian behavior of a peak as shown in equation 3.8.

$$\sigma = \alpha \pm \beta \sqrt{E} \quad (3.8)$$

where E is the energy in keV, σ is standard deviation of Gaussian distribution, β is detector dependent coefficient and α is a correction constant obtained from the fitting line. By using equation 3.8 and experimental value at Table 3.4, Gaussian distributions of peaks were obtained up to 4.5 MeV for BGO detector. The equation 3.8 was used to convert the gamma-ray histograms of MCNP5 and GEANT4 simulations to gamma-ray spectrum of MCNP5 and GEANT4. Therefore, MCLLS measurements were able to achieve more accurately. The response functions were obtained experimentally by using the fitting function of Figure 3.12. The equation 3.9 was used as response function.

$$\sigma = 4.6561 \pm 0.7979 \sqrt{E} \quad (3.9)$$

Table 3.4 : The response functions of BGO detector by using Cs-137, Co-60, Eu-152 sources and C (4.43 MeV).

Energy (keV)	$\sqrt{E}$ (keV)	(σ) (keV)
122	11.03	13.82
245	15.64	17.37
344	18.55	20.34
662	25.7	28.04
779	27.9	23.94
964	31.04	26.4
1173	34.24	28.88
1332	36.49	30.43
1408	37.53	32.97
2504	50.03	61.53
4439	66.62	50.57

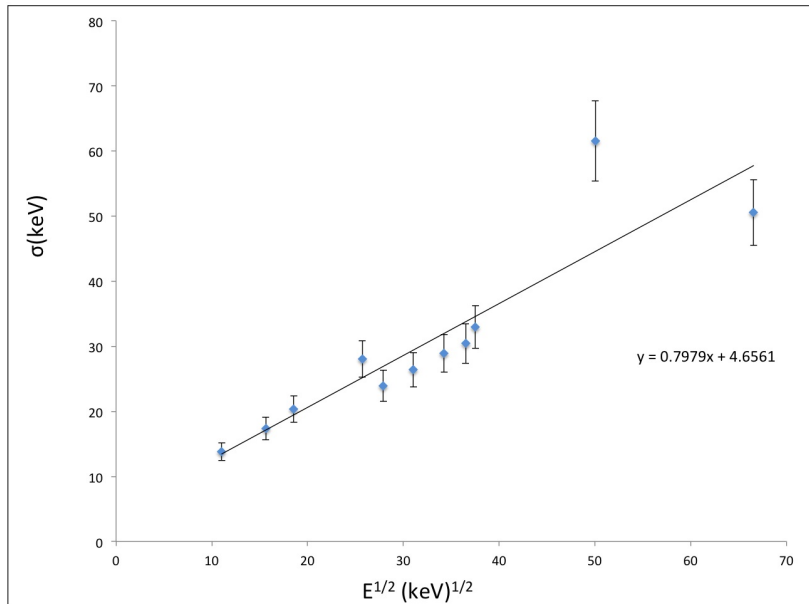


Figure 3.12 : Response functions of BGO detector between 0 and 4.5 MeV.

3.1.6 Efficiency calibration

There is a relation between sensitivity and efficiency of a gamma detection system. Generally, each reaction occurred inside of detector, causes an output signal as a result of interactions of incident particles with detector material. The charged particles can be detected via radiation of ionization or excitation as soon as they reach detector material. They can be detected and counted without any information loss. So, their counting efficiency is assumed to be a value of 100 %. However, charge-less particles expose an important interaction before their detection. Their counting efficiency usually can

not reach to a value of 100 %. Therefore, for an incident gamma-ray, the counting probability of it is said to be efficiency of the detector. There are four types of efficiency explained in detailed.

- Single escape efficiency: The measurement is based on the peak of 511 keV. This peak is obtained by escaping of one annihilation gamma rays from detection.
- Double-escape efficiency: The measurement is based on the peak of 1022 keV. This peak is obtained by escaping of two annihilation gamma rays from detection.
- Full-energy peak efficiency: It is found by using the relation between obtained gamma counts of interested gamma energy of the net peak and emitting gamma counts of the source. The full-energy peak efficiency is calculated with equation 3.10

$$\varepsilon_P(E) = \frac{N_P(E)}{F(E)} = \frac{N_P(E)}{AI_{\gamma(E)}} \quad (3.10)$$

where $N_P(E)$ is the net counts of peak, A is the activity of radioisotope source, $I_{\gamma(E)}$ is the intensity of gamma energy. The full-energy peak can be found as a product of geometrical efficiency and intrinsic efficiency as shown in equation 3.11.

$$\varepsilon_P(E) = \varepsilon_G \varepsilon_I(E) \quad (3.11)$$

where $\varepsilon_P(E)$ is the full-energy peak efficiency, ε_G is geometrical efficiency, and $\varepsilon_I(E)$ is intrinsic efficiency which depends on detector characteristic. The geometrical efficiency is a function of solid angle for a point source as shown in equation 3.12.

$$\varepsilon_G = \frac{\omega}{4\pi}, \omega = 2\pi \left(1 - \frac{d}{\sqrt{d^2 + r^2}}\right) \quad (3.12)$$

where r is the radius of detector, d is the distance of source to detector, and ω is solid angle between source and the detector related only the geometry between source and the detector. The geometric efficiency is defined as the number

of particles incident on the detector in a given interval divided by the number emitted by the radiation source in the same interval depending on the solid angle of source on the detector.

- Total detector efficiency: It is found by using the relation between obtained whole gamma counts of the spectrum and emitting gamma counts of the source as shown in equation 3.13.

$$\epsilon_T(E) = \frac{N_T(E)}{F(E)} = \frac{N_T(E)}{AI_{\gamma}(E)} \quad (3.13)$$

There are three types of total efficiency which are intrinsic, absolute, and relative efficiency as listed below.

- The intrinsic efficiency is described the probability of a gamma-ray emitting from a radioactive source reaching the detector to being recorded by the detector.
- The absolute efficiency is the product of geometric and intrinsic efficiency. It is defined as the counting probability of emitted gamma-ray from a radioactive source.
- The relative efficiency: It is used to compare the efficiency of a detector with NaI at specific distance and value of gamma energy. For instance, The efficiency of germanium detector with a crystal of 3 inch at 1332 keV ^{60}Co and 25 cm distance is defined as equation 3.14.

$$\epsilon_{rel\text{toNaI}}(Ge) = 100 \times \frac{\epsilon_{tot}(Ge, 1332\text{keV})}{\epsilon_{tot}(NaI, 1332\text{keV})} \quad (3.14)$$

3.1.6.1 Intrinsic Efficiency Measurement of BGO

By using equation 3.15, Intrinsic efficiency calibration was performed with ^{137}Cs , ^{60}Co , and ^{152}Eu radioactive point sources at 10 different distances. The intrinsic efficiency is defined as follows:

$$\epsilon(E_{\gamma}) = \frac{P_{TL}}{A \cdot f_{\gamma} \cdot TT} \quad (3.15)$$

where P is the peak area for the corresponding gamma energy value in count number , TT is real time in second, TL is live time in second, A is the activity of the radioactive source in Bq.

In order to calculate the efficiency, the activity values must be determined for the related measurement day. The disintegration of a radioactive element is explained exponentially via a decay process. The relation between a number of given nucleus and disintegrated nucleus is given in equation 3.16.

$$dN(t) = -\lambda N(t)dt, (\lambda = \ln 2/t_{1/2}) \quad (3.16)$$

where N(t) is number of given nucleus and λ is decay constant which is related to half-life ($t_{1/2}$) of the nucleus. By integration of equation 3.16, the number of disintegrated nucleus is found in equation 3.17.

$$N(t) = N_o e^{-\lambda t} \quad (3.17)$$

where N(t) is the number of disintegrated particle at measured time (t), N_o is the number of nucleus before disintegration at time=0. The equation 3.17 is known as the formula of radioactive decay law. The activity of a radioisotope is the average number of disintegrated nucleus per seconds in unit of Curie (Ci) or Becquerel (Bq). The activity relation via decay rate is shown in equation 3.18

$$A(t) = N(t)\lambda = A_o e^{-\lambda t} \quad (3.18)$$

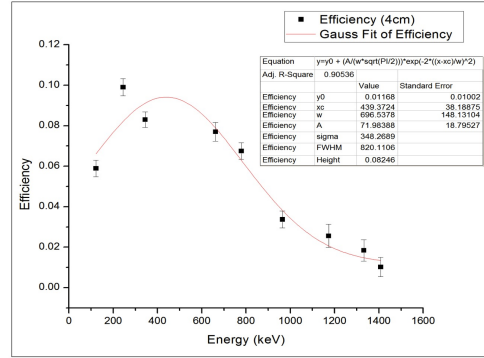
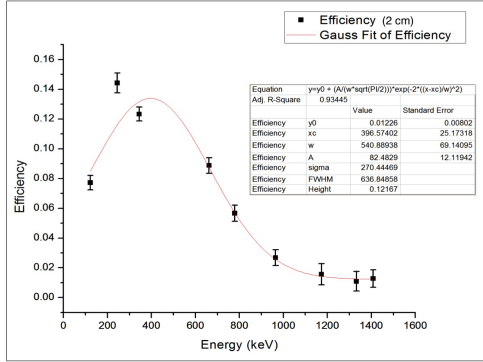
where A_o is the initial activity, t is the elapsed time between calibration date and measurement date. For three radioactive point sources, their activity values and calibration date were shown in Table 3.9.

Table 3.5 : The features of ^{137}Cs , ^{60}Co , and ^{152}Eu sources.

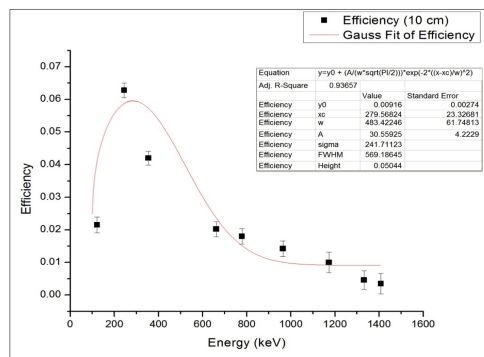
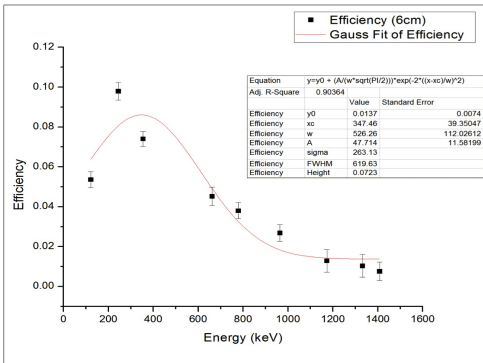
Source	Calibration Date	Activity	Half-life
^{60}Co	15-Nov-06	33000 (Bq)	30.07 y
^{137}Cs	15-Nov-06	34900 (Bq)	5.27 y
^{152}Eu	1-Dec-06	40810 (Bq)	13537 y

The used values for efficiency calculation are area under the peak, true time, live time, intensities of gamma energy, and activity of the source. These parameters were shown between Table A.1 and Table A.10 at appendix section. For three radioactive point sources, the calculated efficiency, total error and uncertainty values at each distances were shown in Table 3.6, Table 3.7, and Table 3.8. The uncertainty of efficiency measurement in quadratic form was shown in equation 3.19. The total error of measurement is contributed from error of area under the peak, true time, intensity of gamma energy and activity of the source. The relation between efficiency and energy were fitted by using exponential function in second order. For each distance, the confidence level of the fit was found greater than 90 % as shown in Figure 3.13, Figure 3.14, and Figure 3.15.

$$\delta\epsilon(E) = \sqrt{\left(\frac{\delta N(E)}{N}\right)^2 + \left(\frac{\delta A}{A}\right)^2 + \left(\frac{\delta I_\gamma(E)}{I_\gamma(E)}\right)^2 + \left(\frac{\delta T}{T}\right)^2 (\epsilon_E)} \quad (3.19)$$

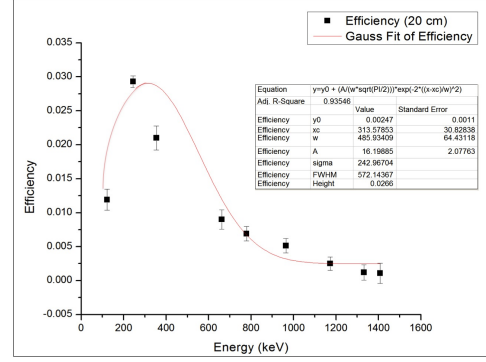
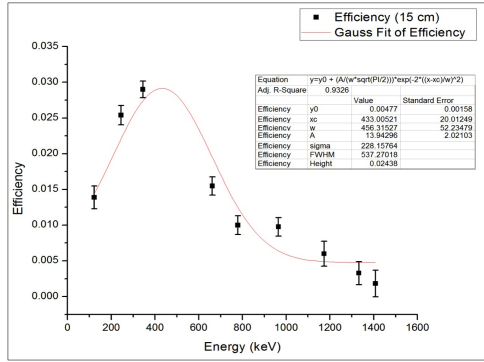


(a) Efficiency measurement at distance of 2 cm. (b) Efficiency measurement at distance of 4 cm.

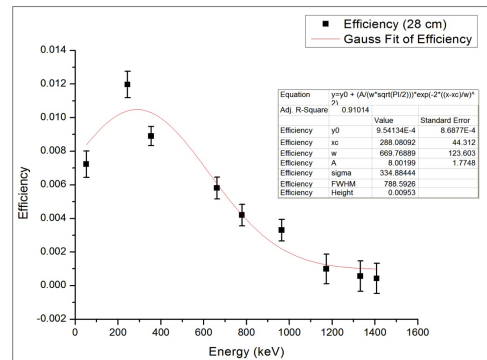
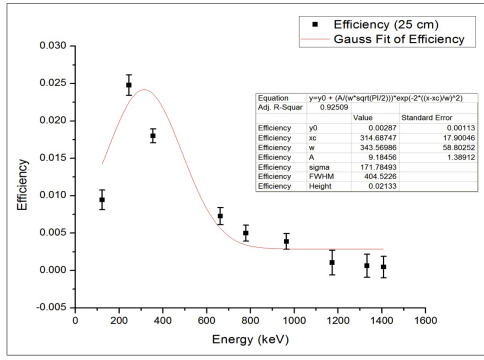


(c) Efficiency measurement at distance of 6 cm. (d) Efficiency measurement at distance of 10 cm.

Figure 3.13 : The relation between efficiency and energy of BGO detector at 2 cm, 4 cm, 6 cm, and 10 cm distances.

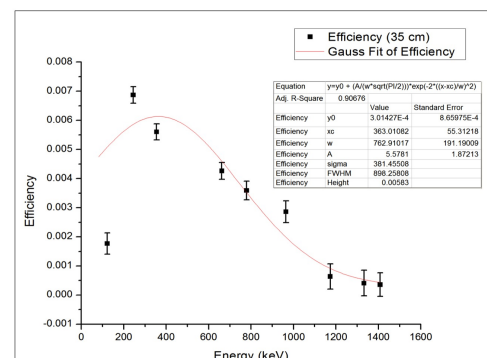
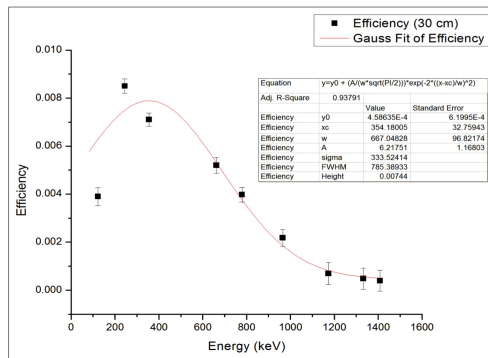


(a) Efficiency measurement at distance of 15 cm.(b) Efficiency measurement at distance of 20 cm.



(c) Efficiency measurement at distance of 25 cm.(d) Efficiency measurement at distance of 28 cm.

Figure 3.14 : The relation between efficiency and energy of BGO detector at 15 cm, 20 cm, 25 cm, and 28 cm distances.



(a) Efficiency measurement at distance of 30 cm.(b) Efficiency measurement at distance of 35 cm.

Figure 3.15 : The relation between efficiency and energy of BGO detector at 30 cm and 35 cm distances.

Table 3.6 : The total errors and uncertainties of efficiency measurements at distances between 2 cm and 10 cm.

E(keV)	Measurement(2cm)			Measurement (4 cm)			Measurement(6 cm)			Measurement(10cm)		
	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε
662	8.88E-02	7.28E-03	6.47E-04	7.7E-02	8.09E-03	8.72E-03	4.52E-02	8.09E-03	3.66E-04	2.02E-02	7.32E-03	1.48E-04
1173	1.56E-02	9.99E-03	1.56E-04	2.56E-02	9.96E-03	2.55E-04	1.28E-02	9.82E-03	1.26E-04	1.0E-02	1.01E-02	6.68E-05
1332	1.09E-02	9.195E-03	1.00E-04	1.84E-02	9.19E-03	1.69E-04	1.03E-02	9.88E-03	1.02E-04	4.6E-02	9.22E-03	4.24E-04
122	7.72E-02	6.86E-03	5.29E-04	5.89E-02	7.13E-03	4.20E-04	5.36E-02	6.91E-03	3.70E-04	2.15E-02	7.55E-03	1.62E-04
2445	1.44E-02	9.38E-03	1.35E-04	9.9E-02	7.42E-03	7.34E-04	9.79E-02	7.63E-03	7.47E-04	6.28E-02	7.19E-03	4.51E-04
344	1.23E-01	6.55E-03	8.08E-04	8.3E-02	6.61E-03	5.48E-04	7.4E-02	6.56E-03	4.86E-04	4.2E-02	6.69E-03	1.94E-04
779	5.67E-02	7.56E-03	4.29E-04	6.75E-02	7.09E-03	4.79E-04	3.8E-02	7.07E-03	2.68E-04	1.8E-02	7.48E-03	1.34E-04
964	2.68E-02	7.44E-03	1.99E-04	3.37E-02	7.31E-03	2.46E-04	2.68E-2	7.25E-03	1.94E-04	1.4E-02	7.58E-03	1.07E-04
1408	1.27E-02	8.08E-03	1.03E-04	1.02E-02	8.19 E-03	8.35E-05	7.56E-03	8.07E-03	6.10E-05	3.5E-03	9.99E-03	3.49E-05

Table 3.7 : The total errors and uncertainties of efficiency measurements at distances between 15 cm and 25 cm.

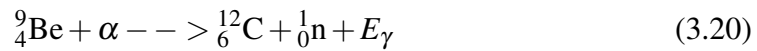
E(keV)	Measurement(15cm)			Measurement (20 cm)			Measurement(25 cm)		
	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε
662	1.549E-02	7.26E-03	1.12E-04	9.0E-03	8.57E-03	7.71E-05	7.28E-03	7.94E-03	5.78E-05
1173	6.0E-03	9.82E-03	5.89-04	2.5E-03	1.023 E-02	2.56E-05	1.06E-03	1.16E-02	1.23E-05
1332	3.28E-03	9.22E-03	2.61E-05	1.19E-03	1.09E-02	1.3E-05	6.31E-04	1.09E-02	6.89E-06
122	1.39E-02	9.13E-03	1.267E-03	1.19E-02	9.1E-03	1.086E-04	9.45E-03	9.18E-03	8.68E-05
245	2.54E-02	7.61E-03	1.93E-04	2.93E-02	9.44E-04	2.766E-05	2.48E-02	9.56E-03	2.37E-04
344	2.9E-02	6.689E-03	1.94E-04	2.1E-02	6.55E-03	1.37E-04	1.8E-02	6.57E-03	1.18E-04
779	1.0E-02	7.512E-03	7.52E-05	6.9E-03	7.24E-03	4.99E-05	5.0E-03	7.46E-03	3.73E-05
964	9.76E-03	7.38E-03	7.21E-05	5.14E-03	7.30E-03	3.75E-05	3.89E-03	7.38E-03	2.87E-05
1408	1.83E-03	1.05E-02	1.93E-05	1.08E-03	8.86E-03	9.55E-06	4.72E-04	1.01E-02	6.44E-06

Table 3.8 : The total errors and uncertainties of efficiency measurements at distances between 28 cm and 35 cm.

E(keV)	Measurement(28cm)			Measurement (30 cm)			Measurement(35 cm)		
	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε	$\varepsilon(E)$	Error(tot.)	δ_ε
662	5.81E-03	7.88E-03	4.58E-05	5.2E-03	8.19E-03	4.26E-05	4.26E-03	7.15E-03	3.05E-05
1173	9.9E-04	1.06E-02	1.05E-05	6.98E-04	1.14E-02	7.96E-06	6.38E-04	1.08E-02	6.89E-06
1332	5.66E-04	1.09E-02	6.64E-06	4.83E-04	1.10E-02	5.34E-06	4.11E-04	1.09E-02	4.51E-06
122	7.23E-03	9.42E-03	6.81E-05	3.9E-03	9.32E-03	3.63E-05	1.77E-03	9.05E-03	1.60E-05
245	1.97E-02	9.50E-03	1.87E-04	8.5E-03	7.54E-03	6.41E-05	6.87E-03	7.15E-03	4.92E-05
344	8.9E-03	6.89E-03	6.1E-05	7.11E-03	6.93E-03	4.92E-05	5.6E-03	6.97E-03	3.90E-05
779	4.2E-03	7.79E-03	3.27E-05	3.98E-03	7.77E-03	3.09E-05	3.59E-03	7.97E-03	2.86E-05
964	3.3E-03	7.68E-03	2.53E-05	2.18E-03	8.72E-03	1.90E-05	2.86E-03	9.31E-03	2.66E-05
1408	4.27E-04	1.08E-02	5.60E-06	3.99E-04	1.08E-02	4.81E-06	3.59E-04	1.02E-02	4.42E-06

3.2 Coal Spectrum Obtained By Pu-Be Source

One of neutron production method is alpha induced-reaction. A radioisotope is used as a source of alpha emission. The radioisotope and an element with a low atomic number are combined to obtain the required interaction. Mostly, ^9Be element is commonly used to interact with alpha particle by reason of having high neutron yield. Other elements used for the reaction are Li, F, and B. The mechanism of neutron production is performed as shown in equation 3.20.



where E_γ is equal to 4.44 MeV which is de-excitation of ^{12}C . The most common radioisotopes and their features are listed in Table 3.9. The selection of radioisotope determines the strength of the neutron source.

Table 3.9 : The comparison of alpha- neutron sources.

Nuclide	Half-life	Neutron yield	Average E_n (MeV)	Gamma dose rate
$^{238}_{94}\text{Pu} / ^{238}_{94}\text{Pu}$	24,114 y.	$2.0 \cdot 10^6$ n/s per Ci	4.2 – 5 (max:11)	0.1 mrem/hr at 1 m/Ci
$^{241}_{95}\text{Am}$	432,2 y.	$2.4 \cdot 10^6$ n/s per Ci	4.2 (max:11)	2.5 mrem/hr at 1 m/Ci
$^{226}_{88}\text{Ra}$	1,600 y.	$15 \cdot 10^6$ n/s. per Ci	3.6 (max:13.2)	9.17 mrem/hr at 1m/Ci

After completion of BGO calibration, measurements of coal sample obtained from Yeniköy were studied. Until customs procedure of neutron generator was completed, First measurements were conducted by using a Pu-Be neutron source as shown in Figure 3.16. Pu-Be was used for alpha-neutron sources.



Figure 3.16 : The experimental set-up of Pu-Be neutron.

The Pu-Be neutron source was placed in a tank of cylindrical volume which is called howitzer. For thermalization of neutrons, howitzer volume was filled with paraffin material (C_nH_{2n+2}). The Pu-Be neutron source was positioned inside of a port which was nearly found at the center of howitzer.

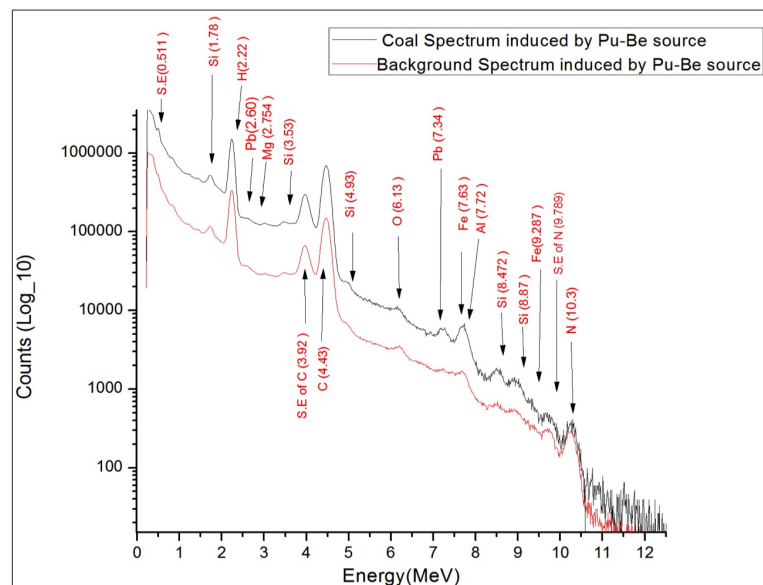


Figure 3.17 : Gamma-ray spectrum of the coal sample induced by Pu-Be neutron source.

The coal sample, BGO detector and neutron source were placed in a geometry of back-scattering. The sample was uninterruptedly irradiated during 24 hours. The collected data by BGO was recorded by using Maestro software of ORTEC which was installed on laptop. The gamma-ray spectrum of coal sample was shown in Figure 3.17. As shown in Figure 3.17 some elements of ash content, C(4.44 MeV), H(2.22 MeV), O(6.13MeV) were observed by using Pu-Be source. Although the identification of coal element was accomplished, observation of gamma-ray spectrum took long time due to the low intensity of the neutron source.

3.3 PFTNA Calibration Process

For PFTNA Process, The neutron production of the neutron generator is performed via D-T fusion reaction which is obtained by interaction with an accelerated beam of ${}^2_1\text{H}$ and ${}^3_1\text{H}$ target as shown in below:



For the prototype apparatus, a neutron generator was used which can operate in pulse mode. The used model of neutron generator was produced by Thermo-Science as shown in Figure 3.18. The neutron generator (ThermoScientific MP320) was equipped with many safety features such as neutron warning lamp, mechanical pressure switch interlock, emergency electrical switch interlock, over/under temperature shutdown, etc.

The neutron yield is about 10^8 n/s with a neutron energy of 14 MeV. Typical lifetime of the neutron generator is 1,200 hours at 10^8 n/s. The neutron generator can be operated in continuous mode if desired. However, it should be kept in mind that non-essential usage of the neutron generator causes run out of operating lifetime earlier. In addition to continuous option, the neutron generator allows to four type of pulse outputs for pulse mode operation which are "SOURCE", "DELAY1", "DELAY2" and "DELAY3". The "SOURCE" pulse output is used to drive the ion source pulse. The neutron generator in itself is adjusted to produce the pulse at frequency of 1 kHz with a 10% duty cycle. It means that it is observed a 100 μs pulse every 1 ms apart. Since pulsing mechanism and emission of neutron cannot happen in same time duration, the input gate signal to detector delays 2 μs which is called latency. The pulse duration is



Figure 3.18 : The neutron generator used for prototype apparatus.

adjusted to cover this latency value. Taking account to this delay, neutron pulse time is adjusted to $6 \mu\text{s}$ for ion current of $60 \mu\text{A}$ and 80kV high voltage.

The adjustable frequency values enable to modify the duration of space between two pulses. Therefore, the duration of interactions induced by thermal and fast neutrons are determined. The control of neutron generator is provided by using a wireless connection via interface. The interface provided by Thermo Fisher Scientific is shown in Figure 3.19.

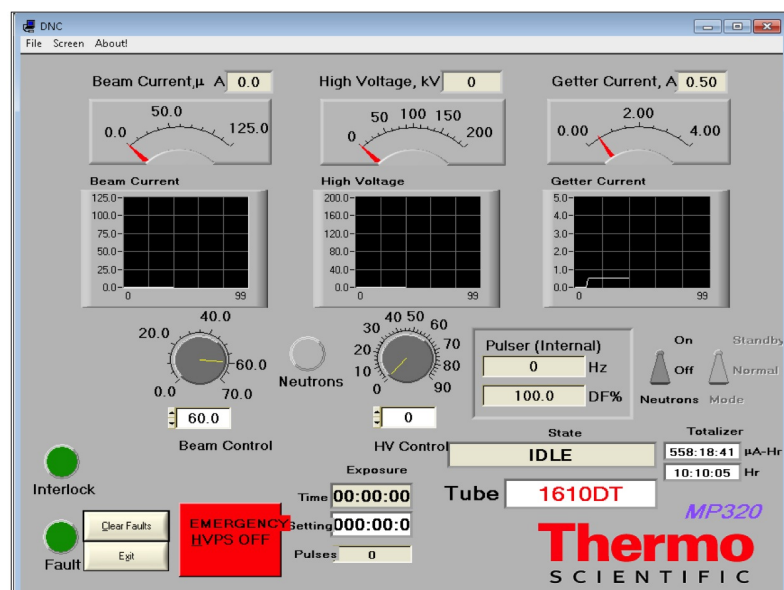


Figure 3.19 : The interface of MP320 neutron generator by Thermo Fisher Scientific.

By using this interface, the operation of the neutron generator is accomplished from far distances without radiation exposure. During operation, the voltage, current and beam current values are observed. For any emergency situation, the warning signs are turned on. In addition to these, operating duration of neutron generator can be set ahead.

The experimental set-up consists of a coal sample, a 14 MeV neutron generator, a BGO (Ortec DigiBASE-E) gamma-ray detection system, blocks of polyethylene material covering the neutron generator for minimizing the radiation, lead shielding for BGO, concrete walls for radiation protection and an industrial pc connection system. The concrete walls were marked from 1 to 6 for the measurements of radiation dose. The experimental system is shown in Figure 3.20.

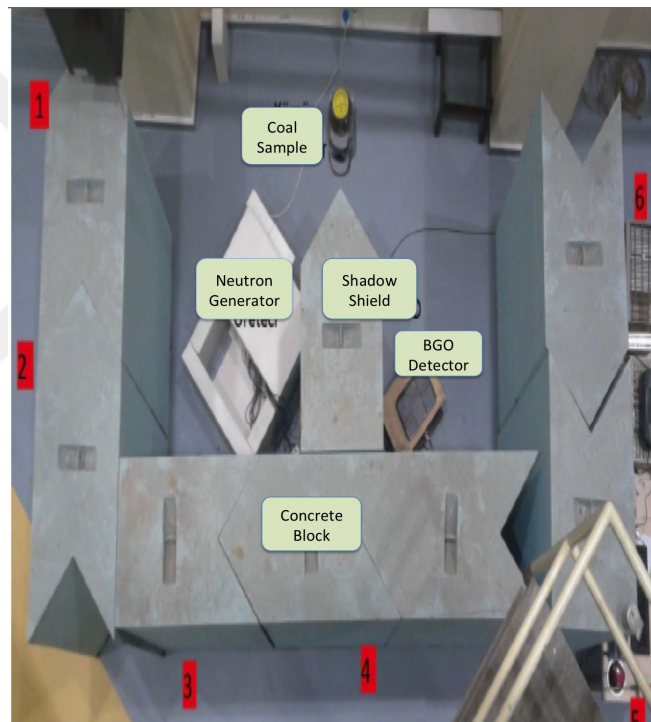


Figure 3.20 : Experimental set-up used for the measurement and the neutron generator.

The output of “DELAY1” of the neutron generator was directly connected to the input of the BGO detector. The industrial PC was to provide the connection between BGO and neutron generator with aid of ports. By using a wireless router adapter, the information exchange was provided among industrial and laptop PC which was installed the interface of neutron generator. Additionally, the installation of the wireless router adapter enables the remote control for radiation protection. The connection set-up of computers is shown in Figure 3.21.

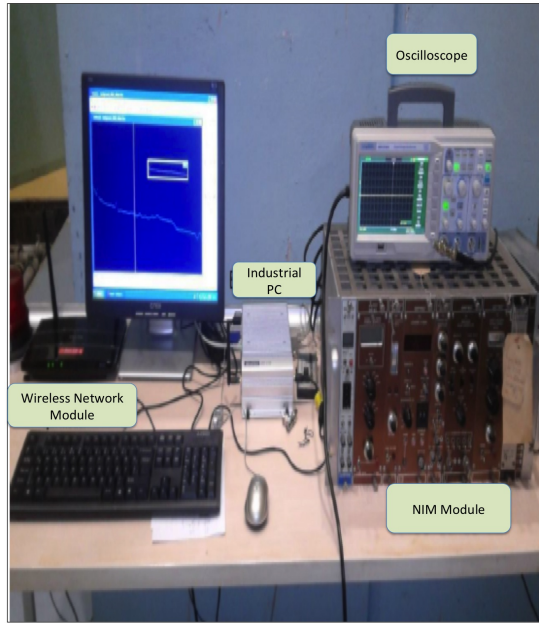


Figure 3.21 : NIM electronics and data acquisition system with the industrial PC.

By considering the elemental contents of the lignite coal reserve in the Aegean Region as shown in Table 3.12, the experimental eight sets of synthetic coal were prepared with varying elemental contents except sulfur element. Same amount of sulfur element was used to prepare the sets of synthetic coal. Each set was weighted to 2 kg without moisture content. The weight percentages and amount of each content of these sets are illustrated in Table 3.10 and Table 3.11.

Table 3.10 : The weight percentages of compounds in synthetic coal sample (%).

Coal	Set-1 (%)	Set-2 (%)	Set-3 (%)	Set-4 (%)	Set-5 (%)	Set-6 (%)	Set-7 (%)	Set-8 (%)
C	89.35	82.65	76.05	69.32	62.672	56.05	49.9	42.73
Ash	6.65	13.35	19.95	26.67	33.33	39.95	4.61	53.26
S	4.00	4.00	4.00	4.00	4.00	4.00	4.00	4.00

Table 3.11 : Amount of carbon element and oxide compounds inside of the sets (g).

Content	Set-1 (g)	Set-2 (g)	Set-3 (g)	Set-4 (g)	Set-5 (g)	Set-6 (g)	Set-7 (g)	Set-8 (g)
C	1787.00	1653.35	1521.00	1386.75	1253.45	1120.96	988.00	854.7
SiO ₂	54.68	109.34	162.67	217.34	271.99	326.66	379.99	434.67
Fe ₂ O ₃	24.01	46.67	70.66	93.33	117.33	141.33	164.00	188.02
Al ₂ O ₃	24.01	49.33	73.53	98.66	122.66	146.86	171.99	196.01
MgO	6.66	11.99	18.66	25.33	32.00	37.33	44.0	50.67
CaO	24.00	49.33	73.53	98.66	122.66	146.86	171.99	196.00
S	80.00	80.00	80.00	80.00	80.00	80.00	80.00	80.00
Total	2000.36	2000.03	2000.09	2000.10	2000.11	2000.03	2000.07	2000.07

Table 3.12 : The study on the lignite coal, collected from different regions of Turkey [99].

Content (wt.%)	North Marmara	South Marmara	Aegan	Black Sea	Mediterranean	Central Anatolia	East Anatolia
Moisture	31.00 ± 11.00	25.00 ± 9.00	25.00 ± 10.00	16.00 ± 9.50	29.00 ± 12.00	22.00 ± 12.00	31.00 ± 21.00
Ash 815 °C	16.20 ± 6.30	22.20 ± 11.60	21.40 ± 9.90	19.50 ± 10.00	19.00 ± 9.30	27.40 ± 19.50	26.00 ± 11.20
Volatile	25.20 ± 5.00	25.90 ± 4.30	27.90 ± 4.76	32.20 ± 7.00	28.20 ± 5.30	25.70 ± 3.00	24.80 ± 5.50
Fixed C	27.60 ± 5.70	27.2 ± 6.50	25.90 ± 7.90	32.30 ± 7.90	24.30 ± 8.20	24.80 ± 3.70	18.00 ± 11.70
Total S	2.60 ± 1.50	3.10 ± 1.50	2.80 ± 1.90	3.50 ± 2.90	3.40 ± 1.40	3.90 ± 2.00	2.40 ± 1.10
Fe in ash	2.80 ± 1.90	1.70 ± 0.97	2.20 ± 1.30	2.00 ± 1.30	2.40 ± 1.60	3.30 ± 2.40	2.30 ± 1.20
Mg in ash	0.53 ± 0.21	0.30 ± 0.29	0.59 ± 0.40	0.35 ± 0.30	0.52 ± 0.32	0.69 ± 0.34	0.93 ± 0.87
Al in ash	1.8 ± 0.95	2.40 ± 1.10	2.30 ± 1.40	1.50 ± 1.30	1.70 ± 1.70	2.90 ± 2.30	2.60 ± 1.40
Si in ash	3.90 ± 2.30	6.20 ± 4.50	5.10 ± 3.20	5.80 ± 4.70	4.10 ± 4.70	6.40 ± 5.40	8.10 ± 4.20
Ca in ash	0.98 ± 0.48	0.79 ± 0.42	2.30 ± 2.80	1.50 ± 1.30	2.20 ± 1.30	1.20 ± 0.61	3.20 ± 1.40



Figure 3.22 : Synthetic coal samples prepared and used for calibration and test purposes.

For the on-line coal analyzing, Pulsed Fast Thermal Neutron Analysis (PFTNA) technique was used to collect the gamma rays separately from fast and thermal neutron spectra. Operating of neutron generator in pulse mode, neutrons could be produced and stopped in between certain period of time intervals. BGO gamma detector gate was connected to the neutron generator source output to obtain both 5V and 0V pulses with $10\ \mu\text{s}$ with 10% duty cycle and $90\ \mu\text{s}$ duration respectively.

Preparing 8 sets of synthetic coal samples with varying elemental contents, which are determined by taking into account coal reserves in Aegean region of Turkey, PFTNA technique was tested to sort as gamma counts induced by fast and thermal neutrons. The gamma-ray spectrum of coal sample induced by thermal and fast neutron were collected by BGO detector with digiBASE-E module. 1024 channels were spanned to 10 MeV energy. The total spectrum of coal set-1 was shown in Figure 3.23. The experimental data was taken in the format of list mode and then the spectrum was analyzed. Thanks to this data format, the data was in raw form and consequently dead time was minimized. The gamma-ray spectrum induced by thermal and fast neutron for coal set-1 was comparatively shown in Figure 3.24.

As shown in Figure 3.24, some of the elements in synthetic coal sample could be identified by analyzing the spectrum induced by fast or thermal neutrons. The experimental results showed that C(3.68 MeV and 3.85 MeV), O(6.91MeV , 7.12 MeV), Si (1.78 MeV) were observed only in the gamma spectrum of fast neutrons

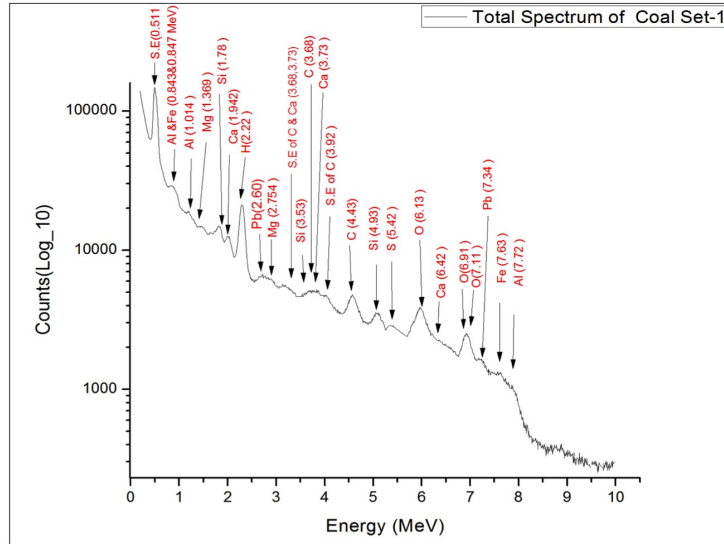


Figure 3.23 : The total gamma-ray spectrum of synthetic coal set-1.

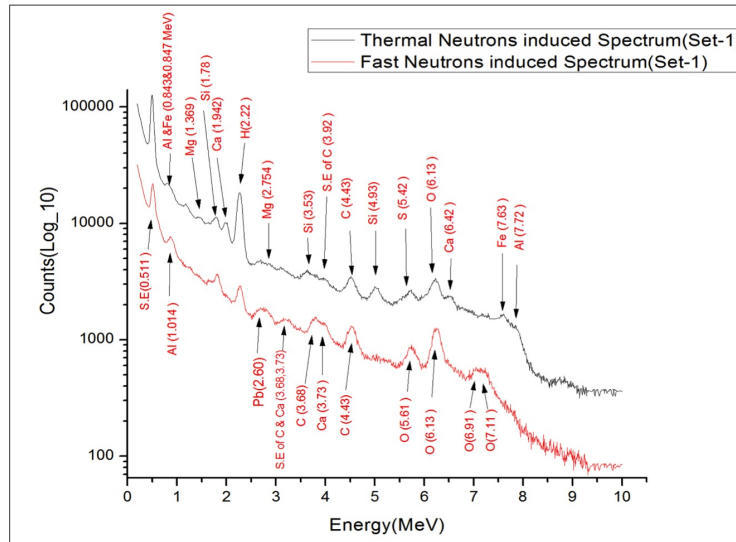


Figure 3.24 : The comparison spectrum of synthetic coal set-1 induced by fast and thermal neutrons.

induced. The gamma energy of elements in ash content which are Fe (7.63 MeV), Al(7.72 MeV), Ca(1.94 MeV, 6.42 MeV), Si(3.53 MeV, 4.94 MeV), Mg(1.38 MeV), were observed only in the gamma spectrum of thermal neutrons induced. In addition, the counts of gamma ray induced by thermal neutrons were more than the ones induced by fast neutrons. Because pulsed-mode duration of neutron generator was longer for thermal neutron output. The irradiation time with thermal neutron was 90 μ s whereas irradiation time of fast neutrons was 10 μ s.

The eight sets of synthetic coal samples were irradiated 10 minutes by 14 MeV neutrons. Total gamma spectrum were shown in Figure 3.25. According to gamma-ray

spectrum in Figure 3.25, there was not noticeable difference among gamma-ray spectra of synthetic coal sets related to their concentrations.

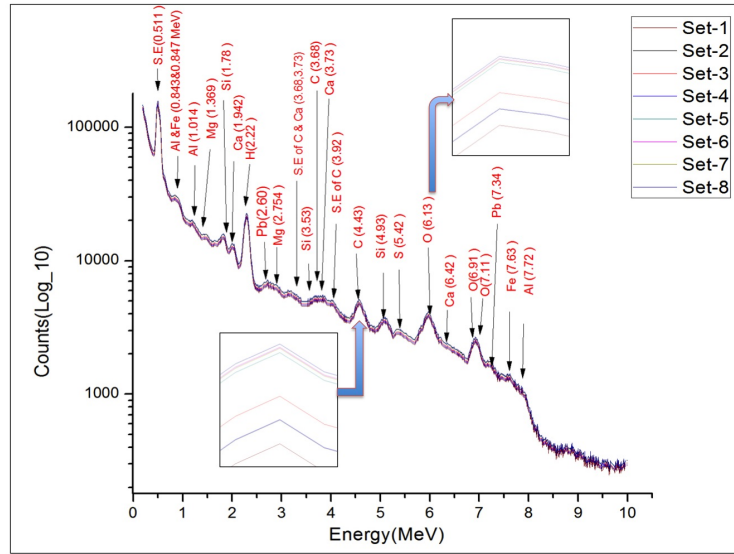


Figure 3.25 : The comparison spectrum of 8 synthetic coal sets induced by fast and thermal neutrons.

The gamma ray counts of C (4.43 MeV) was expected to be decreased from coal set-1 to coal set-8 based on Table 3.10 and Table 3.11. Similarly, the gamma ray counts of O (6.13 MeV) was expected to be increased from coal set-1 to coal set-8 based on contribution from ash content. However the desired relation could not be obtained as seen in Figure 3.25. One of the reason might be thought that the background contribution obtained from concrete walls and polyethylene blocks used to cover the generator surpassed the coal spectrum. It was concluded that amount of synthetic coal sample was not good enough to determine the concentration of element contents in coal sample. So it was decided to increase the amount of coal sample for the later studies. The gamma and neutron dose values of behind the concrete walls in experimental set up of Figure 3.20, were shown in Table 3.13. In order to provide the radiation protection, The experiment was conducted at 50m away of experimental set up by using wireless access.

Table 3.13 : The dose rate of neutron and gamma accumulated on the concrete walls.

Type of Dose	Wall-1	Wall-2	Wall-3	Wall-4	Wall-5	Wall-6
Neutron	90 mRem/h	2.5 mRem/h	1 mRem/h	3mRem/h	0.7mRem/h	0.4 mRem
Gamma	26 μ Sv/h	1.3 μ Sv/h	0.8 μ Sv/h	2.35 μ Sv/h	0.4 μ Sv/h	0.4 μ Sv/h



4. MONTE CARLO SIMULATION METHODS

Monte Carlo methods are used to solve complex problems that are based on the statistical principles in many areas especially for the mathematical and physical problems [88]. Unlike deterministic methods using definite equations to describe the behavior of the interested physical problem, the Monte Carlo methods simulate the real processes based on the probabilistic nature of the physical interactions. The algorithms using the Monte Carlo methods are mostly written in C++ or Fortran programming languages and realize the simulation with the aid of random number generators. The random number generator stochastically chooses a number between 0 and 1 to mimic the event of the physical process including the information of interaction type of particles between each other and medium, material of structure, geometry of the simulation and cross section data. When a particle of the event is created, the history of random number generator is started to collect the whole possible physical information upto ending of the event. The schematic diagram of a random event starting from the neutron scattering to the photon capture is shown in Figure 4.1 [87].

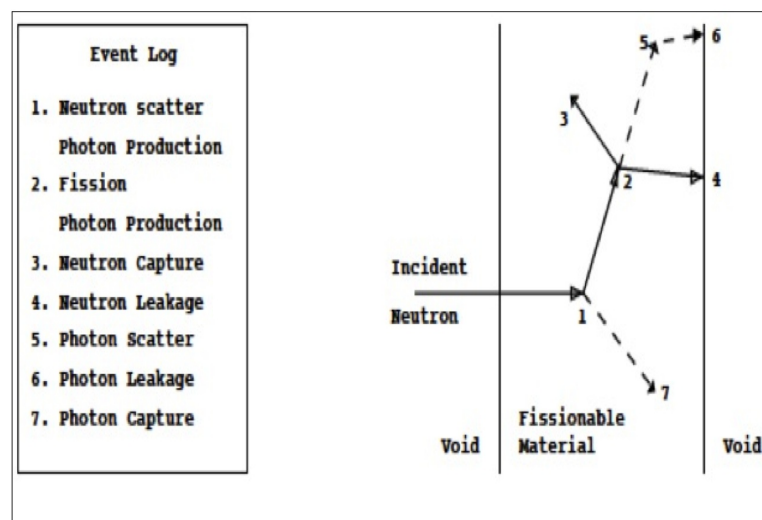


Figure 4.1 : Random history of a neutron incident on a slab of material [87].

The production of a new particle, scattering, capturing and its direction of travel are produced according to the probability of occurring the physical interactions which is represented by random number generator. The procedure of the collection of events is repeated and recorded up to the end of the last history of the created particles.

The most used simulation programs based on Monte Carlo method are CEARCPG, MCNP5, and GEANT4. CEARCPG, written in Fortran language, was developed by Center for Engineering Application of Radioisotopes (CEAR) group at North Carolina State University. CEARCPG having the geometry package of MCNP4B which is old version of MCNP5 can be used for PGNA simulation [89]. Another simulation program based on Fortran language is Monte Carlo N-Particle (MCNP) which was created by a group at the Los Alamos National Lab. It can simulate complicated 3D geometries with the aid of extended widely in cross-section data libraries of continuous energy spectrum [90]. The other simulation programming, GEANT4 (GEometry AND Tracking) is widely used Monte Carlo system implemented in the C++ programming language for simulating the passage through matter which was developed by CERN (European Organization for Nuclear Research) [91].

In this chapter, the structure of the MCNP5 and GEANT4 simulation program was introduced. The simulation of the prototype apparatus and the generating of the element library for analyzing of coal content were done by using MCNP5 and GEANT4.

4.1 The Structure of MCNP5

As a general-purpose, MCNP5 is powerful modelling tool which is defined continuous energy, generalized-geometry, time dependent. It includes various particles in Monte Carlo transport code. These can be neutron only, photon only, electron only, combined neutron/photon or photon/electron transport [87]. The transport code is chosen according to characteristics of the problem of interested.

In order to execute the MCNP5 simulation, the geometry of the simulation including material information, the cross sections of the physical interactions must be well defined in the input text file as data cards. According to the identification of the problem specified by users, the simulation program records the data as a track of an

event coming from the source to the termination in the bounded volume as an output text file. A typical input file of MCNP5 contains three main groups of command lines called as "cards". These are respectively cell cards, surface cards, and data cards whose lines includes 80 columns obligatory having first five blank columns.

- The cell cards define an object, which consist of importance number, the surface information, the number of the material inside of the cell, density of the material in units of g/cm^3 , bounded by surfaces. Each cell has an importance number as 0 or 1. The importance number of the cell provides a variance reduction. The interaction of particles is tracked in the cells with importance numbered by 1 whereas the particles are terminated in the cell numbered by 0.
- The surface cards give the information of the geometry of the problem. The line of surface cards includes the specific parameters of surfaces which are surface number, dimensions and radius in cm in a Cartesian coordinate system. The kind of surface can be a plane, cylinder or sphere that bounds a cell.
- The data cards consist of card combinations which include information about energy, type and location of the source, the information of materials and their cross sections. Also the type of tally according to specification of the problem must be added into data cards with information of cut-offs and variance reductions if needed. The member of data cards are mode cards, material cards, physics cards, tally cards, and history cards.
 - Mode cards allow only the selected transport kind of particle such as "MODE P" for photon transport. If any Mode is not selected, the Mode N runs and only neutron transport is allowed for the simulation.
 - Material cards gives the specification of material which has individually atomic mass of elements and the atomic fraction of the elements of the compound and the information of cross section data. Nearly for one hundred isotopes and elements, there are more than five hundred types of neutron interactions including various temperature information. Photon interactions are also available for all element atomic number from $Z=1$ through $Z=94$. MCNP5 data packages are included with cross section information of dosimetry and activation interactions.

The main data of nuclear interactions is obtained from the Evaluated Nuclear Data Files (ENDF) system [92], Evaluated Nuclear Data library (ENDL)" [93] and Activation Library (ACTL) [94] and collected from Applied Nuclear Science (T-2) [95–97] Group at Los Alamos.

- The physics cards are used to define the photon interactions in detailed. This card allows the energy cut-off with a limitation of maximum energy, selection of information of coherent scattering if requires, and production of electrons for the problem including bremsstrahlung model. One of the seven tally cards is used to obtain the desired result of Monte Carlo calculations by using sources of neutron, photon and electron. While the energy of photon and electron sources are allowed from 1 keV to 1000 MeV, the energy of neutron source varies from 10^{-11} MeV to 20 MeV. Besides the flux calculations and the energy bins from zero energy to energy of source can be set for the related tally. The tally cards and their specifications are given in Table 4.1 [98].
- The history (nps) card includes the information of the number of particles that is created by the source. As the source is reached the end of the history, the MCNP simulation is terminated at the same time.

Table 4.1 : The tally cards and their specifications [98].

Tally Type	Specification	Particle Type
F1	Surface current	n, γ , e
F2	Surface flux	n, γ , e
F4	Track length estimate of cell flux	n, γ , e
F5	Flux at a point	n, γ
F6	Track length estimate of energy deposition	n, γ ,
F7	Track length estimate of fission energy deposition	n
F8	Energy distribution of pulses created	γ , e

The output of MCNP as an ASCII file involves the complete lines of the input file, the calculation results of the desired tally type, the relative error (R) if occurred. The R value of the related tally is inversely proportional to $\sqrt{N}$ where N is history number. The R values play a role for the statistically significance of the desired tally output. Depending on the type of source and geometry of the detector, the reliability of the value is well defined by manual of the MCNP software. Since the N is related to computer time (T), R value is associated with $C/\sqrt{T}$ where C is a positive constant

for the desired tally. In order to reduce R value, either the T is increased or C is reduced. The option of increasing T is limited by computer budget. For instance, If obtaining the R value of 0.1 lasts 3 hours, decreasing it up to 0.01 which is considered reliable for many situation, is needed 300 hours. Because of this, reducing C is applied by using variance reduction techniques. There are several kinds of variance reduction techniques which can be properly used according to type of the problem. The most common ones are energy cutoff, time cutoff, geometry splitting/Russian roulette, energy splitting/Russian roulette, weight cutoff/Russian roulette.

- Energy cutoff: For saving computer time, the user defines the energy range of followed particles.
- Time cutoff: Time cutoff is used based on time in like manner as energy cutoff.
- Geometry Splitting/Russian roulette: The analogy of geometry splitting is based on adjusting of the importance of the geometry in simulation by user. When a particle passes through a cell having a parameter of higher importance, it is separated in two or more particles proportionally importance number of entering and leaving cell. The particles in higher importance cell have lower weight whereas the particles in less importance cell gain higher weight. In this manner, the statistic is improved and the total weight remains same as before.
- Energy splitting/Russian roulette: While the particles having more significant energy ranges can be divided very high frequency at the request of user, the particles having less significant energy ranges are less split.
- Weight cutoff/Russian roulette: The particles having low weight are exposed to Russian roulette. Since they are not significant, they are killed to save the computer time. Other particles survive by having high weight to keep the total weight same as before.

4.2 Design of prototype apparatus

The prototype apparatus was composed of three main parts: polyethylene construction, a pulse mode operating neutron generator, a BGO detector. The schematic view of the prototype apparatus was shown in Figure 4.2. Nearly two tons of polyethylene material was used to cover of whole system in order to provide the biological protection from

the neutron radiation coming from the neutron generator. Besides that, a pure iron

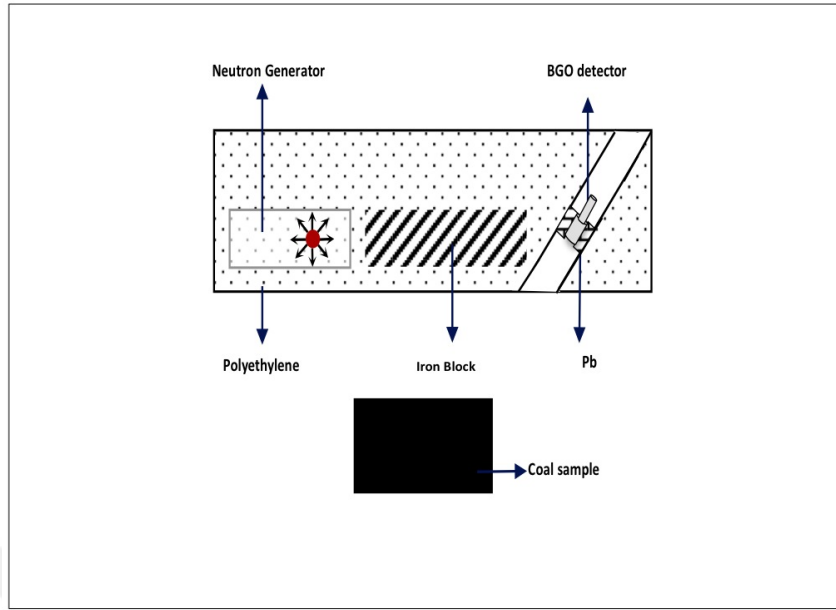


Figure 4.2 : Schematic view of the prototype apparatus.

block which has a length of 60 cm and surface area of $2,25 \text{ dm}^2$, was placed between the neutron generator and BGO detector as a shadow shielding to protect the BGO detector from the damage of 14 MeV fast neutrons. Also, BGO detector was covered with lead shielding to prevent from background radiation of gamma rays.

The BGO detector as a scintillation type detector was preferred the one having a crystal dimension of $3'' \times 3''$. It was placed with an angle of 53° to the ground plate. The placement of the detector was the best possible position to be able to reach the releasing prompt gamma rays of a coal material to the detector. The BGO detector was covered a lead shielding material to avoid the gamma rays coming from interactions of neutrons with polyethylene material and iron block.

The neutron source of prototype apparatus is a D-T neutron generator which can be worked as pulse mode. Because of producing fast neutrons of 14 MeV, the neutron generator is essential to use for prototype apparatus. It was placed to right above the container of the coal sample indicated by a red dot in the schematic view of the virtual experimental set up as shown in Figure 4.2.

4.2.1 MCNP5 simulation of prototype apparatus

The process was aimed to simulate the geometry of the prototype apparatus and interaction of the synthetic coal material with fast and slow neutrons emitted 10^9

neutrons as a total number of particles. In the simulation, neutrons of 14 MeV were emitted from a point source. The neutron generator of prototype apparatus was represented by using this point source.

The geometry of prototype apparatus was obtained by using the cell cards and surface cards of MCNP5. The front (x-z plane) view and the top (x-y plane) view of the geometry of prototype apparatus were shown in Figure 4.3 and Figure 4.4 respectively. The cells being consist of different materials were illustrated with different color codes. Each cell was included the information of material density and importance of the cell. The same numbering was used to represent materials inside of the cells in Figure 4.3

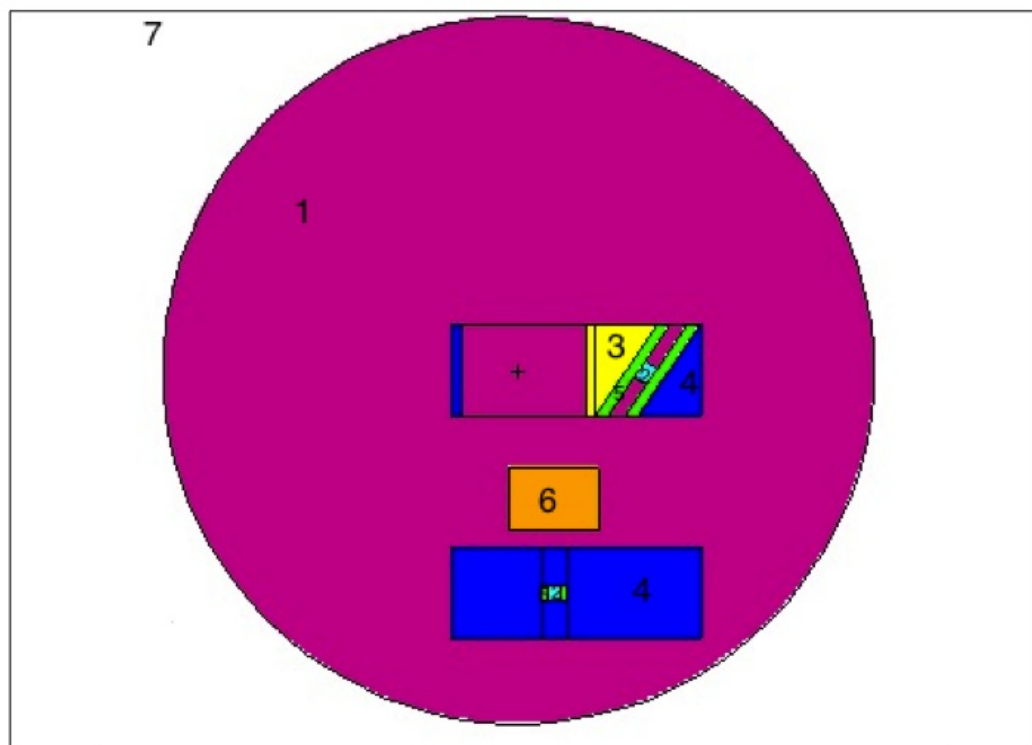


Figure 4.3 : Front view of the prototype apparatus geometry obtained by MCNP5.

and Figure 4.4. The properties of the cells were explained as below:

- Cell#1 was composed from air medium which is the volume outside of prototype apparatus and synthetic coal material.
- Cell#2 represented the crystal volume of BGO detector including material, dimension and position.
- Cell#3 represented the volume of iron block lying between the neutron generator and lead shield of the detector.

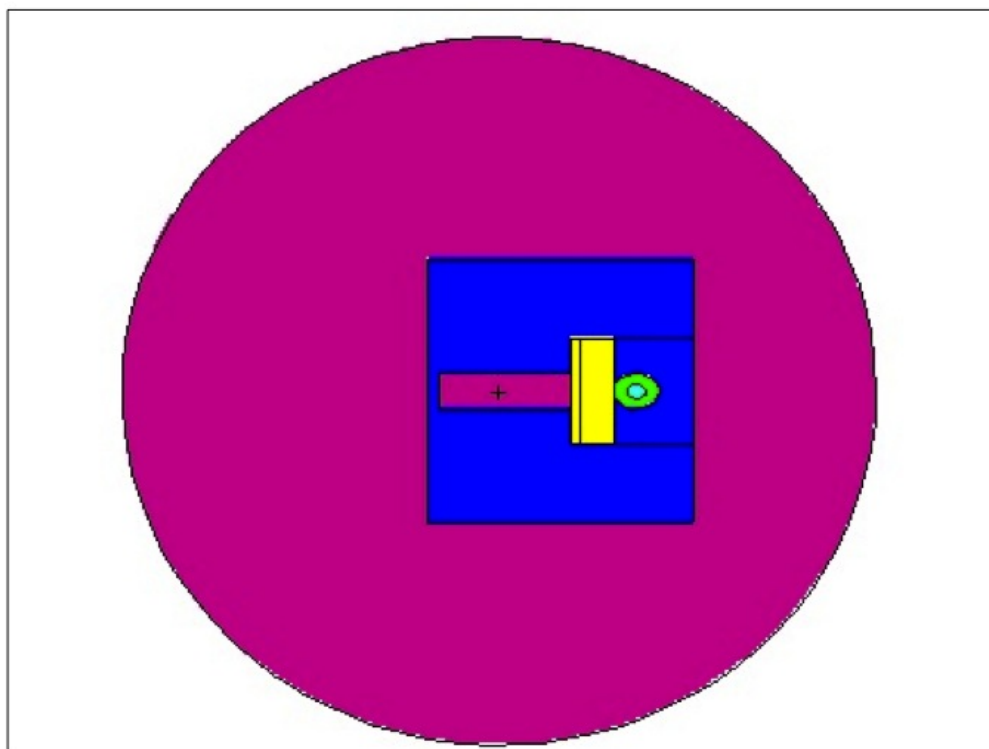


Figure 4.4 : Top view of the prototype apparatus geometry obtained by MCNP5.

- Cell#4 was filled with polyethylene material to provide a biological protection from neutrons of 14 MeV.
- Cell#5 was filled with lead shielding material by surrounding the BGO detector.
- Cell#6 represented the volume filled with synthetic coal material.
- Cell#7 represented the outside region where can not be created any interaction and produced the Monte Carlo simulation. The particle transport was stopped by this spherical border.

The lignite coal inside cell#6 has a density about 1.4 g/cm^3 , and the cylindrical volume of this cell was about 78.571 cm^3 . The elements in coal as the combination of ash, moisture and fixed carbon were weighted 110 kg in this volume. The concentration of the elements in Turkish lignite coal was shown in Table 3.12.

A point source of 14 MeV was placed inside of the data card section of the input file. The position of neutron source was used in same the position with design of the prototype apparatus. The information about PGNAA and NIST processes induced by fast and thermal neutrons was introduced in physics card part of the data card section. The photo-nuclear interaction was mainly constituted for the physics card. The cross

section information of elements was collected from ENDF/B7 nuclear data file. The particle history of the simulation in nps card was entered as total number of particle of the source. The coal sample was irradiated by 14 MeV neutrons until particle histories of 10^9 neutrons were completed. The average cell flux tally of MCNP5 (or “F4 tally”) was used to detect the gamma ray energies between 0 and 10 MeV with 1024 channels in BGO volume. The class diagram of MCNP5 simulation was shown in Figure B.1 at appendix section.

As a variance reduction technique, Geometry Splitting/Roussian roulette and energy cutoff were used to reduce the relative error “R”. The neutron importance and photon importance of each cell were arranged by increasing the importance of cells by power of 2 to reduce the relative error of the simulation. Based on this qualitative analysis, the recommended interpretation of the estimated 1σ confidence interval for the point detectors, the value of the relative error “R” associated with an MCNP tally should be less than 0.05. Since the relative errors of simulation results were found under 0.05 value, the usage of variance reduction technique was found enough for this problem [87]. The output data file of the simulation was given as an Ascii file. The relation between number of particles and energies of the gamma rays was represented as a histogram in the simulation. In order to convert the histogram to Gaussian distribution, the experimental response functions of BGO detector were used. The histogram and spectrum of coal sample were shown in Figure 4.5 and Figure 4.6 respectively.

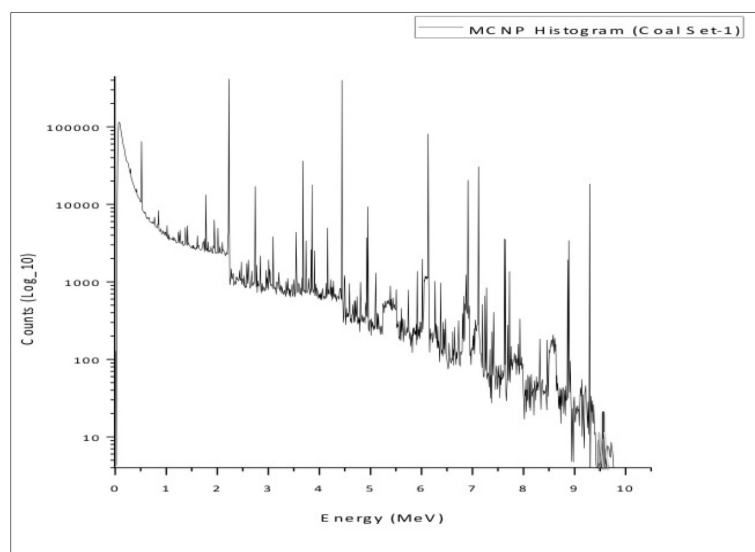


Figure 4.5 : The gamma-ray histogram of the Coal Set-1 obtained by MCNP5.

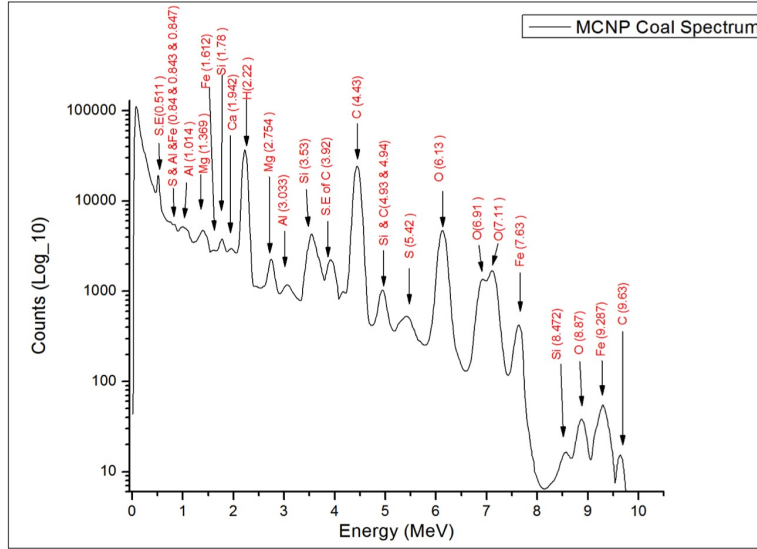


Figure 4.6 : The gamma-ray spectrum of the Coal Set-1 obtained by MCNP5.

As shown in Table 4.2, it is seen that some of elements have greater cross sections of gamma ray energies obtained inelastic scattering interactions of neutrons, whereas some of elements have greater cross sections of gamma ray energies obtained thermal neutron capture interactions.

Table 4.2 : The cross sections of gamma ray energies induced by thermal and fast neutrons for interested elements of coal.

Element	Reaction	Energy (MeV)	Cross-section (barn)	
			Thermal Neutron	Fast Neutron
H-1	(n, γ)	2.223	0.333	0.0002
C-12	(n, $n' \gamma$)	4.438	0.0035	0.427
	(n, γ)	4.940		
N-14	(n, $n' \gamma$)	0.730, 1.634, 2.313	0.0079	0.4
	(n, γ)	1.885, 5.269, 10.829		
O-16	(n, $n' \gamma$)	5.618, 6.129	0.0002	0.508
	(n, γ)	0.870, 1.087, 2.185		
Mg-24	(n, $n' \gamma$)	1.369, 2.754, 3.866	0.066	0.645
	(n, γ)	0.585, 2.828, 3.916		
Al-27	(n, $n' \gamma$)	0.843	0.231	0.017
	(n, γ)	7.724		
Si-28	(n, $n' \gamma$)	1.779	0.172	0.123
	(n, γ)	4.934		
S-32	(n, $n' \gamma$)	1.273, 2.230	0.534	0.379
	(n, γ)	0.841, 3.221, 5.420		
Ca-40	(n, $n' \gamma$)	3.736	0.431	0.066
	(n, γ)	1.942, 6.421		
Fe-56	(n, $n' \gamma$)	0.847	2.56	0.074
	(n, γ)	7.631		

C (4.43 MeV), O (6.13 MeV), and N (10.83 MeV) can be detected by using gamma ray energies induced of 14 MeV neutrons. The elements of ash content, sulphur and moisture which are Fe (7.63 MeV), Si (3.53 MeV), Ca (1.942 MeV), Al (1.014 MeV), Mg (1.369 MeV), S (5.42 MeV), H (2.22 MeV), can be detected by using thermal neutron capture interactions. Some of the elements such as S (0.84 MeV), Al (0.843 MeV) and Fe (0.847 MeV) which gamma energies are close to each other could not be resolved due to the poor energy resolution of the BGO detector.

The BGO detector at transmission geometry was placed the bottom of coal sample volume as shown in Figure 4.3. The comparison of gamma spectrum between transition and back-scattering geometry was shown in Figure 4.7.

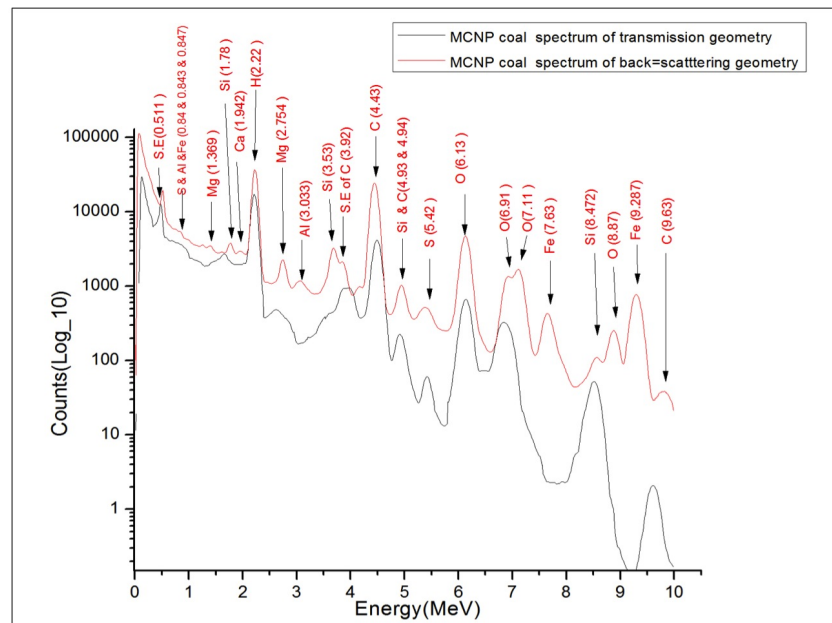


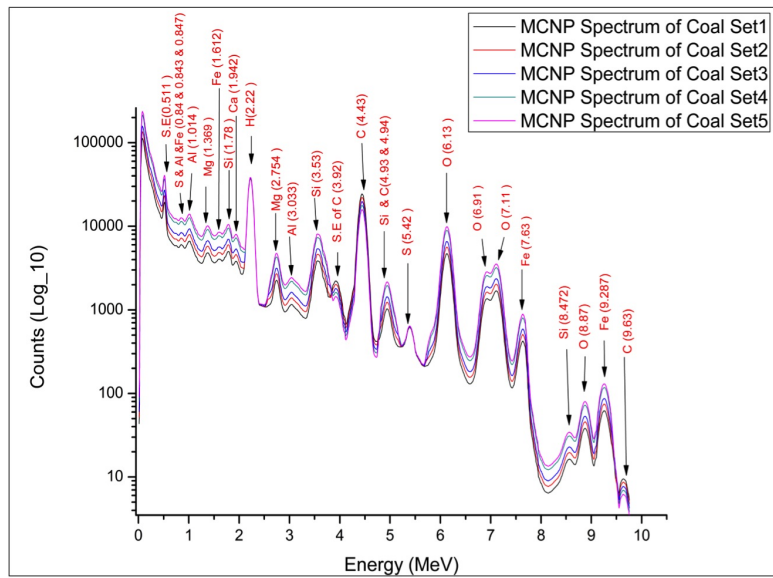
Figure 4.7 : The comparison of back-scattering and transmission gamma rays spectra of the Coal Set-1 obtained by MCNP5.

Because of self-absorption inside of the coal sample, some of gamma energy levels could not be detected by BGO detector at transmission position. Therefore the placement of BGO was decided to back-scattering position for the detection of coal sample content. According to the study of lignite coal on Aegean region [99], five synthetic coal samples were prepared on dry basis. The amount of sulfur content was remained constant. While carbon contents were decreased, ash contents were increased. The contents of five synthetic coal sets were shown in Table 4.3.

Table 4.3 : The concentration of the compounds in each coal set.

Compound	Set#1 (%)	Set#2 (%)	Set#3 (%)	Set#4 (%)	Set#5 (%)
C	76.00	69.34	62.66	56.00	49.34
SiO ₂	8.20	10.86	13.60	16.34	19.00
Fe ₂ O ₃	3.60	4.6	5.86	7.10	8.22
Al ₂ O ₃	3.60	4.94	6.14	7.34	8.62
MgO	1.00	1.26	1.60	1.86	2.20
CaO	3.60	4.94	6.14	7.34	8.62
S	4.00	4.00	4.00	4.00	4.00

The comparison of the gamma-ray spectra of five synthetic coal sets was shown in Figure 4.8. The simulation results of coal sample sets were consistent with carbon, sulfur and ash contents of coal sets.

**Figure 4.8 :** The comparison of gamma-ray spectra of five synthetic coal sets obtained by MCNP5.

From coal sample set-1 to set-5 respectively, the photo peak level of neutron induced gamma energy representing the carbon content was decreased, whereas gamma energies of elements of Ca, Fe, Si, Al, Mg in ash content were observed increase.

Another simulation was performed to calculate the effective dose rate per unit activity per unit area on the surfaces of prototype apparatus which are polyethylene surfaces and additional lead blocks of 5 cm thickness on two lateral surfaces. The F2 tally of MCNP5 was used to obtain the simulation of dose evaluation. The F2 tally describes the particle flux on average surface. The calculation result in rem/h value is converted to Sievert/h dividing by 0.01. The simulation results were shown in Table 4.4.

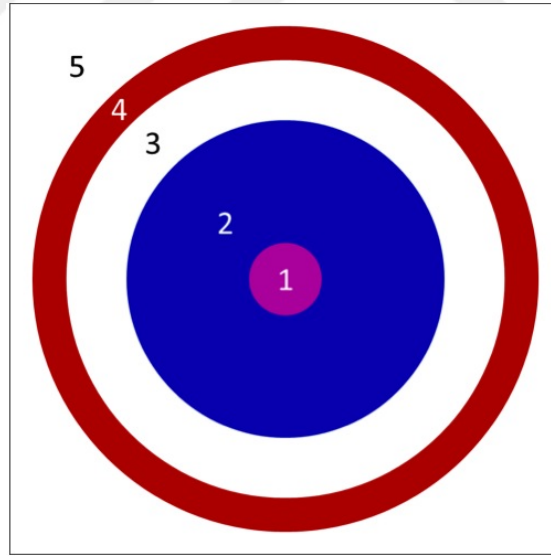
Table 4.4 : Effective dose rate of neutrons on polyethylene and lead surfaces.

Surface	Dose S_{PE}	Dose S_{Lead}	S_{PE} (cm^2)	S_{Lead} (cm^2)	$E_{dose} S_{PE}$ (Sv/h)	$E_{dose} S_{Lead}$ (Sv/h)
S-1	4.860 E-08	5.019 E-08	14040	14560	3.470 E-06	3.447 E-06
S-2	1.346 E-08	1.347 E-05	14040	14560	9.592 E-07	9.252 E-07
S-3	6.065 E-08	9.095 E-08	7280	7280	8.331 E-06	1.25 E-05
S-4	4.515 E-08	6.720 E-08	7280	7280	8.331 E-06	9.231 E-07
S-5	4.525 E-08	4.527 E-07	37800	39200	1.2 E-05	1.15 E-05
S-6	3.02 E-08	3.024 E-05	37800	39200	7.99 E-05	7.71 E-05

Since the maximum dose rate is $10 \mu Sv h^{-1}$ for neutron source the prototype apparatus remains in the level of safe radiation dose without additional lead shielding.

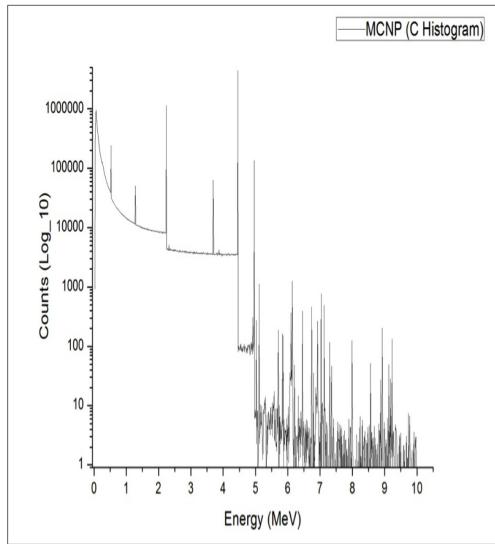
4.2.2 Generating of MCNP5 element library

In order to define the elemental contents of coal sample, MCNP5 Library Least Square method (MCNP5 LLs) was developed. Therefore, the information of gamma ray interactions of each compound induced by neutrons was obtained in a simple simulation geometry. Unlike the geometry of prototype apparatus, each of MgO, Fe₂O₃, CaO, Al₂O₃, SiO₂, S, C, and N compounds was simulated separately in a spherical fixed volume shown in Figure 4.9.

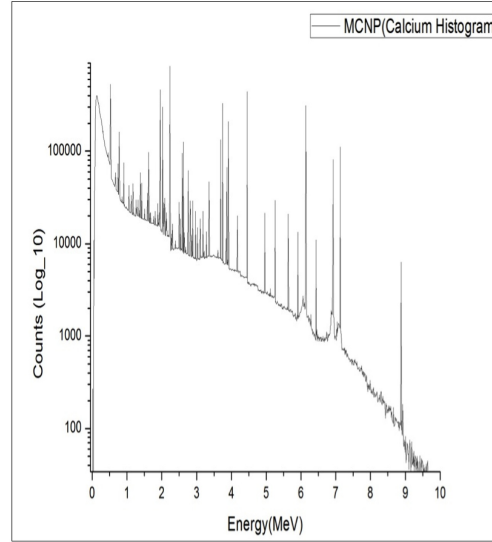
**Figure 4.9** : The spherical fixed-geometry for elemental library spectra.

The point neutron source of 14 MeV was inserted inside the cell#1, which was filled with air medium. The cell#2 was filled with polyethylene material in order to thermalize the neutrons. Each compound was inserted separately to the cell#3 and whole volume was surrounded with BGO detector material inside the cell#4, and the cell#5 represents the vacuum. For this purpose, the MCNP5 average cell flux tally (or

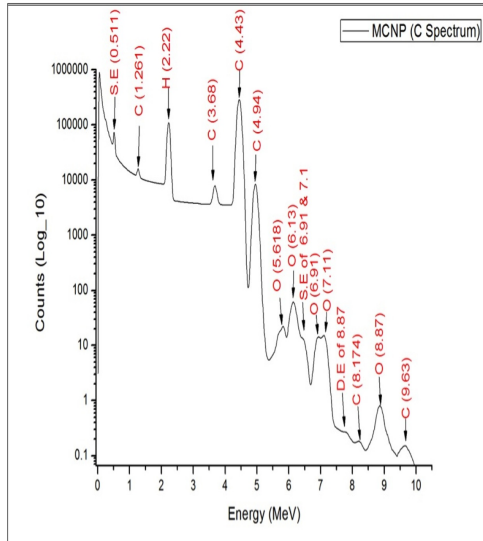
“F4 tally”) was used to obtain library of the single element spectrum. The particle history of the simulation in nps card was entered 10^8 as the total number of particles of the neutron source. The histograms and spectrums of the MCNP5 simulation of MgO, Fe₂O₃, CaO, Al₂O₃, SiO₂, S, C, and N in the fixed spherical geometry were shown in between Figure 4.10 and Figure 4.13.



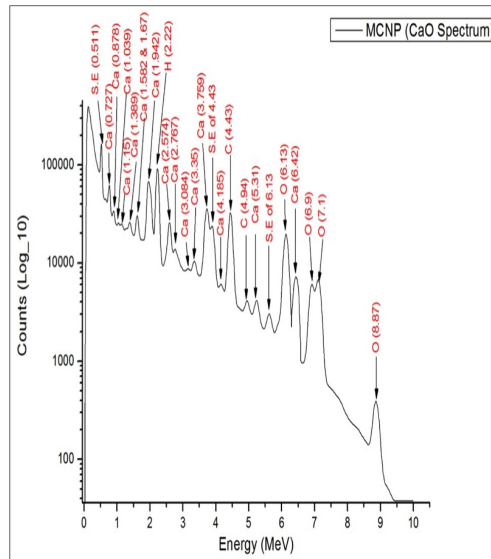
(a) The gamma-ray histogram of C.



(b) The gamma-ray histogram of CaO.

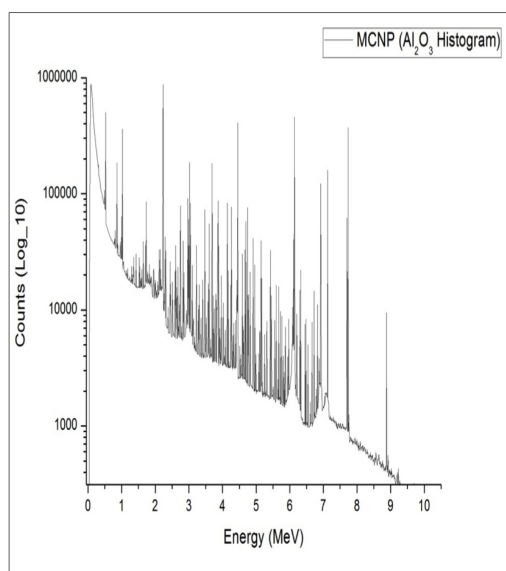


(c) The gamma-ray spectrum of C.

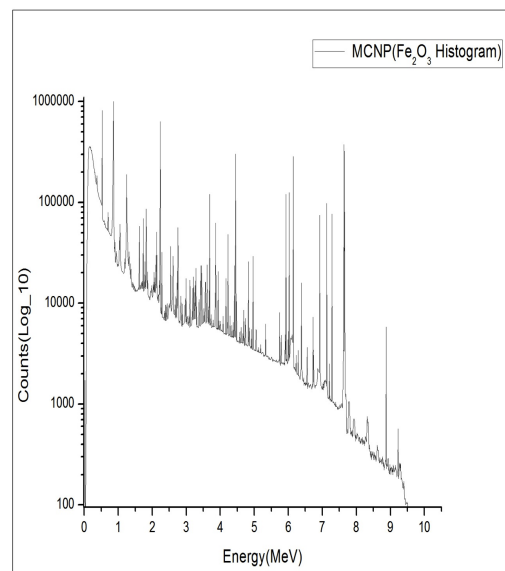


(d) The gamma-ray spectrum of CaO.

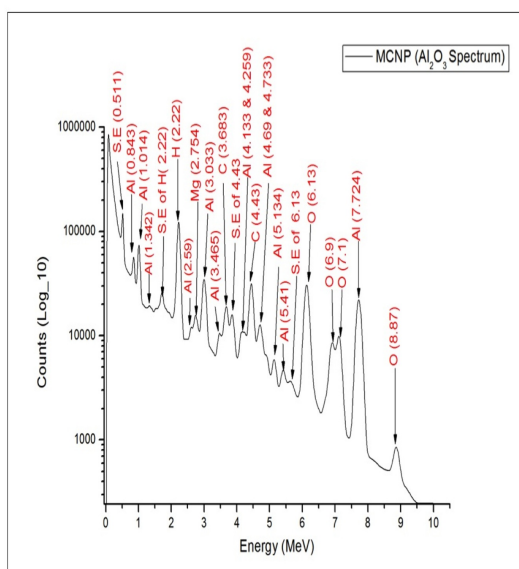
Figure 4.10: The gamma-ray histograms and spectra of C and CaO.



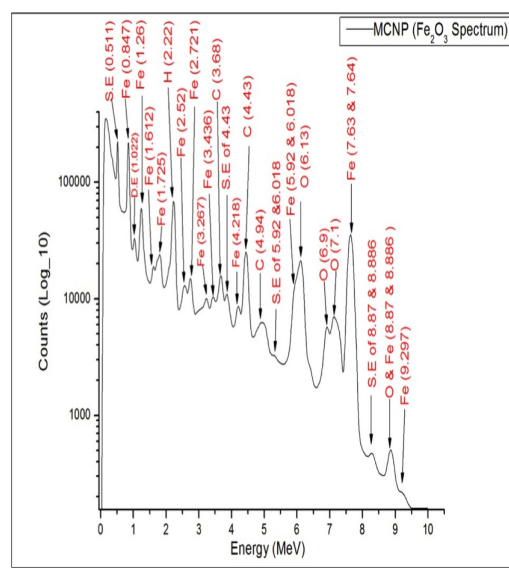
(a) The gamma-ray histogram of Al₂O₃.



(b) The gamma-ray histogram of Fe₂O₃.

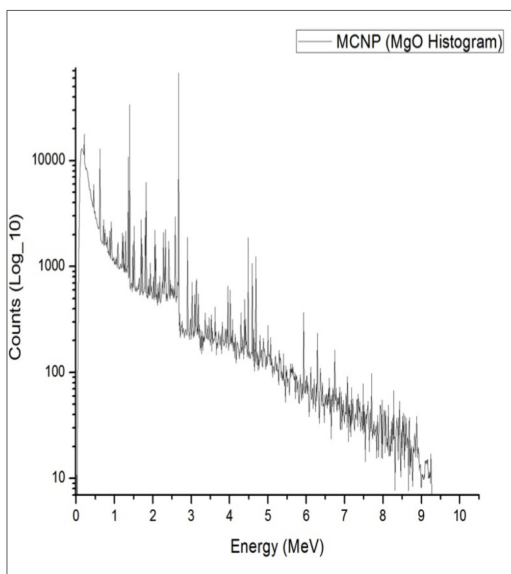


(c) The gamma-ray spectrum of Al₂O₃.

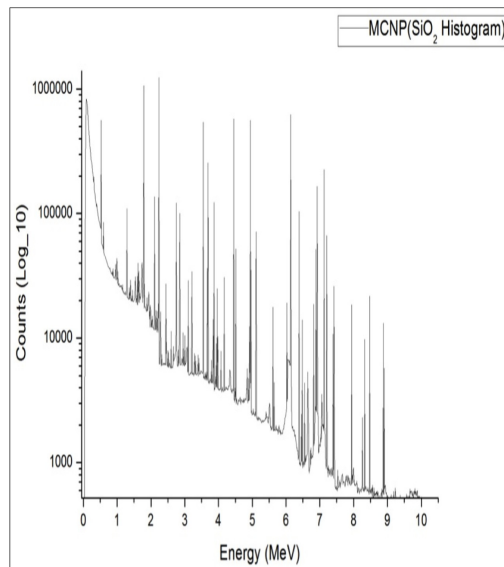


(d) The gamma-ray spectrum of Fe₂O₃.

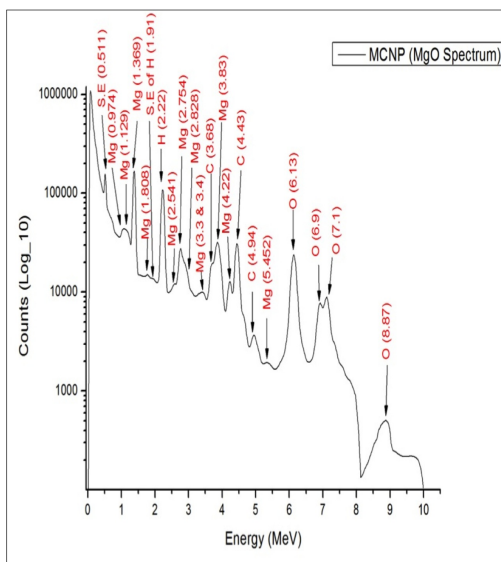
Figure 4.11: The gamma-ray histograms and spectra of Al₂O₃ and Fe₂O₃.



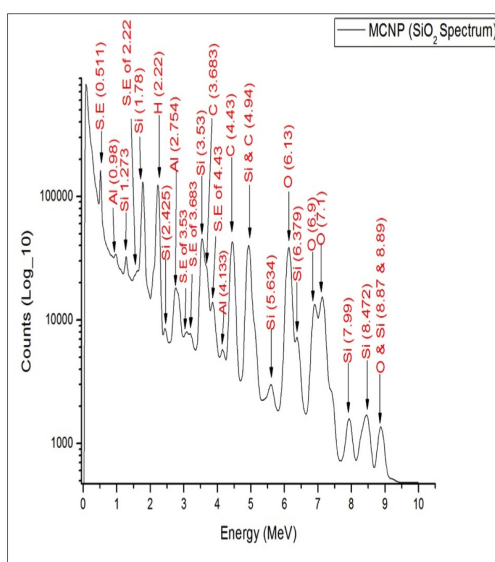
(a) The gamma-ray histogram of MgO.



(b) The gamma-ray histogram of SiO₂.

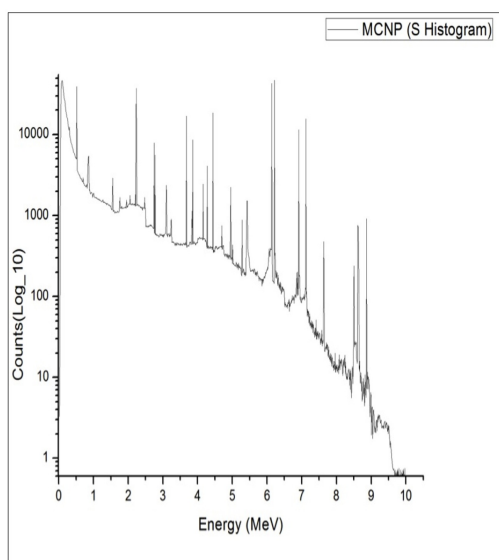


(c) The gamma-ray spectrum of MgO.

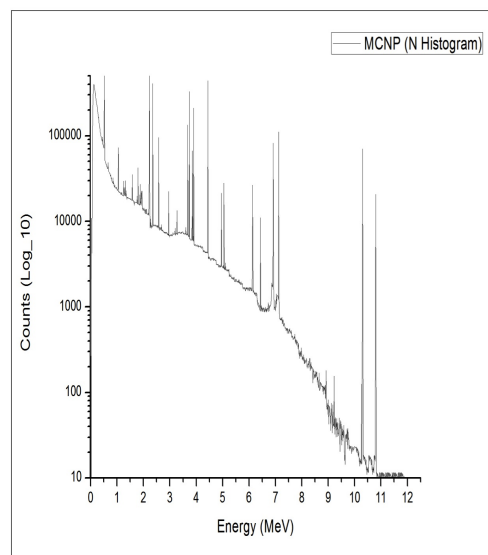


(d) The gamma-ray spectrum of SiO₂.

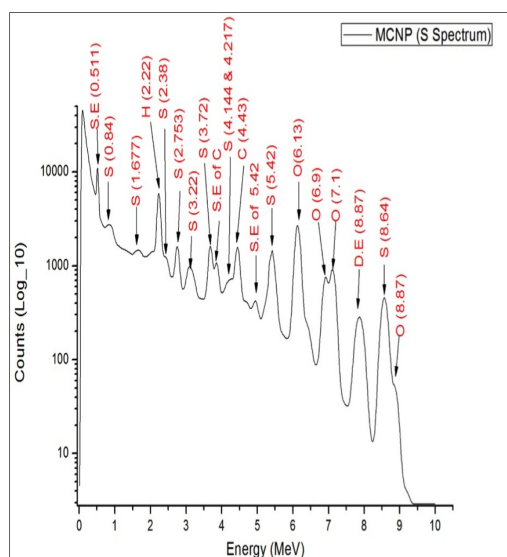
Figure 4.12: The gamma-ray histograms and spectra of MgO and SiO₂.



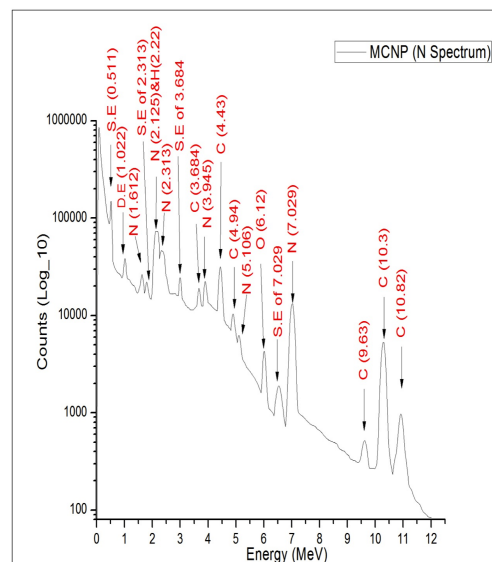
(a) The gamma-ray histogram of S.



(b) The gamma-ray histogram of N.



(c) The gamma-ray spectrum of S.



(d) The gamma-ray spectrum of N.

Figure 4.13: The gamma-ray histograms and spectra of S and N.

4.3 The Structure of GEANT4

GEANT4 [100], a physics simulation toolkit implemented in the C++ language, is used in several fields such as medical physics, particle physics, nuclear physics, accelerator design. In order to describe the physical interactions of particles with matter, the physics processes included hadronic, optical, electromagnetic (EM), atomic relaxations, etc. over a wide energy range from 100 eV to several PeV. In order to give an overall aspect to the structure of simulation, the schematic structure of GEANT4 classes was shown in Figure 4.14.

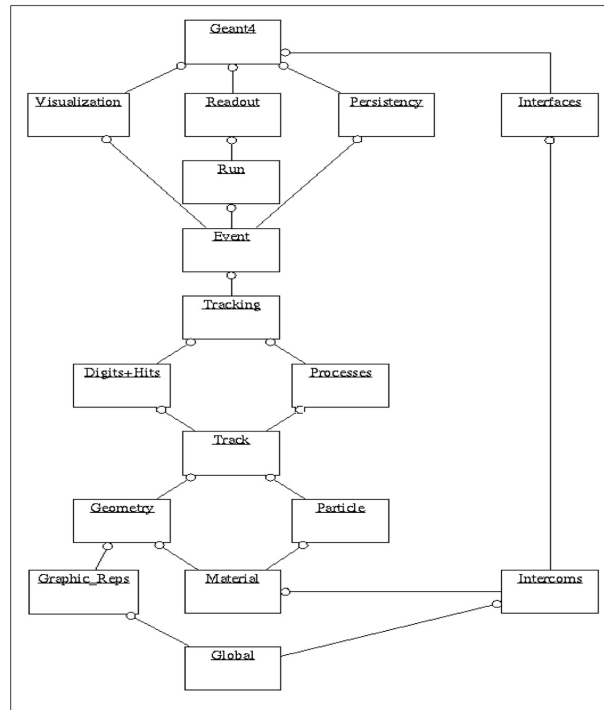


Figure 4.14 : The schematic view of GEANT4 classes [100].

The classes of GEANT4 are classified in two main groups as mandatory user classes and user classes. The mandatory user classes must be derived from three base classes which are `G4_UserDetectorConstruction`, `G4_UserPhysicsList`, and `G4_UserPrimaryGeneratorAction` provided by GEANT4. The classes of GEANT4 are to be associated to the `G4_RunManager`, which controls the overall behavior of the program. In the class of `G4_UserDetectorConstruction`, the detector materials and geometry, scoring regions, as well as visualization specifications are defined by the user.

The GEANT4 simulation program also includes a physics list derived from G4_UserPhysicsList to register the particle types, their associated physics processes, and the production of particles cut-off. The last mandatory class, G4_UserPrimaryGeneratorAction, specifies the energy, direction, dimensions, and type of the primary particles. The class of G4_UserPrimaryGeneratorAction provides to arrange the way of primary particles generation. The primary particle generator should be initiated in the constructor of G4VuserPrimaryGeneratorAction. One of the generators defined by GEANT4 is G4ParticleGun. This class produces the specific type of primary particles with their information such as energy, position, and momentum.

GEANT4 provides five base classes for user control. These classes contain methods that can be invoked at various stages of the simulation process. Users can write their own implementations of these methods to perform various actions, such as defining a histogram, analyzing the results, setting priority control to a track, storing particle trajectories, and terminating a track.

4.3.1 Geometry construction

In GEANT4, the geometry construction is initialized by creating a world volume. The world volume as a mother volume includes all of defined components as daughter volumes without overlapping. The volumes in GEANT4 are created in three steps.

- In the first step, specific shape and dimensions are created for the associated solids. The solid is classified in three categories as Constructive Solid Geometry (CSG), Boundary Represented Solid (BREP) and Boolean solids. CSG includes geometrical information such as boxes, cylinders, cones, spheres, and parallelepipeds. BREP defines the boundary surfaces of the geometrical information. Boolean solids use one of Boolean operations which are union, intersection or subtraction.
- In the second step, the created solids are associated to a logical volume containing the information such as the material, sensitivity.
- At the last step, a physical volume is created from logical volume equipped with specific position included of its mother volume. The same solid class can create

the different logical volumes. Also the multiple physical volumes can be created by using same logical volumes.

4.3.2 Particle Description

Most of particle types known in nature are identified in GEANT4 including such as photons, neutron, mesons, ions. The information of a particle is defined by three levels of classes. These are G4ParticleDefinition, G4DynamicParticle, and G4Track.

- G4ParticleDefinition class contains the static information of the particle type. Some of these information are particle name, mass, lifetime, and interaction type to undergo.
- The G4DynamicParticle class contains the dynamic information of the particle. Some of these information are energy, momentum, and polarization which are obtained on the move of particle. The G4DynamicParticle class is marked with a G4ParticleDefinition object.
- A G4Track class has the information about the particle such as the position, track length, kinetic energy, direction, and weight of the particle representing the end point of the current step as it traverses in matter. The G4Track class is marked with its associated G4ParticleDefinition and G4DynamicParticle classes.

4.3.3 Physics processes

The physics processes of GEANT4 simulations are classified into seven major groups. These are electromagnetic, hadronic, decay, optical, photo-lepton hadron, parameterization, and transportation. A physics process in GEANT4 is originated from the G4VProcess class. A desired physics process is included to simulate at least one of continuous (AlongStep), discrete (PostStep), and at rest (AtRest) actions. The GetPhysicalInteraction Length (GPIL) method limits the step size and defines the interactions. The Dolt method generating the final states of the particle must be defined for the selected action. G4VuserPrimaryGeneratorAction class enables the user to arrange the way primary particles are generated. The actual primary particle generator should be initiated in the constructor of G4VuserPrimaryGeneratorAction. One of the generators defined by GEANT4 is G4ParticleGun. This class produces the specific type of primary particles with the information of energy, position, and momentum.

4.3.4 Tracking and particle transportation

The tracking of a particle along a step is managed by the `G4SteppingManager` class. First of all the step length is determined with regard to the smallest PIL. This class also includes information of the transportation process of related particle. The continuous processes are called the relevant `AlongStepDoIt` to collect the information of track properties. By applying `PostStepDoIt` method, each interaction is called to update the energy information of the track. Before checking the termination of the track, the `G4Track` objects are builded to move the secondary particles on a stack. The particle can be assigned to the next volume on the condition that the boundary of volume limits the step. The information of hits is collected for the sensitive detector volume. The `UserSteppingAction` method of the `G4UserSteppingAction` class is used to complete to the step.

4.3.5 Stack management

By using C++ standard containers, `G4Track` objects, producing secondary particles, are moved on at least one of the stacks which are defined Urgent, Waiting, and PostponeToNext Event. The behavior of the stacks is easily managed by fulfilling a `G4UserStackingAction` class, which contains the methods of `ClassifyNewTrack`, `NewStage`, and `PrepareNewEvent`. The method of `ClassifyNewTrack` is called during a new track is pushed. At that moment, the user can set it to one of the three stacks, or kill the track. If the Urgent stack has no objects, `NewStage` method is used. Objects in the Waiting stack is supposed to store to the Urgent stack. At the beginning of each history, `PrepareNewEvent` is called to reset the classification scheme by the user. The stacking algorithm in GEANT4 provides to speed up the simulation by preventing to record uninterested tracks and keeps the information of the tracks with the highest priority.

4.3.6 Scoring techniques

The information of particle interactions as hits are recorded the user-defined scoring regions. Scoring regions are set as sensitive detectors by identifying their logical volume. In order to describe a user own class, `G4VHit` and `G4VSensitiveDetector`

classes are fulfilled to include the information such as any geometrical information, position of the hit, energy and momentum of the track, the energy deposition along a step. The collected information as a derived from G4THitsCollection class is implemented with G4UserEventAction class which is defined at the end of each particle history of the analysis.

4.3.7 User-interface and visulation

The GEANT4 User-interface (UI) options are provided by either running in a character terminal, or with a Java or Motif-based graphical user interface. By using pre-defined UI commands, the simulation processes can be easily controlled users communicate. Also, if a new command is desired to use, it can be defined in a class of G4_UImessenger. The geometry of the simulation and the trajectories of the particles can be displayed by using the visualization drivers. The most common visualization drivers are OpenGL, DAWN2, and HepRep.

4.4 GEANT4 Simulation of Prototype Apparatus

The geometry set-up of prototype apparatus was placed in the World volume of a rectangular box shape with 40 m dimensions for x,y and z directions as shown in Figure 4.15.

The neutron source was placed at the origin of this volume. 14 MeV neutrons with a solid angle of 4π were modeled within the “PrimaryGeneratorAction.cc” file. A rectangular red box volume of including iron material was placed to near of the neutron generator position as an shielding material. The lead material surrounding the BGO detector volume was created as an green cylindrical volume. The coal sample in blue cylinder was located at the bottom of prototype apparatus volume. All of the detector material with geometrical information were treated as a compound of the atomic numbers, atomic weights, and atomic factions of the elements in the DetectorConstruction.cc file. The class diagram of DetectorConstruction.cc file was shown in Figure B.2 at appendix section. The hadronic physics processes for neutron interactions with matter inelastic scattering and neutron capture for neutron-nucleus interactions were included in a user defined PhysicsList class. The illustration of the production of neutrons in the simulation was shown in Figure 4.16.

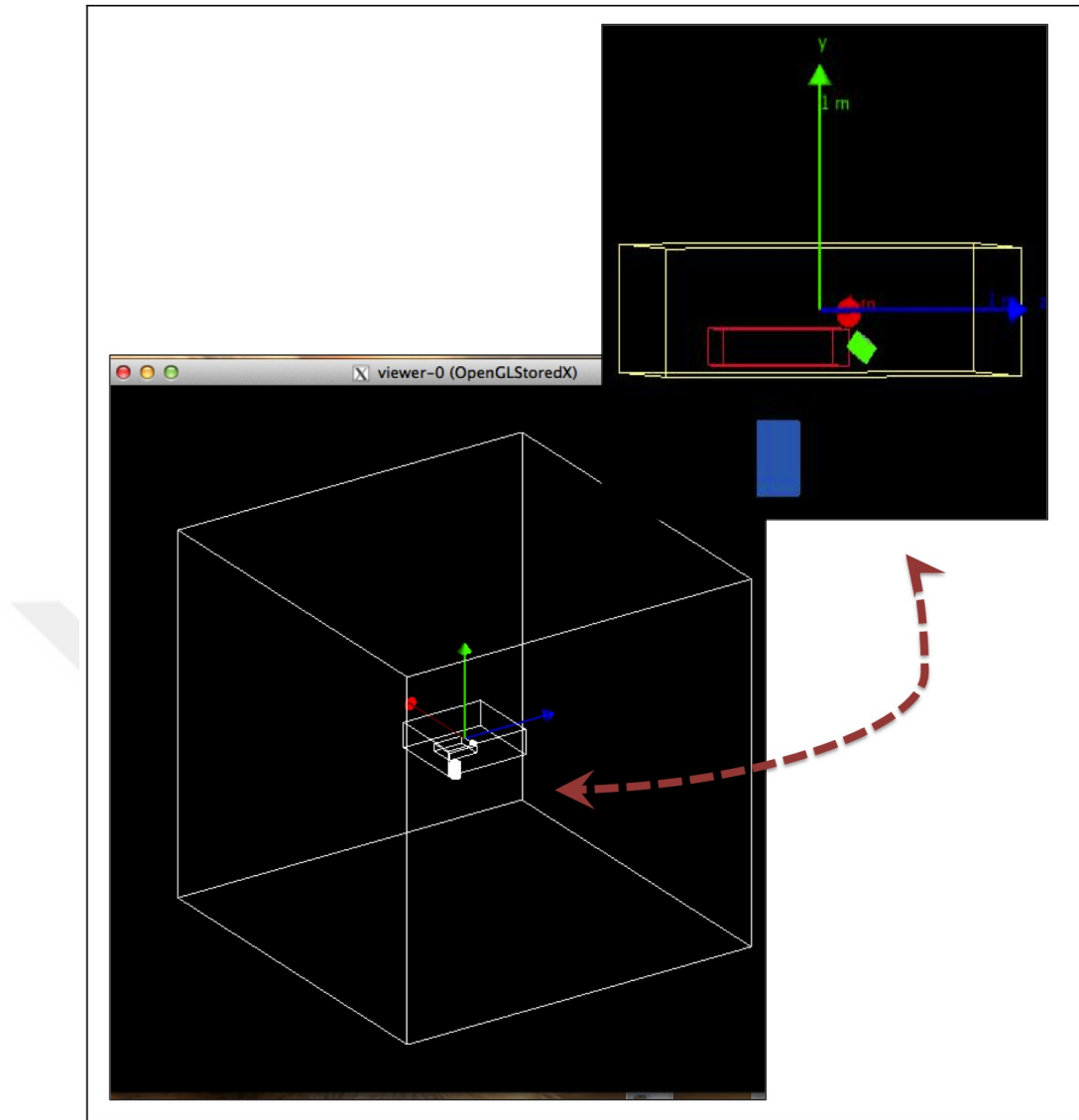


Figure 4.15 : The illustration of the prototype apparatus obtained by GEANT4.

The gamma particles coming from electromagnetic processes were collected in the BGO detector as a sensitive volume. 10^6 neutrons in the run were produced. The resulting gamma rays were recorded to an external file. The output file included the information of energy deposition and number of γ particles. The output file was converted to histogram of coal sample as shown in Figure 4.17. Then, The histogram of coal sample was converted to spectrum by using response functions of BGO detector. The spectrum of coal sample was shown in Figure 4.18

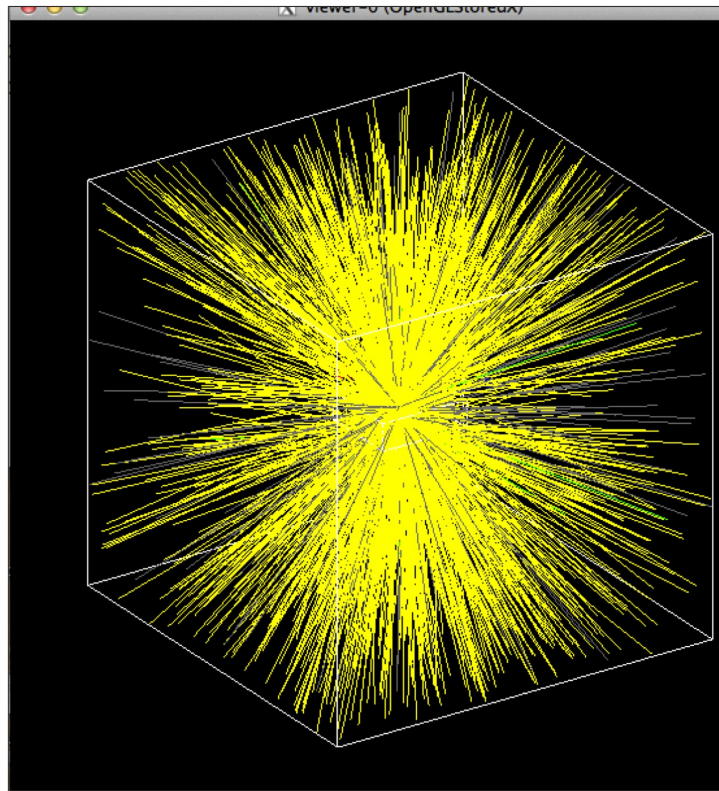


Figure 4.16 : The illustration of production of neutrons obtained by GEANT4.

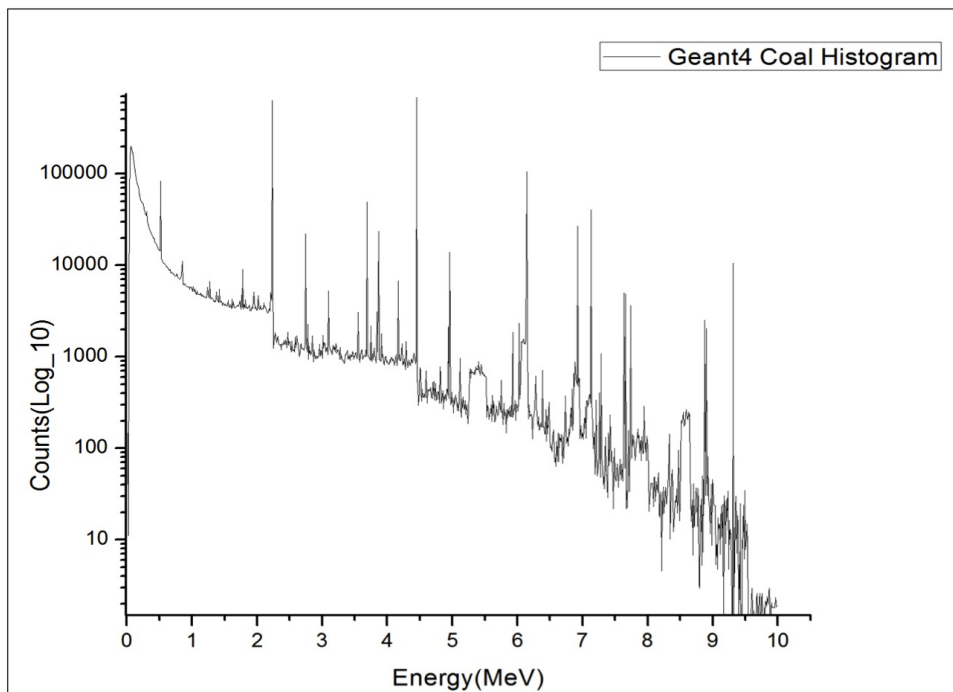


Figure 4.17 : The gamma-ray histogram of coal sample obtained by GEANT4.

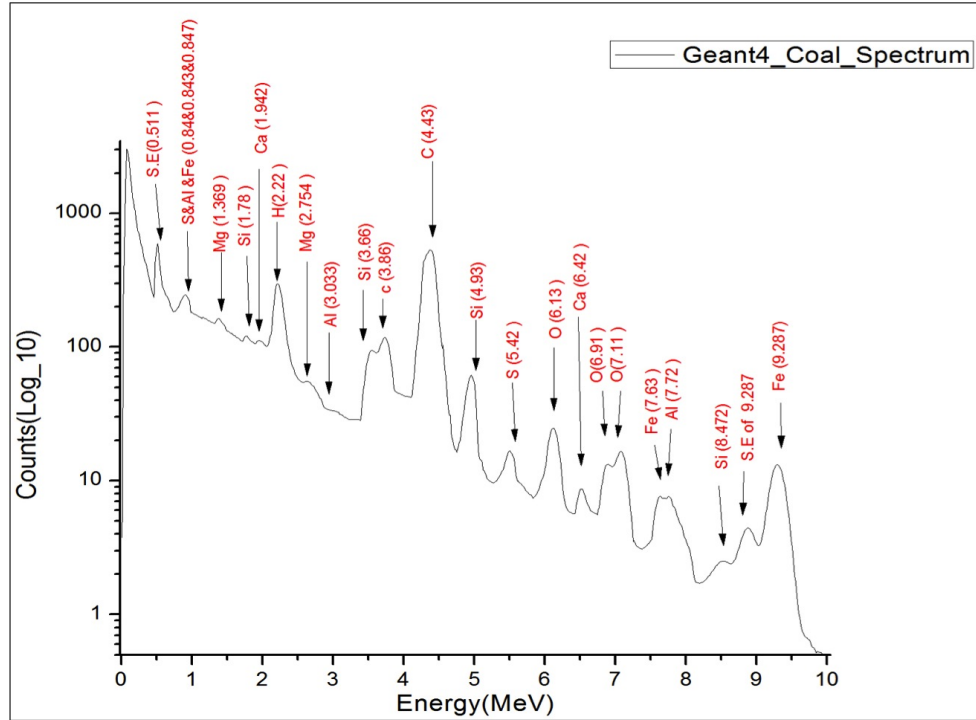
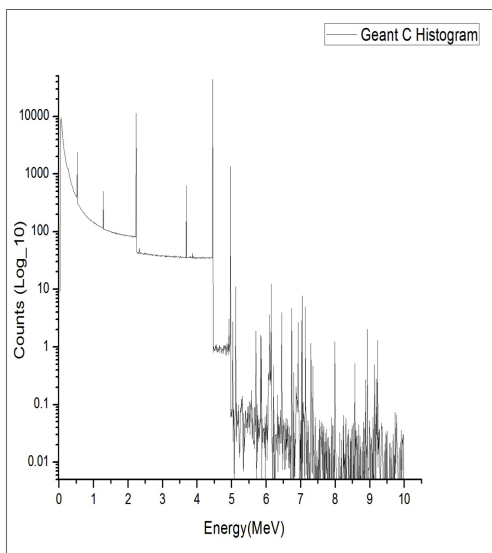


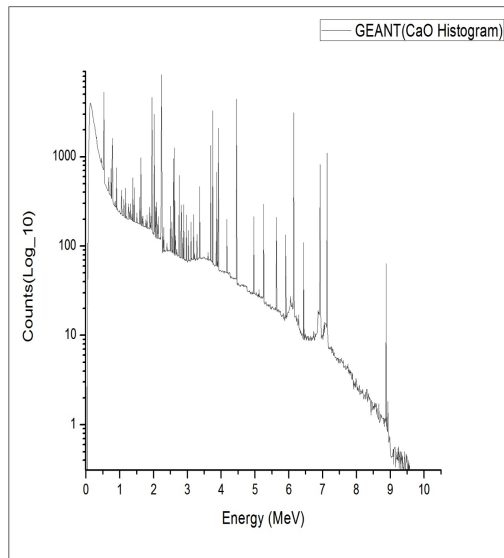
Figure 4.18 : The gamma-ray spectrum of coal sample obtained by GEANT4.

4.4.1 Generating of GEANT4 element library

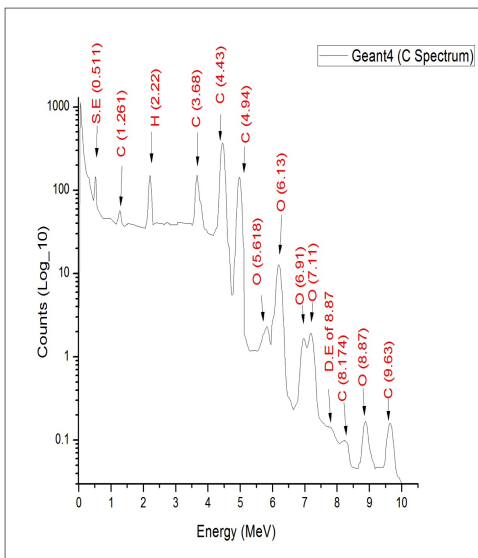
In order to define the elemental contents of coal sample, GEANT4 Library Least Square method (GEANT4 LLS) was developed. Therefore, the information of gamma rays of each compound induced by neutrons was obtained in a simple simulation geometry. Unlike the geometry of prototype apparatus, each of MgO, Fe₂O₃, CaO, Al₂O₃, SiO₂, S, C, and N compounds was simulated separately in a spherical fixed volume shown in Figure 4.9. The resulting gamma particles coming from electromagnetic processes were collected in the BGO detector as a sensitive volume. 10⁵ neutrons in the run were produced and the resulting gamma rays were recorded to an external file to convert to a histogram file including energy deposition and number of γ particles. The gamma ray histograms and spectra of the GEANT4 simulation of MgO, Fe₂O₃, CaO, Al₂O₃, SiO₂, S, C, and N in the fixed spherical geometry were shown in between Figure 4.19 and Figure 4.22.



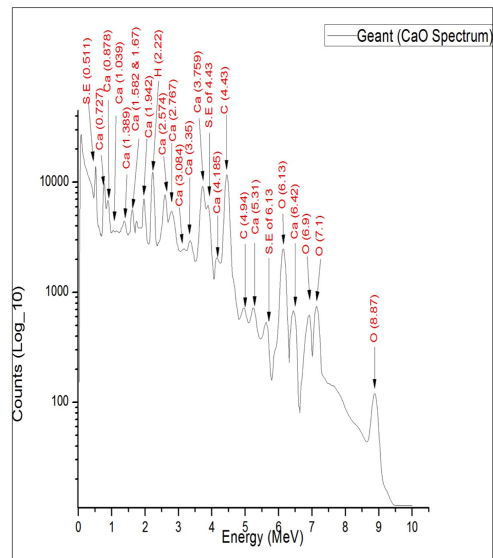
(a) The gamma-ray histogram of C.



(b) The gamma-ray histogram of CaO.

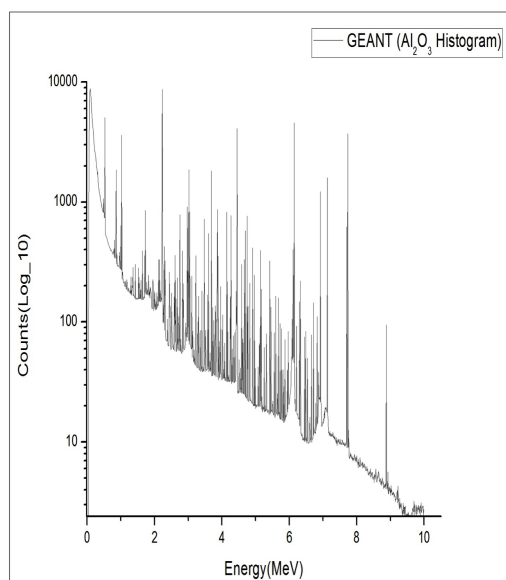


(c) The gamma-ray spectrum of C.

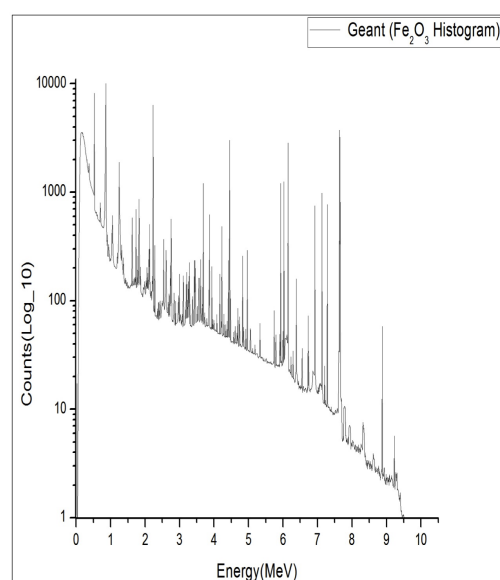


(d) The gamma-ray spectrum of CaO.

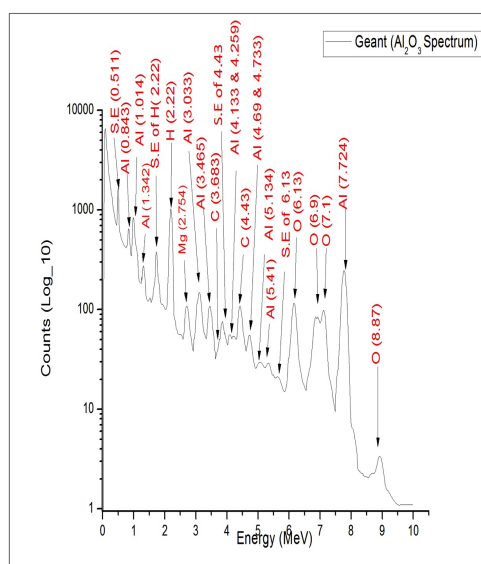
Figure 4.19 : The gamma-ray histograms and spectra of C and CaO.



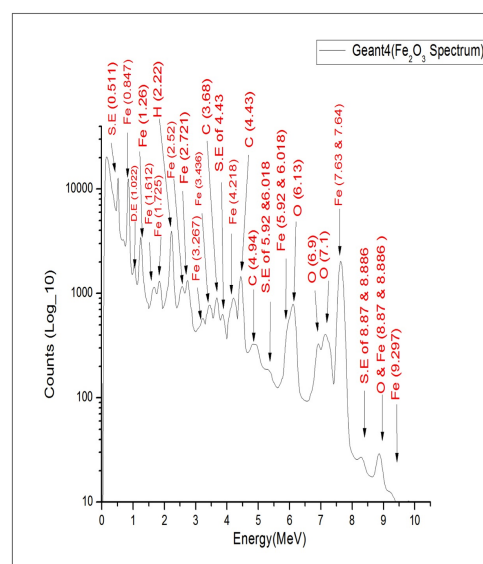
(a) The gamma-ray histogram of Al₂O₃.



(b) The gamma-ray histogram of Fe₂O₃.

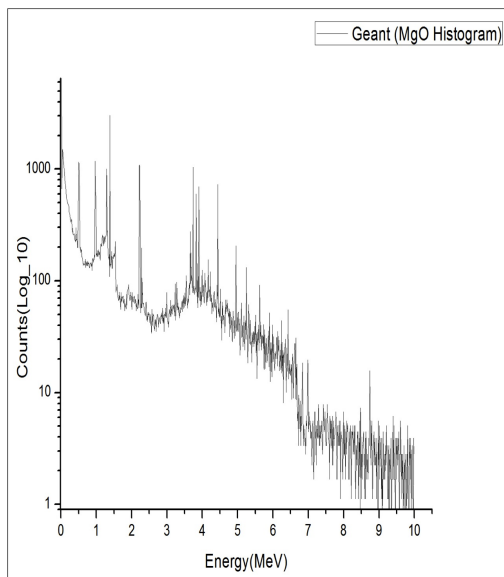


(c) The gamma-ray spectrum of Al₂O₃.

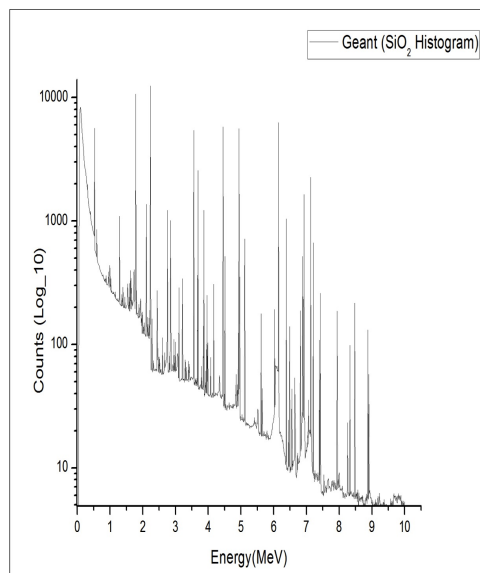


(d) The gamma-ray spectrum of Fe₂O₃.

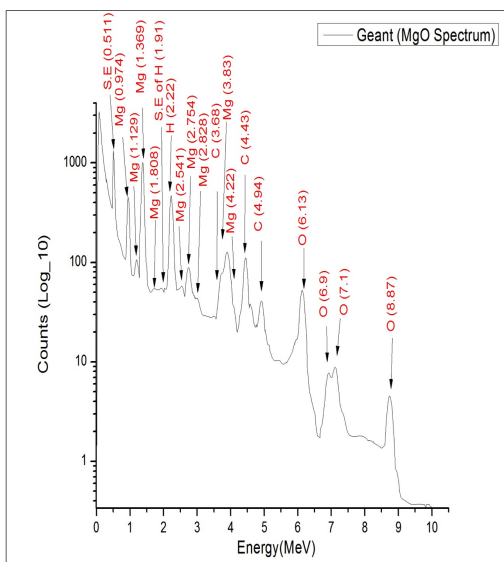
Figure 4.20 : The gamma-ray histograms and spectra of Al₂O₃ and Fe₂O₃.



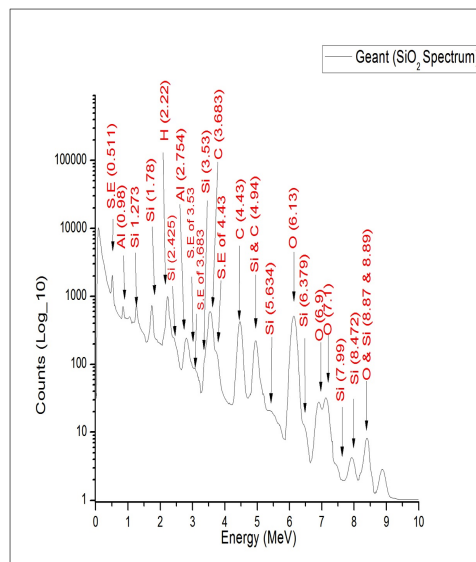
(a) The gamma-ray histogram of MgO.



(b) The gamma-ray histogram of SiO₂.

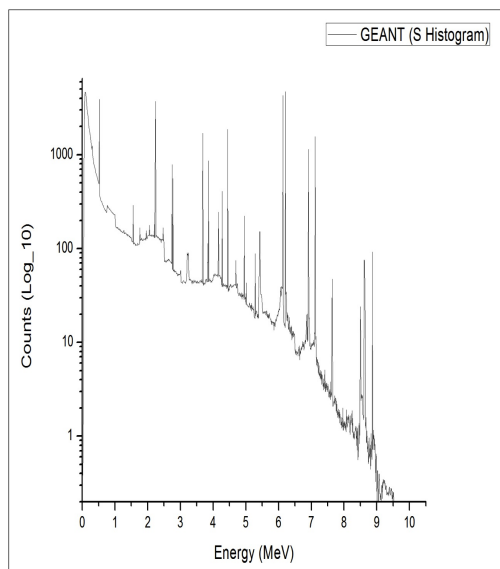


(c) The gamma-ray spectrum of MgO.

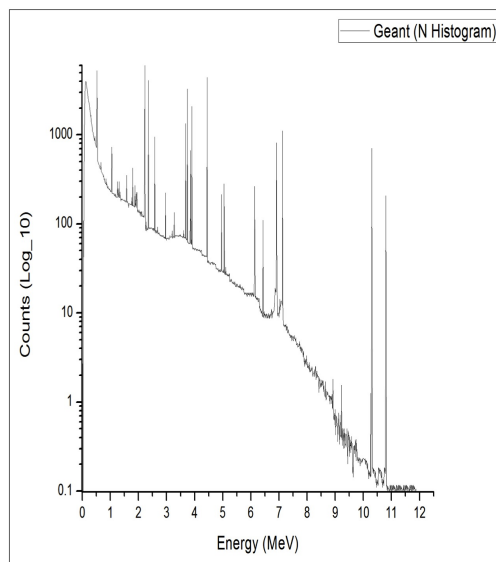


(d) The gamma-ray spectrum of SiO₂.

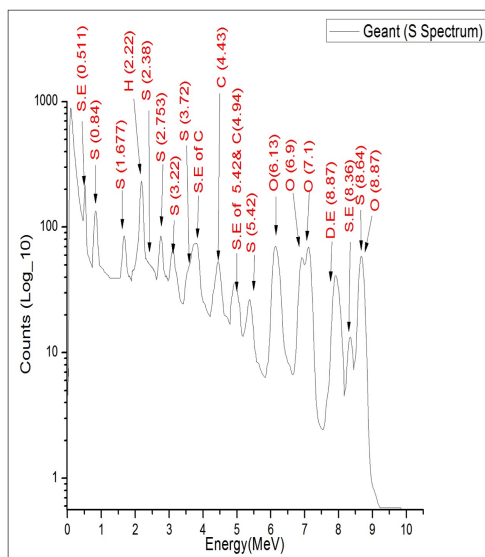
Figure 4.21 : The gamma-ray histogram and spectra of MgO and SiO₂.



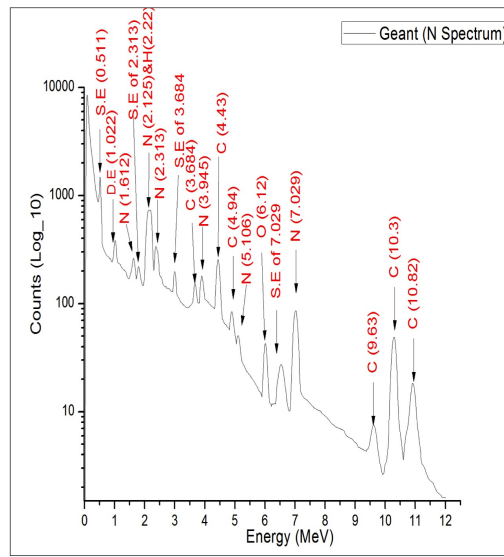
(a) The gamma-ray histogram of S.



(b) The gamma-ray histogram of N.



(c) The gamma-ray spectrum of S.



(d) The gamma-ray spectrum of N.

Figure 4.22 : The gamma-ray histograms and spectra of S and N.



5. RESULTS

Analyzing the obtained spectra from Pulsed Fast and Thermal Neutron Analysis (PFTNA) experiment is very difficult due to its non-linear nature. Library Least Square is a common mathematical approach for treating non-linear radiation effects in cases such as PFTNA. This approach is characterized by the library spectra through MCNP5 and GEANT4 simulations. In this approach the non-linear radiation problem can be treated by iteration and comparison of the estimated and calculated results based on simulation libraries. The simulation LLS method was performed to analyze the elemental content of coal samples such as ash, C, S, H, N, O contents. The moisture value can not be estimated by using H and O elements. The coal as an organic material includes O and H elements independently of moisture. The moisture content was calculated by using weight loss and the result was compared with the variable which was calculated by subtracting of the other elements from the total coal sample. Firstly, the simulation LLS method was tested on the coal samples obtained by MCNP5 simulation. Then the simulation LLS method was applied to experimental spectra. By using Library Least-square method, the content of the coal samples was calculated and compared with the analyzed the coal sample.

5.1 Manufactured Prototype Apparatus

The prototype apparatus including neutron generator, BGO detector, polyethylene and iron shielding material was constructed to be used in the coal mine. The manufactured of prototype apparatus and the six meters of conveyor belt were shown in Figure 5.1

According to laboratory analysis report of coal sample, it includes N element. For that reason, the range of BGO detector was adjust to 12 MeV. The 50 kg of coal sample was irradiated with 14 MeV neutrons for ten minutes. Also, MCNP5 and GEANT4 simulations of coal sample were repeated by adding N element. The gamma-ray spectrum of experimental coal sample was compared with gamma-ray spectra of simulation coal sample obtained by MCNP5 and GEANT4 as shown in Figure 5.2.

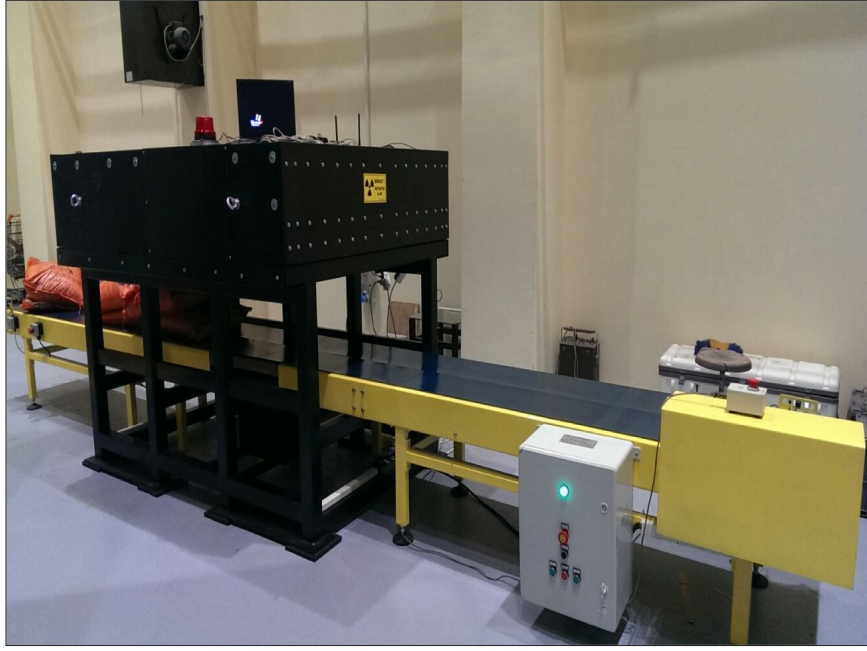


Figure 5.1 : The illustration of manufactured prototype apparatus with 6m conveyor belt.

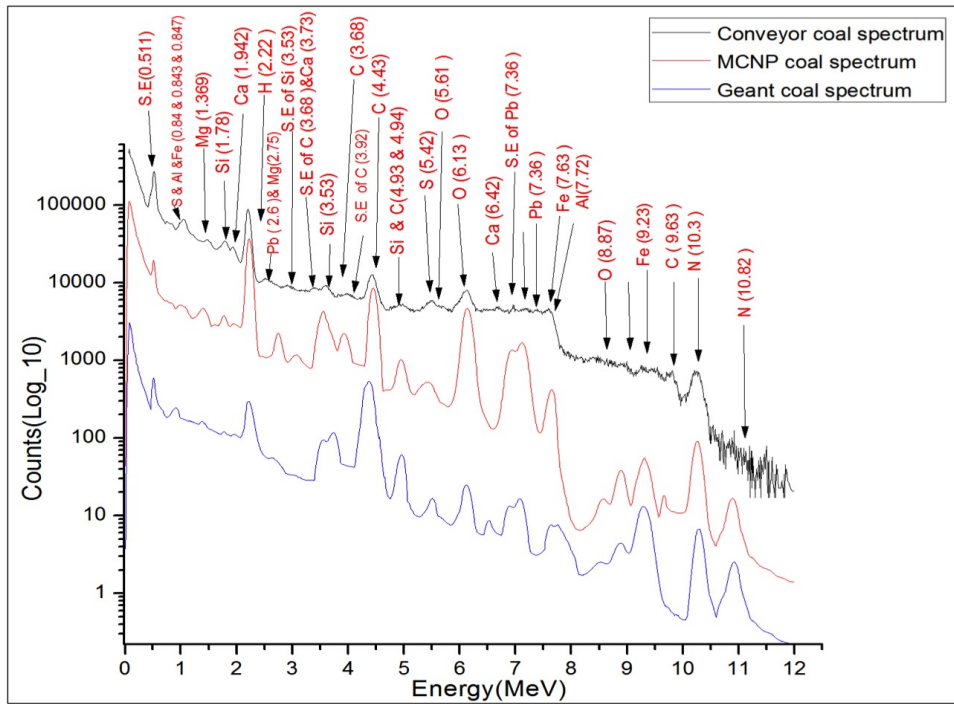


Figure 5.2 : The comparison of gamma-ray spectra of coal sample obtained by MCNP5, GEANT4 and experimental.

The coal sample sets of 2 kg were found similar to each other in terms of counts per channel. Because of this, the amount of coal samples were increased to distinguish better the components of coal content. In order to determine the required amount of

coal sample for calculations, the background of prototype apparatus, coal samples of 30 kg and 50 kg were compared as shown in Figure 5.3

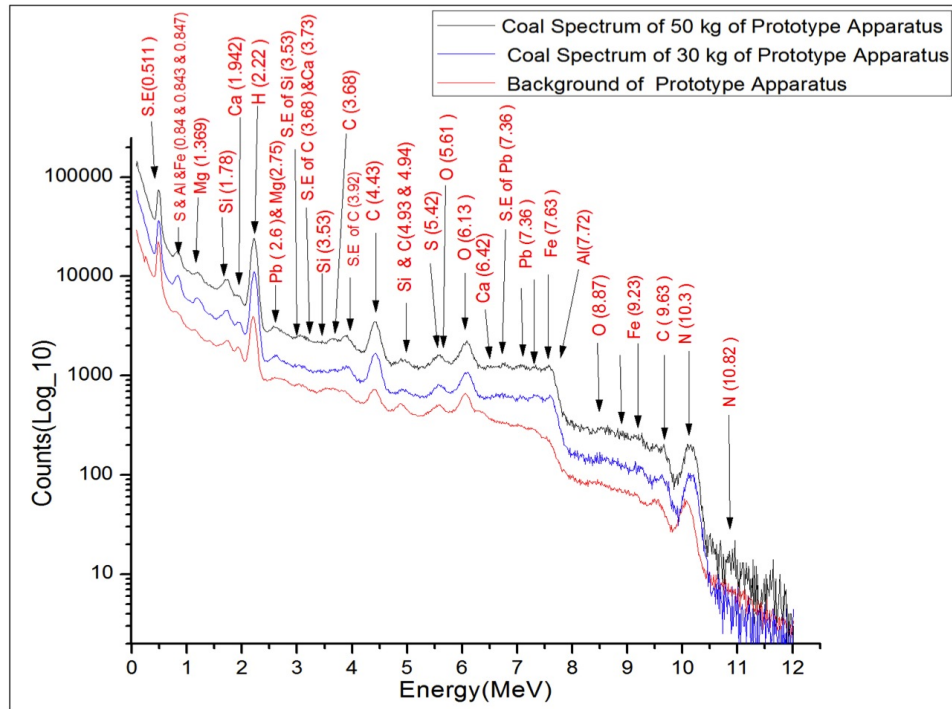


Figure 5.3 : Experimental gamma-ray spectrum of coal sample obtained by manufactured prototype apparatus.

According to Figure 5.3, coal sample of 30 kg was found enough to overcome the background contribution to coal spectrum. For LLS calculations of MCNP5 and GEANT4 library, coal sample sets of 40 kg were prepared.

5.1.1 The MCLLS method for the MCNP5 simulated coal samples

In a spherical geometry, the chemical compounds were simulated by using MCNP and GEANT4. Each spectrum of chemical compounds was used to obtain the element library by utilizing Multiple Linear Regression (MLR) method for experimental spectrum. In order to test the method, The MCNP element library was applied on five coal spectrum of MCNP simulation. After having applied the MLR method on the coal spectrum, the MLR and Coal Set-1 spectra were shown in Figure 5.4.

The regression coefficients obtained from MLR and the regression variables of compounds of each coal set were shown in Table 5.1. The correlation factor between the MLR and the coal sample spectrum was found greater than 0.99 for each coal sample.

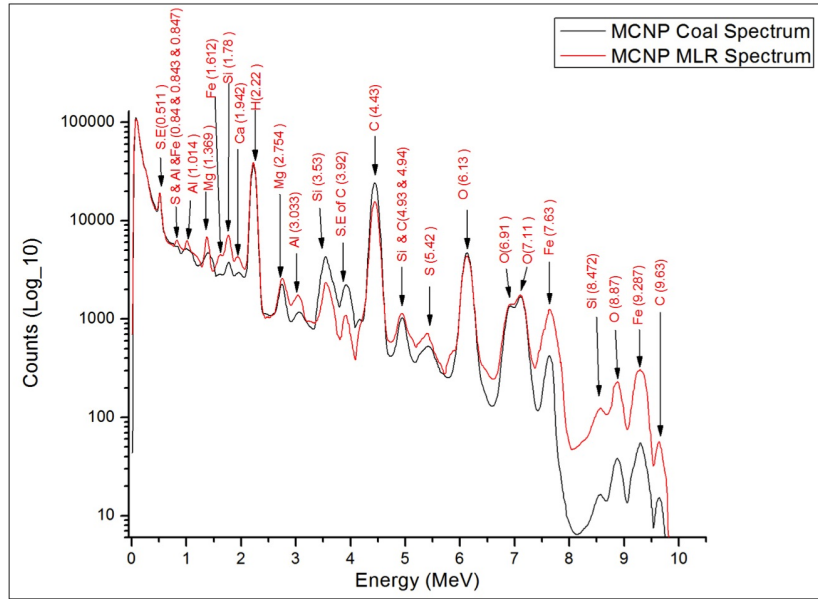


Figure 5.4 : Comparison the gamma-ray spectrum of Coal Set-1 and MLR method obtained by MCNP5.

Table 5.1 : The regression variables of compounds and regression coefficients of each coal set in MLR.

Variable	Set-1	Set-2	Set-3	Set-4	Set-5
a_1 (SiO_2)	31.673	34.495	39.617	54.208	59.304
a_2 (Al_2O_3)	38.309	69.556	96.526	161.826	188.756
a_3 (CaO)	71.799	74.146	76.658	79.760	82.588
a_4 (C)	11.926	3.285	-1.185	-8.813	-12.838
a_5 (Fe_2O_3)	-25.27001	-19.3327	-16.53293	-15.21516	-13.21699
a_6 (MgO)	19.235	27.675	35.541	54.989	62.675
a_7 (S)	-119.463	-134.307	-159.540	-234.502	-261.060
Regression Coefficient	0.99059	0.99527	0.99698	0.99756	0.99897

This result showed that the simulation of coal sample was consistent with simulation of element library. Since the desirable correlation between the gamma-ray spectrum of coal sample and the combination of element library was reached, the concentrations of the element contents for the related coal sample were calculated by using library least squares (LLS) method as given by Equation 2.25 for coal set-1.

The MCNP LLS was applied for five coal sets which were determined contents of coal sample and shown in Table 4.3. The comparison values of prepared element contents with calculated element contents were shown in Table 5.2 for five coal sets. By using Equation 2.25, the X matrix was calculated for each coal spectrum, and the results showing good agreement between prepared elemental contents and calculated values are given in Table 5.2

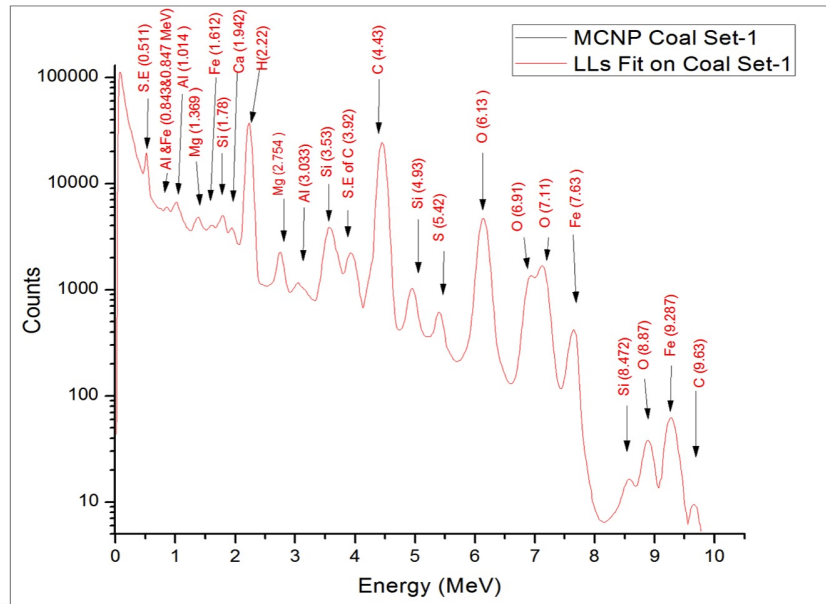


Figure 5.5 : The MCNP5 LLS fit for gamma-ray spectrum of coal Set-1 obtained by MCNP5.

Table 5.2 : The comparison of the concentration of elemental contents between prepared and calculated values for the each coal set.

Compound	Set-1		Set-2		Set-3		Set-4		Set-5	
	Prep.(%)	Cal.(%)	Prep.(%)	Cal.(%)	Prep.(%)	Cal.(%)	Prep.(%)	Cal.(%)	Prep.(%)	Cal.(%)
SiO ₂	8.20	8.07	10.86	10.77	13.60	13.75	16.34	16.44	19.00	19.02
Al ₂ O ₃	3.60	3.56	4.94	4.87	6.14	6.22	7.34	7.40	8.62	8.61
CaO	3.60	3.56	4.94	4.87	6.14	6.23	7.34	7.40	8.62	8.61
C	76.0	75.98	69.34	69.26	62.66	62.72	56.0	56.05	49.34	49.34
Fe ₂ O ₃	3.60	3.51	4.66	4.63	5.86	5.91	7.10	7.10	8.22	8.21
MgO	1.00	0.93	1.26	1.25	1.60	1.61	1.86	1.87	2.20	2.20
S	4.00	3.99	4.00	3.98	4.00	4.02	4.00	4.05	4.00	3.98
Total	100.00	99.60	100.00	99.63	100.00	100.46	100.00	100.31	100.00	99.97

5.1.2 The MCLLS method for the experimental spectra

The LLS method was used to determine the contents of experimental coal samples which have laboratory analysis report. According the analysis report, the coal sample has C, N, O, H, S, ash (MgO, Fe₂O₃, CaO, Al₂O₃, SiO₂), and moisture with different ratios. The moisture content was calculated by weight loss. The contents of analyzed coal samples were shown in Table 5.3.

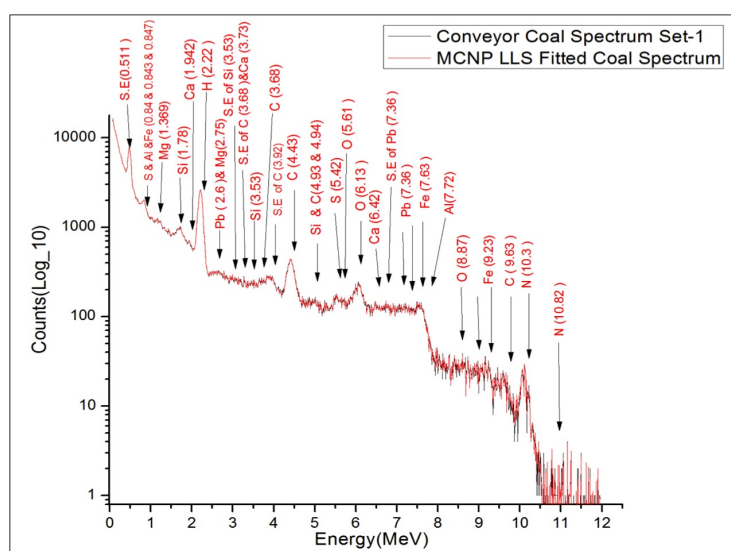
The results of the MCNP5 LLS and GEANT4 LLS were shown in Table 5.4 and Table 5.5. X matrix was found for each of the sets individually after writing a code in MATLAB. Additionally, at this stage MCLLS was created by taking into account values of Table 5.3. For this purpose A matrix was calculated once again. According

Table 5.3 : The element contents of analyzed coal samples.

Set#	C(%)	H(%)	N(%)	S(%)	Ash(%)	O(%)	SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	CaO(%)	MgO(%)	Moist.(%)
Set1	71.09	3.29	1.49	1.57	10.51	2.37	6.02	2.3	2.02	0.25	0.37	9.67
Set2	69.67	3.2	1.44	1.53	12.48	2.3	7.05	2.7	2.38	0.29	0.42	9.38
Set3	68.87	3.14	1.42	1.5	13.58	2.26	7.63	2.92	2.59	0.36	0.51	9.22
Set4	68.42	3.11	1.4	1.49	14.2	2.24	7.95	3.05	2.7	0.38	0.54	9.13
Set5	67.89	3.07	1.39	1.47	14.94	2.21	8.34	3.2	2.84	0.41	0.58	9.02
Set6	67.36	3.03	1.37	1.45	15.68	2.18	8.72	3.35	2.97	0.44	0.61	8.92
Set7	66.65	2.99	1.35	1.43	16.66	2.15	9.24	3.55	3.15	0.47	0.66	8.77
Set8	66.38	2.97	1.34	1.42	17.03	2.14	9.43	3.62	3.22	0.49	0.67	8.74
Set9	66.12	2.95	1.33	1.41	17.4	2.12	9.62	3.7	3.29	0.5	0.69	8.67
Set10	65.85	2.93	1.32	1.4	17.76	2.11	9.82	3.77	3.36	0.52	0.71	8.61
Set11	65.67	2.92	1.31	1.39	18.01	2.1	9.95	3.82	3.4	0.55	0.72	8.57

to Table 5.4 and Table 5.5, after using the LLS method, the calculated value of the concentration of elements was found in excellent agreement with the analyzed value of the concentration of the elements.

The MCNP5 and GEANT4 fitting spectra on Coal Set-1 were shown in Figure 5.6 and Figure 5.7 respectively. For C, H, O, N, S ash and moisture the deviations were found less than 0.5% from the analyzed values of the element contents. The deviation values of C, H, O, N, S ash and moisture of MCNP5 LLS and GEANT4 LLS were shown in Table 5.6. For C, H, O, N, S ash and moisture content, the comparisons of laboratory results with calculation values obtained by MCNP5 LLS and GEANT4 LLS were shown in Figure 5.8, Figure 5.9, Figure 5.11, and Figure 5.10.

**Figure 5.6** : The fitting gamma-ray spectrum on Coal Set-1 by using MCNP5 LLS.

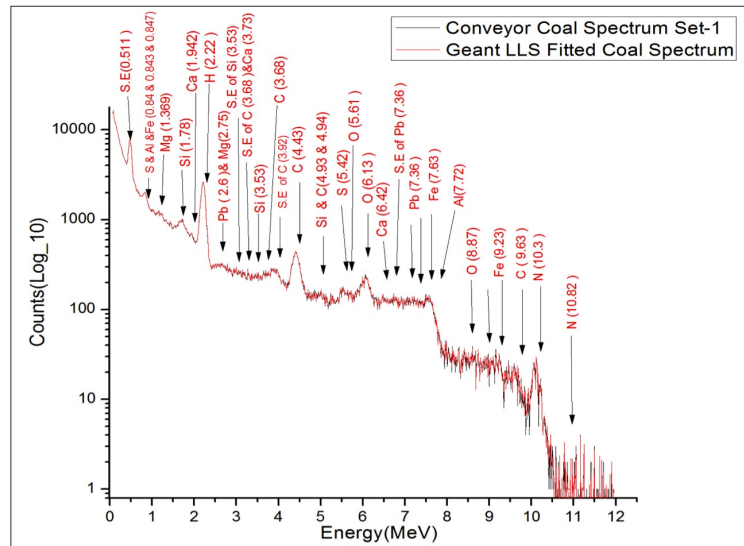


Figure 5.7 : The fitting gamma-ray spectrum on Coal Set-1 by using GEANT4 LLS.

Table 5.4 : The calculation results of element contents of coal samples by MCNP5 LLS method.

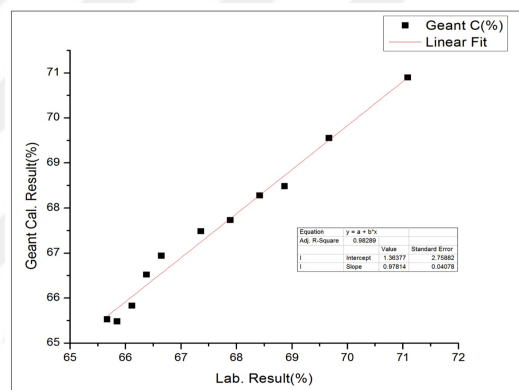
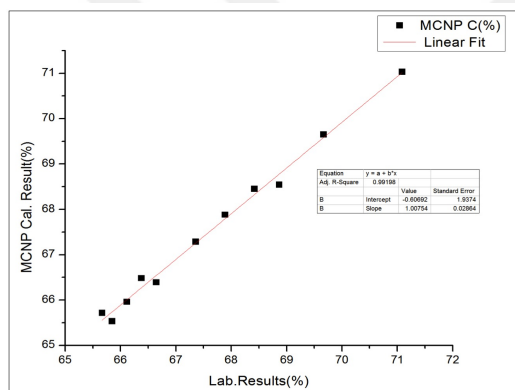
Set#	C(%)	H(%)	N(%)	S(%)	Ash(%)	O(%)	SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	CaO(%)	MgO(%)	Mst.(%)
Set1	71.02	3.288	1.489	1.571	10.86	2.368	5.908	2.269	1.998	0.246	0.365	9.397
Set2	69.64	3.193	1.436	1.533	12.825	2.295	7.058	2.695	2.373	0.285	0.414	9.069
Set3	68.54	3.15	1.425	1.493	13.945	2.268	7.893	2.904	2.576	0.354	0.563	9.234
Set4	68.45	3.118	1.404	1.48	14.68	2.246	7.977	3.107	2.696	0.376	0.608	9.681
Set5	67.88	3.057	1.383	1.463	15.267	2.2	8.25	3.184	2.79	0.407	0.636	8.817
Set6	67.28	3.037	1.374	1.436	16.169	2.185	8.644	3.47	2.943	0.435	0.677	8.546
Set7	66.39	2.999	1.355	1.436	16.57	2.157	8.989	3.444	3.049	0.452	0.636	9.093
Set8	66.48	2.965	1.338	1.418	17.411	2.137	9.467	3.639	3.258	0.48	0.667	8.251
Set9	65.95	2.946	1.328	1.406	17.661	2.117	9.67	3.62	3.122	0.506	0.743	8.616
Set10	65.53	2.926	1.318	1.383	18.087	2.107	9.762	3.69	3.371	0.525	0.739	8.699
Set11	65.716	2.924	1.312	1.386	18.408	2.103	9.85	3.852	3.378	0.559	0.769	8.191

Table 5.5 : The calculation results of element contents of coal samples by GEANT4 LLS method.

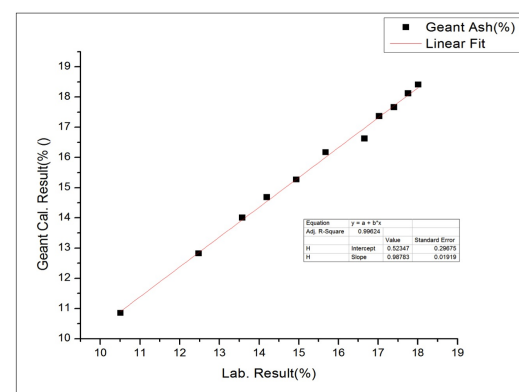
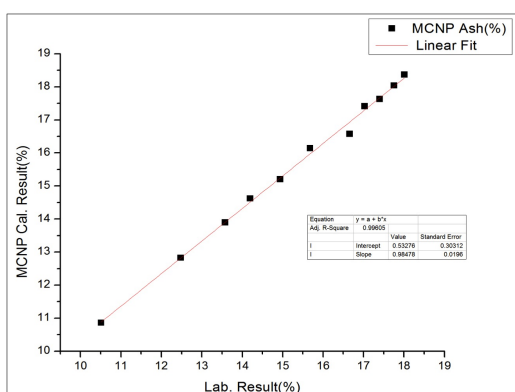
Set#	C (%)	H (%)	N (%)	S (%)	Ash (%)	O (%)	SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	CaO(%)	MgO(%)	Mst.(%)
Set1	70.89	3.302	1.499	1.568	10.848	2.359	5.958	2.277	2.008	0.246	0.359	9.527
Set2	69.54	3.189	1.442	1.537	12.819	2.305	7.048	2.705	2.375	0.287	0.414	9.159
Set3	68.48	3.144	1.417	1.493	13.937	2.251	7.641	2.914	2.582	0.364	0.446	9.213
Set4	68.27	3.121	1.41	1.482	14.621	2.235	7.548	3.067	2.715	0.378	0.484	8.796
Set5	67.73	3.055	1.387	1.46	15.2	2.22	8.3	3.192	2.8	0.413	0.503	8.881
Set6	67.48	2.955	1.254	1.434	16.139	2.15	8.67	3.457	2.951	0.456	0.608	8.555
Set7	66.94	2.987	1.349	1.438	16.623	2.149	8.997	3.467	3.053	0.476	0.653	8.154
Set8	66.52	2.976	1.332	1.405	17.366	2.135	9.397	3.614	3.189	0.481	0.685	8.266
Set9	65.82	2.968	1.323	1.418	17.628	2.123	9.58	3.75	3.127	0.496	0.675	8.678
Set10	65.48	2.92	1.311	1.38	18.068	2.1	9.864	3.7	3.358	0.516	0.65	8.691
Set11	65.52	2.897	1.317	1.394	18.368	2.107	9.888	3.847	3.382	0.556	0.69	8.351

Table 5.6 : The deviation values of C, H, O, N, S ash and moisture obtained by MCNP5 LLS and GEANT4 LLS respectively.

Set	C%	C%	Ash%	Ash%	S%	S%	H%	H%	O%	O%	N%	N%	Mst.%	Mst.%
1	0.063	0.193	0.35	0.28	0.001	0.002	0.002	0.012	0.002	0.011	0.001	0.009	0.273	0.143
2	0.021	0.121	0.349	0.345	0.003	0.007	0.007	0.011	0.005	0.005	0.004	0.002	0.311	0.221
3	0.33	0.39	0.365	0.357	0.007	0.007	0.01	0.004	0.008	0.009	0.005	0.003	0.014	0.007
4	0.03	0.144	0.480	0.421	0.01	0.008	0.008	0.011	0.006	0.005	0.004	0.01	0.449	0.334
5	0.01	0.16	0.335	0.26	0.007	0.01	0.013	0.015	0.01	0.01	0.007	0.003	0.203	0.139
6	0.077	0.123	0.489	0.459	0.014	0.024	0.007	0.005	0.005	0.007	0.004	0.203	0.374	0.365
7	0.26	0.29	0.09	0.037	0.006	0.008	0.009	0.003	0.007	0.001	0.005	0.001	0.323	0.256
8	0.1	0.14	0.381	0.336	0.002	0.015	0.005	0.006	0.003	0.005	0.002	0.008	0.489	0.474
9	0.161	0.273	0.261	0.228	0.004	0.008	0.004	0.018	0.003	0.003	0.002	0.007	0.054	0.008
10	0.32	0.37	0.327	0.308	0.017	0.02	0.004	0.01	0.003	0.01	0.002	0.009	0.089	0.081
11	0.046	0.144	0.398	0.358	0.004	0.004	0.004	0.023	0.003	0.007	0.002	0.007	0.379	0.219

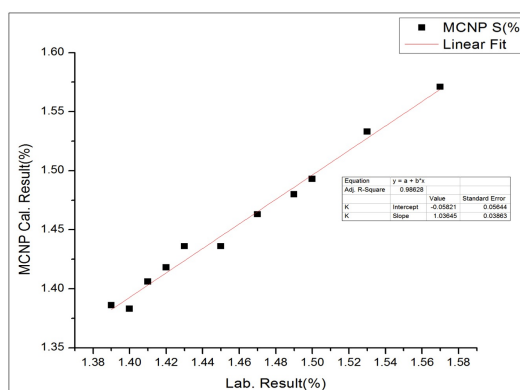


(a) The comparison of MCNP5 LLS with labora- (b) The comparison of GEANT4 LLS with labora-
tory results for C. tory results for C.

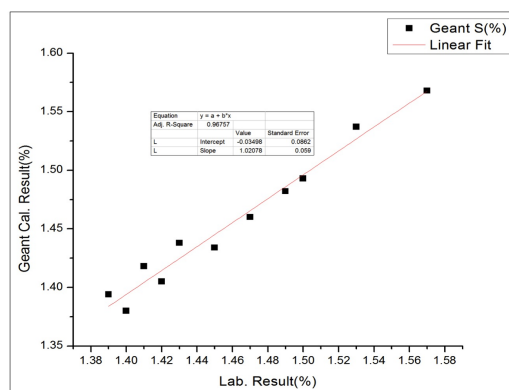


(c) The comparison of MCNP5 LLS with labora- (d) The comparison of GEANT4 LLS with labora-
tory results for ash. tory results for ash.

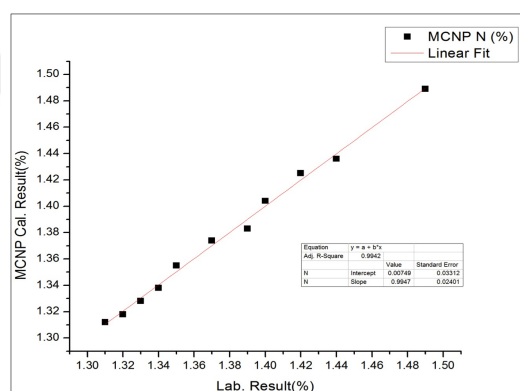
Figure 5.8: The comparison of laboratory results of C and ash contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.



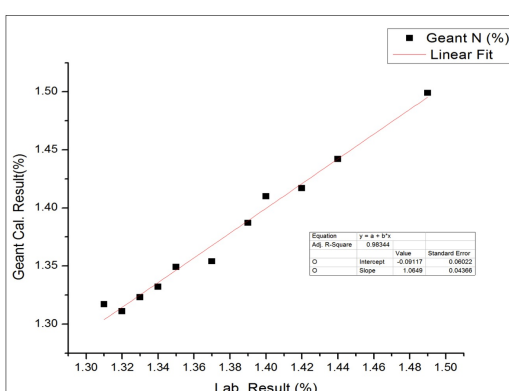
(a) The comparison of MCNP5 LLS with labora-



(b) The comparison of GEANT4 LLS with labora-

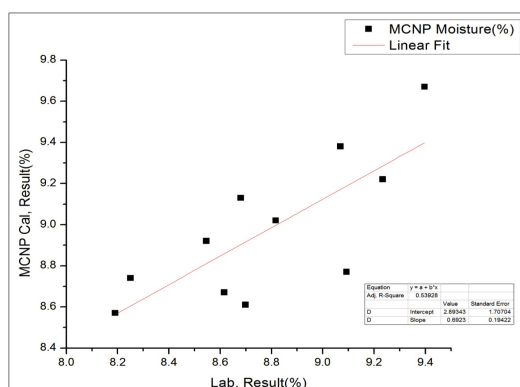


(c) The comparison of MCNP5 LLS with labora-

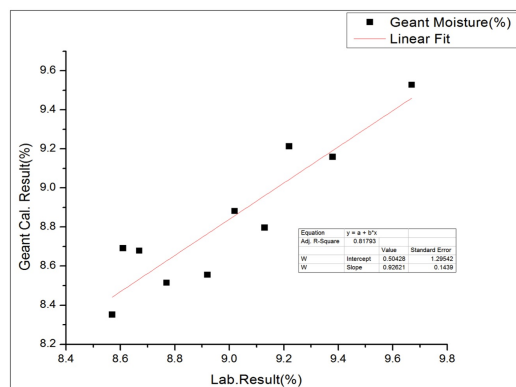


(d) The comparison of GEANT4 LLS with labora-

Figure 5.9: The comparison of laboratory results of S and N contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.

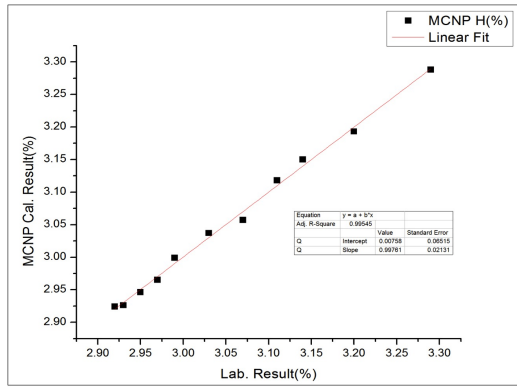


(a) The comparison of MCNP5 LLS with labora-

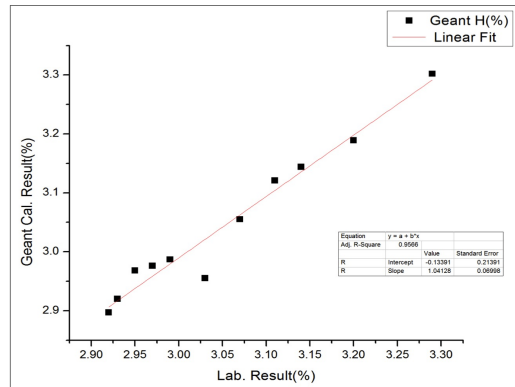


(b) The comparison of GEANT4 LLS with laboratory results for moisture.

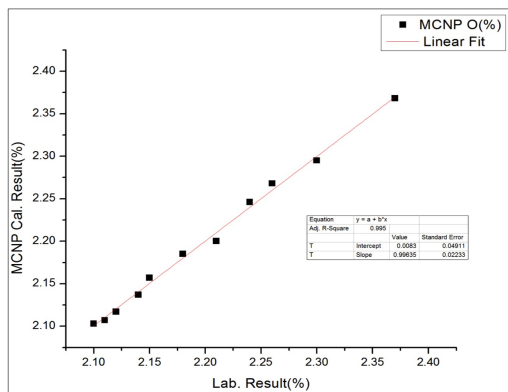
Figure 5.10 : The comparison of laboratory results of moisture contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.



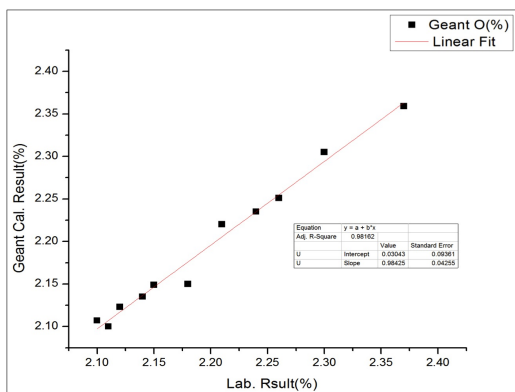
(a) The comparison of MCNP5 LLS with labora-



(b) The comparison of GEANT4 LLS with labora-
tory results for H.



(c) The comparison of MCNP5 LLS with labora-



(d) The comparison of GEANT4 LLS with labora-
tory results for O.

Figure 5.11 : The comparison of laboratory results of H and O contents with calculation values obtained by MCNP5 LLS and GEANT4 LLS.

The comparisons between Figure 5.8 and Figure 5.11 refer to the ratios through set1 to set11 which were assumed as unknown samples. These calculation were in the form of verification. They were observed that these values were so close to the values written in Table 5.3 except moisture content. The moisture contents were calculated by subtracting other contents from the total amount. But still, deviation values from analyzed moisture content were less than 0.5 % which were acceptable for these type of element analysis devices.

In Figure 5.6 and Figure 5.7, there were a fitting procedure between an unknown sample total gamma ray spectrum and MCLLS. It was observed that they are in good agreement with each other. There was not found any comparative study related with MCLLS of coal sample obtained by MCNP5 and GEANT4 in literature, therefore this calculation process has unique importance.

6. CONCLUSION

For our country, the process of improving the quality and preparation of the coal carries great importance to produce the energy efficiently from the coal power plants. For that purpose, a prototype apparatus was aimed to design by using some nuclear techniques. The prototype apparatus was developed to be able to determine the content of the ash, moisture and calorific value of the coal moving on the conveyor belt as on-line.

The aim of this thesis was to simulate a prototype apparatus which was used to analyze the content of moisture, ash, sulfur, and carbon of a coal sample on-static position. Monte Carlo N-Particle 5 (MCNP5) and Geometry And Tracking 4 (GEANT4) simulation programs were used to design the geometry and physical interactions of the prototype apparatus. Both of synthetic coal samples and experimental coal samples including different element contents were simulated and analyzed. Different design options were tested by using both experimental and simulation methods. The coal spectrum obtained from transmission and back-scattering position of BGO detector were compared. Because of self-absorption inside of the coal sample, some of gamma energy levels could not be detected by BGO detector at transmission position. Therefore the placement of BGO was decided to be at back-scattering position for the detection of coal sample content.

In order to obtain the optimum results from the NDA, the gamma-ray spectroscopy BGO detector used in the prototype apparatus was tested to examine the characteristic features. The studies about performance of neutron generator and calibration of BGO were performed. Synthetic coal samples were prepared by taking into account of coal reserves in Aegean region of Turkey. Eight sets with varying elemental contents of 2 kg were irradiated 10 minutes by neutrons of 14 MeV. The obtained gamma spectrum was not enough to distinguish the contents of each set. One of the reasons might be that the background contribution obtained from concrete walls and polyethylene blocks used to cover the generator surpassed the coal spectrum. It was concluded that amount of synthetic coal sample was not good enough to determine the concentration of element

contents in coal sample. So it was decided to increase the amount of coal sample for MCLLS calculations.

In order to analyze the spectrum of PFTNA, Monte-Carlo LLS (MCLLS) was obtained by using MCNP5 and GEANT4. The Monte Carlo technique was designed to produce elemental libraries for interested sample by taking into account the prompt gamma ray contributions from shielding material and mixture of elements. In addition to this, non-linear effects of detection system could be fixed with MCLLS method

In order to define the elemental contents of coal sample, MCNP5 Library Least Square method (MCNP5 LLS) and GEANT4 Library Least Square (GEANT4 LLS) were developed. Therefore, the information of gamma ray interactions of each compound induced by neutrons was obtained in a simple simulation geometry. Unlike the geometry of prototype apparatus, each of SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , MgO , S, C, and N compounds was simulated separately in a spherical fixed volume.

Firstly, MCLLS was applied for simulation coal spectrum of MCNP5. The desirable correlation between the coal spectrum and the combination of element library was reached. Then the LLS method was used to determine the contents of experimental coal samples which have laboratory analysis report. According the analysis report, the coal sample has C, N, O, H, S, ash (SiO_2 , Al_2O_3 , Fe_2O_3 , CaO , MgO), and moisture with different ratios. For applying MCNP5 LLS and GEANT4 LLS, eleven coal sample sets of 40 kg were prepared for calculating contents of ten elements. By using the LLS method, the calculated value of the concentration of elements was found in excellent agreement with the analyzed value of the concentration of the elements.

Except the moisture content, a linear relationship was found between calculated values and laboratory results. The moisture could not be calculated independently from other elements. Since O element may belong either ash content or moisture. H element may belong to either hydrocarbon content or moisture. Because of this reason, the moisture content was calculated by subtracting the other elements from the total coal sample. Thus the moisture content was effected directly from the deviation values of the other contents of coal sample. As the deviation values of ash contents were investigated, the results of GEANT4 LLS method were found to have closer values to laboratory values. As the deviation values of carbon contents were investigated, the

results of MCNP5 LLS method were found to have closer values to laboratory values. The deviation values of C, H, O, N, S and moisture were found less than 0.5% from the analyzed values of the element contents. By comparing other PGNAA devices, the accuracy and precision values of measurements were found satisfactory.





REFERENCES

- [1] **Url-1**<<https://www.worldenergy.org/wp-content/uploads/2016/10/World-Energy-Resources-Full-report-2016.10.03.pdf>>, date retrieved 10.11.2017.
- [2] **Url-2**<<http://www.enerji.gov.tr/en-US/Pages/Electricity>>, date retrieved 10.11.2017.
- [3] **Kirchner, A.T., Maude, C.M.** (1994). *On-line analysis of coal - symposium review.*, IEA Coal Research, London, UK.
- [4] **Sowerby, B., D.** (1991). Online nuclear techniques in the coal industry, *Nuclear Geophysics*, 5, 491-506.
- [5] **Eiji, Yoshida., Kiyoshi, Shizuma., Satoru, Endo., Takamitsu, Oka.** (2002). Application of neural networks for the analysis of gamma-ray spectra measured with a Ge spectrometer, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 484(1-3), 557-563.
- [6] **İ., A., Reyhancan., B., Untu., M., N., Erduran.** (2016). MCNP5 Element Library Least Squares Method for On-line Coal Analysis, *Journal of Nuclear Sciences* 3(2), 27-34.
- [7] **Fusheng, Li., Weijun, Guo., Robin., P., Gardner.** (2008). Implementation Of Monte Carlo-Library Least Squares Approach to Energy Dispersive X-Ray Fluorescence Analysis, *International Centre for Diffraction Data*. Retrieved from <https://pdfs.semanticscholar.org>
- [8] **Alfassi, Z., B.** (1990). *Activation Analysis*, Volumes I and II., CRC Press, Boca Raton, FL .
- [9] **E., V., Sayre., R., W., Dodson., and Dorothy, Burr, Thompson.** (1957). Neutron Activation Study of Mediterranean Potsherds, *American Journal of Archaeology* 61(1), 35-41.
- [10] **Perlman, I., and F., Asaro.** (1969). Pottery analysis by neutron activation, *Archaeometry* 11, 21-52.
- [11] **Bishop, Ronald, L.** (1975). *Western Lowland Maya Ceramic Trade: An Archaeological Application of Nuclear Chemical and Geological Data Analysis*, (Doctorate dissertation), Southern Illinois University, Carbondale. Ann Arbor: University Microfilms International.
- [12] **Bieber, A., M., Jr., D., W., Brooks., G., Harbottle., and E., V., Sayre.** (1976). Application of multivariate techniques to analytical data on Aegean ceramics, *Archaeometry* 18, 59-74.
- [13] **Harbottle, G.** (1982). *Provenance Studies Using Neutron Activation Analysis: The Role of Standardization In Archaeological Ceramics*, (pp. 67-77), Smithsonian Institution Press, Washington, D.C.

- [14] **Alfassi, Z., B.** (1998). *Instrumental Multi-Element Chemical Analysis*, Kluwer Academic Publishers, Dordrecht, Netherlands.
- [15] **Sowerby, B., D., Cutmore, N.** (2008). Personal communication, Retrieved from <https://csiropedia.csiro.au/coalscan-on-line-analysis-instruments-for-the-coal-industry>.
- [16] **Lim, C., S.** (2004). Recent developments in neutron-induced gamma activation for on-line multielemental analysis in industry, *Journal of Radioanalytical and Nuclear Chemistry* 262, 525-532.
- [17] **Lim, C.S., Sowerby, B., D.** (2005). On-line bulk elemental analysis in the resource industries using neutron-gamma techniques, *Journal of Radioanalytical and Nuclear Chemistry* 264, 15-19.
- [18] **Lim, C.S., Sowerby, B.D., Abernethy, D.A.** (2002). On-belt Analysis of Ash in Coal, *Coal Preparation*, 22(3), 165-179.
- [19] **Jing, S., Gu, D., Qiao, S., Liu, Y., Liu, L.** (2005). Development of pulse neutron coal analyzer, *Review of Scientific Instruments* 76(4), 5110, doi: 10.1063.1/1889232.
- [20] **Borsaru, M., Berry M., Biggs., M., Rojc A.** (2004). In situ determination of sulphur in coal seams and overburden rock by PGNA, *Nuclear Instruments and Methods in Physics Research B* 213, 530-534.
- [21] **Dep, L., Belbot, M., Vourvopoulos, G., Sudar, S.** (1998). Pulsed neutron-based on-line coal analysis, *Journal of Radioanalytical and Nuclear Chemistry* 234, 107-112.
- [22] **Woodward, R., C., Evans, M., P., Empey E., R.** (2008). *A major Step Forward for On-line Analysis*, Thermo Electron Corporation, Electric Power Research Institute (EPRI).
- [23] **Roald, A., Schrack., Charles, D., Bowman.** (1975). *Nuclear Cross Sections and Technology: Proceedings of a Conference*, (March 3-7), Washington, D.C., Department of Commerce, National Bureau of Standards, Institute for Basic Standards, Center for Radiation Research.
- [24] **Kenneth, S., Krane.** (1987). *Introductory Nuclear Physics*, John Wiley and Sons Ltd, New York, United States.
- [25] **W., H., Hallenbeck.** (1994). *Radiation Protection*, Lewis Publishers, Florida.
- [26] **Glenn, F., Knoll.** (2000). *Radiation Detection and Measurement*, Wiley.
- [27] **Url-3**<<http://www.nuclear-power.net>>, date retrieved 10.05.2017.
- [28] **Paull, Reuss.** (2008). *Neutron Physics*, EDP Sciences, France.
- [29] **P., Rinard.** (1991). Neutron interactions with matter, (pp. 357-377), *In Passive Nondestructive Assay of Nuclear Materials*.
- [30] **Url-4**<https://www.medphysics.wisc.edu/courses/mp501/ne408/handoutsSentMaterials/neutron_inter.pdf>, date retrieved 11.05.2017.
- [31] **Url-5**<http://www.kayelaby.npl.co.uk/atomic_and_nuclear_physics/4_7/4_7_2f.html>, date retrieved 11.05.2017.

- [32] **Alevra, A., V., and Thomas, D., J.** (2003). Neutron spectrometry in mixed elds: multisphere spectrometers, *Radiation Protection Dosimetry* 107(1–3), 37–72.
- [33] **Url-6**<http://archaeometry.missouri.edu/naa_overview.html>, date retrieved 10.08.2017.
- [34] **Url-7**<https://three.jsc.nasa.gov/articles/Heilbronn_Neutron_Supplement.pdf>, date retrieved 11.05.2017.
- [35] **Fritz, A., R., Schmidt.** (1970). *The Attenuation Properties Of Concrete For Shielding Of Neutrons Of Energy Less Than 15 MeV*, Oak Ridge National Laboratory, Tennessee.
- [36] **A., V., Astin.** (1957). Protection Against Neutron Radiation Up To 30 Million Electron Volts, *National Bureau Of Standards Handbook-63* 41(17), Los Alamos Scientific Laboratory, Washington 25, D. C.
- [37] **Sowerby, B., D.** (2009). Nuclear techniques for the On-Line Bulk Analysis of Carbon in Cola-Fired Power Stations, *Applied Radiation and Isotopes* 67, 1638-1643.
- [38] **Womble, P. C., Paschal, J. and Moore, R.** (2005). Cement Analysis Using d + D neutrons, *Nuclear Instruments and Methods in Physics Research B* 241(1-4), 765-769.
- [39] **Shypailo, R. J., Ellis, K., J.** (2005). Design Considerations for a Neutron Generator-Based Total-Body Irradiator, *Journal of Radioanalytical and Nuclear Chemistry* 263(3), 759-765.
- [40] **James, W., D., and Fuerst, C., D.** (2000). Overcoming Matrix Effects in the 14-MeV Fast Neutron Activation Analysis of Metals, *Journal of Radioanalytical and Nuclear Chemistry* 244(2), 429-434.
- [41] **Wielopolski, L., Hendrey, G., Johnsen, K., Mitra, S., Prior, S., Rogers, H., and Tolbert, H.** (2008). Nondestructive System for Analyzing Carbon in the Soil, *Soil Science Society of America Journal* 72, 1269-1277.
- [42] **Nikitin, A., and Bliven, S.** (2010). Needs of Well Logging Industry in New Nuclear Detectors, (pp. 1214-1219), *Nuclear Science Symposium Proceedings*, (Oct. 30 – Nov. 6), Knoxville, TN.
- [43] **Maglich, B., C., and Nalcioglu, O.** (2010). ONCOSENSOR for Noninvasive High-Specificity Breast Cancer Diagnosis by Carbogen-Enhanced Neutron Femto-Oximetry, (pp. 57-58), *ASME Conference Proceedings: Congress on Nano-Engineering for Medicine and Biology*, New York, N.Y.
- [44] **Vourvopoulos, G., and Womble, P.** (2001). Pulsed Fast Thermal Neutron Analysis: A Technique for Explosives Detection, *Talanta* 54(3), 459-468.
- [45] **Reber, E., L., Blackwood, L.G., Edwards, A., J., Jewell, J., K., Rohde, K., W., Seabury, E., H. and Klinger, J., B.** (2005). Idaho Explosives Detection System, *Nuclear Instruments and Methods in Physics Research B* 241(1-4), 738-742.
- [46] **Barzilov, A., and Womble, P., C.** (2003). NELIS - A Neutron Inspection System for Detection of Illicit Drugs, *AIP Conference Proceedings* 680, 939-942.

- [47] **D., T., Shyu, C., M., Sullivan, R., A., and Vourvopoulos, Holstin., G.** (2006). PELAN for Non- Intrusive Inspection of Ordnance, Containers, and Vehicles, *Proceedings of SPIE* 6213, 621307.
- [48] **Alexander, P., Barzilov, Ivan, S., Novikov, and Phillip C., Womble.** (2012). Material Analysis Using Characteristic Gamma Rays Induced by Neutrons, *Gamma Radiation*, doi: 10.5772/36372 .
- [49] **B., K., Das, and A., Shyam.** (2008). *Review of Scientific Instruments* 79, 123305.
- [50] **Duderstadt, J., J., and Moses, G., A.** (1982). *Inertial Confinement Fusion*, Wiley-Interscience Publication.
- [51] **Bishnupriya Nayak, and S., V., G., Menon.** (2012). *Laser and Particle Beams*, 30 517–523, doi:10.1017/S0263034612000365.
- [52] **Gozani, T.** (1995). Understanding the Physics Limitations of PFNA - The Nanosecond Pulsed Fast Neutron Analysis, *Nuclear Instruments and Methods in Physics Research B*, 99, 743-747.
- [53] **Strellis, D., A., Gozani, T., and Stevenson, J.** (2009). Air Cargo Inspection Using Pulsed Fast Neutron Analysis, *IAEA Proceedings Series: Topical Meeting on Nuclear Research - Applications and Utilization of Accelerators*, Paper SM/EN-05, International Atomic Energy Agency, Vienna, Austria.
- [54] **Overley, J.C., Chmelik, M.S., Rasmussen, R.J., Schofield, R.M.S., Sieger, G.E., and Lefevre, H.W.** (2006). Explosives Detection via Fast Neutron Transmission Spectroscopy, *Nuclear Instruments and Methods in Physics Research Section B*, 251, 470-478.
- [55] **NNMAB-482-6** (1999). *The Practicality of Pulsed Fast Neutron Transmission Spectroscopy for Aviation Security*, National Academies Press.
- [56] **Koltick, D.,S., Kane, S.,Z., Lvovsky, M., Mace, E.,K., McConchie, S.,M., and Mihalcz, J., T.** (2009). Characterization of an Associated Particle Neutron Generator With ZnO:Ga Alpha-Detector and Active Focusing, *IEEE Transactions on Nuclear Science*, 56, 1301-1305.
- [57] **Cooper, J.,C., Koltick, D.,S., Mihalcz, J.,T., and Neal, J., S.** (2003). Evaluation of ZnO(Ga) Coatings as Alpha Particle Transducers within a Neutron Generator, *Nuclear Instruments and Methods in Physics Research A*, 505(1-2), 498-501.
- [58] **Perret, G., Perot, B., Artaud, J-L., and Mariani, A.** (2006). EURITRACK Tagged Neutron Inspection System, *Journal of Physics Conference Series*, 41, 375-383.
- [59] **Eleon, C., Perot, B., Carasco, C., Sudac, D., Obhodas, J., and Valcovic, V.** (2010). Experimental and MCNP Simulated Gamma-Ray Spectra for the UNCOSS Neutron-Based Explosive Detector, *Nuclear Instruments and Methods in Physics Research A*, 629, 220-229.
- [60] **Vourvopoulos, G., and Womble, P.** (2001). Pulsed Fast Thermal Neutron Analysis: A Technique for Explosives Detection, *Talanta*, 54, 459-468.
- [61] **Womble, P.C., Paschal, J., and Moore, R.** (2005). Cement Analysis Using d + D neutrons, *Nuclear Instruments and Methods in Physics Research B*, 241(1-4), 765-769.

- [62] **Attix, F., H.** (1986). *Introduction to Radiological Physics and Radiation Dosimetry*, John Wiley & Sons, New York.
- [63] **Url-8**<<http://hyperphysics.phy-astr.gsu.edu>>, date retrieved 11.05.2017.
- [64] **E., Frytag.**(2006). *Strahlenschutz an hochenergiebeschleunigern*, G.Braum, Karlsruhe, 1072.
- [65] **Berger, M., J., Hubbell, J., H., Seltzer, S., M., Coursey, J., S., and Zucker, D., S.** (1999). *Nistxcom: Photon Cross Sections Database*, Retrieved from <http://physics.nist.gov/PhysRefData/Xcom/Text/XCOM.html>.
- [66] **Url-9**<<https://radiologykey.com/radiation-detectors>>, date retrieved 11.05.2017.
- [67] **J., Eberthaj, and J., Simpson.** (2008). From Ge(Li) detectors to gamma-ray tracking arrays—50 years of gamma spectroscopy with germanium detectors, *Progress in Particle and Nuclear Physics* 60(2), 283-337.
- [68] **Renato Turchetta.** (2016). *Analog Electronics for Radiation Detection*, CRC Press.
- [69] **Doug Reilly, Norbert Ensslin and Hastings Smith., Jr.** (1991). *Passive Nondestructive Assay of Nuclear Materials*, Los Alamos National Laboratory.
- [70] **Url-10**<<http://www.kip.uni-heidelberg.de>>, date retrieved 11.05.2017.
- [71] **B., S., Chauhan.** (2008). *Problems and Solutions in Engineering Chemistry*, Laxmi Publications.
- [72] **Url-11**<<https://www.hofstragroup.com/product>>, date retrieved 11.05.2017.
- [73] **Url-12**<<http://www.cnstn.rnrt.tn/afra-ict/NAT/gamma/html/pulsespectrum.htm>>, date retrieved 11.05.2017.
- [74] **Url-13**<<http://file.wiener-d.com/documentation>>, date retrieved 11.05.2017.
- [75] **Url-14**<<https://www.britannica.com/science>>, date retrieved 11.05.2017.
- [76] **James, G., Speight.** (2005). *Handbook of Coal Analysis*, USA.
- [77] **Url-15**<<http://www.uky.edu/KGS/coal-analyses-proximate.php>>, date retrieved 10.09.2017.
- [78] **Url-16**<<http://www.uky.edu/KGS/coal-analyses-ultimate.php>>, date retrieved 10.09.2017.
- [79] **Zhang, Jinzhao., Tuo, Xianguo.** (2013). PGNAA Neutron Source Moderation Setup Optimization, *Chinese Physics C*, Retrieved from <https://arxiv.org/abs/1309.1308v1>.
- [80] **Tsoufanidis N.** (1995). *Measurement and Detection of Radiation*, (2nd ed.), USA.
- [81] **Holloway, J.,P., and Akkurt, H.** (2004). The fixed point formulation for large sample PGNAA—Part 1: Theory, *Nuclear Instruments and Methods in Physics Research A* 552, 529–544.

- [82] **Gardner, R.P., Sood, A., Wang, Y.Y., Liu, L., Guo, P. and Gehrke, R.J.** (1997). Single Peak Versus Library Least-Squares Analysis Methods for the PGNAA Analysis of Vitrified Waste, *Applied Radiation and Isotopes*, 48(10-12), 1331-1335.
- [83] **Aleksandr, Kavetskiy., Galina, Yakubova., Stephen, A., Prior., and Henry, Allen, Torbert.** (2017). Neutron-Stimulated Gamma Ray Analysis of Soil, *New Insights on Gamma Rays*, Dr. Ahmed Maghraby (Ed.), doi:10.5772/68014.
- [84] **Url-17**<<https://www.crystals.saint-gobain.com/products/bgo>>, date retrieved 10.09.2017.
- [85] **Url-18**<http://www.omegapiezo.com/crystal_scintillators.html>, date retrieved 10.09.2017.
- [86] **Gordon., R., Gilmore.** (2008). *Practical Gamma-ray Spectrometry*, (2nd ed.), John WileySons Ltd.
- [87] **J, K, Shultis., and R, E, Faw.** (2011). *An MNCP Primer*, Dept. of Mechanical and Nuclear Engineering, Kansas State University Manhattan.
- [88] **X-5 Monte Carlo Team.** (2003). *MCNP – A General Monte Carlo N-Particle Transport Code*, LA-UR-03-1987, Version 5.
- [89] **Han, X. G.** (2005). *Development of Monte Carlo code for coincidence prompt gamma-ray neutron activation analysis*, (Doctoral dissertation). Retrieved from <http://www.lib.ncsu.edu/resolver/1840.16/3224>.
- [90] **Adjei, Christian, Amevi.** (2012). *MCNP5 and GEANT4 comparisons for preliminary fast neutron pencil beam design at the University of Utah TRIGA system*, (Master's thesis). Retrieved from <https://collections.lib.utah.edu/details?id=195791>.
- [91] **Arce, Pedro., Ignacio Lagares, Juan., Harkness, Laura., Pérez-Astudillo, Daniel., Cañadas, Mario., Rato, Pedro., De Prado, María., Abreu, Yamiel., De Lorenzo, Gianluca., Kolstein, Machiel. and Díaz, Angelina.** (2014). Gamos: A framework to do Geant4 simulations in different physics fields with an user-friendly interface, *Nuclear Instruments and Methods in Physics Research A Accelerators, Spectrometers, Detectors and Associated Equipment*, 735, 304-313.
- [92] **R., Kinsey.** (1979). Data Formats and Procedures for the Evaluated Nuclear Data File ENDF, *Brookhaven National Laboratory Report BNL-NCS-50496*, ENDF 102, (2nd ed.) (ENDF/B-V), Brookhaven National Laboratory.
- [93] **R, J. Howerton., D, E. Cullen., R, C. Haight., M, H. MacGregor., S, T. Perkins. and E, F. Plechaty.** (1975). The LL Evaluated Nuclear Data Library (ENDL): Evaluation Techniques, Reaction Index, and Descriptions of Individual Reactions, *Lawrence Livermore National Laboratory Report UCRL-50400, 15*, Part A, Lawrence Livermore National Laboratory.
- [94] **M.A., Gardner., and R. J., Howerton.** (1978). Evaluated Neutron Activation Cross-Section Library-Evaluation Techniques and Reaction Index, *Lawrence Livermore National Laboratory Report, UCRL-50400, 18*, Lawrence Livermore National Laboratory.

- [95] **E.D., Arthur., and P.G., Young.** (1980). Evaluated Neutron-Induced Cross Sections for Fe-54,56 to 40 MeV, *Los Alamos Scientific Laboratory report LA-8626-MS*, (ENDF-304), Los Alamos Scientific Laboratory.
- [96] **E.D., Arthur, P.G., Young, A.B., Smith, and C.A., Philis.** (1981). New Tungsten Isotope Evaluations for Neutron Energies Between 0.1 and 20 MeV, *Trans. Am. Nucl. Soc.* 39(793), Lawrence Livermore National Laboratory.
- [97] **D.G., Foster. Jr., and E.D., Arthur.** (1982). Average Neutronic Properties of Prompt Fission Products, *Los Alamos National Laboratory Report LA-9168-MS*, Los Alamos National Laboratory.
- [98] **Judith, F., Briesmeister.** (2010). *MNCP - A General Monte Carlo N-Particle Transport Code*, (Version 4C), LA-12625-M.
- [99] **Curtis, A. Palmer., Ertem, Tuncali., Kristen, O. Dennen., Timothy, C. Coburn. and RobertB, Finkelman.** (2004). Characterization of Turkish coals: a nationwide perspective, *International Journal of Coal Geology* 60, 85-115.
- [100] **Geant4 Collaboration.** (2007). *Geant4 User's Guide for Application Developers*, (8th ed.).



APPENDICES

APPENDIX A : The Data For Efficiency Calibrations

APPENDIX B : Class Diagrams Of MCNP5 And GEANT4 Simulations



APPENDIX A

Table 1 : The data used for efficiency calibration at distance of 2 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_{γ}	$\delta_{I_{\gamma}}$
¹⁵² Eu/122	622	600	289000	830	26864	163.89	0.2858	6 E-4
¹⁵² Eu/245	622	600	204390	154	26864	163.89	0.0758	19 E-5
¹⁵² Eu/344	622	600	527590	980	26864	163.89	0.265	4 E-4
¹³⁷ Cs/662	612	600	1237508	7281	27302	165.233	0.85	77 E-4
¹⁵² Eu/779	622	600	109688	462	26864	163.89	0.12942	19 E-5
¹⁵² Eu/964	622	600	63104	252	26864	163.89	0.14605	21 E-5
⁶⁰ Co/1173	610	600	110019	419	11835	108.788	0.99	7 E-5
⁶⁰ Co/1332	610	600	76626	273	11835	108.788	0.99	4 E-5
¹⁵² Eu/1408	622	600	38316	198	26864	163.89	0.21005	24 E-5

Table 2 : The data used for efficiency calibration at distance of 4 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_{γ}	$\delta_{I_{\gamma}}$
¹⁵² Eu/122	625	600	271417	703	26832	163.834	0.2858	6 E-4
¹⁵² Eu/245	625	600	120874	409	26832	163.834	0.0758	19 E-5
¹⁵² Eu/344	625	600	354660	770	26832	163.834	0.265	4 E-4
¹³⁷ Cs/662	607	600	981451	3574	27291	165.199	0.85	77 E-4
¹⁵² Eu/779	625	600	140773	463	26832	163.834	0.12942	19 E-5
¹⁵² Eu/964	625	600	79338	298	26832	163.834	0.14605	21 E-5
⁶⁰ Co/1173	758	750	51521	258	11805	108.65	0.99	7 E-5
⁶⁰ Co/1332	758	750	861910	298	11805	108.65	0.99	4 E-5
¹⁵² Eu/1408	625	600	137008	424	26832	163.834	0.21005	24 E-5

Table 3 : The data used for efficiency calibration at distance of 6 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_{γ}	$\delta_{I_{\gamma}}$
¹⁵² Eu/122	759	750	308467	767	26836	163.817	0.2858	6 E-4
¹⁵² Eu/245	759	750	149507	571	26836	163.817	0.0758	19 E-5
¹⁵² Eu/344	759	750	395950	759	26836	163.817	0.265	4 E-4
¹³⁷ Cs/662	604	600	629342	1884	27301	165.23	0.85	77 E-4
¹⁵² Eu/779	759	750	99678	323	26836	163.817	0.12942	19 E-5
¹⁵² Eu/964	759	750	78753	286	26836	163.817	0.14605	21 E-5
⁶⁰ Co/1173	608	600	90129	312	11834	108.784	0.99	7 E-5
⁶⁰ Co/1332	608	600	72610	264	11834	108.784	0.99	4 E-5
¹⁵² Eu/1408	759	750	31999	165	26836	163.817	0.21005	24 E-5

Table 4 : The data used for efficiency calibration at distance of 10 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
$^{152}\text{Eu}/122$	904	900	148792	564	26829	163.795	0.2858	6 E-4
$^{152}\text{Eu}/245$	904	900	114983	327	26829	163.795	0.0758	19 E-5
$^{152}\text{Eu}/344$	904	900	167669	692	26829	163.795	0.265	4 E-4
$^{137}\text{Cs}/662$	603	600	282403	1756	27289	165.193	0.85	77 E-4
$^{152}\text{Eu}/779$	904	900	58814	239	26829	163.795	0.12942	19 E-5
$^{152}\text{Eu}/964$	904	900	50375	224	26836	163.817	0.14605	21 E-5
$^{60}\text{Co}/1173$	904	900	55871	179	11788	108.572	0.99	7 E-5
$^{60}\text{Co}/1332$	904	900	48579	213	11788	108.572	0.99	4 E-5
$^{152}\text{Eu}/1408$	759	904	69870	239	26829	163.795	0.21005	24 E-5

Table 5 : The data used for efficiency calibration at distance of 15 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
$^{152}\text{Eu}/122$	1507	1500	160073	475	26817	163.759	0.2858	6 E-4
$^{152}\text{Eu}/245$	1507	1500	77563	293	26817	163.759	0.0758	19 E-5
$^{152}\text{Eu}/344$	1507	1500	313881	714	26817	163.759	0.265	4 E-4
$^{137}\text{Cs}/662$	1507	1500	538999	1829	27291	165.199	0.85	77 E-4
$^{152}\text{Eu}/779$	1507	1500	55020	227	26817	163.759	0.12942	19 E-5
$^{152}\text{Eu}/964$	1507	1500	57541	224	26817	163.759	0.14605	21 E-5
$^{60}\text{Co}/1173$	1206	1200	48469	229	11788	108.572	0.99	7 E-5
$^{60}\text{Co}/1332$	1206	1200	39643	204	11788	108.572	0.99	4 E-5
$^{152}\text{Eu}/1408$	1507	1500	15478	132	26817	163.759	0.21005	24 E-5

Table 6 : The data used for efficiency calibration at distance of 20 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
$^{152}\text{Eu}/122$	3618	3600	328485	779	26791	163.679	0.2858	6 E-4
$^{152}\text{Eu}/245$	3618	3600	21464	153	26791	163.679	0.0758	19 E-5
$^{152}\text{Eu}/344$	3618	3600	538051	996	26791	163.679	0.265	4 E-4
$^{137}\text{Cs}/662$	2405	2400	322899	1863	27276	165.154	0.85	77 E-4
$^{152}\text{Eu}/779$	3618	3600	86452	311	26791	163.679	0.12942	19 E-5
$^{152}\text{Eu}/964$	3618	3600	72678	271	26791	163.679	0.14605	21 E-5
$^{60}\text{Co}/1173$	2408	2400	70185	251	11771	108.494	0.99	7 E-5
$^{60}\text{Co}/1332$	2408	2400	25446	183	11771	108.494	0.99	4 E-5
$^{152}\text{Eu}/1408$	3618	3600	21865	138	26791	163.679	0.21005	24 E-5

Table 7 : The data used for efficiency calibration at distance of 25 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
¹⁵² Eu/122	3618	3600	260486	815	26791	163.679	0.2858	6 E-4
¹⁵² Eu/245	3618	3600	61728	253	26791	163.679	0.0758	19 E-5
¹⁵² Eu/344	3618	3600	461384	879	26791	163.679	0.265	4 E-4
¹³⁷ Cs/662	1803	1800	166938	764	27291	165.199	0.85	77 E-4
¹⁵² Eu/779	3618	3600	62593	252	26791	163.679	0.12942	19 E-5
¹⁵² Eu/964	3618	3600	54976	214	26791	163.679	0.14605	21 E-5
⁶⁰ Co/1173	2408	2400	29740	196	11758	108.434	0.99	7 E-5
⁶⁰ Co/1332	2408	2400	14852	115	11758	108.434	0.99	4 E-5
¹⁵² Eu/1408	3618	3600	12876	103	26791	163.679	0.21005	24 E-5

Table 8 : The data used for efficiency calibration at distance of 28 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
¹⁵² Eu/122	3618	3600	199292	753	26788	163.67	0.2858	6 E-4
¹⁵² Eu/245	3618	3600	144021	571	26788	163.67	0.0758	19 E-5
¹⁵² Eu/344	3618	3600	230350	651	26788	163.67	0.265	4 E-4
¹³⁷ Cs/662	2403	2400	184520	825	27270	165.1363	0.85	77 E-4
¹⁵² Eu/779	3618	3600	52505	242	26788	163.67	0.12942	19 E-5
¹⁵² Eu/964	3618	3600	47076	209	26836	163.817	0.14605	21 E-5
⁶⁰ Co/1173	3618	3600	41709	193	11754	108.415	0.99	7 E-5
⁶⁰ Co/1332	3618	3600	23708	178	11754	108.415	0.99	4 E-5
¹⁵² Eu/1408	3618	3600	10524	93	26788	163.67	0.21005	24 E-5

Table 9 : The data used for efficiency calibration at distance of 30 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_γ	δ_{I_γ}
¹⁵² Eu/122	3618	3600	107502	376	26791	163.679	0.2858	6 E-4
¹⁵² Eu/245	3618	3600	152250	283	26791	163.679	0.0758	19 E-5
¹⁵² Eu/344	3618	3600	182247	529	26791	163.679	0.265	4 E-4
¹³⁷ Cs/662	1803	1800	159285	797	27270	165.1363	0.85	77 E-4
¹⁵² Eu/779	3618	3600	49746	228	26791	163.679	0.12942	19 E-5
¹⁵² Eu/964	3618	3600	30840	187	26791	163.679	0.14605	21 E-5
⁶⁰ Co/1173	3618	3600	29312	181	11771	108.494	0.99	7 E-5
⁶⁰ Co/1332	3618	3600	20531	165	11771	108.494	0.99	4 E-5
¹⁵² Eu/1408	3618	3600	9717	82	26791	163.679	0.21005	24 E-5

Table 10 : The data used for efficiency calibration at distance of 35 cm.

Source/E (keV)	TT (sn)	LT (sn)	Area	δ_{Area}	Activity	$\delta_{Activity}$	I_{γ}	$\delta_{I_{\gamma}}$
$^{152}\text{Eu}/122$	4020	4000	54135	147	26784	163.658	0.2858	6 E-4
$^{152}\text{Eu}/245$	4020	4000	55790	154	26784	163.658	0.0758	19 E-5
$^{152}\text{Eu}/344$	4020	4000	159229	478	26784	163.658	0.265	4 E-4
$^{137}\text{Cs}/662$	2403	2400	126037	378	27300	165.227	0.85	77 E-4
$^{152}\text{Eu}/779$	4020	4000	42540	209	26784	163.658	0.12942	19 E-5
$^{152}\text{Eu}/964$	4020	4000	22968	158	26784	163.658	0.14605	21 E-5
$^{60}\text{Co}/1173$	3618	3600	40593	202	11754	108.415	0.99	7 E-5
$^{60}\text{Co}/1332$	3618	3600	20221	120	11754	108.415	0.99	4 E-5
$^{152}\text{Eu}/1408$	4020	4000	9814	79	26784	163.658	0.21005	24 E-5

APPENDIX B

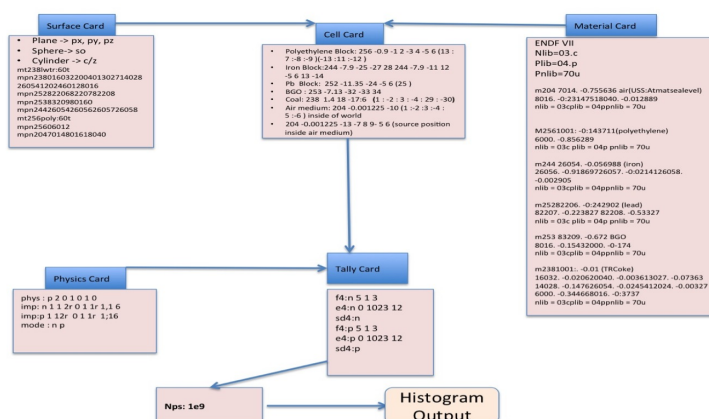


Figure B.1 : Class diagram of MCNP5 for prototype design.

Figure 1 : Class diagram of MCNP5 Simulation For Prototype Design

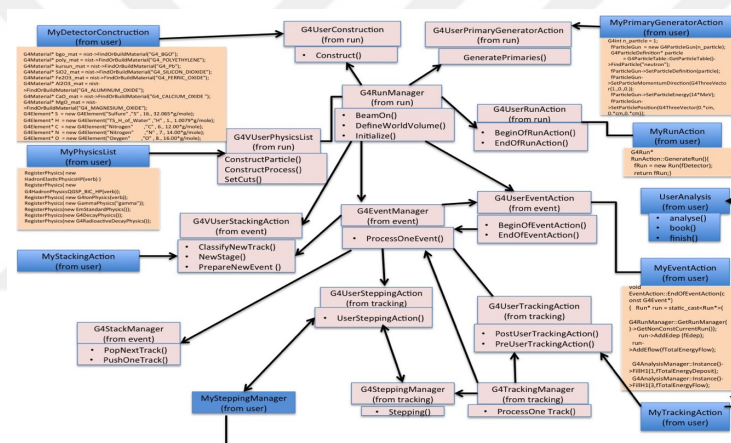


Figure C.1 : Class diagram of GEANT4 for prototype design.

Figure 2 : Class diagram of GEANT4 Simulation For Prototype Design

CURRICULUM VITAE

Name Surname: Başak Untu

Place and Date of Birth: Mardin - 19.07.1981

E-Mail: untuc.basak@itu.edu.tr

B.Sc.: 2006, Gaziantep University, Engineering of Physics

M.Sc.: 2009, Gaziantep University, Engineering of Physics, A Monte Carlo Study Of Long-Lived Particles (Neutralinos) in CMS at LHC.

Ph.D.: 2018, Istanbul Technical University, Physics Engineering, Design Of A Prototype Apparatus For On-Line Elemental Analysis Using Nuclear Techniques,

Professional Experience and Rewards:

- Gaziantep University Rectorate Outstanding Achievement Award (Ranking 1 in department).
- Intern at Turk TELEKOM, Adana, July/2004.
- Intern at Sanaem, TAEK, Ankara, July/2005.
- Intern at Oncology Department in Education and Research Hospital, Gaziantep, August/2005.
- Customer Relationship Officer in Sehitkamil Branch at Halkbank, Gaziantep, July/2007-April/2008.
- MSc Thesis Study in Sofia University, Bulgaria, 2008-2009.
- Scholarship of Summer School Student in CERN, June/August-2009.
- Researcher at "Search for Physics BSM And Detector Upgrade Studies At The CERN CMS EXPERIMENT" Project, June-December/2011.
- ITU Research assistant in General Physics Laboratories, 2010 – 2011, Summer School 2015.
- Research assistant supported TBİTAK Project (Scholarship), 2014 - 2015.
- Research assistant in General Physics Laboratories, Turkish Air Force Academy, 2015-2016.

PUBLICATIONS/PRESENTATIONS ON THE THESIS

- İ., A., Reyhancan., **B., Untu**. (Corresponding Author), M., N., Erduran. (2016). MCNP5 Element Library Least Squares Method for On-line Coal Analysis, *Journal of Nuclear Sciences* 3(2), 27-34. (Article Instance)

- Reyhancan, İ.A., **Untuğ B.**, Angin, N. (2015). MCNP Simulation Of a Prototype For The On-line Coal Analysis. *9th International Physics Conference of the Balkan Physical Union*, August 24 – 27 Istanbul, Turkey. (Poster Presentation Instance)
- Reyhancan İ.A., Angin N., **Untuğ B.** (2015). A Prototype for the On–line Element Analysis Using Nuclear Techniques. *9th International Physics Conference of the Balkan Physical Union*, August 24 – 27 Istanbul, Turkey. (Presentation Instance)

