

İSTANBUL TECHNICAL UNIVERSITY ★ GRADUATE SCHOOL OF SCIENCE
ENGINEERING AND TECHNOLOGY

**GAMMA RAY SPECTRA ANALYSIS FOR PULSED FAST THERMAL
NEUTRON ACTIVATION (PFTNA) OF BULK COAL SAMPLES**



M.Sc. THESIS

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Department of Physics Engineering

Physics Engineering Programme

JUNE 2017

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**KÖMÜR ÖRNEKLERİNDEN PFTNA METODU İLE AKTİVASYON SONUCU
ELDE EDİLEN GAMA SPEKTRUMLARININ ANALİZİ**

YÜKSEK LİSANS TEZİ

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To my spouse and my parents,



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ABBREVIATIONS

BG	: Background
BGO	: Bismuth Germanium Oxide
EC	: Electron Capture
FWHM	: Full Width at Half Maksimum
MeV	: Mega Electron Volt
MCLL	: Monte Carlo Library Least square
MCA	: Multi Channel Analyzer
MLR	: Multiple Linear Regression
NG	: Neutron Generator
NIS	: Neutron Inelastic Scattering
NISTA	: Neutron Inelastic Scattering and Thermal Neutron Capture
PMT	: Photo Multiplier Tube
PGNAA	: Prompt Gamma Neutron Activation Analysis
PFTNA	: Pulsed Fast Thermal Neutron Activation
SVD	: Singular Value Decomposition
TNC	: Thermal Neutron Capture



SYMBOLS

σ	: Cross section
γ	: Gamma ray
$d\Omega$	: Solid angle
n	: Neutron
α	: Alpha particle
A	: Atomic number
I_a	: Incident beam current
R_b	: Detection rate of incoming particles
R	: Radius of a nucleus
b	: Barn
θ	: Angular distribution angle theta
ϕ	: Angular distribution angle phi
$d\sigma/ d\Omega$	: Differential cross section
σ_t	: Total cross section
Ci	: Curie
E	: Neutron energy
v	: Neutron velocity
R	: Reaction number per second
n	: Neutron number
t	: Thickness of the target material
N	: Nuclei number per m^3
a	: Surface area of neutron beams
$\Sigma(m^{-1})$	: Macroscopic cross section
M	: Ion mass
N_A	: Avagadro's number
Z	: Atomic number
ρ	: Material density
T	: Kinetic energy
E_γ	: Gamma photon energy
B_e	: Binding energy
ns	: Nano second
μs	: Micro second
H	: Proton
D	: Deuterium
keV	: Kiloelectron volt



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GAMMA RAY SPECTRA ANALYSIS FOR PULSED FAST THERMAL NEUTRON ACTIVATION (PFTNA) OF BULK COAL SAMPLES

SUMMARY

In this study it is aimed to make a real time analysis of coal samples. The coal analysis is conducted in terms of its calorific value, sulphur content, ash and amount of moisture. Coal materials are classified for better combustion and minimum detrimental effects to environment. These beneficial features of coal samples are determined by burning the material before they are classified. Nuclear techniques conduct such determinations in non-destructive ways. Therefore, these methods are important in coal industry.

The main principle of this array of experiments is based on the interaction of neutrons with coal material. Coal materials exposed to neutrons are radio actively excited and disintegrate by emitting gamma rays. These gamma rays have energy values which are the fingerprint of the elements presenting in coal. The elements are identified thanks to their certain gamma ray energy values. For the coal analyzing, Pulsed Fast Thermal Neutron Activation Analysis (PFTNAA) is utilized. This technique gives precise result even in a short period of time. Coal material interacts with neutrons and there are neutron inelastic scattering and thermal neutron capture processes. One of these kind of interaction branches are dominated for some of the elements. PFTNAA method also gives more precise results and information for some critical elements such as carbon and oxygen. There are other kinds of neutron sources used for neutron activation process, however these neutron sources such as Cf^{252} are not sufficient for identifying some certain elements. Carbon has specific energy value at 4.44MeV and this certain energy value is identified via fast neutron interaction mechanism. Neutron generator emits fast neutrons with 14 MeV energy and therefore it is detected carbon elements in material if they exist. In PFTNAA technique it is obtained two separate spectra. In these spectra some elements belong to fast and some of them belong to thermal neutron induced gamma spectra. They are identified via thermal and fast neutron interaction mechanisms. Therefore, it is

needed to separate thermal and fast neutron induced gamma ray spectra. For this purpose, collected gamma ray data is in the raw form. List mode data acquisition system allows for the data collection when the neutron generator is ON and OFF state. When the neutron generator is ON, resulted gamma rays belong to the fast neutron induced spectrum, when the neutron generator is OFF the neutrons are thermalized through the material and obtained spectrum is thermal neutron induced gamma ray. In the experiment, BGO detector gate port is directly connected to the neutron generator. In this way, BGO detector is triggered according to the ON/OFF mode of neutron generator. In order to allow fast neutrons to be thermalized, it is needed a certain duration to be waited without neutron emission. In this period of time neutrons are thermalized and they lose their energy. Operation time and duty cycle are adjusted via neutron generator interface. Voltage and current values are also set with the aid of interface control of neutron generator. PFTNAA technique is also useful in terms of radiation concern. The radiation only produced when the neutron generator is ON. Additionally, during experiment it is used WiFi router so as to study far away from the experimental setup therefore detrimental effects of the radiation. By the aid of remote desktop connection, both BGO detector and neutron generator are controlled confidentially. After obtaining the data collected by BGO detector, they are classified as two separate spectra. In order to obtain these spectra, it is written a C++ decoding code reading 32-word data coming from each of gamma ray entering detector. Raw data is analysed according to bitwise operators and decoding instruction of digiBASE-E module. Each bit carries its information about peak data or time of gamma ray photon striking the detector. The instruction manual says that digiBASE-E module is active when 32-bit word first bit is 1 and it is at its OFF state when the first bit is 0. After decoding the list mode file the gamma ray photons are converted from histogram to a spectrum data. Each event corresponds to the same amplitude value are collected and stored in a channel where they belong to. In this way spectrum data is obtained. After obtaining the separate spectra, elements in coal material are identified clearly. The peaks appear individually without overlapping as in the total gamma ray spectra. The most dominant cross section values for the energy of elements are recognized in the apparent peaks in the spectra. By separating fast and thermal neutron induced gamma ray spectra element libraries are created. This database gives the information about the behavior of elements in the energy versus amplitude domain. This data store is used for a calibration process and

determination of elemental content in the coal sample. This element library can be used for different coal samples but similar weights. By using library least square method element concentration of coal sample can be found easily.

After data acquisition system is completed, offline analyzing methods are used for identifying the elements in coal and calculating their concentrations. There are various kinds of methods used in determination of element content. In this work it is conducted some experimental studies including both synthetic and real coal materials. There are two types of techniques are adopted for creating element library. They are; experimental studies and semi – experimental studies. These hybrid works are done with some mathematical formulation sets as well. Experimental studies can be grouped as synthetic coal sample experiments and real coal sample experiment arrays as stated. The synthetic coal sample concentration is known and therefore the system can be calibrated according to known values. Afterwards, a linear set of equations are created by using calibration coefficients in order to obtain unknown coal sample contents. The element library is found for the purpose of element concentration. This data base is found by different kinds of methods. Firstly, it is used least square method by taking inverse of a matrix obtained from a coal sample that has a known concentration. Obtained matrix includes the information for element library for coal samples with similar weights and type. In order to use this element library for another coal sample, working conditions and neutron emission durations should be the same as the experiment conducted for finding element library. Real coal samples are used for the identification and determination of some critical elements appearing in the bulk coal sample. It is tested that obtained element library works well or not by using real coal sampels. Element library data set can be found by using both experimental data and some mathematical algorithms as well. Obtained gamma ray spectra includes background information and element energy peaks are located just over this background data. These energy peaks are specific for each element and it is known their peak distribution shapes. By using these information, special Gaussian peaks are created correspond to certain energy values for the elements in coal sample. These peaks are summed with background data calculated by the aid of ROOT Background algorithm. Calculated background data gets the source data as gamma ray spectrum for real coal sample. After obtaining library set for each element, they are fitted to a real coal sample gamma ray spectrum

in order to check their consistency to the real data. Both matrix inversion operation and background calculation algorithm used as fundamental techniques for elemental responses are fitted well with experimental gamma ray spectrum.

Deconvolution process is other part of the offline analysis of this study. BGO detector cannot resolve accurately the energy peaks of elements. There are some multiplets appearing in the spectrum. Gamma ray – material interaction mechanisms coming from the environment interfere with fundamental nuclear reactions and then certain energy peaks cannot be identified easily. At this stage ROOT Deconvolution algorithm is used so as to separate multiplets. This mathematical formulation takes gamma ray spectrum as a source data for input and a response function for the relevant element as a second input. The result gives a spectrum for individual elements in energy – amplitude scale. They can be treated also as responses for elements.

In this experimental study, it is conducted a non – destructive testing of coal material. Some critical element contents are determined in the aim of reaching calorific value, moisture, ash and sulphur concentration. These parameters have crucial importance in coal industry in terms of classification of coal. This procedure is performed by burning of coal. Our study helps to find contents of coal non – destructively and in a more accurate way. All of these experimental and numerical efforts are spent for this purpose.

KÖMÜR ÖRNEKLERİNDEN PFTNA METODU İLE AKTİVASYON SONUCU ELDE EDİLEN GAMA SPEKTRUMLARININ ANALİZİ

ÖZET

Bu çalışmada kömür numunelerinin gerçek zamanlı analizinin yapılması hedeflenmiştir. Kömür analizi, kömür malzemesinin kalori değeri, kükürt oranı, kül oranı ve nem oranı açısından değerlendirilmektedir. Kömürün tasniflenmesi, yandığında çevreye en az hasarı verecek şekilde olması ve daha iyi yanma özelliklerine sahip olması kriterlerine uygun olacak şekilde gerçekleştirilir. Tüm bu olumlu özelliklerin belirlenebilmesi için, kömür sınıflandırılmadan önce yakılıra analizi yapılır. Tam bu noktada nükleer teknikler kömrün sınıflandırılmadan önce hasar almadan analizinin yapılıp değerlendirilmesini sağlar. Bu yüzden, kömür endüstrisinde nükleer teknikler büyük önem arz etmektedir.

Tezde geçen deneylerin temel prensibi üretilen nötronların malzeme ile etkileşimi üzerine dayanmaktadır. Nötrona maruz kalan kömür malzemesindeki çekirdekler uyarılır ve gama fotonu salarak bozunurlar. Gözlemlenen bu gama enerji değerleri kömür iersinde bulunan elementlere ait karakteristik enerjileri vermektedir. Analiz metodu olarak darbeli hızlı termal nötron aktivasyon (PFTNAA) tekniği kullanılmaktadır. Bu metod kısa bir zaman içerisinde bile daha hassas sonuçlar verebilmektedir. Kömür malzemesi nötronlarla etkileşime girdikçe termal yakalama ve nötron esnek olmayan saçılma reaksiyonları açığa çıkar. Bu reaksiyonlar bazı elementlerde baskınken bazılarında değildir. PFTNA tekniği karbon ve oksijen gibi bazı kritik elementler için bilgi vermektedir. Aktivasyon işlemi için Cf^{252} gibi nötron üreticinden başka kullanılan kaynaklar da mevcuttur, fakat bu kaynaklar bazı elementlerin belirlenebilmesi için yetersiz kalmaktadır. Karbon elementinin 4.44MeV'deki enerji değeri hızlı nötronların malzemeye etkileşimi sonucu açığa çıkmaktadır. Nötron üretici ise 14MeV enerji değerine sahip nötronlar üretmektedir ve bu yolla karbon elementi tespit edilmektedir. PFTNAA tekniği ile ikş ayrı spektrum elde edilmektedir. Bu spektrumlardaki element enerji değerlerinin bazıları hızlı nötronların indüklemesiyle açığa çıkan gama fotonlarına aitken, bir kısım enerji değerleri ise termal nötronların indüklemesiyle oluşan gama ışınlarına aittir. Bu

yüzden termal ve hızlı nötron ile indüklenmiş malzemeye ait gama ışını spektrumları ayrı olarak ele alınmalıdır. Bunun için deney esnasında yapılan ölçümler ham veri olarak depolanmıştır. Ham veriler kullanılarak liste modu veri kazanım yöntemi benimsenmiştir. Bu yolla nötron üretici kapalı ve açık konumdayken elde edilen sayımlar değerlendirilmektedir. BGO dedektörü üreticinin hem açık hemde kapalı olduğu durumda sayım yapmaya devam etmektedir. Nötron üretici açık konumdayken elde edilen gama ışını spektrumları hızlı nötronların sebep olduğu fotonları kapsarken, üretici nötron ışımayı durdurduğunda elde edilen spektrum ise termal nötronların indüklemiş olduğu fotonlardan ibarettir. Deney sırasında kullanılan BGO dedektörünün kapı port girişi doğrudan nötron üreticisine bağlanmaktadır. Bu yolla nötron üreticinin kapalı/açık durumuna göre BGO dedektörü tetiklenmektedir. Hızlı nötronların termalize olup enerjilerini kaybetmeleri için bir miktar süre nötron üretimi olmadan beklenmelidir. Bu süre zarfında hızlı nötronlar malzemeye etkileşime girerek enerjilerini kaybeder ve termalize olurlar. Deney için gerekli süre ve görev döngüsü nötron üreticinin sağlamış olduğu arayüz ile ayarlanabilmektedir. Voltaj ve akım değerleri de bu arayüz yardımı ile ayarlanıp deney esnasında gözlemlenebilmektedir. PFTNAA yöntemi radyasyon konusu düşünüldüğünde de daha avantajlı bir durumda görülmektedir. Radyasyon nötron üretici açık olduğunda başlamaktadır, kararsız çekirdeklerin yayımladığı gama ışınlarında olduğu gibi kontrol edilemez değildir. Bunlara ek olarak deney esnasında Wi-Fi modem kullanılarak deney düzeneğinden uzakta çalışılmıştır. Bu yolla radyasyonun zararlı etkilerinden kaçınılmış olmaktadır. Uzaktan kontrol sistemi ile hem BGO dedektörü hem de nötron üretici kontrol edilebilmektedir. BGO dedektörü tarafından toplanan veriler iki ayrı spektrum olarak ayrıştırılmaktadır. Bunun için C++ kodlama yardımıyla dedektöre her gelen gama fotonunun 32-bit veri içerisindeki bilgileri analiz edilmiştir. Bitwise işlemciler yardımıyla diGiBASE-E modülünün talimatındaki bilgiler ışığında ham veriler analiz edilir. Dedektöre ulaşan her gama fotonu zaman bilgisiyle birlikte tepe enerji bilgisini 32 bitlik word veri formatında taşımaktadır. Kullanım talimatına göre nötron üretici açıksa 31. bit 1, kapalı ise 0 vermektedir. Liste modundaki dosya analiz edildikten sonra gama fotonlarına ait veriler histogram formatından spektrum formatına çevrilir. Aynı genlik değerine sahip fotonlar toplanır ve ait oldukları kanalda depolanır. Spektrumları ayrı ayrı elde ettikten sonra kömür numunesi içerisinde bulunan elementlere ait enerji foto tepeleri daha net olarak gözlemlenebilmektedir. Toplam gama spektrumunda olduğu gibi

çoklu tepeler üst üste gelmemiş olurlar. Meydana gelen reaksiyonlar arasında tesir kesit değeri hangi reaksiyonda hangi enerji değeri için yüksekse o enerji değeri görülmektedir. Hızlı ve termal nötronların indüklediği gama ışınlarına ait spektrumlar ayrı ayrı değerlendirilerek element kütüphanesi oluşturulmaktadır. Bu veri tabanı elementlerin enerji – genlik tanım kümesindeki davranışlarının belirlenmesinde yardımcı olur. Elde edilen element kütüphanesi yardımıyla sistem kalibre edilir ve matris denklemlerinde yerine yazıldığında en küçük kareler yöntemi kullanılarak içeriği bilinmeyen kömür numunelerinin element yüzdelikleri belirlenmiş olur. Bu kütüphane farklı kömür numuneleri için kullanılabilir fakat örneklerin ağırlıklarının kalibre edildiği örneğin ağırlığına yakın olması gerekmektedir. En küçük kareler yöntemi kütüphanesi yöntemi kullanılarak kömür numuneleri için element konsantrasyonu kolayca bulunabilir.

Veri kazanım sistemi tamamlandıktan sonra bir takım analizler yapılarak element yüzdelikleri hesaplanır. Bunun için literatürde pek çok yöntem olduğu görülmüştür. Yapılan çalışmada sentetik ve gerçek kömür numuneleri ile elde edilen deneysel veriler dikkate alınmıştır. Element kütüphanesinin oluşturmak içise temelde deneysel ve yarı – deneysel olmak üzere iki farklı yöntem kullanılmıştır. Yapılan bu hibrid çalışmalar matematiksel formulasyon alt yapısı ile de beslenmiştir. Deneysel çalışmalar ise sentetik ve gerçek kömür numunesi ile yapılan ölçümler olarak ikiye ayrılabilir. Sentetik kömür numunelerinin element yüzdelikleri bilinmektedir ve sistem bu değerler kullanılarak kalibre edilebilir. Sonrasında ise oluşturulan lineer denklem setleri kalibrasyon katsayıları kullanılarak çözülür ve içeriği bilinmeyen kömür örneği için element yüzdelikleri bulunmuş olur. Elementlerin konsantrasyonunu hesaplamak için de kullanılan element kütüphanesinin bulunması yapılan bu çalışmada iki farklı yöntemle gerçekleştirilmiştir. Birinci yöntem en küçük kareler yöntemini kullanarak oluşturulan matris sisteminin tersini alarak yapılır. Burada içeriği bilinen kömür numunesinin gama spektrumu sisteme girdi olarak verilmektedir. Oluşturulan element kütüphanesini farklı numunelerde kullanabilmek için kütüphanenin eldesinde kullanılan deney düzeneğindeki nötron emisyon süresiyle diğer çalışma koşulları içeriği bilinmeyen numunenin koşulları ile aynı olmalıdır. Gerçek kömür numuneleri ise kömürdeki bazı kritik elementlerin tanınması ve elde edilen element kütüphanesinin doğru çalışıp çalışmadığını test etmek için kullanılmaktadır. Element kütüphanesi deneysel ve bazı matematiksel

algoritmalar kullanılarak da bulunmaktadır. Deneysel spektrumlarda yüksek miktarda çevresel radyasyon (Background) görülmektedir. Bu yüzden elementlerin enerji foto – tepeleri spektrumunda temel bir hat (Baseline) üzerinde oturmaktadır. Bu enerji değerleri her element için karakteristiktir ve bu foto – tepelerin dağılım şekilleri de matematiksel olarak bilinmektedir. Bu bilgiler kullanılarak her elementin baskın olan enerji değerleri oluşturulan Gausyen fonksiyonunda merkez enerji değeri olarak konumlandırılmış ve temel yanıt fonksiyonları oluşturulmuştur. Kömür numunelerinden elde edilen gama spektrumu için çevresel radyasyon değeri ROOT Background algoritması kullanılarak elde edilen değerlerle toplanarak her elementin yanıt fonksiyonu bulunmuştur. Hesaplanan çevresel radyasyon için algoritma girdi olarak gerçek kömür gama spektrumunu almaktadır. Her element için ayrı ayrı yanıt fonksiyonu bulunup kütüphane elde edildikten sonra, gerçek kömür örneğinin gama spektrumuna fit edilmiştir. Elde edilen sonuçlar ise kütüphanenin gerçek sonuçlara oldukça uyumlu olduğunu göstermektedir. Matris tersi ve çevresel radyasyon hesabı ile elde edilen element kütüphaneleri deneysel sonuçlara uyum içersinde çıkmıştır. Dekonvolusyon ise bu çalışmanın bir diğer analiz kısmıdır. BGO dedektörü bazı elementleri enerji ekseninde yeterince ayırtıramamaktadır. Bu yüzden foto – tepelerin birbirinden ayrılması için matematiksel bir algoritmaya ihtiyaç vardır. BGO dedektörünün enerji çözünürlüğü istenildiği oranda yüksek olmadığı için bazı foto – tepeler ayırt edilememektedir. Bu yüzden birbirine yakın foto – tepeleri ayırt edebilmek için ROOT Dekonvolusyon algoritması kullanılmıştır. Sistemin girdisi olarak kömür gama spektrumu ve ilgili elementin yanıt fonksiyonu kullanılmıştır. Sonuç olarak ise enerji – genlik skalasında her element için yanıt fonksiyonu bulunmaktadır.

Yapılan bu çalışmada kömür örneklerinin hasarsız muayenesi gerçekleştirilmiştir. Kömürün kalori, nem, kül ve kükürt değerlerini hesaplayabilmek için bazı kritik elementler izlenmiştir. Kömürün tasniflenmesi açısından bu parametreler kömür endüstrisinde büyük önem arz etmektedir. Standart laboratuvar testleri uygulanacak olursa bu değerlerin bazıları kömürü yakarak elde edilmektedir. Bu çalışma sayesinde tez kapsamında bahsedilen yöntemler aracılığıyla kömür parametreleri hasarsız olarak bulunmakta ve bu işlem daha net sonuçlar verecek şekilde uygulanmaktadır. Yapılan tüm deneysel ve matematiksel çalışmalar ise bu amaca hizmet etmek için gerçekleştirilmiştir.

1. INTRODUCTION

Large bulky samples such as cement, coal and industrial minerals are measured by nuclear techniques which include neutrons especially.[1] When a nucleus captures slow neutrons, there is a compound nucleus which emits at least one gamma ray. Additionally, fast neutrons are also used for the gamma ray sources but their production mechanism is via inelastic scattering. Compound nuclei emit gamma rays having certain energy values. The elements are characterized by their energies. [1] Generally, the element content of samples such as cement, coal, soil and rock are measured by time consuming and difficult methods which last about 8h to accomplish.

Prompt Gamma Neutron Activation Analysis (PGNAA) is a very useful method for determining some elements appearing with unknown content in samples. Elemental analysis, coal calorific value, sulphur content, ash and moisture content are measured by the aid of real time analysis of coal quality. It is also useful in making decisions about minimizing the operation cost, optimization of process control as well as resource value maximization. PGNAA device system allows the measurement of real time and therefore sampling error is minimized. PGNAA measurement technique becomes more popular since in industrial areas, it is a common method allowing in-situ analysis. PGNAA method sensitivity is better than other kinds of techniques such as gravimetry, spectrometry and calorimetry. [1]

In the experiments it is common to use ^{252}Cf as a neutron source. This technique produces reactions such as inelastic scattering $(n,n'\gamma)$, $(n,p\gamma)$ and neutron capture interaction. ^{252}Cf isotope produces neutrons with energy maximum 2 MeV. Thus the system is highly dominated by thermal neutron capture interaction through the (n,γ) reaction. Carbon element plays a significant role in a coal material. (n,γ) reaction type produces 4.43 MeV energy and identified from this energy level. This reaction type is suppressed by other gamma ray energies such as Si (4.93MeV) and weak relative to the other interactions. In order to handle with this trouble, it is needed a deuterium – tritium (D – T) fusion neutron generator. It produces fast neutrons

having 14 MeV energy. They are more expensive relative to the isotopic neutron sources however, they emit neutrons with higher mono energetic fluxes. Additionally, isotopic neutron sources produce low energetic neutrons. So, it is not possible to identify C, H, O and N elements at the same time. On the other hand, for isotopic neutron sources, radiation protection is a more crucial point since it emits neutrons continuously. In contrast to isotopic sources, Neutron Generator NG produces neutrons with a duration and periodicity demanded by an operator. [1]

NG works in pulse mode operation by emitting neutrons when it is ON and stops for a duration when it is OFF. The neutrons are fast during the pulse and they are thermalized when the NG is OFF. At this time, they interact with coal material and experiences a large number of interactions. Therefore, there are both thermal and fast neutrons in medium. In this way carbon peak at 4.43MeV is identified. [1]

Interpreting the gamma ray spectra is a difficult problem to solve. Because, there are lots of non-linear effects in the system coming from detection mechanism. There are a large number of methods used in gamma spectrum analyzing issues. Some of them are, Monte Carlo Library Least Square (MCLL), peak area and neural network. [2]

In experiments, BGO (Bismuth Germanium Oxide) detector gate input is connected to the neutron generator and detector is adjusted to collect data in list mode data acquisition. In this way, data is stored in its raw form and dead time is minimized. Each data coming to the detector carries its information including the time and amplitude. These information are found in the 32 – bit words. The bits are encoded with a special information referring to amplitude and time. 29. bit in the system is called as memory routing bit and it is 1 when the neutron is ON, if it is 0, it means neutron generator is OFF. By taking this into consideration, a C++ decoding code is written for separating thermal and fast neutron induced material emitting gamma rays. After separating 2 spectra, element identification becomes more precise. However, in each of the spectrum, there are also some multiplets due to the lower energy resolution of BGO detector. Therefore, element library is created from semi experimental methods. This library is useful in examining each element behavior in coal sample without superposing with other elements. Element library formation and identification of elements in sample is conducted by some mathematical techniques. These methods include CERN ROOT background and deconvolution algorithms. It is calculated the baseline for a total coal spectrum. On the other hand, dominant

energy peak values are taken into account for creating Gaussian peaks for each of the element. Afterwards, they are summed to form a characteristic behavior of element at energy scale individually. After all of the calculations are done, total library is fitted to the experimental coal gamma ray spectrum. The result is satisfactory because, the library is greatly fitted to the coal data. Another data analyzing method is deconvolution. It helps to recognize certain peak in spectrum by introducing a response vector as an input to the system. Several overlapping peaks are identified. [3] For this purpose ROOT programming kit uses a very powerful method called as Gold algorithm. This technique produces result with non-negative components and it is the most stable method among others. As an addition to these techniques, Multiple Linear Regression (MLR) tool is applied to the coal spectra in order to calibrate the system firstly and then use obtained results to find out the elemental concentration of an unknown coal sample. This is actually a verification procedure also and work very well. It is observed that calculated coefficients can be used confidently in finding the contents of an unknown material composition. The results are in a very good agreement with each other.

By using stated techniques, coal gamma ray spectra are analysed and elemental composition of bulky coal material is determined.

1.1 Purpose of Thesis

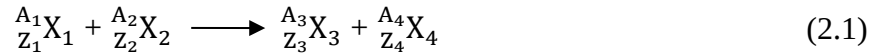
The thesis subject is based on the array of nuclear physics experiments in the aim of conducting a non-destructive analysis of bulk coal samples. Elemental composition of coal material is a significant issue in terms of the classification of coal. According to the obtained information for the coal quality, coal material is used in different areas and applications.

The main objective of this study is identifying the elements at which energy value they present and calculating their concentration in coal samples. In order to accomplish this goal, gamma ray spectra are analysed neatly with some special techniques and it is utilized both experimental data and theoretical calculations. That is one of the outstanding features of this study.

2. THEORY

2.1 Nuclear Reactions

Nuclear reaction is defined as the interaction between at least two particles. Bombarding the particles which are projectile, stationary or slower target. As a result of the reaction there may be two or more particles created. Equation 2.1 defines the reaction of two particles X_1 and X_2 they are colliding particles, resultant products are X_3 and X_4 .



Some fundamental quantities are conserved during a nuclear reaction. Conserved quantities are charge, mass number, total energy and linear momentum respectively:

$$Z_1 + Z_2 \longrightarrow Z_3 + Z_4 \quad (2.2)$$

$$A_1 + A_2 \longrightarrow A_3 + A_4 \quad (2.3)$$

$$E_1 + E_2 \longrightarrow E_3 + E_4 \quad (2.4)$$

$$P_1 + P_2 \longrightarrow P_3 + P_4 \quad (2.5)$$

Generally the nuclear reaction process occurs through the compound nucleus creation. This nucleus is formed after the collision of X_1 and X_2 . Compound nucleus is highly excited and its life time is about 10^{-12} to 10^{-14} s. After this time interval is elapsed, the compound nucleus decays to the products which are X_3 and X_4 . This compound may decay more than one mode and there are more than one way for compound nucleus to decay. [4]

2.1.1 Cross section

Cross section is a term that defines a probability of a reaction to happen. If it is placed a detector in a direction to the particle beam, it is defined a solid angle $d\Omega$ at target

nucleus by the detector. The incident beam current I_a is measured per unit time and N defines the number of nuclei per unit area of target. If particles coming from the target are detected at rate R_b , the cross section is expressed as:

$$\sigma = \frac{R_b}{I_a N} \quad (2.6)$$

According to the equation 2.6 cross section unit is in terms of are per nucleus. Figure 2.1 represents the basic components of nuclear reaction with cross section consideration.

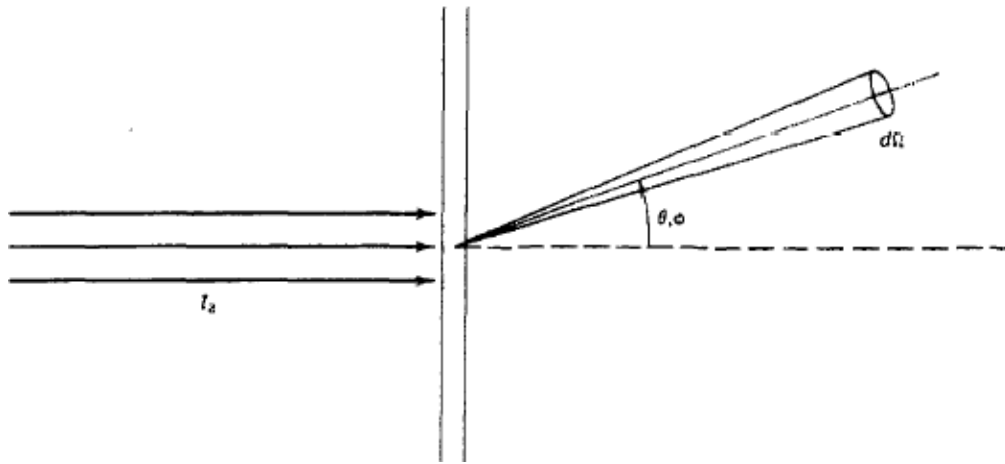


Figure 2.1 : The basic components of nuclear reaction with cross section consideration

If it is considered a typical radius of a nucleus $R = 6$ fm, πR^2 which is the geometrical surface area is calculated as 1 barn. If this calculation is repeated for neutron capture reaction for the Xe^{135} element, the result is calculated as 10^6 b. The cross section term can be thought as if it has a dimension of area, however it is proportional to the probability of nuclear reaction to occur.

Since our detector observes the outgoing particle at the solid angle $d\Omega$, it cannot be detected all of these particles, only some part of the particles are recorded expressed as a fraction dR_b . Additionally, these emitted particles do not release uniformly in all directions. They have angular dependencies represented as θ and ϕ . If the angular distribution is expressed by an arbitrary function $r(\theta, \phi)$, incoming particle at rate R_b is defined as equation 2.7 implies:

$$\frac{d\sigma}{d\Omega} = \frac{r(\theta, \phi)}{4\pi l_a N} \quad (2.7)$$

$d\sigma / d\Omega$ ratio is called as differential cross section. This equation is useful since it gives an information about the angular distribution of the products occurred after reaction. The overall cross section can be calculated by integrating differential cross section over all of the angles $d\Omega = \sin\theta d\theta d\phi$.

$$\sigma = \int \frac{d\sigma}{d\Omega} d\Omega = \int_0^\pi \sin\theta d\theta \int_0^{2\pi} \frac{d\sigma}{d\Omega} d\phi \quad (2.8)$$

$d\sigma / d\Omega$ is independent of the angle and therefore it is constant, the integral thus gives $\sigma = 4\pi(d\sigma / d\Omega)$.

It is not deal with the exact probability of a particle which is emitted from target being at certain angle in most of nuclear physics processes. It is aimed to find the exact energy of the nucleus which is yielded as a result of nuclear reaction. Therefore, cross section term should express the angular dependency of the particles as well as the energy range term dE_b . This added dependency makes the definition of differential cross section to be doubly differential cross section $d^2\sigma / dE_b d\Omega$.

Table 2.1 : Reaction Cross Sections

Cross Sections	Symbol	Technique	Possible Application
Total	σ_t	Attenuation of beam	Shielding
Reaction	σ	Integrate over all angles and all energies of b	Production of radioisotope Y in a nuclear reaction
Differential(Angular)	$d\sigma / d\Omega$	Observe b at (θ, ϕ) but integrate over all energies	Formation of beam of b particles in a certain direction
Differential (Energy)	$d\sigma / dE$	Don't observe b, but observe excitation of Y by subsequent γ emission	Study of decay of excited states of Y
Doubly differential	$d^2\sigma/dE_b d\Omega$	Observe b at (θ, ϕ) at a specific energy	Information on excited states of Y by angular distribution of b

There is another cross section term which is called as total cross section σ_t . It is added cross section σ to all of the probable different kinds of particles b, to the new term total cross section for the specific incoming particle a. Such an approximation can explain the reaction possibility of “between” an incident particle with the target material. Total cross section is found by measuring the intensity loss in the particle beam while they are passing through the target material having known thickness [5]. All of the cross section types are summerized in Table 2.1. It shows the type of cross section its technique and possible application.

2.2 Neutron Physics

2.2.1 Neutron sources

Neutron beams are created by different nuclear reactions. Neutrons cannot be accelerated just like other charged particles. However, it can be worked with high – energy neutrons and their energy can be diminished with other interacting materials. Decreasing the energy of neutron is called as moderating the neutrons. As a result of the nuclear reaction, neutrons have very low energies. Table 2.2 shows the general neutron classification according to their energy values [6].

Table 2.2 : Neutron Energies

Type of Neutron	Energy
Thermal	0.025 eV
Epithermal	1 eV
Slow	1 keV
Fast	100 keV – 10 MeV

2.2.1.1 α - Beryllium sources

The reaction which was used in the discovery of neutron can be utilized for the creation of neutrons in proper conditions. Be^9 atom has relatively low neutron bonding energy. If a helium atom (α particle) collides with the beryllium atom a neutron is released. Represntation of nuclear reaction is shown in equation 2.9. Q value for this reaction is equal to 5.7 MeV.



If an α emitting particle is mixed together with beryllium element, there is a neutron production. There are alpha particles which are emitted from nucleus such as Ra^{226} , have energy values ranging from 5 to approximately 8 MeV. Therefore it can be possible to obtain neutrons having energies up to 13 MeV. The neutrons are monoenergetic particles because; there are many alpha particle groups and alpha particles are slowing in interacting material as well as different kinds of emission directions can happen for the neutrons with respect to the alpha particles and the last reason is the possibility of the C^{12} atom is at its excited state after the production. The neutron energy is found as 5 MeV most likely and there are 10^7 neutrons created per second.

2.2.1.2 Photoneutron sources

This procedure resembles to the (α, n) reaction mechanism. In this process, it is utilized (γ, n) reaction. The benefit of photoneutron source is producing more monoenergetic neutrons by the aid of used monoenergetic gamma ray source. For instance, Na^{24} nucleus emits gamma rays with energy of 2.76 MeV and beryllium nucleus absorbs this photons. As a consequence, this gamma ray energy overcomes the neutron binding energy of beryllium and a neutron is released from Be^9 . There are 2×10^6 neutrons per second per Ci alpha activity, however produced neutrons are shorter – lived particles. Their life time is about 15 hours and the energy value is 0.8 MeV.

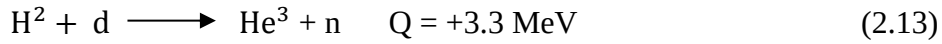
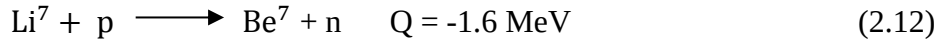
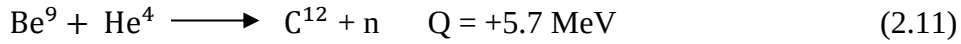
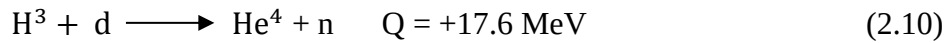
2.2.1.3 Spontaneous fission sources

Spontaneous fission is another used neutron source, Cf^{252} is one of the common examples of this type of source. The neutrons are produced via fission process. Fission is only 3% of the decay process of a nucleus, main part of the disintegration process is α decay. The rate of neutron production is 2.3×10^{12} neutrons / s per gram of Cf^{252} atom. The energy of produced neutrons are continuous and specific in the range between 1 and 3 MeV.

2.2.1.4 Nuclear reaction sources

There are lots of nuclear reactions resulting in neutron production. It is necessary to accelerate particle beams to start the reaction, therefore it is more practical to use radioactive – decay nucleus in order for obtaining neutrons. If the incident energy

and neutron angle values are carefully adjusted, it can be obtained a monoenergetic particle beams. Some of the fundamental reactions are written in equations below.



2.2.1.5 Reactor sources

Nuclear fission reactor core has a high rate of neutron production in the order of 10^{14} neutrons / cm^2 / s. The energy scale ranges between 5 and 7 MeV. Most of the neutrons are thermalized in the reactor, however there are fast neutrons together with the thermal neutrons in reactor. Experiments are conducted by cutting a hole in reactor and permitting neutrons to be extracted. Radioisotope production can be operated by using high fluxes of neutron through neutron capture process, just as neutron activation analysis works.

2.2.2 Absorption and moderation of neutrons

As the neutron beams are travelling through the bulk material, their intensity decreases. There are many kinds of reactions such as (n, p), (n, α) or (n, 2n) for fast neutrons. For thermal or slow neutrons the reaction mechanism occurs via neutron capture process (n, γ) reaction. In general, the cross section value of thermal neutron capture process is larger relative to the other reactions. When the neutrons are out of resonance state, their cross section decreases as a result of increasing velocity. Since inelastic and elastic scattering of neutrons take place in medium, neutrons slow down therefore absorption mechanism gets easier. Neutrons with energies about 1 MeV experience a large number of scattering processes and their energy value decreases to the eV state.

When neutron beams cross a material with thickness dx, they meet ndx atoms per unit surface because, n represents number of atoms per unit volume for the target material. The intensity loss is calculated in equation 2.14 with total cross section term including possible scattering phenomena.

$$dI = -I\sigma_t ndx \quad (2.14)$$

Intensity decreasing behavior is exponential just equation 2.15 points out.

$$I = I_0 e^{-\sigma_t nx} \quad (2.15)$$

Equation 2.15 is valid only for monoenergetic neutrons. Intensity decreases according to the equation 2.15. As a result there is a creation of low energetic neutrons with different cross section values, however equation 2.15 does not include the scattering effects. Therefore, it is not reliable to use this formula so as to calculate the loss of neutrons in material.

If there is an elastic collision between an atom with atomic mass A and a neutron having energy of E as well as velocity value of v , the ratio of final energy to initial neutron energy is calculated by using conservation of laws. Equation 2.16 represents the mathematical formulation of this approach.

$$\frac{E'}{E} = \frac{A^2 + 1 + 2A\cos\theta}{(A + 1)^2} \quad (2.16)$$

Here θ is considered in center of mass system and it is the scattering angle. In special cases, if $\theta = 0$, E' / E equals 1 as expected. For the head – on collision the scattering angle is 180° and the ratio E' / E equals to:

$$\left(\frac{E'}{E}\right)_{\min} = \left(\frac{A - 1}{A + 1}\right)^2 \quad (2.17)$$

If the colliding atom has atomic number $A = 1$, the neutron gives all of its energy. The scattering depends highly on the scattering angle θ , in the case of neutron energies are about 10 MeV. Since the neutrons scatter repeatedly, it is a must to calculate energy loss in each cases. The scattered neutrons are not considered as monoenergetic, if the second scattering process takes place in material [6].

2.2.2.1 Types of neutron interaction

Main neutron interaction mechanisms of neutrons are considered under two classifications; scattering and absorption.

Elastic or inelastic scattering are two possibilities of scattering process. The total kinetic energy is conserved in elastic scattering, however for the inelastic scattering case kinetic energy is not conserved. The part of the energy is converted to the excitation energy for the colliding nucleus. This nucleus emits gamma rays in order to return back to its ground state level after the collision. Scattering makes the neutrons slow down through the medium.

In the absorption process, the neutron disappears and there are other particles created as a result of the interaction [7].

2.2.2.2 Neutron reaction cross sections

Imagine that a neutron beam particles hit a target material having a thickness t . The reaction number per second R is expressed by a formula as equation 2.18 implies.

$$R \text{ (reaction / s)} = \left(\frac{\text{neutrons per m}^2\text{s}}{\text{hitting the target}} \right) \left(\frac{\text{targets exposed}}{\text{to the beam}} \right) \times \left(\frac{\text{probability of interaction}}{\text{per n/m}^2\text{per nucleus}} \right) \quad (2.18)$$

Or it can be written as

$$R = I [n / (\text{m}^2\text{s})][N (\text{nuclei} / \text{m}^3)][a (\text{m}^2)][t (\text{m})][\sigma (\text{m}^2)] \quad (2.19)$$

I , a and t terms written in equation 2.19 express neutron number per m^2s , surface area of neutron beams hitting the target and the thickness of target material respectively. Cross section term defines the interaction probability in the unit of per target nucleus per neutron per unit surface area which hit the target.

The cross section unit is given in terms of barn (b).

$$1 \text{ b} = 10^{-24} \text{ cm}^2 = 10^{-28} \text{ m}^2 \quad (2.20)$$

The energy of neutron and the atomic weight of target material are the main factors that highly affect neutron cross section.

The cross section expressed as σ (b) is named as microscopic cross section. Another type of the cross section is macroscopic cross section symbolized as $\Sigma (\text{m}^{-1})$. Mathematical expression of macroscopic cross section is given in equation 2.21.

$$\sum_i = \text{probability of an interaction form i occurring per unit distance of neutron} \quad (2.21)$$

Linear attenuation coefficient can be considered as analogous to the macroscopic cross section. If a monoenergetic neutron beam collides the target material with thickness t , the number of neutrons which don not interact anything through matter is calculated with equation 2.22.

$$I(t) = I(0)e^{-\Sigma t} \quad (2.22)$$

The mean free path that defines the distance between two interactions occurring successfully:

$$\lambda = \frac{\int_0^\infty x e^{-\Sigma x} dx}{\int_0^\infty x e^{-\Sigma x} dx} = \frac{1}{\Sigma} \quad (2.23)$$

2.2.2.3 The neutron flux

The neutron flux is utilized for the neutron reaction rate calculations. The neutron beams do not emerge parallelly in most of practical applications. There is an energy distribution of neutrons and they are emitted in all directions.

If it is considered a neutron group which have the same speed value v and move in all directions. Interaction rate is calculated with equation 2.24.

$$R = \left(\text{in } 1 \text{ m}^3, \text{the travelled distance by neutrons} \right) \left(\text{probability of interaction conducted by one neutron per unit distance} \right) \quad (2.24)$$

The neutron flux refers to the number of neutrons hitting 1 m^2 in 1 s only if the neutron beams emerge in parallel behavior [7].

2.3 Interaction of Radiation with Matter

2.3.1 Heavy charged particles

Energy loss of a fast charged particle beam occurs during the passage of beams through the matter. Almost all of the energy is lost to the electrons in most of the cases. The nuclei and the electrons in medium are exposed to the Coulomb force because of the changing fields. As the ions pass, it exerts an impulse that transfers

momentum. The much more smaller the mass of the electron, the more energy is carried away from particles than heavy nuclei do.

In a head-on collision with an ion having a mass M and energy value of E and an electron at rest has energy change:

$$\Delta E = E(4m/M) \quad (2.25)$$

α -particle with 4-MeV has this energy value about 2.2keV. The energy loss will be generally much less than this (2.2 keV).

During the passage of particles through the material they excite and ionize the atoms. The more energetic the colliding particles, the more secondary electrons are generated.

2.3.2 Bethe – Bloch formula

The rate of the energy loss of a particle per unit length is known as the stopping power of the sample. A formula which is derived with the quantum-mechanical effect including relativistic factors is called as Bethe-Bloch formula.

$$-\frac{dE}{dx} = \left(\frac{Ze^2}{4\pi\epsilon_0}\right)^2 \frac{4\pi Z\rho N_A}{Amv^2} \left[\ln\left(\frac{2mv^2}{I}\right) - \ln(1 - \beta^2) - \beta^2 \right] \quad (2.26)$$

$v = \beta c$ is the velocity of ion, ze is the electronic charge, m is the mass of an electron, N_A is Avagardo's number and Z , A and ρ are the atomic number, atomic mass number and density of the material respectively. The term β^2 in Equation 2.26 expresses the relativistic factors. In the case of particles propagating with speed which is less than c , then the term β^2 is so small that it can be neglected. The average energy required to ionize an atom in material is expressed as I in formula. It is difficult to obtain a certain value of this; however it is taken as 11 ZeV for I . This value of energy is an approximate and empirical value. For example, $I = 86$ eV for air. Since the particles lose their energy as they travel through material, the sign of the dE/dx term becomes negative, therefore energy loss rate ($-dE/dx$) is positive.

2.3.3 Gamma rays

Gamma ray photons are generated as a result of nuclear transitions and they are energetic. The energy values of the gamma rays are (considered in this chapter) above few tens of keV and the wavelengths are shorter than most atomic dimensions.

There are three main interaction mechanisms of gamma rays or X-rays with matter. They are photoelectric effect, Compton scattering and pair production.

The term mass attenuation coefficient μ_m is a criteria that measure the possibility of the gamma ray photons interacting with material. The dependencies are atomic number of material Z and the gamma ray energy E_γ . All of the three interaction mechanisms contribute to this term, however their relative contributions are different.

2.3.3.1 Photoelectric effect

Photoelectric effect occurs in the vicinity of an atom in which the energy of photon is totally converted into a releasing electron from inner shells of atoms. The photoelectron generates a kinetic energy expressed in Equation 2.27.

$$T = E_\gamma - B_e \quad (2.27)$$

B_e expresses the binding energy of the electron.

An electron which is less tightly bound to the atom may be released. This atom lost its electron before releasing this electron. The electrons that are emitted by this process are called as Auger electrons. The vacancy in the inner shell may be filled by an emission of X-ray photon from higher shells of the atom. This process is known as X-ray fluorescence.

The photoelectric mass attenuation coefficient changes with gamma ray energies E_γ and Z . For example, there is a sharp increase in the mass attenuation coefficient for lead at low energy values. This increase is due to the innermost shell contributions. K-shell electrons have 90 keV binding energy B_K in lead. These electrons do not participate in photoelectric absorption below this energy value, because there is not enough energy to eject electrons from the atom. The discontinuity in the energy dependence of mass attenuation coefficient μ_m is called as K edge and the energy is

proportional to Z^2 . Photoelectric absorption edges depend on the less bound shells of an atom (L, M...) at lower photon energies.

Relative contribution of two K shell electrons is greater than all of the rest electrons located in the atom. This is a surprising case because they occupy only a small fraction of the volume in the atom. However conservation of both energy and momentum forbids the photoelectric effect from happening on a free electron. The smaller the binding energy of an electron, the more an electron has the probability of a free electron and the smaller it is the possibility of the photoelectric effect. That is why most tightly bound electrons participate in the most of the photoelectric effects. There is a relation showing that the dependency between photoelectric cross section and atomic number with gamma ray energy. It is expressed for the energy values about few hundred keV.

$$\sigma_{PE} \propto Z^5/E_\gamma^{3.5} \quad (2.28)$$

2.3.3.2 Compton scattering

In this interaction type, gamma ray photon having energy E_γ , scatters from an electron considered as being free. The consequence is a photon with a lower energy E_γ' and an electron with kinetic energy T depending on scattering angle. Figure 2.2 shows the general mechanism of Compton scattering. If relativistic kinematic is used, conservation of energy for kinetic energy of electron gives the relation in Equation 2.29.

$$T = E_\gamma - E_\gamma' = E - mc^2 \quad (2.29)$$

E is the total energy of scattered electron with its rest mass energy mc^2 .

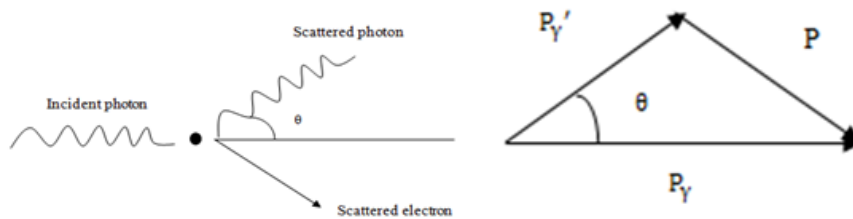


Figure 2.2: The general mechanism of Compton scattering

Figure represents the general mechanism how Compton scattering occurs in terms of vectors. Conservation of momentum gives the relation; $p_\gamma = E_\gamma/c$, $p_\gamma' = E_\gamma'/c$ and p is the momentum of the electron. Equation 2.29 is obtained by using cosine rule:

$$(pc)^2 = E_\gamma^2 + (E_\gamma')^2 - 2E_\gamma E_\gamma' \cos\theta = E^2 - m^2 c^4 \quad (2.30)$$

To obtain this equation p_γ and p_γ' terms are substituted and $E^2 = p^2 c^2 + m^2 c^4$ relation for electron is used. After the elimination of E term from equation 2.29 and 2.30 scattered photon energy is found as:

$$E_\gamma' = \frac{E_\gamma}{1 + \left(\frac{E_\gamma}{mc^2}\right)(1 - \cos\theta)} \quad (2.31)$$

This energy value takes its minimum value at $\theta = 180^\circ$. When E_γ is large E_γ' (min) approaches to $mc^2/2$

Compton scattering is less dependent on E_γ and Z than photoelectric effect. Compton scattering is directly dependent on the density of electrons in material and density is multiplied by Z/A . However, Z/A is approximately constant along the periodic table and Compton contribution to the mass attenuation coefficient is not dependent on Z .

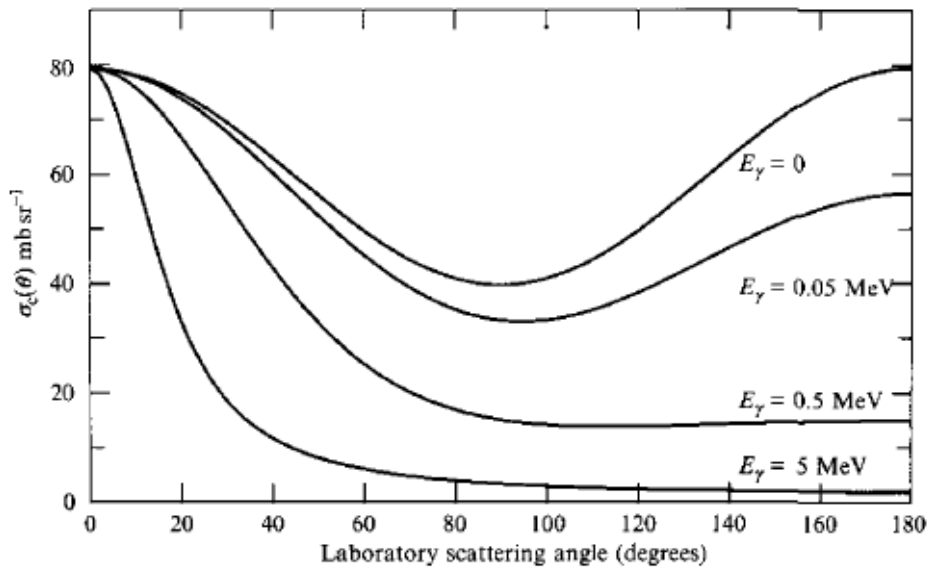


Figure 2.3 : The angular distribution of differential cross section changes with gamma ray energy E_γ [8].

The formula expressing the differential cross section for Compton scattering predicts that the overall Compton cross section should be decrease with gamma ray energy

E_γ . The angular distribution of differential cross section changes with gamma ray energy E_γ . It is represented in Figure 2.3.

2.3.3.3 Pair production

The whole energy of a photon is converted into a pair including electron and positron in this process. The kinetic energy is given by Equation 2.32.

$$T_- + T_+ = E_\gamma - 2mc^2 \quad (2.32)$$

In order to conserve both energy and momentum, pair production process needs a heavy body. Therefore, there is some dependence on Z value. There is threshold energy level $2mc^2 = 1.022 \text{ MeV}$ and pair production process occurs above this energy value.

At the energy levels of a few MeV, the main absorption cross section component is fusion. In this process neutron is captured by forming a compound nucleus. This nucleus is excited with the initial kinetic energy and binding energy of neutron. This energy value equals approximately to 7-8 MeV. Subsequently, this energy is released in the form of gamma rays, neutrons or charged particles.

2.3.3.4 Attenuation

If the photon is collimated properly, all of the three interaction mechanisms cause the photon to be attenuated while it passes through the material. Photoelectric effect and pair production altogether give rise to photon to be disappeared. Compton scattering results in deflection of photons which reach the detector placed in forward direction. Figure 2.4 shows the attenuation process. If the incident beam is not collimated, it is harder for particles to reach the detector.

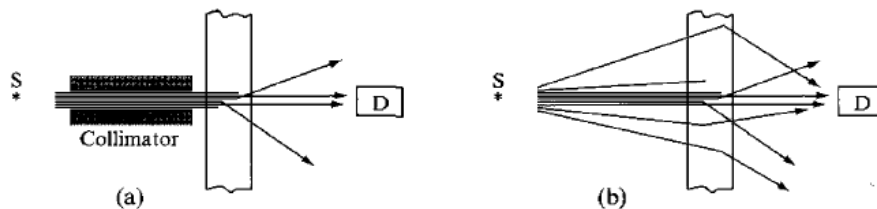


Figure 2.4 : The attenuation process [8]

3. PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS (PGNAA)

After the discovery of neutron in 1932, nuclear reactions and applications were investigated thoroughly. It was observed gamma rays having high penetration feature, during the neutron capture reaction studies in 1934. It was the first detected prompt gamma radiation. Now it is known that this reaction is $H^1(n, \gamma)H^2$ and the resulting gamma ray energy equals to 2.22 MeV.

In 1936 György Hevesy suggested that conducting elemental analysis with the aid of neutrons interacting materials. The neutron activation analysis technique is still one of the most significant methods.

Gamma ray energies, spectra plots and intensities published as tables helped researchers to improve applications.

Isenhour and Morrison conducted the first reactor – based Prompt Gamma Activation Analysis in 1966. They obtained neutron beam from a reactor and detected gamma radiation with a NaI(Tl) detector.

A main turning point is the invention of germanium semiconductor detectors in 1960's. It has twenty or more times greater energy resolution than a NaI detector which has the best energy resolution.

The activation method improved with the germanium detectors counting high energetic gamma rays with enlarged volumes. [9]

3.1 Neutron Capture or (n, γ) Reaction

The most significant reaction is neutron capture process for the PGAA method. Compound nucleus has excitation energy which composed of both binding energy and neutron kinetic energy, when the nucleus absorbs a neutron. 80% of the stable nuclei have 6 – 10 MeV binding energy values. This value increases with increasing Z until it reaches to 22, after this value binding energy decreases. The nucleus emits gamma rays to reach its stable ground state level in a very short of time about 10^{-9} - 10^{-12} s.

If the decay time of the nucleus is in the order of 10 ns to 10 μ s, gamma rays are called prompt gamma. The prompt gamma radiation is a sign for each nuclide, they give an opportunity to identify them and their intensities relate with the number of atoms.

3.2 Neutron Capture of Epithermal Neutrons

This reaction occurs in the case of epithermal neutrons existing in neutron spectrum. The energy of the capture state is not exactly defined as in the case of slow neutrons since these kind of neutrons have broad band of energy spectrum. The kinetic energy of neutrons increases the gamma transition energy states. In the resonance case, the capture state matches with the quasi-stationary level of the compound nucleus. This nucleus has 2-3 order longer life time than off-resonance nuclear level. The branching ratios of de-excitation of gamma rays may differ from the pure slow neutron particles. Therefore, resulting prompt gamma ray spectrum will be different and it has new gamma energy values or different relative intensity peaks. For more sensitive analysis, resonance capture may be used.

3.3 Activation

If the excitation is not stable, radiative decay radiation which are especially will be emitted. This detection system is the basis of neutron activation analysis method (NAA), however some nuclides are identified by using PGNA method too.

3.4 Isomeric Transition (IT)

Prompt gamma radiation can be delayed by metastable levels. Isomeric transitions are defined as gamma transitions coming from metastable levels. Radiative decays from metastable level are not only to ground state but also to another nucleus. If the half-lives of transitions are greater than 1 μ s, they are considered as isomeric. The general half-life durations span in the range between seconds to hours.

3.5 Neutron Capture with Charged Particle Emission

There are element capture possibilities rather than neutron capture processes. The most significant cases are: $\text{He}^3(\text{n,p}) \text{H}^3$, $\text{Li}^6(\text{n,t}) \text{He}^4$, $\text{N}^{14}(\text{n,p}) \text{C}^{14}$, gamma radiation

is not emitted, the other cases are; $B^{10}(n,\alpha) Li^7$ and $B^{10}(n,\alpha) Li^7$ with 3837 b cross section they have branching ratios 6% and 94% respectively. All of these reactions have greater possibility than (n,γ) reactions. There are other kinds of interactions having less importance, they are: $Ar^{37}(n,\alpha) P^{34}$, $Ni^{59}(n,\alpha) Fe^{56}$, $Ni^{59}(n,p) Co^{59}$.

3.6 Elastic Scattering

In this case, incident neutron interacts with the nucleus. It does not excite the nucleus but neutron alters its direction and losses its kinetic energy due to recoiling of nucleus. The energy loss is the basis of neutron thermalization.

The wave effects become important when the wavelength of neutron is approaching the distance between atoms at lower energy values. Bragg scattering effect appears in crystalline materials just like X-Ray Bragg scattering. This can be utilized for creating mono-energetic neutron beam. In addition, neutron guides can be produced by using total reflection of neutrons from surface in the case of Ni^{58} usage. If this material is used, it is obtained a huge coherent elastic scattering cross section.

Bragg condition is satisfied in all directions for polycrystalline materials therefore neutrons are scattered isotropically. However, the Bragg condition is not valid under the condition that neutron wavelength is greater than a critical value. As a result, the macroscopic cross section of the matter decreases to a lower level and therefore the neutron beam can easily pass through the material. This process is used in order to produce low energetic neutron beam by filtering the neutrons.

3.7 Thermalization

Consecutive elastic scattering processes causes neutrons to decrease their energy in a large volumes of media. As they are colliding with particles, the energy of neutron beams approach to the energy of atoms in the media. The most effective moderators are H and D. They thermalize the fast neutrons after 20-25 scattering interactions. The scattering cross section of D nuclei is much lower, thus the scattering probability of D with neutrons is lower. If hydrogenous materials are used, it is observed that the energy distribution of neutrons can alter.

3.8 Inelastic Scattering of Fast Neutrons

Inelastic scattering occurs only when the incident neutron have energy value higher than the first excited state of scattering nucleus. This type of reaction is treshold reaction. Observing the fast neutrons can be conducted by using inelastic scattering of fast neutrons. For example, In^{115} has isomeric transition from metastable level with 336 keV energy. This nucleus can be excited only when incident neutron have energy above 336 keV.

From secondary reactions or cosmic based fast neutrons may lead to inelastic scattering in gamma ray detectors. This reactions are apparent indicators of fast neutrons in materials.

3.9 Secondary Reactions

X-rays can be also produced during the activation of materials or afterwards. The main mechanism of X-ray production is internal arragement of nucleus. There is a conversion from low energy states which can handle with the gamma ray photons. The electron capture (EC) can sometimes be a significant process, then daughter nuclide ($Z-1$) emits X-ray photons. Low energetic gamma rays may be resulted in X-ray fluorescence in the material. X-ray radiation can be utilized for the quantitative analysis of materials, however fluorescence and absorption make this analysis hard to conduct. Therefore, this type of analysis cannot be quantified by using only X-ray radiation. When β particles which are emitted from radioactive products and electrons created in Compton scattering are decelerated in material, there is X-ray bremsstrahlung phenomena.

Secondary fast neutrons may be produced in the case of fast charged particle beams are created as a result of neutron capture reactions.

During irradiation, an important annihilation radiation having energy value of 511 keV is always produced. One of the most important source of the annihilation radiation is pair production came from the high energy prompt gamma radiation absorption. The other possible contribution may be β^+ particle annihilation of radioactive products.

Low energy gamma ray radiation can be increased with the multiple Compton scattering of gamma ray photons in the materials used as shielding. The low energetic gamma ray energy components are observed with higher possibility when powerful gamma radiation sources are close to the detection system. This effect is due to the so-called build-up factor.

The neutron spectrum may be distorted because of the fissile materials like U^{235} . In the sample there are 2-3 fast fission neutrons are emitted per capture, therefore neutron spectrum is not clearly obtained [10].



4. GAMMA RAY SPECTROMETRY

The fundamental equipment for detection of gamma rays is gamma ray spectrometry. The basic component is the detector that converts the gamma ray energy to an electronic signal. Gamma peaks are produced after the signal amplification, shape edition, digitization and storage in a histogram [10].

In the detector there are a large number of interactions for photons. However there are only three of them play a significant role. These are Compton scattering, photoelectric interaction and pair production which are explained in advanced.

4.1 Detectors

The energy range for the prompt gamma radiation spans from zero to 12 MeV. There are two types of common detectors which are used for this high energy range. These are; scintillation and germanium semiconductor detectors. It must be considered several critical points when choosing one of these detectors. For example industrial applications need durable, vibration and shock resistant, easy to use mechanism that can be used in a wide range of temperature, pressure and humidity conditions. All of these parameters can be satisfied by scintillation detectors. These type of detectors are also capable of detection of the identical objects. Semiconductor detectors has superior energy resolution which makes them appropriate for sensitive analysis of sample identification [10].

4.2 Scintillation Spectrometry

Scintillation detection has been utilized since the early radioactive application terms. These type of detection systems are capable of the identification of alpha – beta particles, gamma rays, neutrons as well as mesons and leptons. For the standard gamma ray spectrometry it is common to use sodium iodide material in detector.

According to the band structure primary electrons causes secondary electron to be created in conduction band, thus there are holes in the valence band. In some

situations, the amount of energy is not sufficient for electron to raise to the conduction band. Then the hole and electron are electrostatically attracted to each other. This entity is called as exciton [11].

If the electrons are de-excited by falling back to the valence band, they emit electromagnetic radiation as a result of energy conservation. This radiation is detected by a photomultiplier tube if it corresponds to optical wavelength. This procedure basically describes the scintillation detector working principle. Scintillation detector material (scintillator matter) must include following properties in order to operate detection of gamma rays and conducting spectrometric studies [11]:

- There should be reasonable number of electron-hole pairs which are created per unit of energy of gamma ray
- It is desirable for scintillator material to have a high stopping power (it means that the material has high atomic number and density in practical cases for gamma radiation)
- The response must be proportional to the energy for spectrometric cases
- Scintillator material must be transparent to the light which is emitted
- Decay time must be so short that allowing high count rates
- In order to permit proper coupling to photomultipliers, refractive index of the material should be near to the glass

The materials for gamma ray measurement are all inorganic crystals: cesium iodide (CsI), sodium iodide (NaI), calcium fluoride (CaF_2) and bismuthgermanate (BGO). NaI is the most important one for these applications.

4.3 Scintillation Activators

The band gap of the sodium iodide material is large and the emission of photons by de-excitation process produce wavelength that is out of visible range. Therefore, it is difficult to detect the light. Additionally, the bulk material absorbs the photons before they strike into the photomultiplier. Activator using solves both of the problem. For NaI this material can be thallium and for CsI it is sodium of thallium. Activated scintillators are expressed as NaI(Tl) and CsI(Na).

The introduction of impurity material about 10^{-3} mol fraction puts extra energy levels between valence and conduction bands. Figure 4-1 shows the scintillator band gap structure.

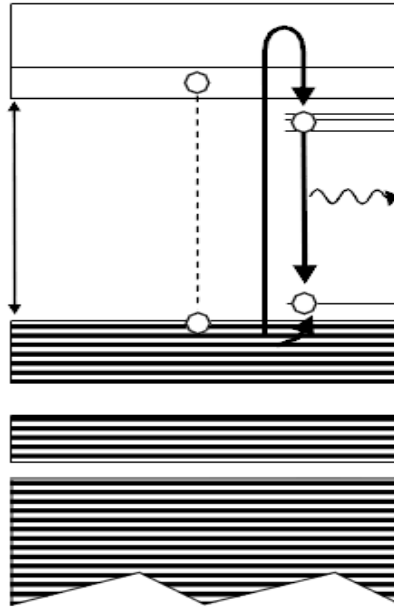


Figure 4.1 : The scintillator band gap structure [12]

The ground state of these activator materials places just above the valence band and the excited states are placed below the conduction band. When an electron-hole pair is created, the hole goes to the nearest activator region. There is a high probability of the capture of electrons in conduction band by the excited activator levels. This gives rise to the emitted light wavelength to correspond visible range. As an addition to this mechanism, emission will not correspond to the absorption properties of the scintillator and there is less information is lost before the measurement by photomultiplier. This procedure can be summarized as:

- Gamma rays are absorbed and there are primary electrons are produced
- Primary electrons cause formation of electron-hole pairs
- Excitons are created and electrons pass to the conduction band leaving a hole in valence band
- Activator energy levels are dominant with electron captures, holes and excitons
- De-excitation of photons emit visible light

- Light is recorded and measured by photomultiplier tube and then it is processed to produce an electronic signal

All of the energy which is absorbed from the gamma radiation is emitted back as scintillation photons. Na(Tl) has maximum efficiency when light output is considered, it releases only 12% of the total emission [11]. The energy left is stored as lattice vibrations or heat. The compatibility of light response of detector and the scintillation spectrum affects the output signal.

4.4 Lifetime of Excited States

The lifetime of an excited state is relatively short approximately $1\mu\text{s}$. The direct emission is called as luminescence. Short decay time gives rise to the short detector pulses. Generally, there is one dominant excited mode is observed however there are other excited states also. For instance, bismuth germanate material's luminescence properties are dominated by two components 60 ns and 300 ns.

Some of the transition from any of the levels to the ground state can be forbidden. One of the electrons placed in one of the states is excited with thermal effects and passes to the allowed energy level. The lifetime of these levels are longer than the normally excited states. When these long-lived states are de-excited and there is emission of light as a result of this process and this emission is called as phosphorescence. Actually, this may cause a problem in the form of a background to the normal detector pulses.

4.5 Scintillator Detector Materials

Different materials which are used in gamma radiation measurement are listed in Table 2.1. The light output is expressed as the number of photons emitted per unit of gamma-ray energy that is absorbed. NaI(Tl) is known as the most efficient inorganic scintillator material. There are 38000 photons per MeV energy. In most cases, efficiency measurements are conducted relative to the NaI(Tl). For example, CsI(Na) is noted as 85%. This value is expressed as relative conversion efficiency or scintillation conversion efficiency. The efficiency term describes the conversion of gamma ray energy to the light and the conversion of light energy to the

photoelectron as well. The number of photoelectrons are highly dependent on the detector output signal and therefore it is utilized to measure the pulse height ratio.

The conversion efficiency describes the comparison of efficiency values and this comparison is meaningful if the scintillator detector has similar features to NaI(Tl). As Table 2.1 implies, the CsI(Tl) has misleading value of RCE since its properties do not resemble to NaI(Tl). The real amount of light which is emitted by a CsI(Tl) scintillator is 52000 photons per MeV energy of gamma radiation absorbed compared to NaI(Tl) scintillator.

4.6 Sodium Iodide – NaI(Tl)

This scintillator material is the most common used material. It is obtained easily and cheap. The detector is produced with maximum 0.75m diameter. In most cases 76 mm diameter by 76 mm high cylindrical NaI detector was used for many years in the aim of gamma-ray spectrometry.

The iodide atoms of scintillator material has high gamma photon absorption coefficient thus it has high intrinsic efficiency. There is a high rate of absorption at low energy values. Since NaI(Tl) has the greatest light output relative to the other traditional scintillator materials, it has the best energy resolution as well.

Although NaI(Tl) has lots of benefits, it has some disadvantages. It is sensitive to the environmental conditions, for example it is brittle and affected easily by thermal changes. It is also hygroscopic therefore it must be captured all of the time. Additionally, long afterglow exhibition creates problems especially at high count rates. At these situations, pulses can pile up and high count rate data collection can be limited. [11]

4.7 Bismuth Germanate – BGO

BGO which has a chemical formula $\text{Bi}_4(\text{GeO}_4)_3$ is a relatively novel material in the scintillator spectrometry. It has lower output of light relative to the NaI(Tl) but it has greater density which provides a high stopping power. This feature leads to the detector to be an ideal choice for active shielding mechanism. Figure 4.2 represents the intrinsic efficiency of peak power for both NaI(Tl) and BGO.

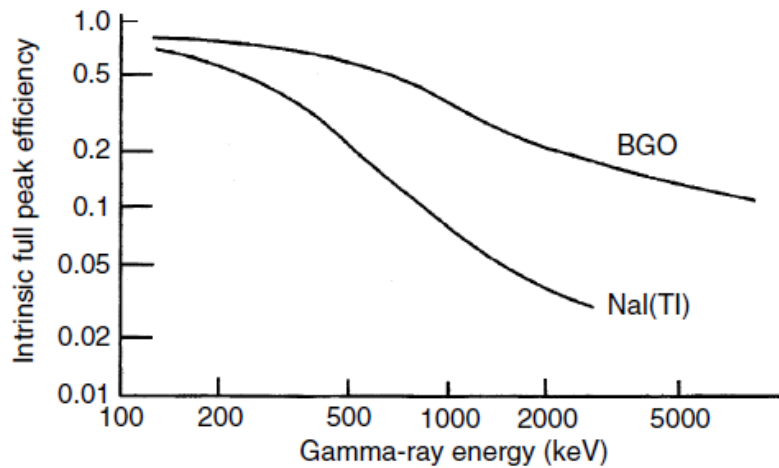


Figure 4.2 : The intrinsic efficiency of peak power for both NaI(Tl) and BGO [12].

This does not take into consideration incomplete interaction mechanisms. They improve the efficiency of BGO detector for the background elimination. Gamma photons having 150 keV energy is 90% absorbed by BGO material with the 2.3 mm of geometric size. In other words, BGO detector with dimension of 1/16th size of the NaI(Tl) detector provides the same efficiency. On the other hand, the BGO detector has lower light output and this situation causes lower resolution. If it is desired to make distinct detection of gamma rays, it is not a proper choice.[11]

BGO material is used without activator material unlike NaI(Tl). There is a large shift between the emission states of Bi^{+3} and the optical states, therefore there is little self-absorption occurs in the material. There is transparency of detector crystal to its own emission. It has the relative efficiency of only 15-20%. BGO material is inert and is not hygroscopic unlike NaI(Tl), thus it is not necessary to be encapsulated all the time.

4.8 Photomultiplier Tubes

The output of a detector is needed to be converted to a electrical signal since it is in the form of light. This procedure is conducted by the aid of photomultiplier tube (PMT). The main representation is shown in Figure 4.3. The mechanism is arranged as:

- The photon strikes the material sensitive to the light this layer is called as photocathode and there is photoelectron emission

- The photoelectrons are directed to the dynodes which are the first stage of electron multiplier. They amplify the signal and they emit more electrons than they receive
- The electrons are multiplied at the first dynode and they pass to the other dynode by multiplying themselves in each event
- The signal which is amplified is collected at the anode and then this signal is transferred to the measurement system

Photomultiplier working principle representation is shown.

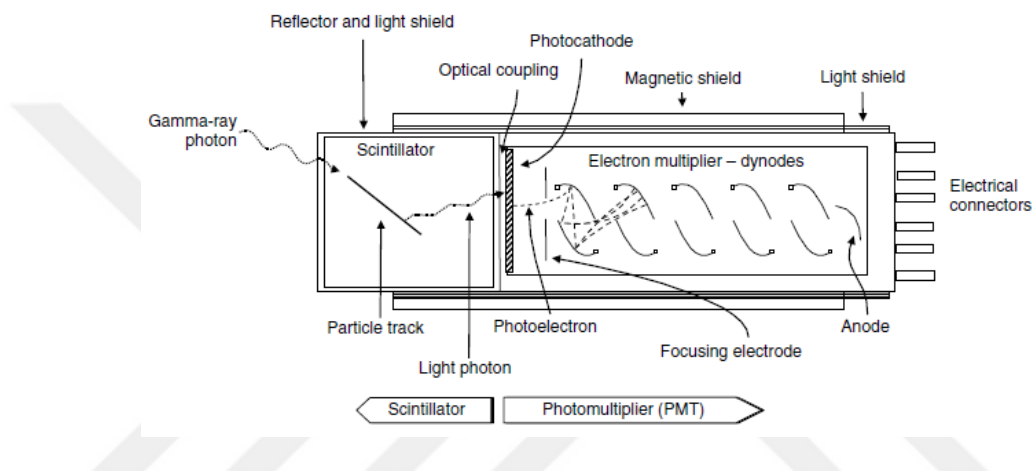


Figure 4.3 : Photomultiplier working principle representation [12]

The design of the photomultiplier tubes depends on the aim of usage. These detectors are designed with end-window structure as well as photocathode inside the tube. The whole structure is encapsulated in an evacuated glass material.

4.9 The Photocathode

This part of the detector works as a light detecting element of PMT. It has a low work function value which allows electrons to release at lower threshold energy. The most common photocathode materials are Na_2KSb activated with caesium, called as multialkali coating and Na_2CsSb activated with caesium and oxygen named as bialkali coating. In scintillation processes it is often used bialkali type materials.

Not every photon received produces a photoelectron. Typical scintillation photon energy is about 3 eV in the blue part of the spectrum. The electrons which are excited by these photons pass to the surface of photocathode and overcome the work function value that is in the order of 1.5 to 2 eV. This limits the thickness of photocathode. It

is a few tens of nanometer range. At this level photocathode is only semi-transparent to the scintillation light.

The quantum efficiency of the photocathode material is about 20 – 30 % at the ideal photon energy level. Quantum efficiency is obtained by taking into account that the number of photoelectrons emitted per incident photon. In NaI(Tl) materials, detector absorbs 26eV energy to produce one photon. If quantum efficiency is taken into account, it is needed 100eV energy to produce electrical signal at the photocathode. [11]

4.10 Dynode Structure in Multiplier Mechanism

These structures are coated with a material that emits some secondary electrons as a result of received single electron. Dynode coating materials can be magnesium oxides, beryllium or Cs_3Sb . Nowadays, gallium phosphide is considered as modern alternatives. This material has negative electron affinity. They have no surface barrier of energy due to their natural band structure. As a consequence, there are more secondary electrons per electron striking the electrode.

The multiplication factor is the ratio of primary and secondary electrons and it depends on the potential difference between consecutive dynodes. Dynode voltages are 80 – 120V and there are approximately 10 – 12 stages. The incoming electron must have adequate amount of energy to overcome bandgap energy value and therefore produce electrons.

Not all of the electrons which escape from dynode reach the next dynode and produce further secondary electrons.

4.11 Complete Detector Construction

Figure 4.4 shows the general structure of scintillation gamma ray detector. The scintillator is coupled to the photomultiplier. The light that is not propagating in the direction of photocathode does not contribute to the detector signal due to the losses at surface. This is reduced by surrounding the detector by a reflector. Scintillation crystal is coated however its end face either magnesium or aluminium oxide. The crystal is encapsulated in a thin aluminium shield.

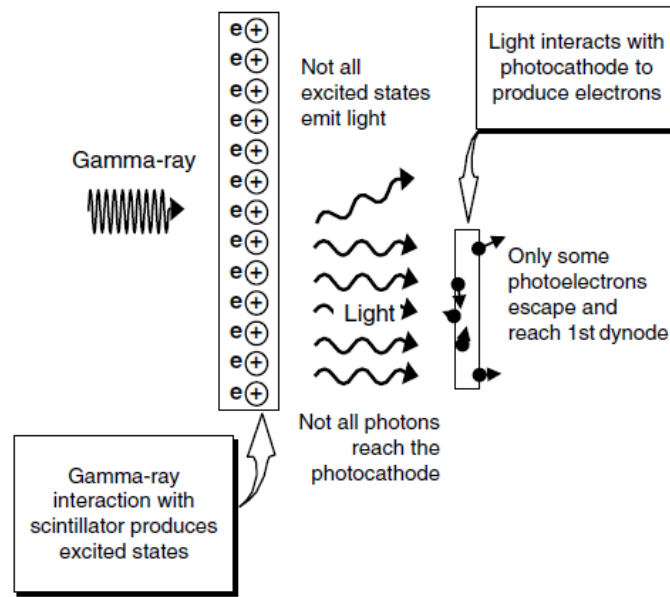


Figure 4.4: The general structure of scintillation gamma ray detector [12].

It is needed to seal the scintillator to avoid hazardous effects if the scintillator is a type of hygroscopic matter.

The passage of electrons from dynode to dynode is probably affected by the magnetic field around the detector. Therefore, photomultipliers are magnetically shielded with high permeability materials. Photomultiplier tube has pins at the end of its casing. The tube connects with the outside world via these pins. Through this connection, dynodes and anode are biased supply.

Photomultipliers are sensitive to the mechanical shock and vibration thus they should be protected. If there is a vibration probability, ruggedized mechanisms can be utilized.

4.12 Scintillation System Resolution

The resolution of scintillation detectors is greater than semiconductor detectors. The resolution is described by full width at half maximum (FWHM) of an energy spectrum in terms of energy generally in unit of keV. The resolution is expressed as:

$$W = \text{FWHM} \times 100/E \quad (4.1)$$

In semiconductor systems, there are uncertainty factors affecting the width of the peak in the spectrum. These are electronic noise and collection of signal. In

scintillation detectors, these uncertainty factors are more than semiconductor detectors. These factors are classified as equation 4.2 implies:

$$W^2 = W_S^2 + W_i^2 + W_D^2 \quad (4.2)$$

W_S expresses the statistical effects and may be due to the reasons listed bellow:

- Electron – hole pair creation
- Number of photoelectrons produced by the photocathode for each of the photon striking
- Multiplication factor for the first dynode
- Collection at the anode and the multiplication factor for each consecutive dynode

W_i term expresses the intrinsic effective line width and it is due to the non-linearity in the conversion of energy to the light. The other term W_D includes the various characteristics of the detector. The factors that are independent of energy are listed as follows:

- Uniformity and purity of detector matter
- Passage of light to the photocathode
- Photocathode and dynode uniformity
- Noise in the electronic measurement

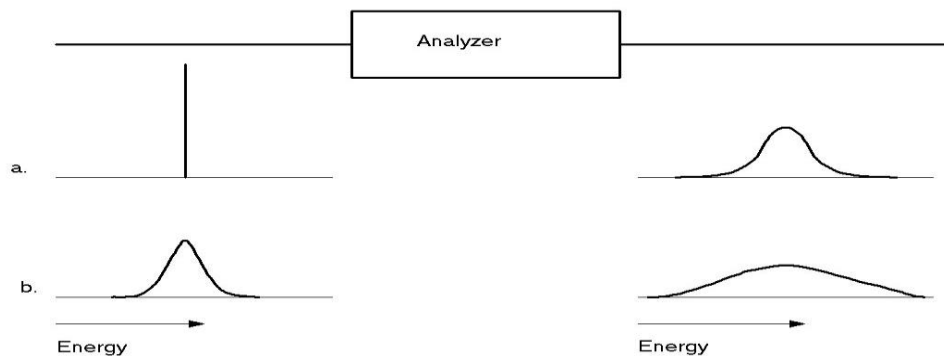


Figure 4.5: The conversion of gamma-ray energy to photoelectron energy.

Electronic noise is a significant problem for the semiconductor detectors, however in scintillation detection systems it is not a major problem for the resolution of the

detector. Figure 4.5 shows the conversion of gamma-ray energy to photoelectron energy.

4.13 Statistical Uncertainties in Detection Systems

The factors that are mentioned before are the most important statistical factors. The resolution is determined by these uncertainty factors. The uncertainty is deal with number of photoelectrons emitted by the photocathode.[11]

The width of scintillation detector peak is highly dependent upon the W_i term that is a square root of gamma-ray energy. W_i term is slightly dependent on the energy and W_D term is not dependent on the energy. As a result, there is an approximate relation written in the equation 4.3.

$$W = a + b/\sqrt{E} \quad (4.3)$$

a and b are the empirical constants. It is important to obtain the resolution of a scintillation detector as an equation varying with energy.

4.14 Deconvolution Algorithm for Gamma Ray Spectrum

Deconvolution algorithms are used in many applications especially in experimental science. They are very useful in the determination of intensity and position of gamma ray multiplets. Deconvolution solves many of the critical problem.[13] Operation of deconvolution improves the resolution quality of gamma ray spectra.

There are some methods which are applied for deconvolution process, however the most suitable non-oscillating method is Gold algorithm. This method is utilized for decomposition of gamma-ray multiplets. This algorithm has a basic feature that it has always non-negative solutions. This property is very crucial because spectrum evaluation has physical meaning in considering gamma ray spectrum multiplets. On the other hand, Gold algorithm is a time consuming process. It is needed so many numerical operations.[13] The convergence is an important problem especially in two and multi dimensional spectra. The method is based on the optimization of time and memory.

Deconvolution analysis in nuclear physics problems deals with identification of existence of peaks and their intensities.[14] The peaks that are fundamental

characteristics of spectra are so close to each other. This causes incorrect information due to limited resolution of the detection system, electronic noise and overlapping signal formation. Deconvolution process handles this problem in terms of the resolution of data. [14] Figure 4 – 5 shows the smearing effect of an imperfect detection system. A system identification methods are based on the thoretical information and the direct measurement with a calibration source.[14]

For a discrete system measured value $x(i)$ and result of the measurement which is a raw data $y(i)$ are correlated with the equation 4.4.[15]

$$y(i) = \sum_{k=0}^i x(k)h(i - k), \quad i = 0, 1, \dots, N - 1 \quad (4.4)$$

$h(i)$ expresses the impulse response. It can be written in the matrix form as follows:

$$\mathbf{y} = \mathbf{H} \cdot \mathbf{x} \quad (4.5)$$

The $\mathbf{H}$ matrix has the dimension $N \times M$, $\mathbf{y}$ vector has length of N and the $\mathbf{x}$ vector has length of M . The estimation of the $\mathbf{x}$ vector is expressed by the equation 4.6. [15]

$$\mathbf{x} = (\mathbf{H}^T \mathbf{H})^{-1} \mathbf{H}^T \mathbf{y} \quad (4.6)$$

$\mathbf{A} = \mathbf{H}^T \mathbf{H}$ matrix is called as Toeplitz matrix. When using this algorithm and there can be small errors that can cause huge oscillations in the result. Finding the $\mathbf{x}$ vector with the known matrices $\mathbf{H}$ and $\mathbf{y}$, there is a discrete ill posed problem and it is needed a regularization method to get the proper solution. This technique is based on iterative and direct solution. [15]

Regularized approximate solution of the system is expressed as:

$$\mathbf{x} = (\mathbf{H}^T \mathbf{H} + \alpha \mathbf{Q}^T \mathbf{Q})^{-1} \mathbf{H}^T \mathbf{y} \quad (4.6)$$

α and $\mathbf{Q}$ are the regularization parameters and matrix. For $\mathbf{Q} = \mathbf{E}$ it is obtained zeroth order or Tikhonov regularization. [15] The fundamental form of van Cittert iterative algorithm so as to solve equation 4.7 is expressed as:

$$\mathbf{x}^{(n+1)} = \mathbf{x}^{(n)} + \mu(\mathbf{y} - \mathbf{A}\mathbf{x}^{(n)}) \quad (4.7)$$

Here n represents the iteration number and μ is the relaxation factor. μ should satisfy the equation written in the equation 4.8.

$$0 < \mu < 2/\lambda_{\max} \quad (4.8)$$

In the formula $\lambda_{\max}$ shows the greatest value of A . This is estimated with the aid of the formulation written in equation 4.9.

$$\lambda_i \leq \sum_{m=0}^{M-1} |A_{im}|, \quad i = 0, 1, \dots, M-1 \quad (4.9)$$

If it is chosen a local variable factor of relaxation:

$$\mu_i = \frac{x^{(n)}(i)}{\sum_{m=0}^{M-1} A_{im} x^{(n)}(m)} \quad (4.10)$$

and we use it in the equation of Van Cittert algorithm it is obtained:

$$x^{n+1}(i) = \frac{y(i)}{\sum_{m=0}^{M-1} A_{im} x^{(n)}(m)} x^{(n)}(i) \quad (4.11)$$

That is Gold deconvolution algorithm:

$$x^{(0)} = [1, 1, \dots, 1]^T \quad (4.12)$$

Equation 4.12 is the initial solution and if the vectors y and h are positive, the result is always positive.[15] The positive and definite deconvolution methods are based on the iterative solution of linear set of equations. Therefore it must be altered the particular solution $x^{(L)}$ in such a way and it is needed to repeat after each equation until it converges. In order to change the relations between elements in the solution, it should be applied a non-linear boosting function. The algorithm for the boosted deconvolution Gold method:

1. Setting the initial solution $x^{(0)} = [1, 1, \dots, 1]^T$
2. Adjusting the needed repetition number R and iteration L
3. Adjust the repetition number $r = 1$
4. According to the equation 4.11 (Gold algorithm), find the solution $x^{(L)}$ for $k = 0, 1, \dots, L-1$
5. Stop the iteration if $r=R$, otherwise;

- a. Use boosting operation for example, $x^{(0)}(i) = [x^{(L)}(i)]^p$, $i = 0, 1, \dots, N-1$, and p is the boosting coefficient > 0 ;
- b. $r = r + 1$
- c. Go to the step 4 [15]

In order to study with the deconvolution algorithm, it is selected a synthetic data having 256 channels which are very close to each other and wide peaks $\sigma = 5$ having noise. [15] The spectrum with thin lines show the Gaussians, thick line represents the virtual spectrum with added noise. If the system is solved by least square method without any regularization, there is huge oscillation obtained at the end of the process.

Even for the Tikhonov regularization there can be observed some oscillations. Deconvolution method enhances the resolution of gamma ray spectra. [15] Gold deconvolution algorithm is an efficient tool for this purpose that gives non-negative solutions by taking square of the negative values. There is boosted and improved method of Gold algorithm used in ROOT TSpectrum class. It is very useful in implementing low resolution spectra. [15]

5. EXPERIMENT

5.1 PGNAA in Coal Industry

Improving the product quality and reducing the cost are the main criteria for the operation of many industries. Social demands force some methods to be sustainable to environment. Economical factors including environmental issues point out that on-line measurement systems are essential.[16]

On-line measurement methods have benefits of supplying real time measurement opportunity and there is no need to take samples. [16] There is especially growing need for the on-line methods in order to obtain elemental analysis in a short period of time.

5.1.1 Coal material ingredients and quality

The main purpose of coal analysis is to obtain the information about coal quality as well as its own characteristic features. [17] All of these data are used for utilisation of coal in different applications. Standard laboratory analysing methods are classified as follows: [17]

1. Proximate analysis: Ash content, moisture content, fixed carbon and volatile matter
2. Ultimate analysis: Hydrogen, nitrogen, carbon, oxygen, sulphur
3. Ash analysis: Coal ash and its major and minor components
4. Calorific value: Specific energy or heating value

Coal material is a combustible matter that is created naturally and its composition changes from coal type to type and even there are alterations in between samples of coal. [18]

The compositions of coal can be classified as 2 groups: 1) organic part which can be also divided into two group as soluble and insoluble parts, 2) inorganic part that is

generally identified as ash content for combustion. Due to the complex structure of coal, there is a variation of coal property even in a specific type of coal. [18]

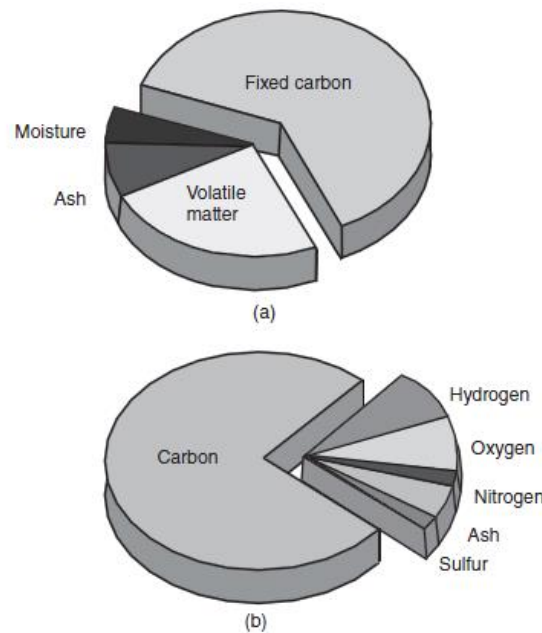


Figure 5. 1 : Representation of the data kinds comparison from the analysing methods: (a) proximate analysis (b) ultimate analysis [18] (Source: US Department of energy, EIA Coal Data).

Depending on the properties of coal, coals are divided into three classes: (1) hard coking coal, (2) semi-soft coking coal and (3) thermal coal. The first type of coal is considered as the production of strong coke coal. This criteria is based on the yield, strength and distribution size parameters. These parameters depend on the plastic properties and rank of the coal. Semi-soft coking coal type is a weaker version of the coking coal and it has lower coke quality due to its higher impurity. The last type of the coal is commonly used for generation of electricity. The thermal coal is fired and used as pulverized fuel. [18]

Evaluation of the coal is conducted by determination of some critical properties. There are other kinds of organic materials that determine the environmental behavior of coal. [18]

5.1.2 On-line coal quality determinations

There are lots of applications which use on-belt measurement techniques, such as controlling of mine grade and coal preparation places, blending of products, observation systems at coal shipping ports. [16] The main characteristic parameters

that define the coal quality are ash content and calorific value of coal (specific heat). Both of these parameters are determined after the burning of coal. Ash is described as a residue after the coal is burnt. Both the non-destructive and direct measurement of these parameters is not possible. Therefore they are inferred from the measurement data obtained from mineral materials (Si, Al, Fe and Ca) and organic/inorganic materials (H, C and O elements). [16]

Prompt gamma neutron activation analysis (PGNAA) is becoming a common measurement methods in coal industry. This analysis technique is traditionally based on the Cf^{252} neutron source with NaI detection systems in a transition geometry placed relative to the sample. PGNAA method gives the coal quality by collecting data of mineral matters in coal such as Al, Si, Fe, Ca dominantly. [16]

5.1.3 Neutron induced gamma ray techniques

Neutron induced analysing methods are becoming popular techniques for multi-element analyses. With this method, the sample under test is exposed to the neutrons. As a result of the neutron – material interaction, gamma rays are emitted. These gamma rays are used for the interpretation of elemental composition of the material under investigation due to the fact that emitted gamma photons have characteristic energy values for each of the nuclei. [16]

Neutron-gamma methods have high penetrating radiation feature that allows non-destructive testing of materials. This technique creates data sets of measurement which are averaged over a bulk volume of matters. The unique useful features of neutron induced gamma ray method outweigh the hazardous effects deal with high-intensity neutron source. Although there is a known concern about radiation danger, neutron-induced methods are used commonly for the on-line analysis. [16]

There is a method which uses both neutron inelastic scattering and thermal neutron capture processes (NISTA) [16]. This technique is utilized for the mineral, energy and coal industries. The main elements considered in the determination of coal quality are C, H, O, Si, Fe, Al and Ca. These are measured by using neutron – induced methods.[16] In our experiments it is used NISTA technique in order to obtain on – line measurement data from coal materials passing on a conveyor.

5.1.4 Prompt neutron – gamma interaction mechanism

For the on – line measurement systems, the most crucial point is prompt interactions of the neutrons with nuclei. Generated prompt gamma – rays are dominantly thermal neutron capture (TNC) and neutron inelastic scattering (NIS). [16]

TNC (n, γ) reactions result in nucleus with energy about 8 MeV which is above the ground state. There are some specific elements that response to the TNC reactions are basically H, Si, Fe, Ca, Cl, S and Ti. [16]

NIS(n,n' γ) reactions need higher energetic neutrons for the on-line analysing techniques. This reaction is observed via Am²⁴¹ – Be neutron source or (d,t) neutron generators. Fast neutrons scatter inelastically from the elements' nucleus in the material. There are gamma rays produced during the inelastic process and these gamma ray photons have certain energies for each of element. NIS method is a distinctive way for conducting analysis of some specific elements which are O, C, Al, Fe and Si. In contrast to TNC reactions, NIS method is simpler and there are no complicated slowing down and thermal capture processes. [16]

Table 5.1 shows the most dominant element gamma ray energies when their nuclei interact with neutrons via 2 different mechanisms TNC and NIS.

Table 5.1 : The most dominant element gamma ray energies [19].

Isotope	Energy (MeV)	TNC	NIS
H-1	2.22	X	
Mg-24	2.83/3.91	XX	
Fe-56	7.63	X	
Ca-40	3.73		X
Ca-40	1.94/6.42	XX	
Si-28	4.93	X	
Si-28	1.78		X
Al-27	7.72	X	
O-16	5.61/6.12		XX
C-12	4.44		X

5.1.5 Pulsed fast thermal neutron activation method

In this technique, neutron generators produce fast neutrons with energies 14MeV. [3] Neutron generator emits neutrons isotropically. The reaction starts with the collision of fast neutrons with the coal material and fast neutron reactions produce some information of certain elements such as C and O. Characteristic gamma rays are

obtained from the coal material. These data are detected by the BGO (Bismuth Germanium Oxide) detector. In the experiment it is used BGO detector instead of traditional NaI detectors, because BGO detector has high efficiency and has the capability of detect high energy values such as C with 4.44MeV energy as well as its low internal background [3].

The detector gives the energy values as a result of the fast neutron interactions and fast neutron gamma induced gamma spectrum is obtained. The neutrons are thermalized through the material and they loose their energy, after the neutron generator stops. [3] Fast neutrons scatter with light elements such as H, S and Cl there are (n, γ) reactions. These information are stored as thermal energy spectrum. The gamma rays are also stored when the neutron generator is off and this kind of spectrum is recorded as activation spectrum. [3]

By seperating the spectra such as fast and thermal neutron induced gamma ray, the certain element energies are detected clearly and some background information is eliminated.

The reasons why it is applied PFTNA method instead of PGNAA for the on-line coal analysis:

- (n, γ) reactions give specific information for the key elements and PGNAA cannot give such data [3]
- There is less radiation since the neutron generator is turned off periodically [3]
- Transmission of fast neutrons is utilized for the measurement of moisture in coal material [3]

5.2 digiBASE-E Module and Data Collection

In the experiment BGO detector crystal is connected to the digiBASE-E module. It has the capability of digital signal processing and is compatible to the most of photomultiplier tubes. There are 14-pins for the digital signal processing unit. It is used for ideally for the scintillation detectors including NaI(Tl), LaBr₃(Ce). It has also capability of pulse hight analysis (PHA) as well as list mode data acquisition with 32-bit input of counter. There is an opportunity to connect other spectrometric

devices with the aid of its gating features. [20] This module is proper for BGO crystal and pulsed fast thermal neutron activation analysis. Neutron generator is connected to the GATE input of the module and it is also directly connected to the BGO detector.

Obtained data from the sample was conducted by activating the “routing” option in MAESTRO software. According to this mode, neutron generator operation signal is adjusted properly. If the data is collected while list mode is active, the detector continues detecting gamma photons both neutron generator is on and off. The signal of the neutron generator is square signal represented below.

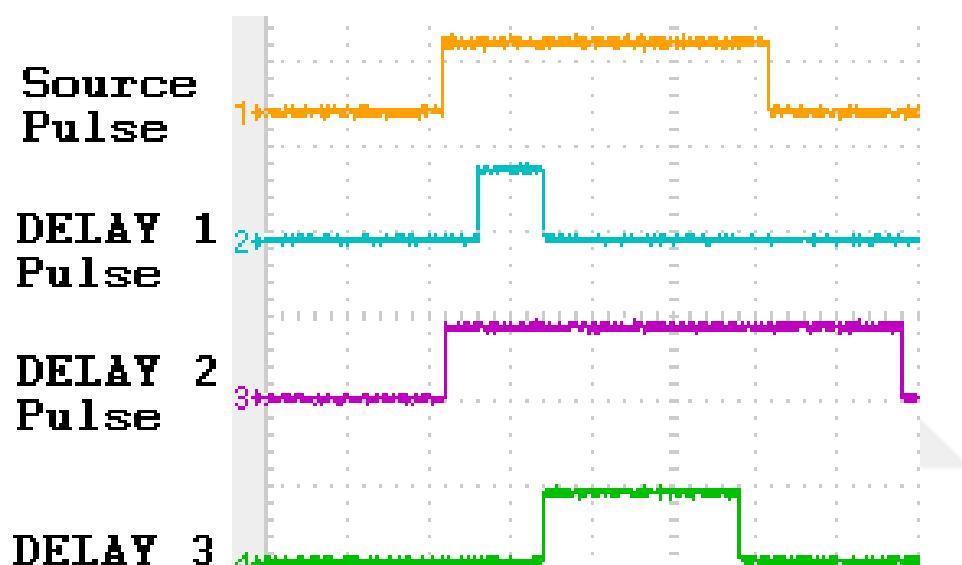


Figure 5. 2 : The electroniz output signals of the neutron generator

According to the signal of the neutron generator, 14MeV energy neutrons are emitted for a duration adjusted manually. Neutrons are produced for a certain duration then neutron generator is off. While the generator is off mode, the neutrons are thermalized through the material. The spectrum file (list mode file) gained from the module, is in the form of both the neutrons are fast and thermal. Therefore, there are two spectra are obtained as a result of data collection. These files are created with ASCII language. In order to separate the spectra as neutron induced gamma rays (thermal & fast), it was written a C++ code by using the bitwise operators.

Figure 5.2 shows the neutron generator signal durations and the elements conducted analysis. In experiment it was adjusted neutron generator signal with 10kHz and 10% duty cycle. This signal is used as an input for GATE. The data are obtained as list mode format and this is obtained with the aid of MAESTRO software. DigiBASE-E

module obtains data as 32-bit words and these words have meaning bit by bit as explained below:

Peak data

Data[31:30] = 3

Data[29] = Memory Routing Bit

Data[28:17] = Conversion Data[11:0]

Data[16:0] = RealTime PreScale [17:1]

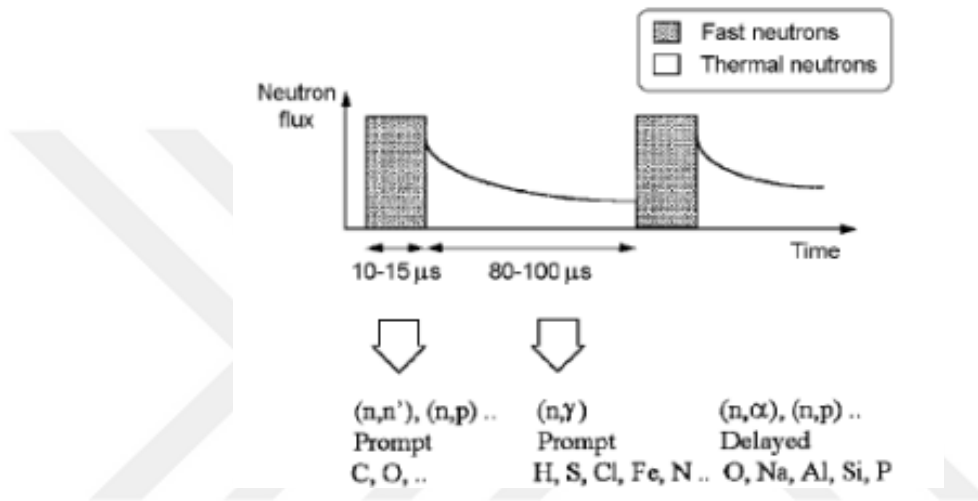


Figure 5.3: The neutron generator signal durations and the elements conducted analysis [3]

These data are created in the form of little Endian. It is inferred from the table that 31. and 30. bits represent the data belong to the “peak data”, 29.bit expresses the meaning of whether triggering signal is active of pasive (routing). The signal is active (there are fast neutrons in media) if memory routng bit is 1, the signal is pasive (there are thermal neutrons in media) if memory routing bit is 0. Bits that are in the interval between 28 – 17, give the information for the amplitude of gamma rays. Total number of channels used in the data acquisition is 1024 and “conversion data” gives the information in which channel gamma rays are collected therefore their amplitude.

Since the obtained data are in the form of list mode, each of the data delivered to the detector is transferred to the computer as raw data. These raw data are classified as fast and thermal neutron induced gamma ray spectra by decoding words in C++ environment.

5.3 Neutron Generator

The generator system is based on popular series of A320 neutron generator from Thermo Electron MP320. This neutron generator is able to produce pulsed and continuous output and the number of neutrons is approximately 10^8 neutrons per second. It is controlled by the RS 232 port and their programmable part is adjusted by a terminal programme or LabVIEW interface. [21]

The neutron generator produces neutrons by accelerating a beam of deuterium ions toward a target including tritium. Produced neutrons have energy of 14MeV. The reaction takes place as equation 5.1 expresses.



Resultant neutrons have 14MeV energy. Figure 5.4 shows the compact neutron generator.



Figure 5. 4: Compact neutron generator [21]

The technical details are: [21]

Maximum accelerating voltage = 90 kV

Frequency = 250 Hz – 20kHz continuous signal

Duty cycle = 5% - 100% minimum pulse width duration is 5µsec

Neutron generator has the opportunity of controlling some parameters with the aid of graphical user interface (GUI). This platform is utilized if the terminal is not used. The GUI interface has the ability of connecting buttons to the certain functions which are sent to the system as commands. While sending the commands to the system, some of the values are observed during operation. GUI interface window is represented in Figure – 5.5.

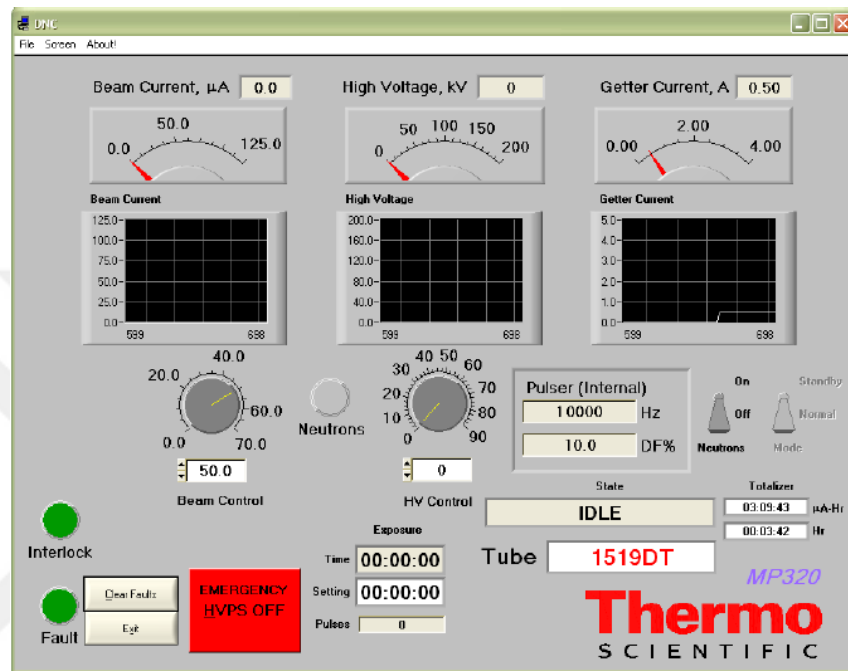


Figure 5.5 : GUI interface window

It can be monitored beam current, voltage value and current value while generator is in operation.

5.4 List Mode Data Acquisition

In List mode data acquisition system, each event is sent to the computer with the information of time indicating the pulse arrival. [22]

In this mode, digiBASE-E transfers one 32-bit word for each detectable peak data. The system is complex to use for digiBASE-E. [23] The whole time of the detector can be recorded and afterwards it is reproduced for the offline analysis. Every detection of radiation is recorded with its time stamp. This opportunity has the advantage of making analysis of arbitrary part of the spectrum after the data

collection ends. This method is used with the ability of managing DAT file binary file format. [23]

Although the time stamps are deal with the event, it is not possible to obtain the “true” time stamp without looping at the precision level of digiBASE-E. It has also a larger time stamps which include much longer time ranges. The time stamp with the data word is combined with the larger time stamp then, this information gives the real time of the event data relative to the device starting time. [23]

Data words are transferred by digiBASE-E via the “GetRawData” command. Four – byte words in the 2048-word long data type is recorded in the first channel. The left data 2047 words can be gained by each transfer. This factor should be taken into consideration due to the fact that whole system of digiBASE-E can crash. Therefore, it must be regularly checked to prevent the overflow of buffer. [23]

digiBASE-E has four kinds of word data. Each of these types includes 2-bit flags in bit 30-31. [23]

The zeroth kind of data is a time sync stamp signal. Sync time stamps roll over every 8 seconds. [23]

The first kind of data is live time stamp. It is constructed in 10ms ticks. Whereas this will excite with the same periodicity as a real time stamp, it will be left behind the real time beause of the dead time that detector experineces. Live time stamp shows the real time at that moment and it accompanies a real time stamp. [23]

The second type of data is real time stamp. It is too long to count every 10ms. This type basically counts for the time.

The third data type is a real event. It includes the memory routing bit at 29.bit. Between the bits 28-17, one can reach the information for channel number or energy bin where the event is belong to. The remaining bits correspond to the time stamp in 80ns units. [23]

In our experiments, we used list mode data acquisition system since it minimizes dead time and gives the opportunity for managing the spectrum analysis in a desired time interval.

5.5 Fast and Thermal Neutron Induced Gamma Ray Spectra Analysis Using C++ Code

In the experiments, neutron generator was used in pulsed mode configuration. Therefore, thermal and fast neutron induced gamma ray spectra are obtained. It is written a C++ code to separate the data when the neutron generator is on or off. If it is on (if neutrons are emitted) resulted gamma ray photons belong to fast neutron interaction, if it is off the neutrons are thermalized through the matter and resulted gamma photons belong to the thermal neutron interaction. In the programme, it is obtained only amplitude information which is obtained by using bitwise operators.

All of the data obtained from detectors are represented 0 or 1. [24] For most of the systems, an array of eight bits create a one byte, and the standart type of storage unit is char. Other kinds of data types need larger number of bytes to be stored. Bitwise operators such as AND, inclusive OR and exclusive OR compares the two data bit by bit. The operator AND sets the bits to 1 if both of the bit data are . Inclusive bitwise OR operator sets the bit to 1 if either or both the bits are equal to 1. Exclusive one sets the result as 1 exactly one of the operants bit number ie equal to 1. [24] The left shift operator shifts the position bit by bit by the amount of bit written in the right handside of the operator. The left shift operator operates just as right shift operator but to left side of the operand. The bitwise complement operator sets all of the bits to 0 in the operand 1 bits, and sets all the bits 0 in operand to 1. [24] Bitwise operator symbols and their names are written in the table.

These operators were used to obtain the needed information from the 32-bit word data such as time and amplitude. These are placed in a certain bit number given in digiBASE-E peak data information. In order to select needed information masking procedure is applied in the programme. It is utilized when hiding some bits and selecting other bits. For example if AND operator is used in mask operation it is obtained such results:

00000000 00000000 11111101 11101000 (value)

10000000 00000000 00000000 00000000 (MASK)

00000000 00000000 00000000 00000000 (result = value & MASK)

When AND operator is used with MASK procedure, all of the bits are hidden (written as 0) if both the operands are not 1 or 0. It is resulted 1 if the both operands are 1. Another example explains the last case.

```
11111101 11101000 11111101 11101000 (value)
10000000 00000000 00000000 00000000 (MASK)
```

```
-----
10000000 00000000 00000000 00000000 (result = value & MASK)
```

By utilizing the peak data information of digiBASE-E detector SPEC_fast and SPEC_thermal spectrum arrays are created and added in each peak data event. Firstly, data are read by getting information as char. Then, it is returned to the data with masking operation and gained the needed physical data at which channel the gamma rays go into. Both fast and thermal neutron induced gamma spectra arrays are written in separate input files.

In this way some element energies are identified easily in separate spectra (thermal or fast).

5.6 Energy and Shape Calibration of BGO detector

The maximum energy limit of gamma rays in NISTA technique is 10MeV. There is a need for energy calibration for gamma rays corresponding to MCA (Multi Channel Analyzer) channels which is connected to the BGO detector.

As the photo-peak gamma energies increase, the width of the gamma ray energies (FWHM) also increase. However, there is no standart gamma ray energy source in the energy range between 3-10MeV. In order to obtain trusted results, it should be used lots of data point as much as possible. Standart sources are Cs-137, Co-60, Eu-152. In order to reach the 3MeV energy value, 2.75MeV gamma energy is produced. An aluminum foil is radiated with Pu-Be neutron source, and there is 2.754 MeV gamma energy from Na-24. It was used radioisotopes of Na-24 for the calibration as well. The calibration was conducted for multi channel analyzer having 1024 channel. Energy values and some quantities related with the calculation of calibration are represented in the table below.

Table 5. 2 : Energy values and some quantities related with the calculation of calibration

Energy (keV)(E)	Channel No.	$E^{1/2}$ (keV) ^{1/2}	Sigma (keV)	FWHM(keV)
121	12	11	14	32
244	22	16	17	41
344	34	19	20	48
661	64	26	28	66
778	72	28	24	56
964	90	31	26	62
1173	114	34	29	68
1332	130	36	30	72
1408	132	38	33	78
2504	244	50	62	145
2754	268	52	94	221

Calibration is based on the Gaussian distribution function. [25] It is expressed by an equation:

$$f(x) = a \cdot \exp\left(-\frac{(x - x_0)^2}{2\sigma^2}\right) \quad (5.2)$$

$$\text{FWHM} = 2.355\sigma$$

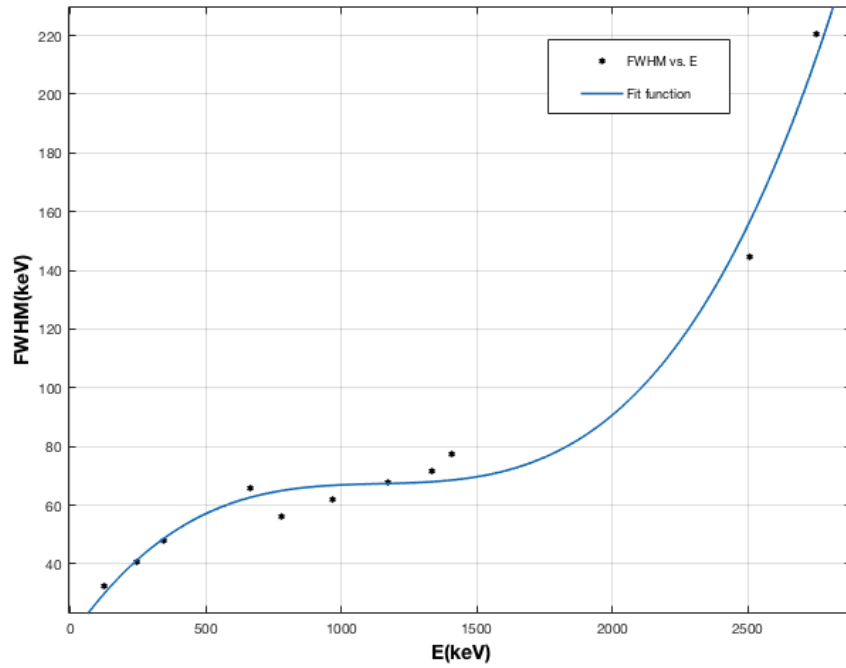


Figure 5. 6 : The shape calibration of the detector

The function parameters are as follow:

$$\text{FWHM}(E(\text{keV})) = p_1 E(\text{keV})^3 + p_2 E(\text{keV})^2 + p_3 E(\text{keV}) + p_4 \quad (5.3)$$

$$p_1 = 3.37 \times 10^{-8}$$

$$p_2 = -0.0001152$$

$$p_3 = 0.1333$$

$$p_4 = 15.09$$

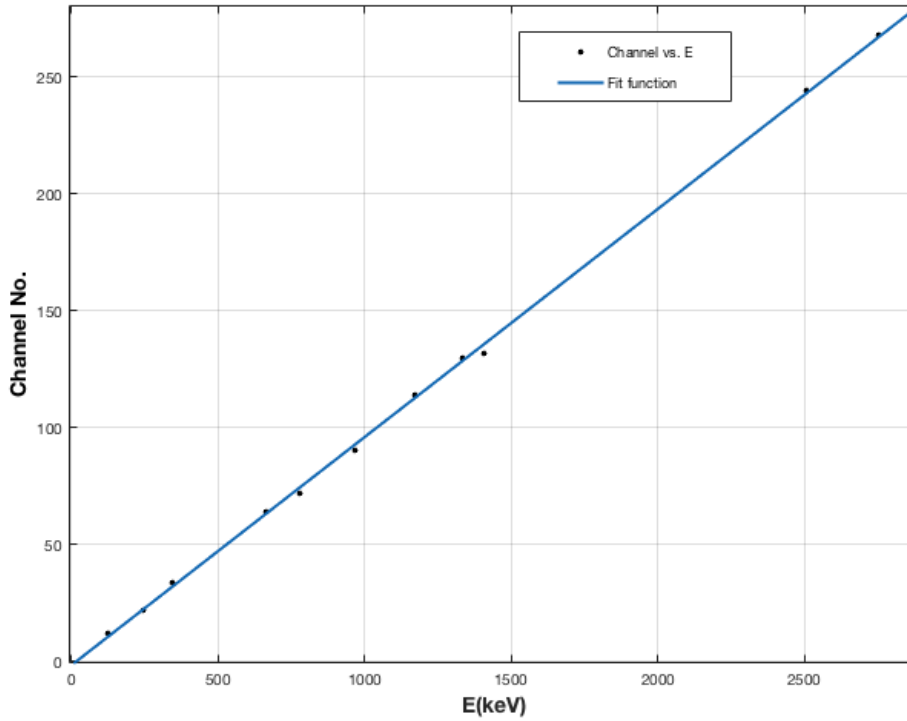


Figure 5. 7 : The channel versus energy calibration

$$\text{FWHM}(E(\text{keV})) = p_1 E(\text{keV}) + p_2$$

$$p_1 = 0.0976 \quad (5.4)$$

$$p_2 = -1.553$$

5.7 Experimental Setup and Data Analysis

In the experiment neutron generator output is connected to the BGO detector gate input. Generator works in pulser mode and neutrons are produced isotropically with

14MeV energy. Fast neutrons (14MeV) are needed for our experiment because some elements have characteristic gamma ray energies (for example, C element has 4.44MeV) when it interacts with fast neutrons. Therefore, it is identified clearly with NIS operations. General experimental geometry including BGO detector, neutron generator and the coal samples.

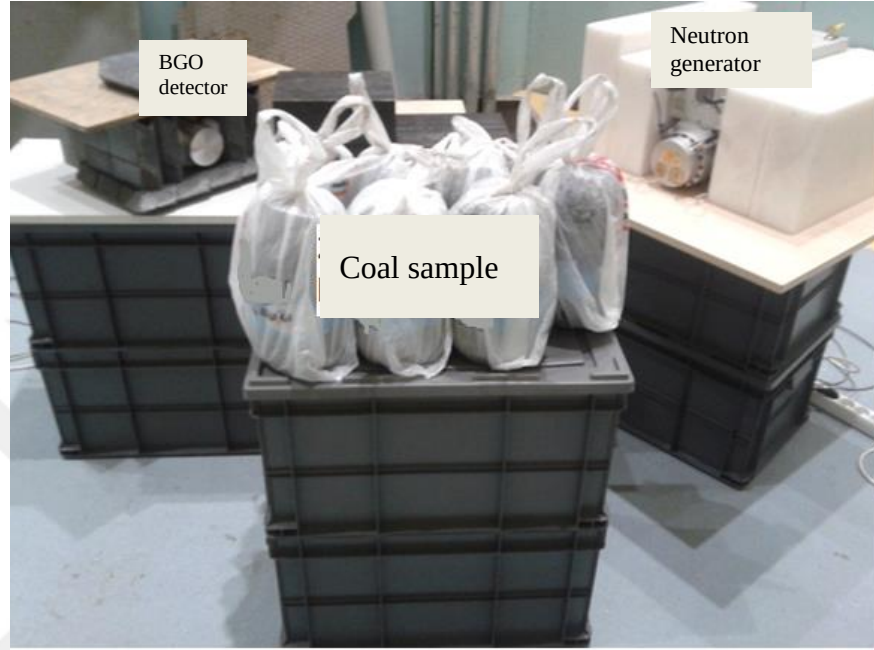


Figure 5. 8 : Experimental geometry

BGO detector is shielded with lead blocks in order to prevent it from high energetic and scattered neutrons. BGO crystal can interact with these fast neutrons therefore shielding mechanism is a must. Additionally, neutron generator is shielded with a polyethylene. Neutron generator produces a high radiation field thus there is needed a shielding material. Neutrons thermalized mostly with nuclei having relatively lower mass. Therefore, a material with hydrogen weighted is preferred. It makes the fast neutrons to be slow down and thermalized. These kind of materials are generally polyethylene, concrete, or even water. [26] In our experimental setup it is used a compact polyethylene shielding material was used and it is observed that thermal neutrons are absorbed by this material as a result of this interaction there is another gamma ray source represented as equation 5.5.



This energy is observed in the experimental spectra and it is considered as background information.

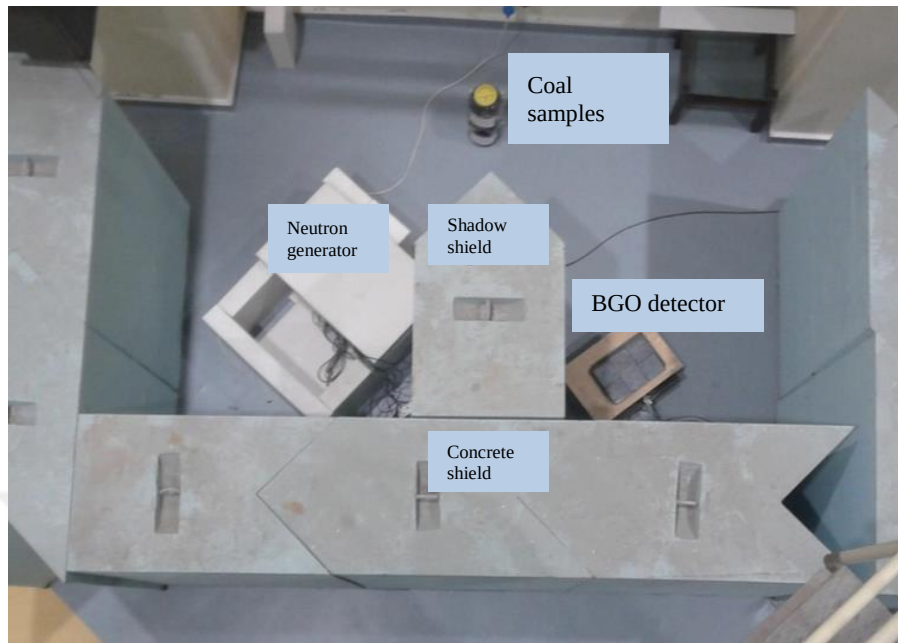


Figure 5. 9 : The top view of the general experimental setup

Figure 5.9 shows the top view of the general experimental setup. Concrete shieldings are used for the biological shielding materials. Concrete walls are also a source of gamma rays, however they are used with this position in each of the experiment. It is considered as a background information as well.

The experiments were conducted with remote desktop operations. A WiFi router was used in order not to be exposed the radiation. The representative acquisition system is shown in Figure 5.10.

Neutron generator and BGO detector are both connected to the computer via RS232 and USB ports respectively. Wireless router was used to construct a connection between computer (Industrial PC) and laptop PC on the purpose of studying far away from nuclear reactions taking place between neutrons and coal as well as the environment surrounding the setup. By this way safety conditions are satisfied.

The picture just below the first figure represents the general experimental setup and total connection of detector, neutron generator and pc as well as their communication.

The experimental results from a 10 kg coal sample and a signal $100\mu\text{s}$ - $900\mu\text{s}$ pulse. Neutron generator produces neutrons with a duration of $100\mu\text{s}$ and waits for $900\mu\text{s}$.

As a result of the interaction it is obtained two separate spectra as thermal and fast neutron induced coal sample.

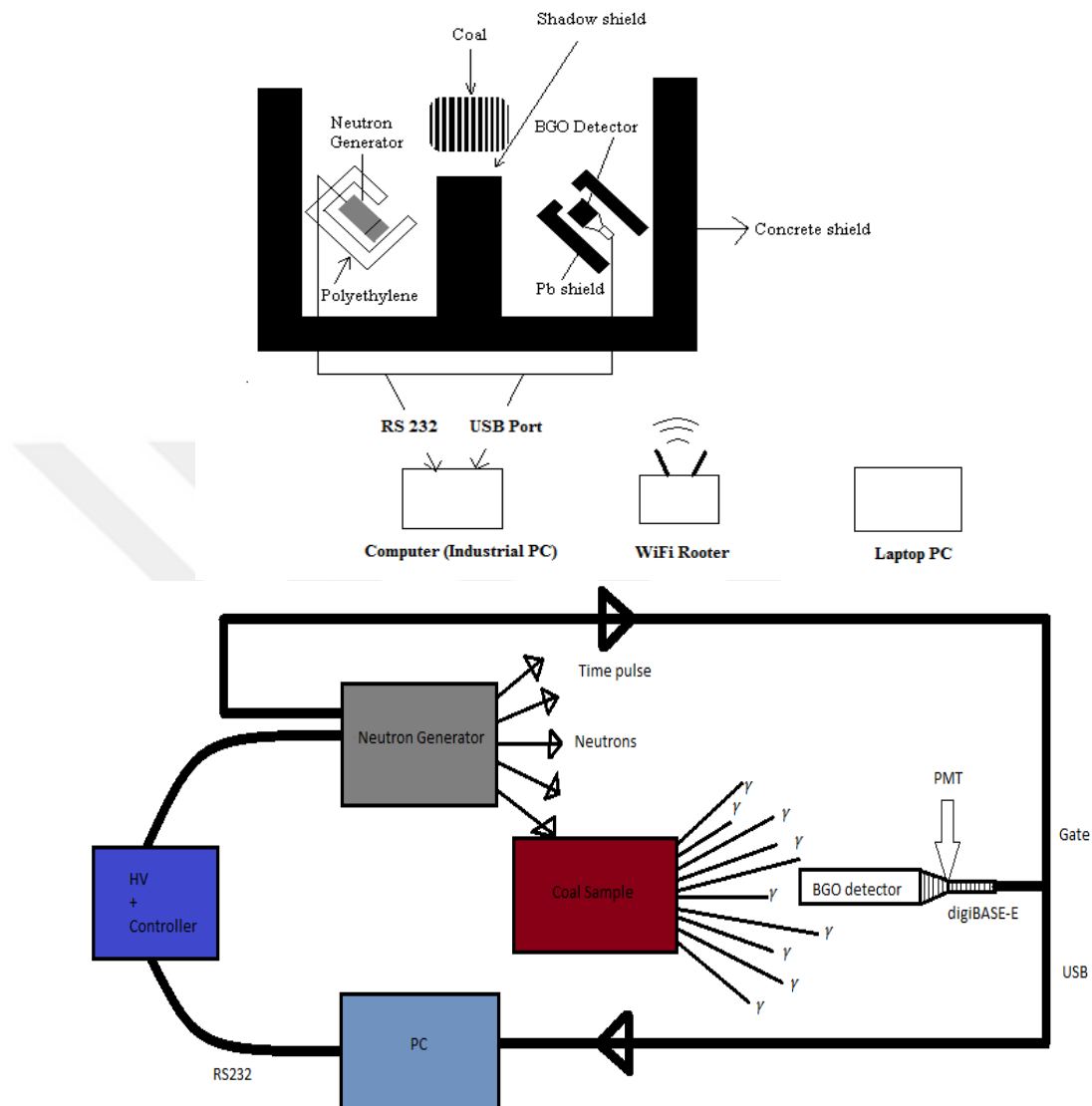


Figure 5. 10: The representative acquisition system

Thermal and fast neutron induced gamma ray spectra separation.

As the graph indicates, it some element energies are identified from the NIS or TNC reaction spectrum. For instance, H, Pb, Al, Si, Ca and Fe are identified from the thermal neutron induced coal gamma spectrum. Their photo-peaks are observed in this spectrum while they are disappeared in the fast neutron induced spectrum. On the other hand, Ca, C and O peaks appear in the gamma spectrum which is resulted from the fast neutron. There are also other kinds of elements which coal does not include in that amount of. They are Pb and N elements. Pb is observed from the

shielding materials around the BGO detector. It interacts with both fast and thermal neutrons. As a result of the reactions there are gamma ray energies at 2.61MeV fast and 7.36MeV thermal. N element is observed dominantly in the spectrum and it is thought that it comes from the air.

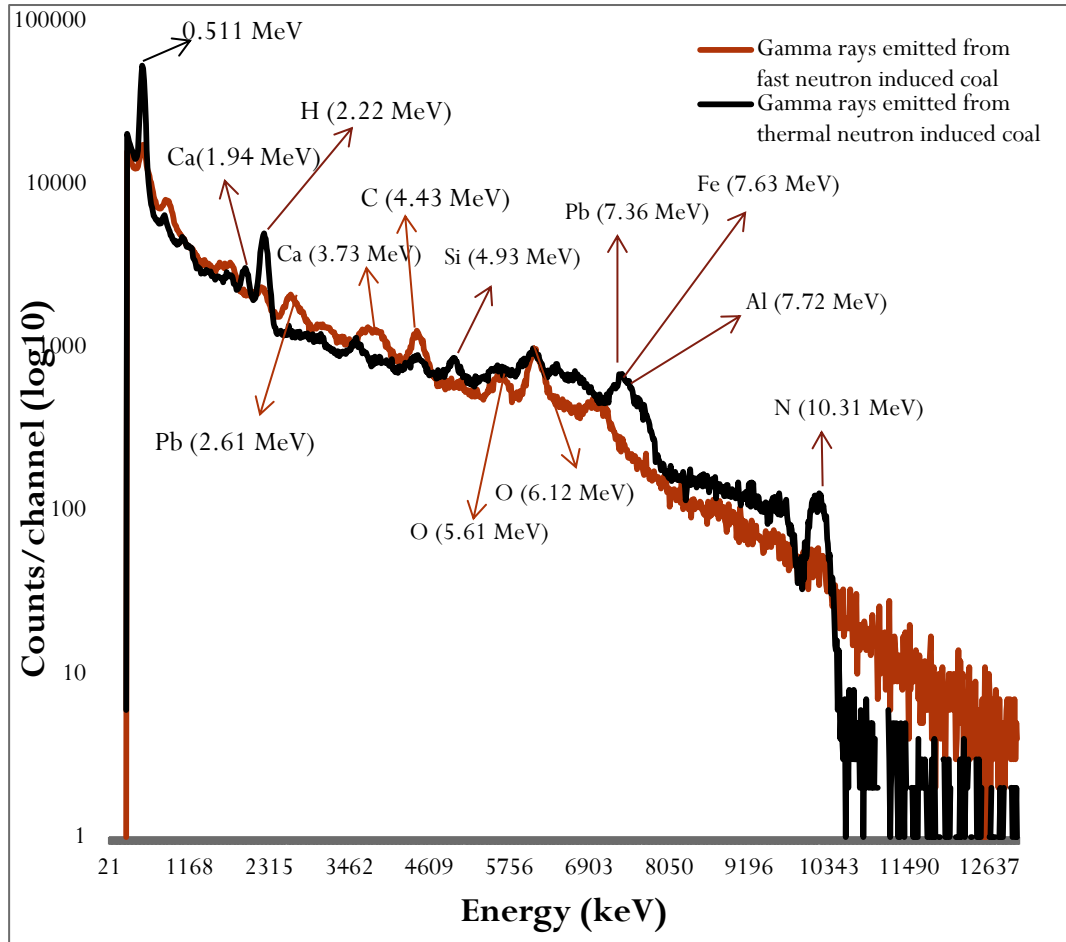


Figure 5. 11: Separation of thermal and fast neutron induced coal spectra

5.7.1 Coal sample measurements

Prepared coal sample constituent weights are determined by taking into account the percentages of coal components (Ash and carbon weights) in Aegean region lignite coal samples. The moisture ratio (25%) and sulphur ratio (4%) are kept constant while the other constituents are changed by a certain ratio. Each sample set is 2000gr. Table 5.3 shows the percentages of the elements in the synthetic coal sample.

The coal samples are radiated with fast neutrons and the signal is in pulse mode. In order to separate fast and thermal neutron induced gamma ray spectra, it is used pulse mode. Resulted gamma rays are collected via BGO detector having digiBASE-E

module. Neutron generator signal is set to duration of 10 μ s neutron emission and 10% duty cycle with 10kHz frequency. However, input signal is not synchronous with the neutron emission. There is a latency between the pulse mode and neutron output. Therefore, there is a latency to the detector. If this is taken into account, the signal duration is adjusted to 6 μ s including 2 μ s latency.

Table 5. 3: Percentages by weight of carbon, ash and sulphur dioxide

	Set-1	Set-2	Set-3	Set-4	Set-5	Set-6	Set-7	Set-8
C %	89	86	76	69	63	50	50	43
Ash %	7	13	20	27	33	40	26	56
SO₂ %	4	4	4	4	4	4	4	4

Table 5. 4: Prepared coal samples and their compositions

	Set-1	Set-2	Set-3	Set-4	Set-5	Set-6	Set-7	Set-8
C	1787	1653	1521	1387	1253	1121	988	855
SiO₂	54	109	16	218	272	327	380	435
Fe₂O₃	24	46	70	93	117	141	164	188
Al₂O₃	24	49	73	99	123	147	172	196
MgO	6	12	18	25	32	37	44	51
CaO	24	49	73	99	123	147	172	196
S	80	80	80	80	80	80	80	80
Total	1999	1998	1851	2001	2000	2000	2000	2001

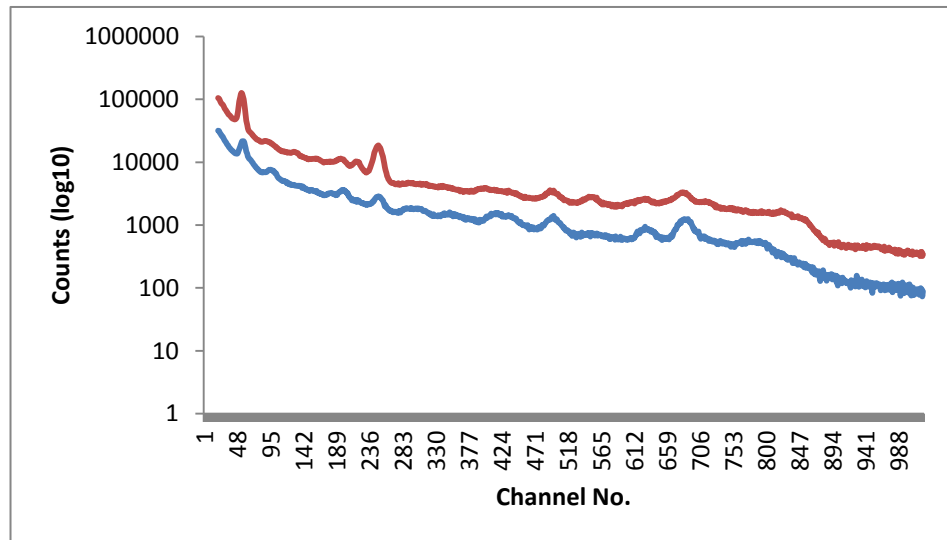


Figure 5. 12 : Set-1 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

After all of the adjustments was conducted, gamma spectra are recorded with list mode data acquisition. Counts per each channel information are collected and expressed by figures below.

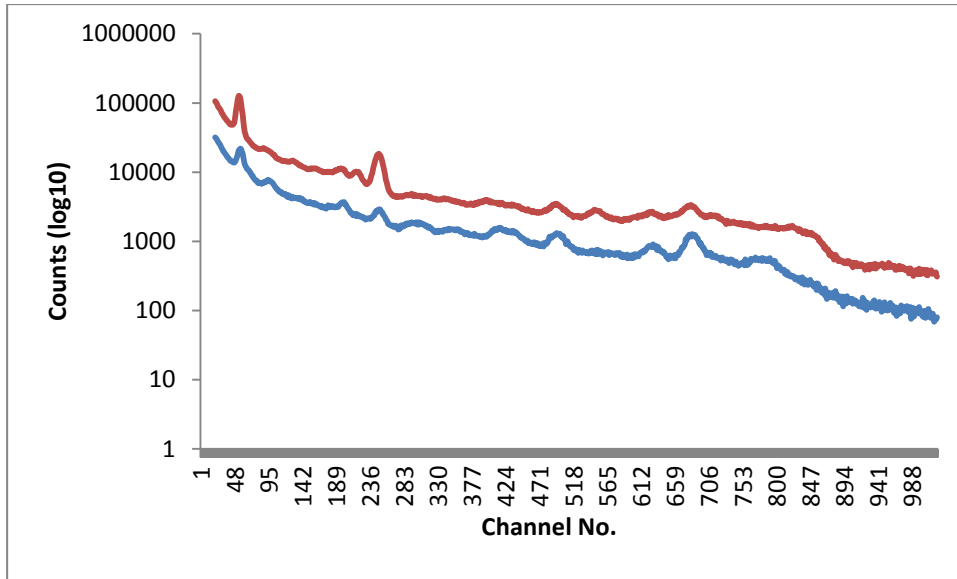


Figure 5.13: Set-2 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

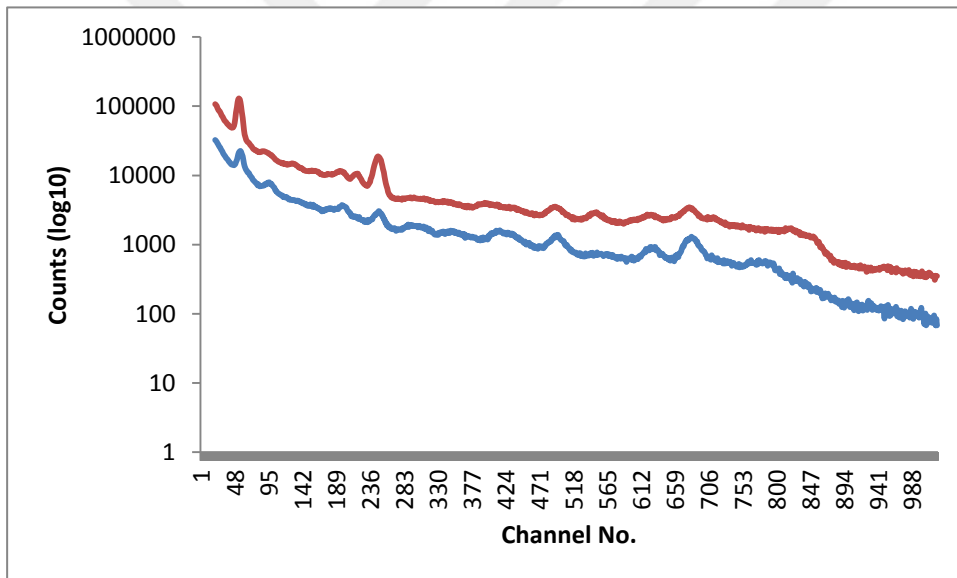


Figure 5. 14: Set-3 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

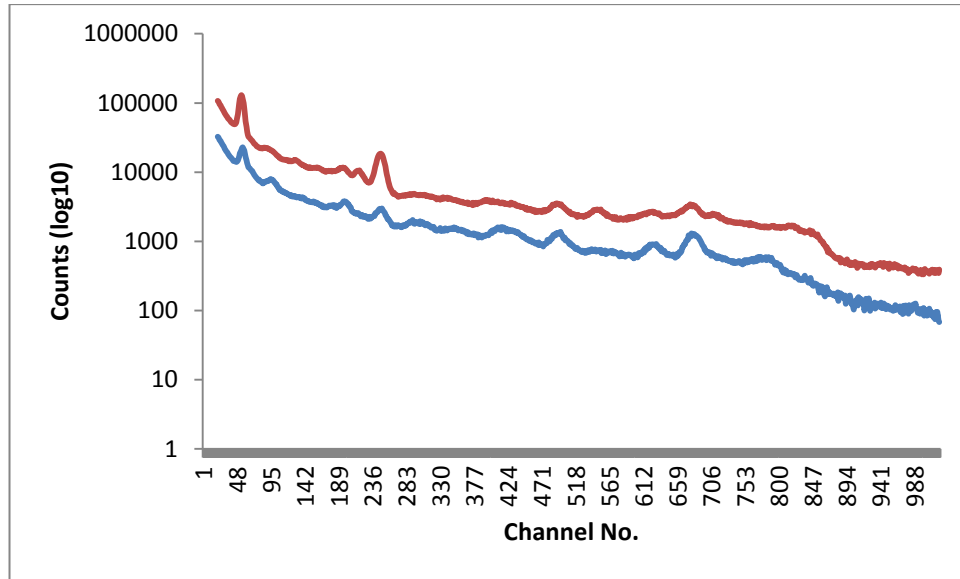


Figure 5. 15: Set-4 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

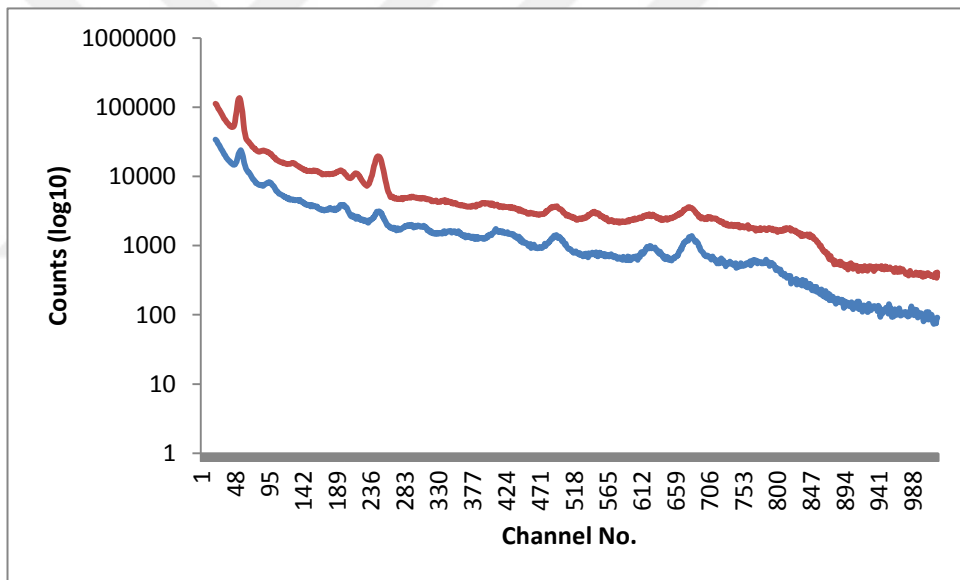


Figure 5. 16: Set-5 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

According to these data for the first 4 sets, the counts per channel values are very similar to each other. The general behaviour and count numbers resembles to each other. In order to be sure about the count number consistency among with other samples, number of coal set is increased. It is thought to be that 8 sets of coal is sufficient for interfering a statistical result. Other gamma ray spectra results are shown below.

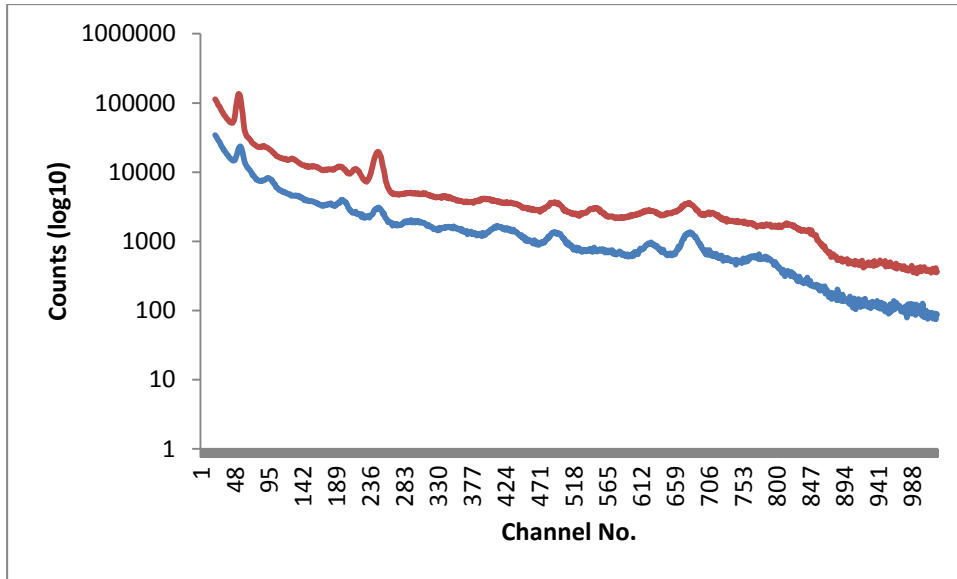


Figure 5.17 : Set-6 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

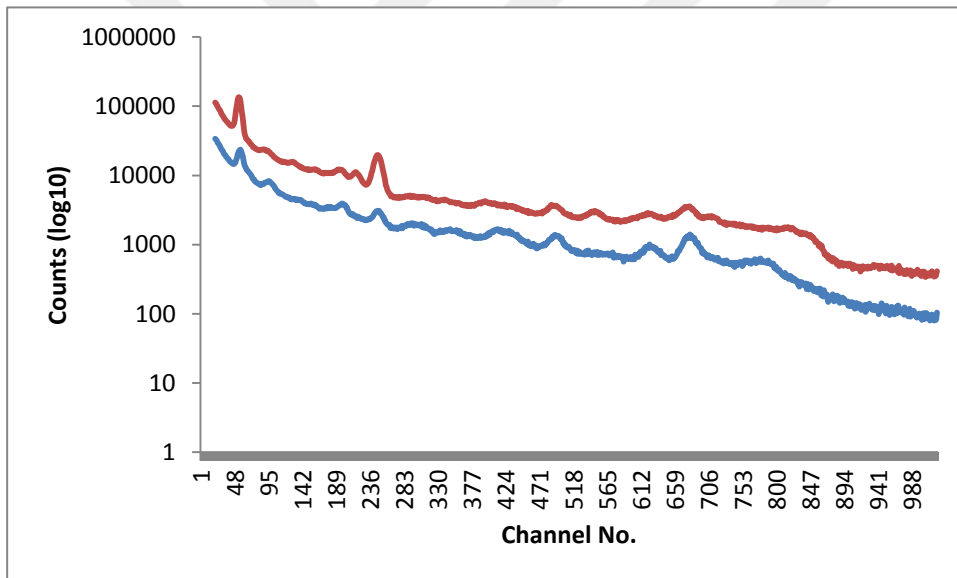


Figure 5.18 : Set-7 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

According to the results above, thermal and fast neutron data for all of the sets are similar to each other in terms of their counts per channel. This might be caused by sample weights and/or high background radiation coming from environmental conditions. The sample amount is not sufficient for correct measurement of the element recognition.

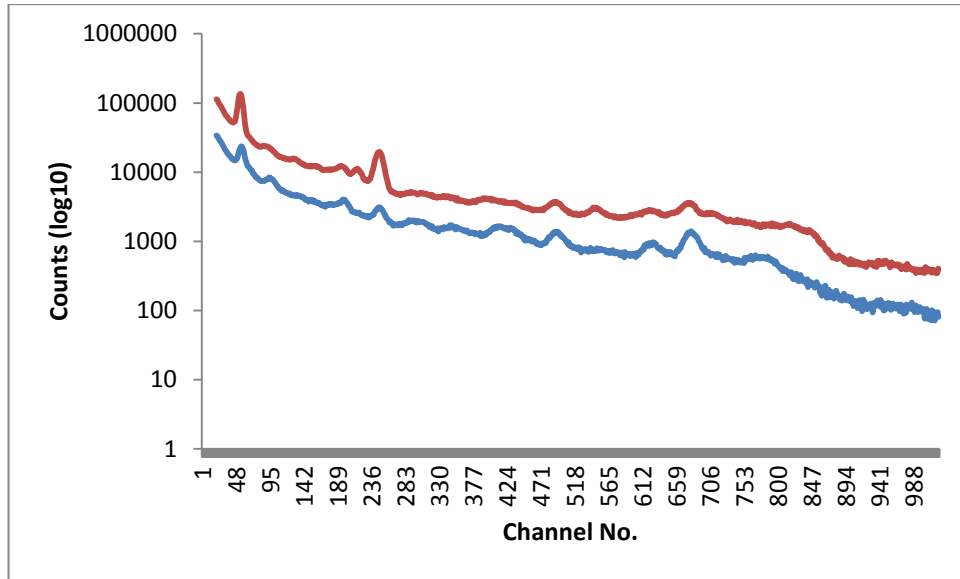


Figure 5.19: Set-8 gamma ray spectra. Red line shows fast neutron induced spectrum and blue one is for the thermal neutron induced spectrum in coal

On the otherhand there is background addition such as biological shielding material concrete wall, detector shielding material lead blocks and polyethylene surrounding neutron generator. Lead blocks and polyethylene dominantly contribute to the gamma spectrum peaks with their energies at 2.61 MeV and 2.22 MeV respectively. Therefore, in the progressive experimental studies it was used 10kg/25kg/50kg coal samples and background radiation was eliminated by using some mathematical methods.

5.7.2 Experiment for response function of BGO detector and element library

Shape calibration of BGO detector expressed by the formula written in 5.2 equation is not sufficient for the correct calibration process since the elements in coal have energy values up to 12 MeV. Therefore, it was needed further energy source in order to make energy calibration in a wide range. In this experiment it was used C-12(n,n') inelastic neutron scattering reaction gamma ray energy at 4.43 MeV for the calibration. By using this energy source as an addition to the previous standard sources, it was found a relation between square root of energy and standard deviation of Gaussian. In this way BGO detector was calibrated up to the 4.5MeV energy. Sigma (keV) versus square root of energy (keV).

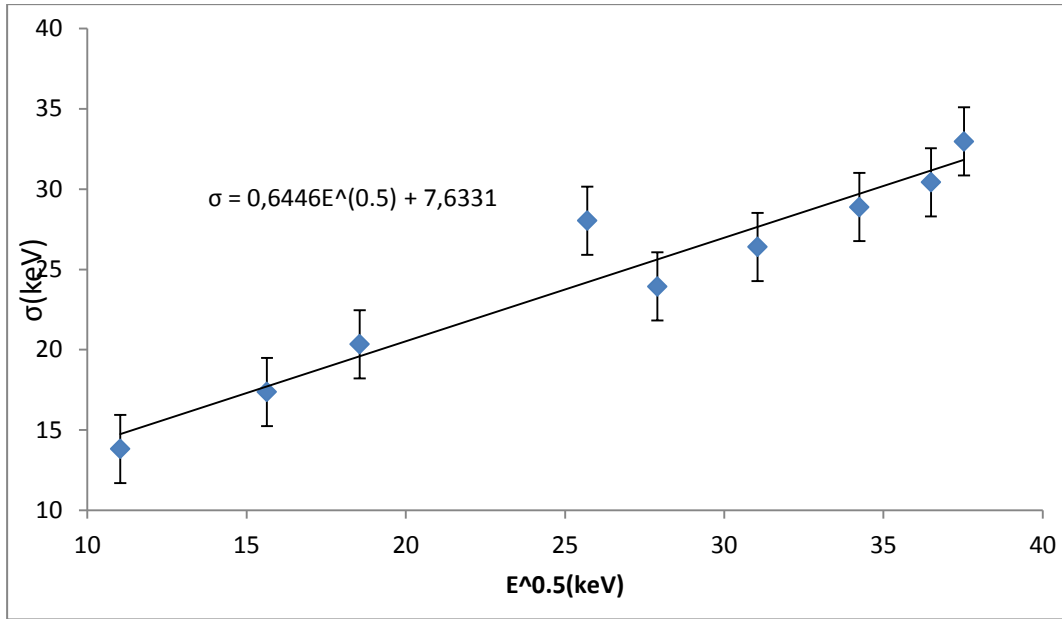


Figure 5. 20: Sigma (keV) versus square root of energy (keV)

It is created a linear fit written in the formula:

$$\sigma(\text{keV}) = 0.6\sqrt{E} + 7.63 \text{ (keV)} \quad (5.6)$$

After this relation was obtained, Gaussian curves were created to obtain a characteristic element curves in coal. These curves have peak energy value based on the dominant energy values written in the Table 5.1. These are the values for the specific energies corresponding to the NIS and TNC reactions.

After creating the Gaussian curves background information is added to the pure gaussians. This background information is obtained via some mathematical algorithms. For this purpose CERN ROOT background algorithm called as “Sensitive Nonlinear Iterative Peak Clipping” was utilized. It was added to the Gaussian peaks after the background information is obtained. Resulted peaks are the certain responses of the elements in coal material.

5.7.2.1 ROOT background algorithm

CERN ROOT TSpectrum Class have features for the advanced spectra processing. It has functions listed as: [27]

- One – dimensional background estimation
- One – dimensional peak search

- One – dimensional smoothing
- One – dimensional deconvolution

This class has some special functions used for the application above. For the background estimation it was used;

```
TH1 *TSpectrum::Background (const TH1 * h, Int_t niter = 20, Option_t* option = “
“)
```

One – dimensional background estimation function.[27] This function calculates a baseline just below the spectrum h. Parameters for this function are described as follows:

- h: 1-D input histogram
- numberIterations, it is adjusted to 20 by default. If numberiterations is increased the result is smoother and lower
- option contains one of the choices below:
 - there is direction parameter and it can be changed to “BackIncreasingWindow”, because its default setting is “BackDecreasingWindow”
 - order of clipping the filter can also be adjusted by using values “BackOrder4”, “BackOrder6”, “BackOrder8”. Default value = “BackOrder2”
 - if “nosmoothing” is selected, the result is not smoothed. By default it is smoothed
 - width of the smoothing window can also be changed as; “BackSmoothing5”, “BackSmoothing7”, “BackSmoothing9”, “BackSmoothing11”, “BackSmoothing13”, “BackSmoothing15”, default value = “BackSmoothing3”
 - if “Compton” is picked, the estimation algorithm takes into account the Compton edge
 - if “same” option is selected the result is superimposed to the current data appearing in the pad [27]

There is a function that calculates the background from the source data. [27]

```
const char* TSpectrum::background(Double_t* spectrum, Int_t ssize, Int_t
numberIterations, Int_t direction, Int_t filterOrder, bool smoothing, Int_t
smoothWindow, bool compton)
```

The main purpose is to separate the useless information from the spectrum. It is deal with the useful information left. [27] The method used in this function is Sensitive Nonlinear Iterative Peak (SNIP) clipping algorithm. In each iteration, new value in i -th channel is calculated. Before the SNIP algorithm application, it is useful to apply LLS operator. [28]

$$v(i) = \log[\log(\sqrt{y(i)} + 1) + 1] \quad (5.7)$$

i shows the channel number, $y(i)$ represents the counts in the i -th channel. This operator allows working over a few orders of magnitude and the square root operation is used for the improving of small peaks in spectrum, $v(i)$ is the histogram data storage vector. [27] It is calculated from vector $y(i)$. According to the given parameter m which is free, $v(i)$ vector is calculated step by step as follows; $v_1(i)$, $v_2(i)$, $v_3(i)$..., $v_m(i)$. In the p -th iteration for new channel i , the result is obtained by taking the average of the values $v_{p-1}(i - p)$, $v_{p-1}(i + p)$ with comparison $v_{p-1}(i)$. It is accepted the minimum of these values. [27]

$$v_p(i) = \min v_{p-1}(i), \frac{[v_{p-1}(i - p) + v_{p-1}(i + p)]}{2} \quad (5.8)$$

Figure 5.21 represents the graphical version of the minimum value formula. Additionally, another case for the region which is outside of the peak region is shown in Figure – 5.22.

After the calculation of $v_m(i)$, it is a must to apply inverse LLS operator in order to get baseline data. [27]

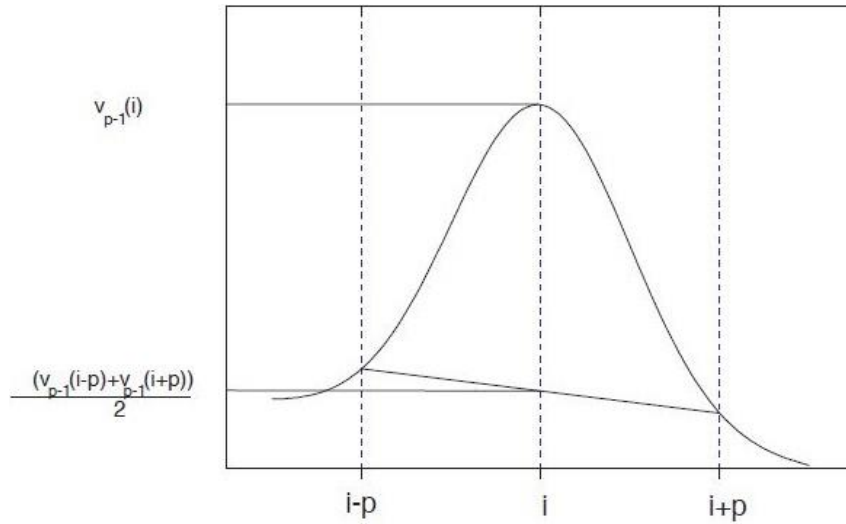


Figure 5.21: The graphical version of the minimum value formula [27]

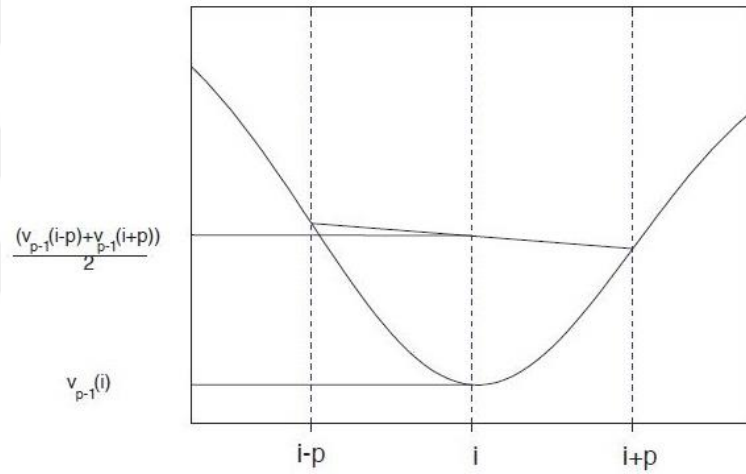


Figure 5.22: Another case for the region which is outside of the peak region [27]

This algorithm was applied our gamma spectrum of coal material for 920 channel data range. Since there are high amplitude photon energies coming from Compton continuum and escaping peaks resulted from pair production in the lower energy range, their data were not taken into account. Because, they give some incorrect mathematical results in the calculation of ROOT Background algorithm. The baseline found with the aid of background algorithm is shown in Figure 5.23.

Red curve is baseline for the gamma spectrum of coal sample. This data was added to the pure Gaussian peaks to obtain element library. Created Gaussian peaks are represented in the same graph with the real gamma spectrum. Figure 5.23 shows the elemental responses before the background addition.

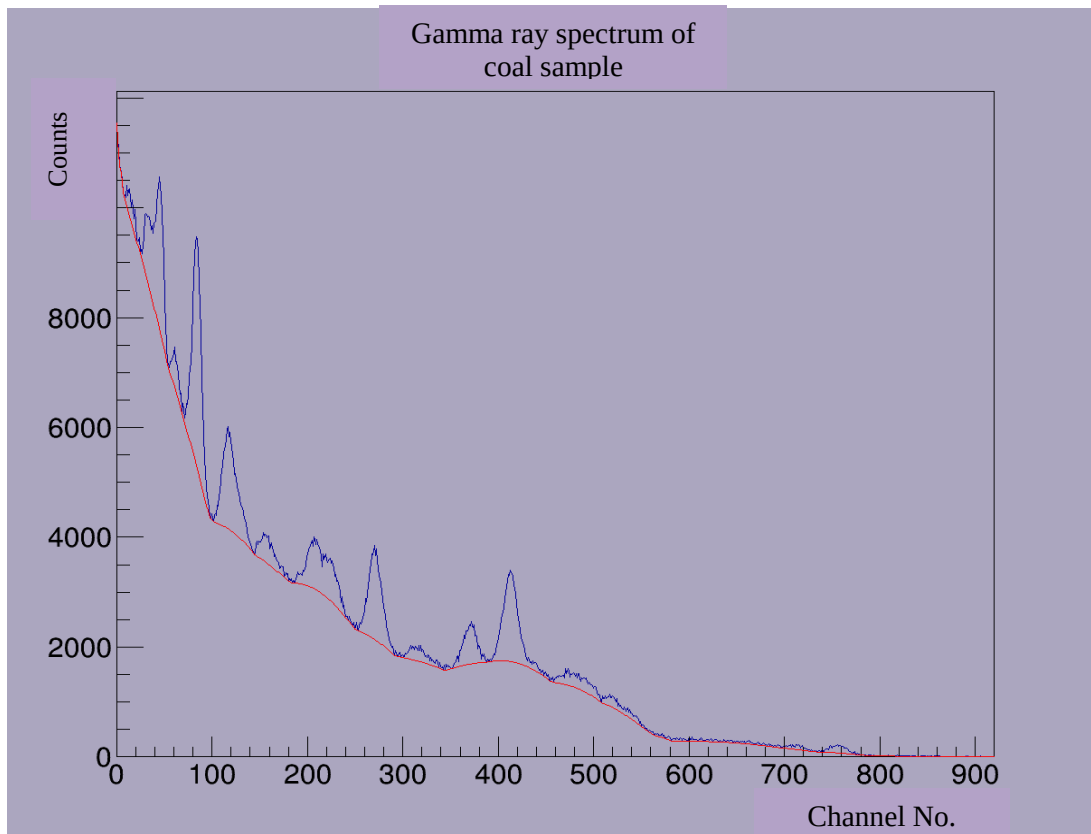


Figure 5.23: The baseline found with the aid of background algorithm

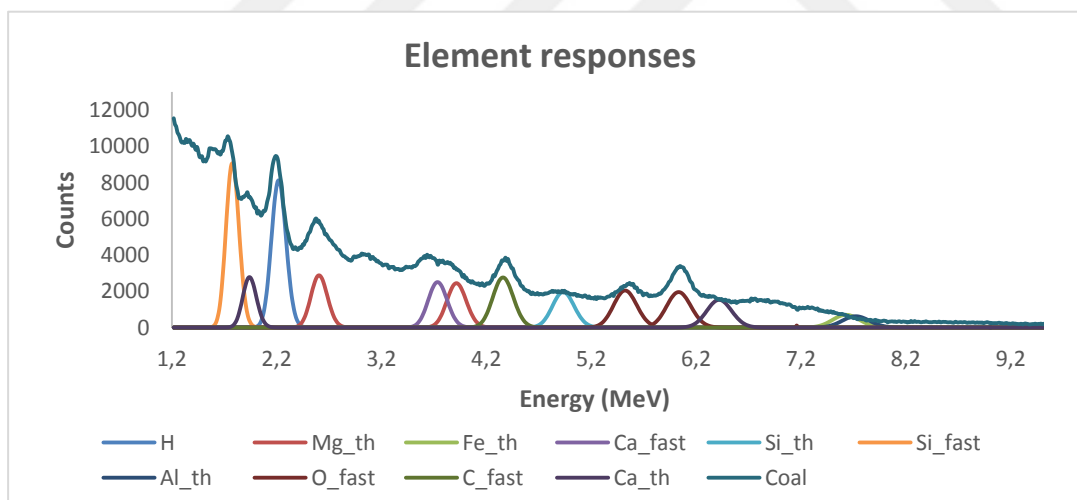


Figure 5. 24: The elemental responses before the background addition

The blue line represents coal data. Abbreviations are:

X_fast: it is the response function of the fast neutron induced gamma ray spectrum

X_th: it is the response function of the thermal neutron induced gamma ray spectrum

5.7.2.2 Element library

It was created two separate element library: fast neutron induced gamma ray spectrum based on the nuclear reaction NIS and the other one is thermal neutron induced gamma ray spectrum originated from the reaction TNC. If the background data is added to the Gaussian peaks, separate element libraries are obtained.

Element library responses from the TNC reactions:

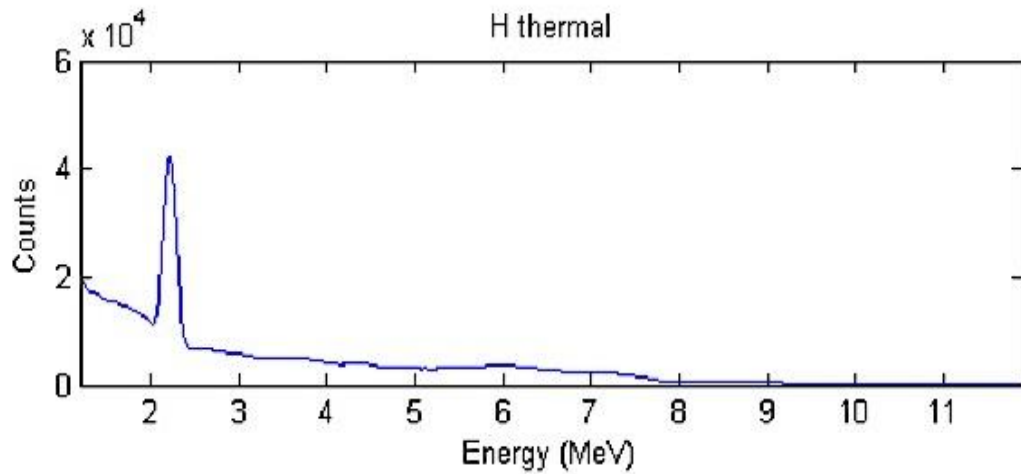


Figure 5. 25 : Response for hydrogen

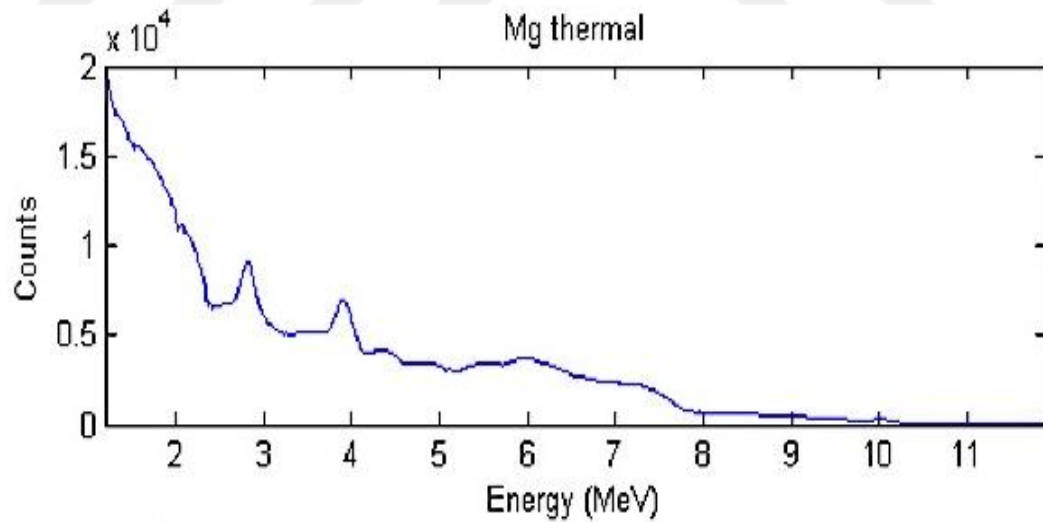


Figure 5. 26 : Response for magnesium

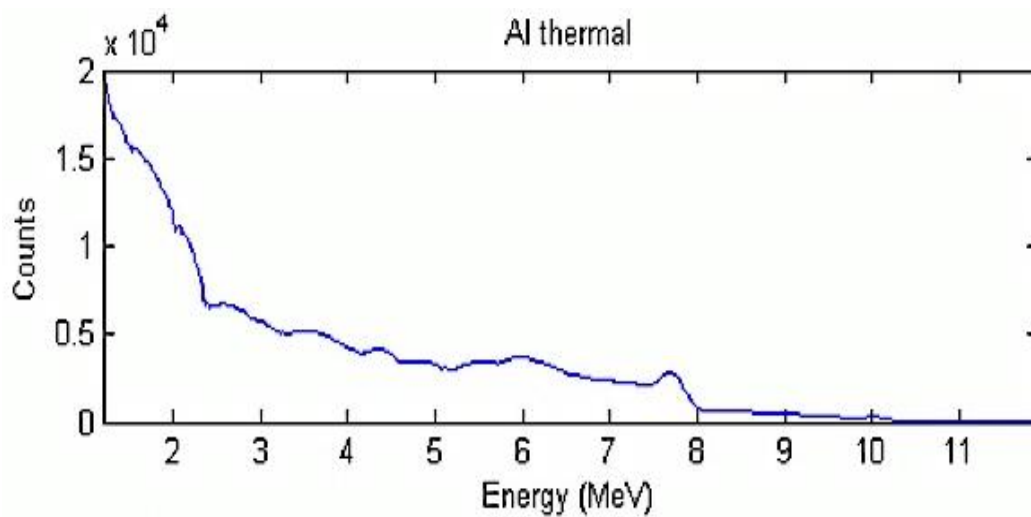


Figure 5.27: Response for aluminum

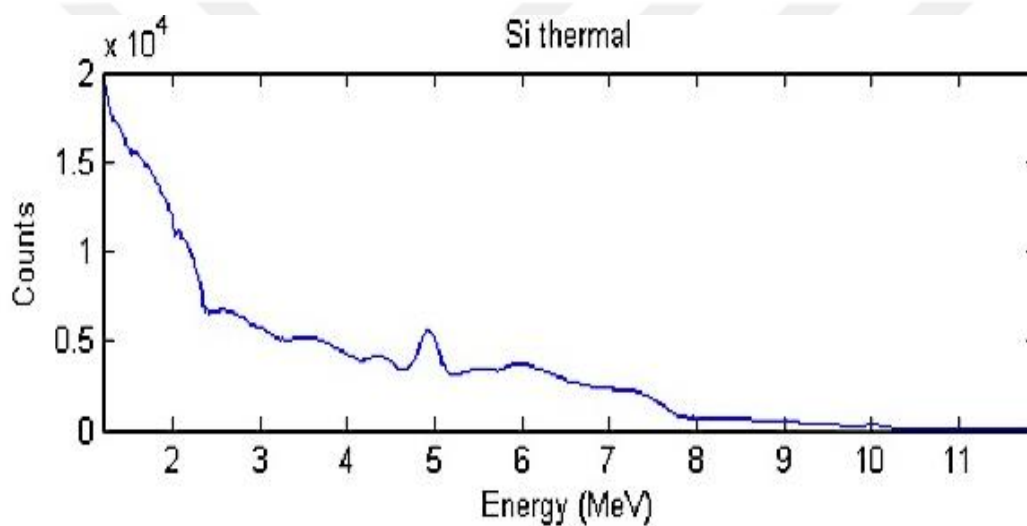


Figure 5.28: Response for silicone

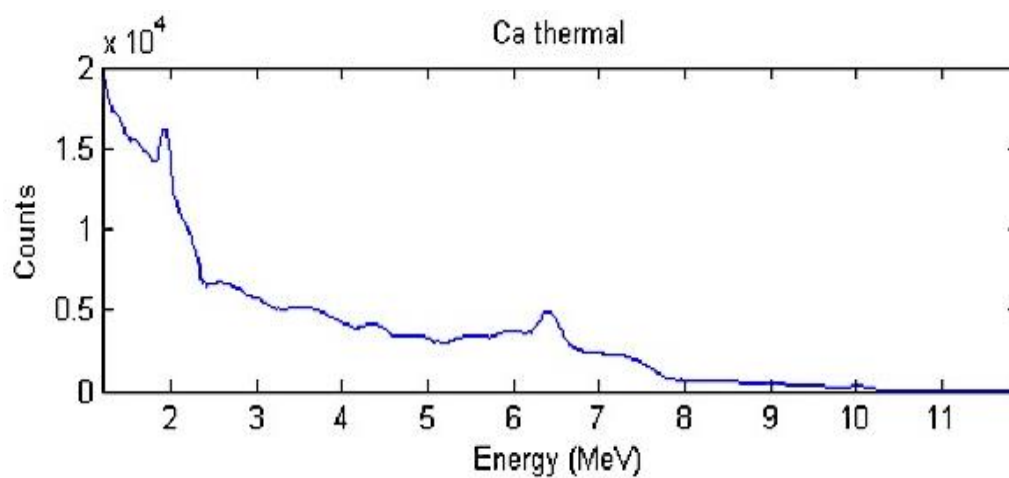


Figure 5.29: Response for calcium

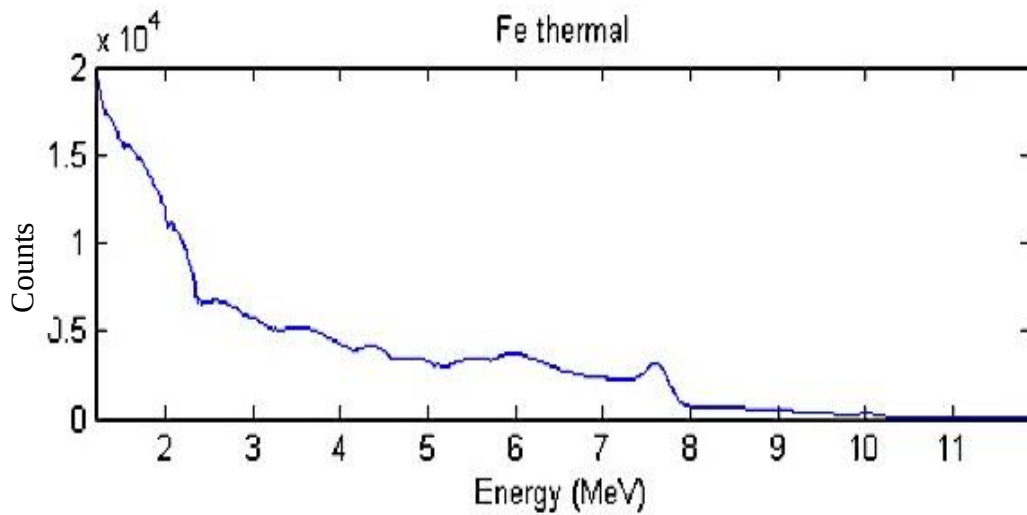


Figure 5.30: Response for iron

Element library responses from the NIS reactions are represented in the figures below. The element library is divided into two groups as thermal neutron induced gamma ray spectra and fast neutron induced gamma ray spectra.

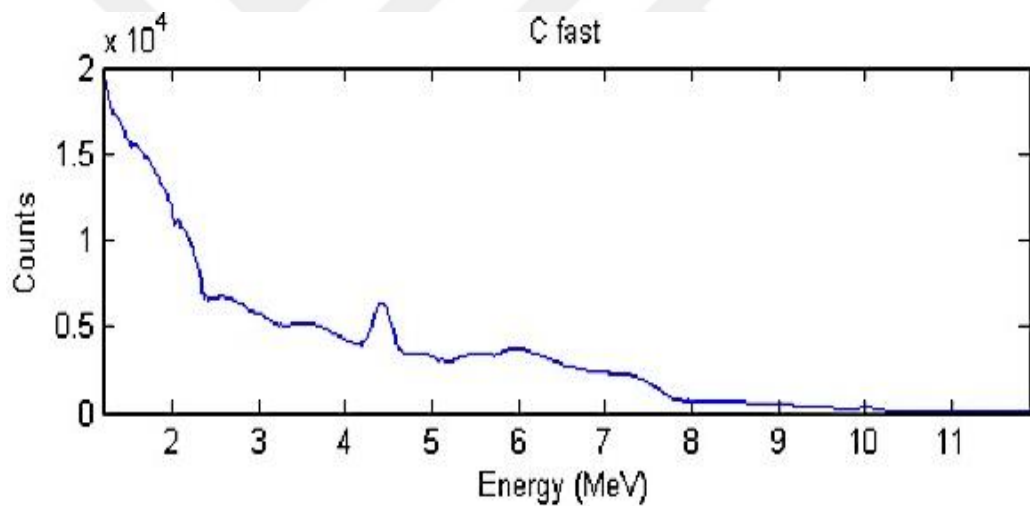


Figure 5.31 : Response for carbon

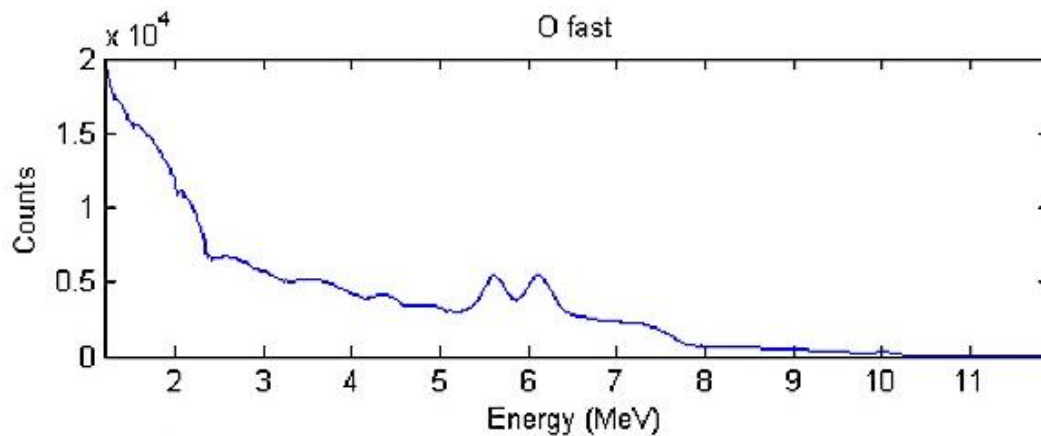


Figure 5.32 :Response for oxygen

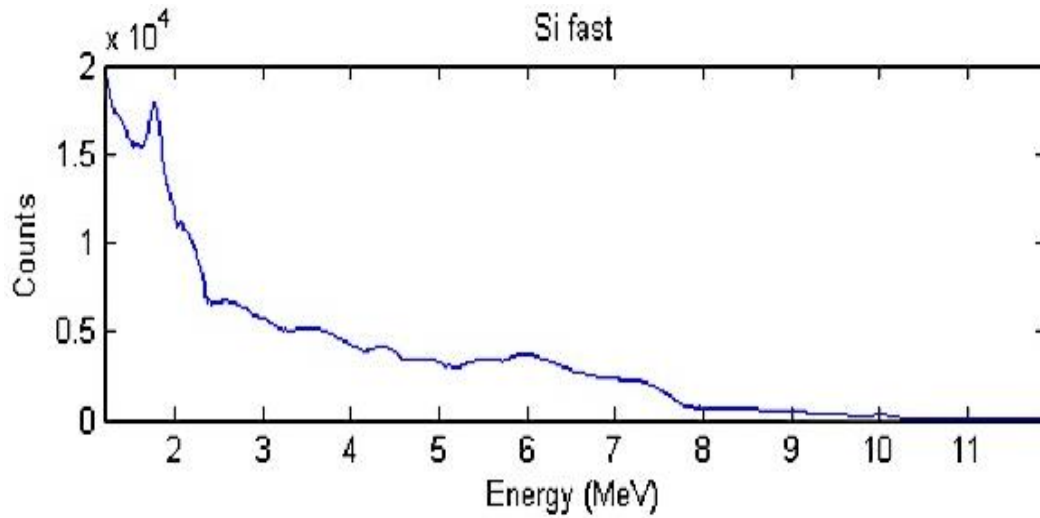


Figure 5.33: Response for silicone

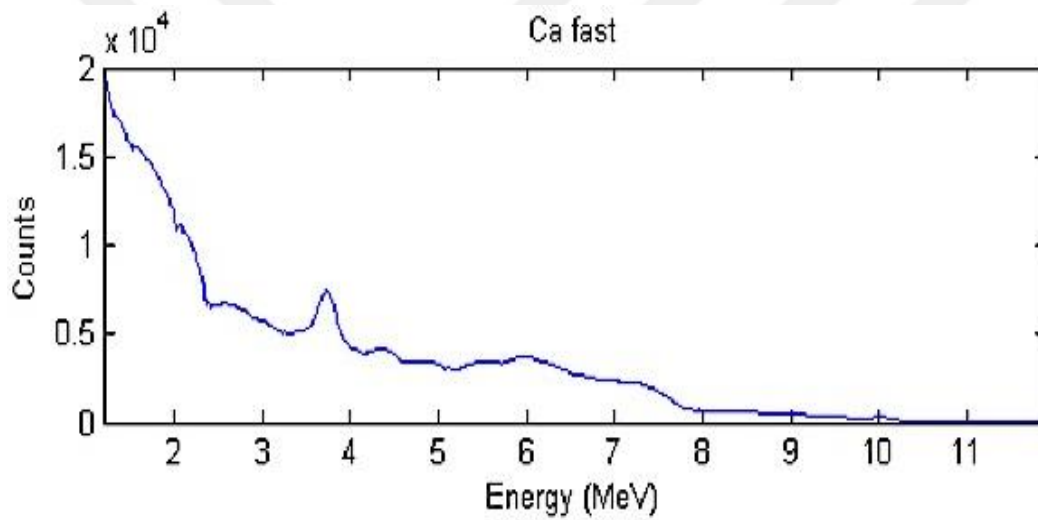


Figure 5.34: Response for calcium

After obtaining these data, multiple linear regression is applied.

5.7.2.3 Multiple linear regression (MLR)

MLR method is used for the determination of coefficient coming from unknown material by using element response library as well. [29] After the MLR coefficients are found, the results are applied to the element response library. At the end coefficient multipliers are determined, then it can be interpreted that the levels of the materials in coal sample. [29]

In this algorithm, coal sample data is the linear summation of each element responses.

$$y_i = \sum_{k=1}^N a_{ik} x_k + e_i, i = 1,2,3,\dots,1024, N = \text{number of elements in sample} \quad (5.9)$$

In equaton 5.9, x values represent data coming from each element appearing in coal, a values are the coefficients indicating the weights of element in coal and e is the intercept where the regression line intersects the axis. The method is based on the least square technique. Unknown coeffcents are found by taking derivative of the error function relative to the coefficients assuming that they are variables. In order to minimize the error and find the most accurate result equation 5.10 is used fundamentally.

$$\text{err} = \sum_{i=1}^N (y_i - a_0 - a_1 x_{1i} - a_2 x_{2i} - a_3 x_{3i} - a_4 x_{4i} - a_5 x_{5i})^2 \quad (5.10)$$

If an example with 5 elements is considered, general solution procedures are as follows:

$$y = a_0 + a_1 x_1 + a_2 x_2 + a_3 x_3 + a_4 x_4 + a_5 x_5 + a_6 x_6 \quad (5.11)$$

It is used least square fit method to obtain unknown coefficients. In order to find the most accurate results, err term is minimized, its derivative equals to zero.

$$\frac{\partial \text{err}}{\partial a_i} = 0 \quad (5.12)$$

The matrix equation represents the symbols given for each new summation. N expresses the number of data.

$$\begin{pmatrix} N & T2 & T3 & T4 & T5 & T6 \\ T2 & T8 & T9 & T10 & T11 & T12 \\ T3 & T9 & T14 & T15 & T16 & T17 \\ T4 & T10 & T15 & T19 & T20 & T21 \\ T5 & T11 & T16 & T20 & T23 & T24 \\ T6 & T12 & T17 & T21 & T24 & T26 \end{pmatrix} \begin{pmatrix} a_0 \\ a_1 \\ a_2 \\ a_3 \\ a_4 \\ a_5 \end{pmatrix} = \begin{pmatrix} T1 \\ T7 \\ T13 \\ T18 \\ T22 \\ T25 \end{pmatrix} \quad (5.13)$$

It is the basic mathematical theory example of the Multiple Linear Regression (MLR) method. It is used with the interpretation of some variables linearly correlated with each other. There is a linear relationship between variable x and the response y . [30] This technique is also conducted by an easy process using Excel data regression model. Summary output of the MLR process is in the Table 5.5 and Table 5.6.

Table 5. 5: Output of the MLR process

Regression	Statistics
Multiple R	0,9966212
R Square	0,9932537
Adjusted R Square	0,9931795
Standart error	219,41432
Observation	920

Table 5.6: Coefficients for the elemental response found by the method in Chapter 5.4.2.4 for the coal sample with 920 channel data

Coefficients		
Variable 1	0.31	H(n, γ)
Variable 2	-0.01	Mg(n, γ)
Variable 3	0.09	Fe(n, γ)
Variable 4	0.098	Ca(n;n', γ)
Variable 5	0.02	Si(n, γ)
Variable 6	0.14	Si(n;n', γ)
Variable 7	-0.01	Al(n, γ)
Variable 8	0.21	O(n;n', γ)
Variable 9	0.27	C(n;n', γ)
Variable 10	-0.1	Ca(n;n', γ)
Intercept	1.58	

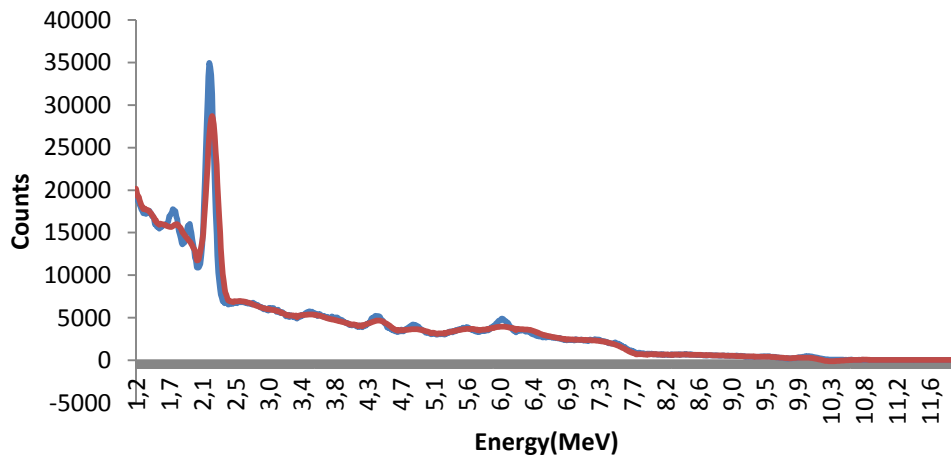


Figure 5. 35 : The real coal data and fitting data are compare

After finding the coefficients, they are substituted in the fitting equation then, the real coal data and fitting data are compared in Figure 5.35.

The blue line shows the coal set data and red one represents the fitting data.

5.7.2.4 Analysis of gamma spectra by using deconvolution ROOT algorithm

In gamma spectra some elements have energy values coming from TNC/NIS reactions are so close to each other. Sometimes detector energy resolution cannot resolve and separate the energy values. In our experiments, such cases are experienced more often since it is used BGO detector in data collection. BGO detector resolution is less than germanium and Sodium Iodine detectors. In order to observe photo peaks more accurate, second method deconvolution is applied.

There are lots of methods used for deconvolution technique. [30] SVD (Singular value decomposition), maximum entropy, linear regularisation, Fourier, VanCittert and gold algorithm which is explained in chapter 4.14. Resolution of gamma spectra is enhanced by Gold algorithm used in CERN ROOT programming packet.

ROOT programming codes calculate deconvoluted spectrum by using Gold algorithm. It uses a response function to deconvolute the spectrum. [31] A source pointer stores the resulting vector. Additionally, a boosting operation can be applied and repeated. There are some parameters used in ROOT:

- source: source spectrum vector pointer
- response: response spectrum vector pointer
- numberIterations
- ssize: length of the response and source spectra
- numberRepetitions, for repeated boosted deconvolution
- boost: coefficient for boosting [31]

It is aimed to decompose the multiplets with this function. The mathematical expression is written in equation 5.14.

$$y(i) = \sum_{k=0}^{N-1} h(i - k)x(k), i = 0, 1, 2, \dots, N-1 \quad (5.14)$$

Impulse function is $h(i)$, x and y are input and output vectors respectively length of the x and h vectors is N . If this is expressed in matrix form:[31]

$$\begin{pmatrix} y(0) \\ y(1) \\ \dots \\ y(2N-2) \\ y(2N-1) \end{pmatrix} = \begin{pmatrix} h(0) & 0 & 0 & \dots & 0 \\ h(1) & h(0) & 0 & \dots & \dots \\ \dots & h(1) & h(0) & \dots & \dots \\ \dots & \dots & h(1) & \dots & \dots \\ \dots & \dots & \dots & \dots & \dots \\ h(N-1) & \dots & \dots & \dots & 0 \\ 0 & h(N-1) & \dots & \dots & h(0) \\ 0 & 0 & h(N-1) & \dots & h(1) \\ \dots & \dots & \dots & \dots & \dots \\ 0 & 0 & 0 & \dots & h(N-1) \end{pmatrix} \begin{pmatrix} x(0) \\ x(1) \\ \dots \\ x(N-1) \end{pmatrix} \quad (5.15)$$

It is assumed that response and output vectors are known. Deconvolution solves the overdetermined system of linear equations. It calculates the x vector. Deconvolution process is time consuming and a critical operation. Gold algorithm works very well without oscillating just as other techniques do not. [32]

In this algorithm the response function is one dimensional vector. Therefore, it is allowed to separate the photo peaks one by one according to given response functions. Total 13 Gaussian peaks are considered for the process. Each element characteristic energy range is taken individually. Element library calculated from deconvolution algorithm is represented in figures below:

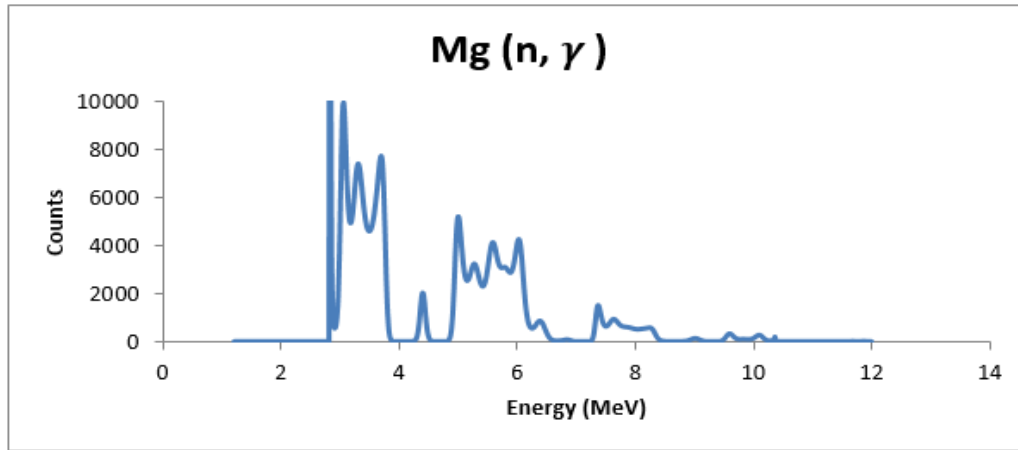


Figure 5. 36 : Deconvolution for magnesium (thermal neutron induced)

Deconvoluted spectra shows a general behaviour that they begin from the energy value where they have a peak. The main photo – peak shape changes according to the response function which is given as an input for deconvolution algorithm in ROOT programming language packet. These photo – peaks appear at an exact energy value

of their characteristic gamma photon energy. The response function theoretically has an exact energy value corresponds to (n,γ) or $(n;n',\gamma)$ reactions. Other peaks have no physical meaning they are created as a result of the mathematical formulation. If it is desired that other kinds of element effects to be appeared in spectra, the response function should be a multidimensional matrix, since it includes other kinds of elements' energy responses. In our case it is used a one – dimensional deconvolution algorithm.

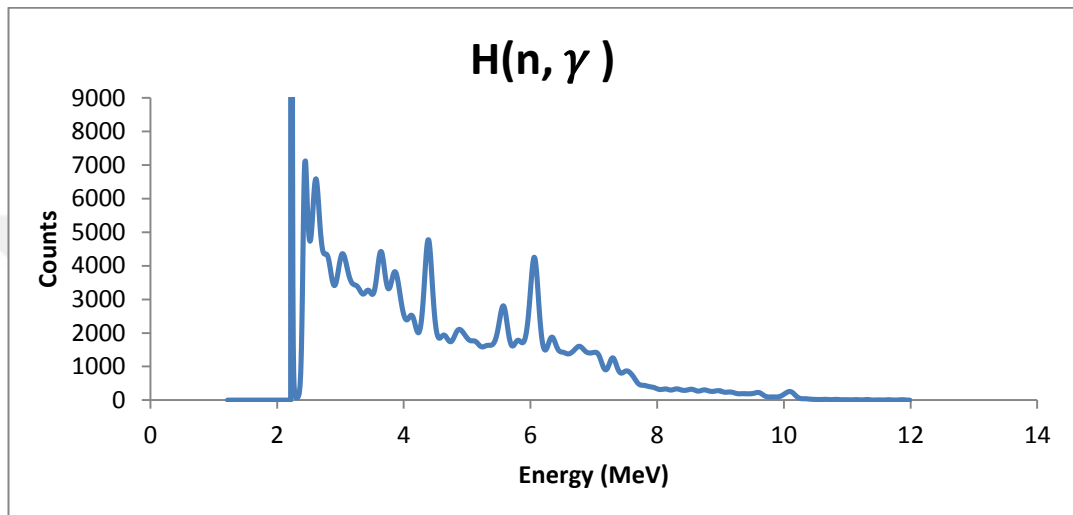


Figure 5.37: Deconvolution for hydrogen (thermal neutron induced)

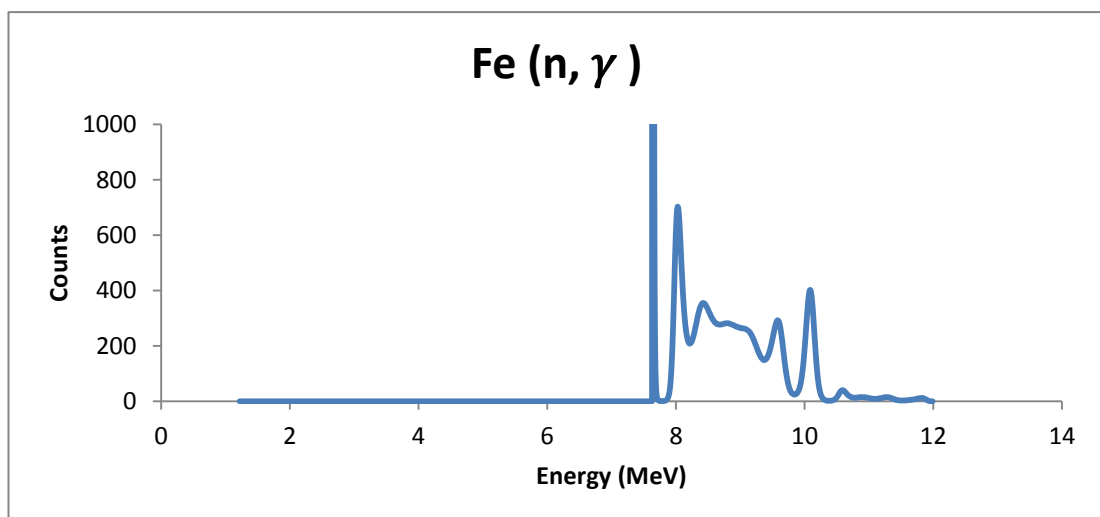


Figure 5. 38 : Deconvolution for iron (thermal neutron induced)

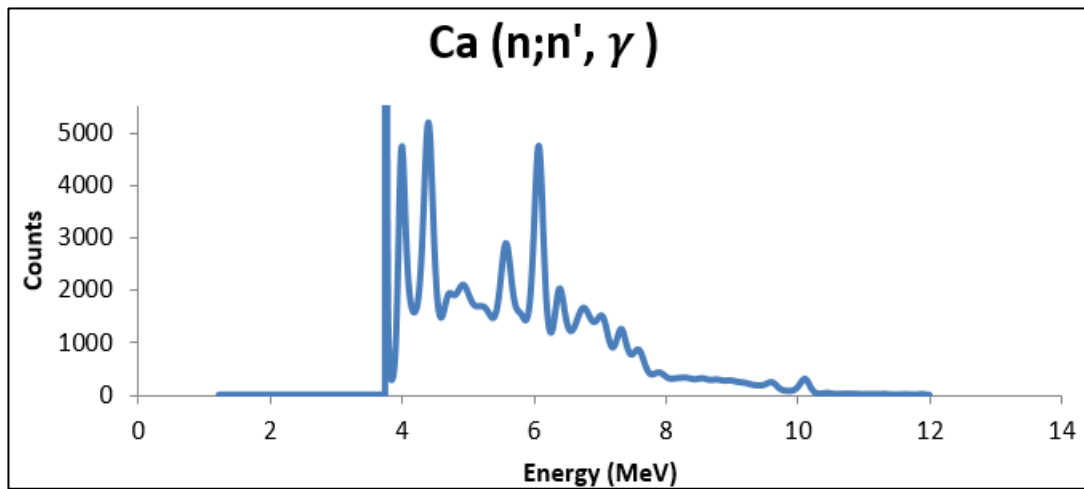


Figure 5. 39: Deconvolution for calcium (fast neutron induced)

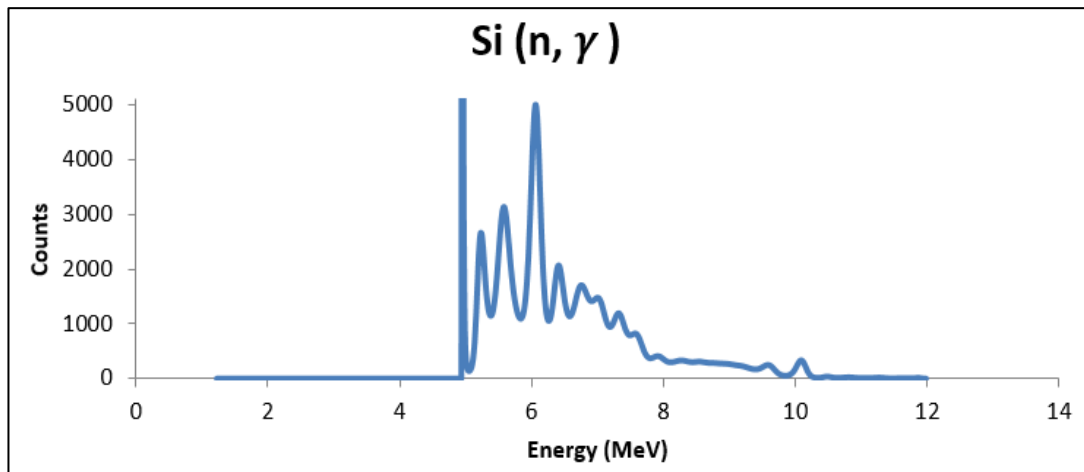


Figure 5. 40: Deconvolution for silicone (thermal neutron induced)

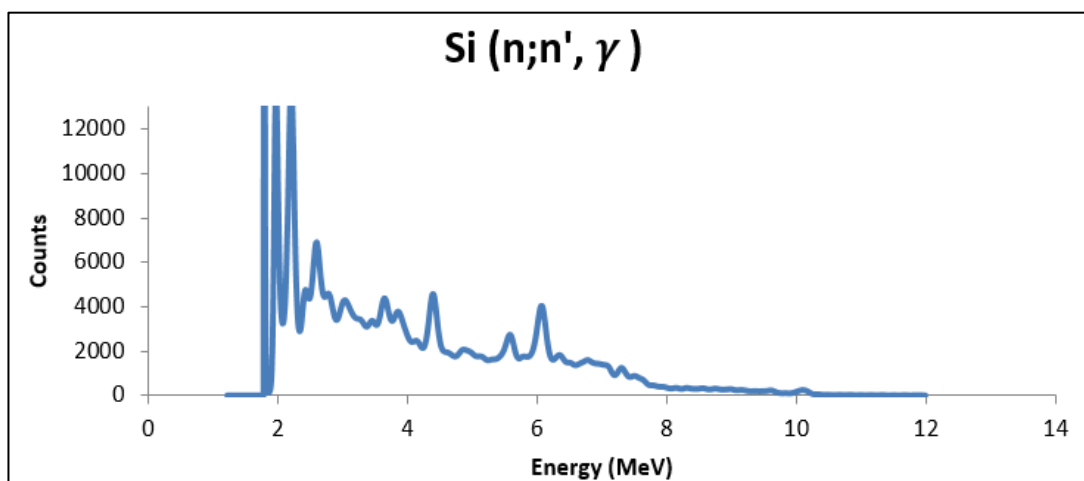


Figure 5. 41: Deconvolution for silicone (fast neutron induced)

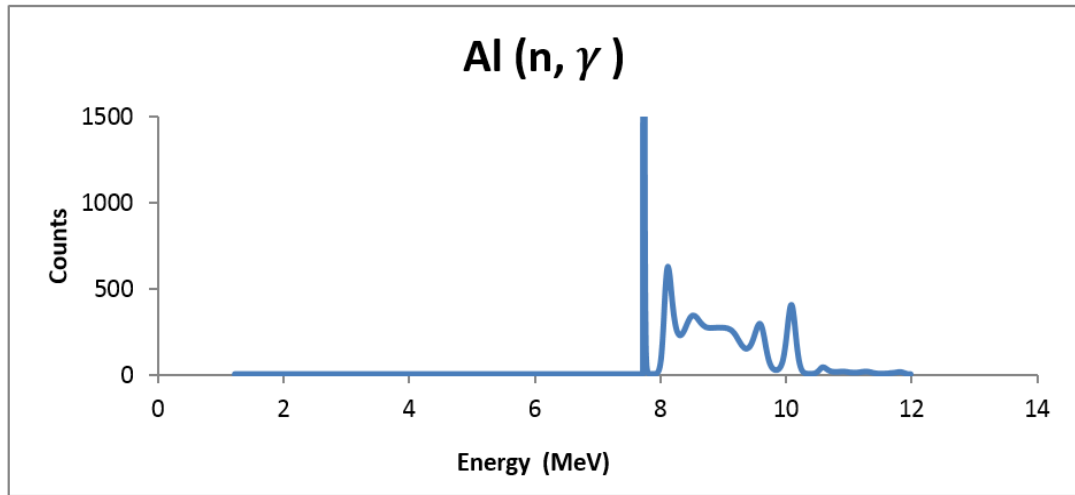


Figure 5.42: Deconvolution for aluminum (thermal neutron induced)

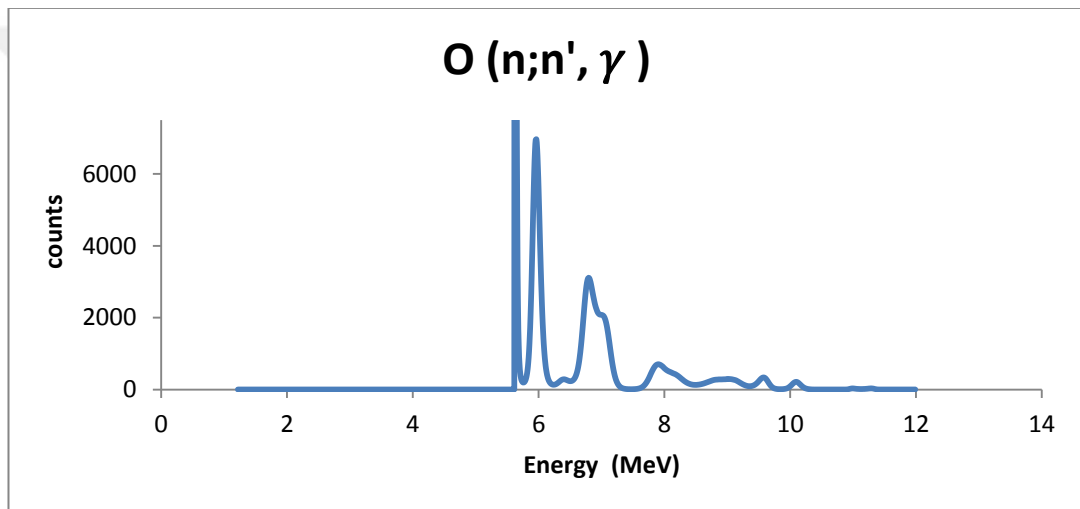


Figure 5.43: Oxygen deconvoluted (fast neutron induced)

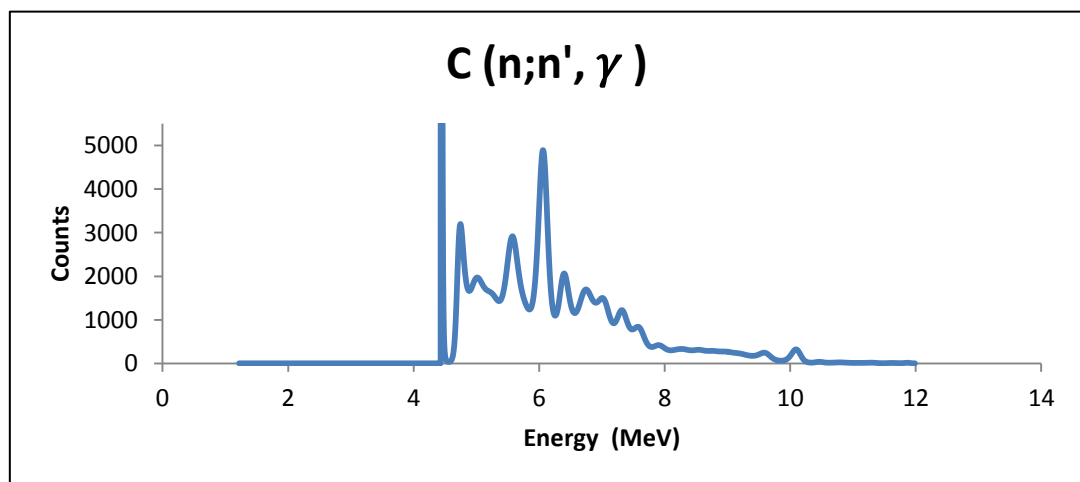


Figure 5.44: Carbone deconvoluted (fast neutron induced)

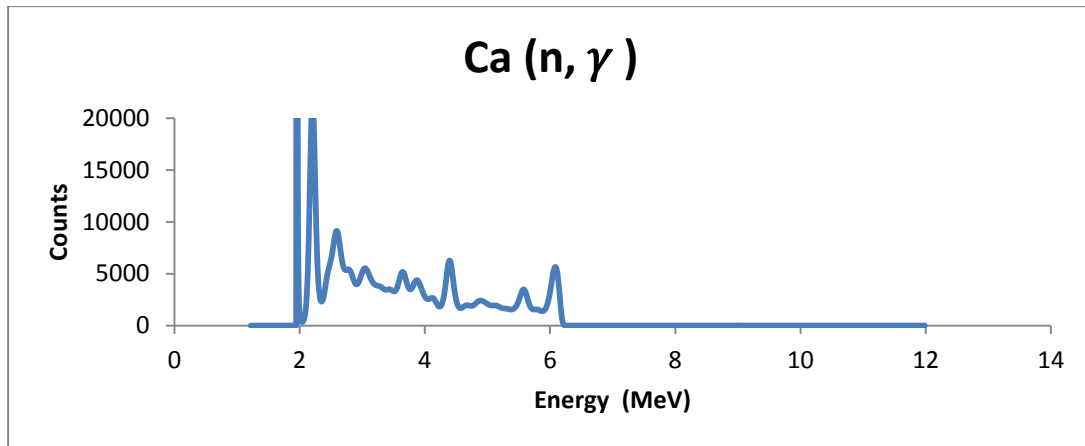


Figure 5. 45: Calcium deconvoluted (thermal neutron induced)

The element library found by deconvolution process and coal spectrum comparison are represented below.

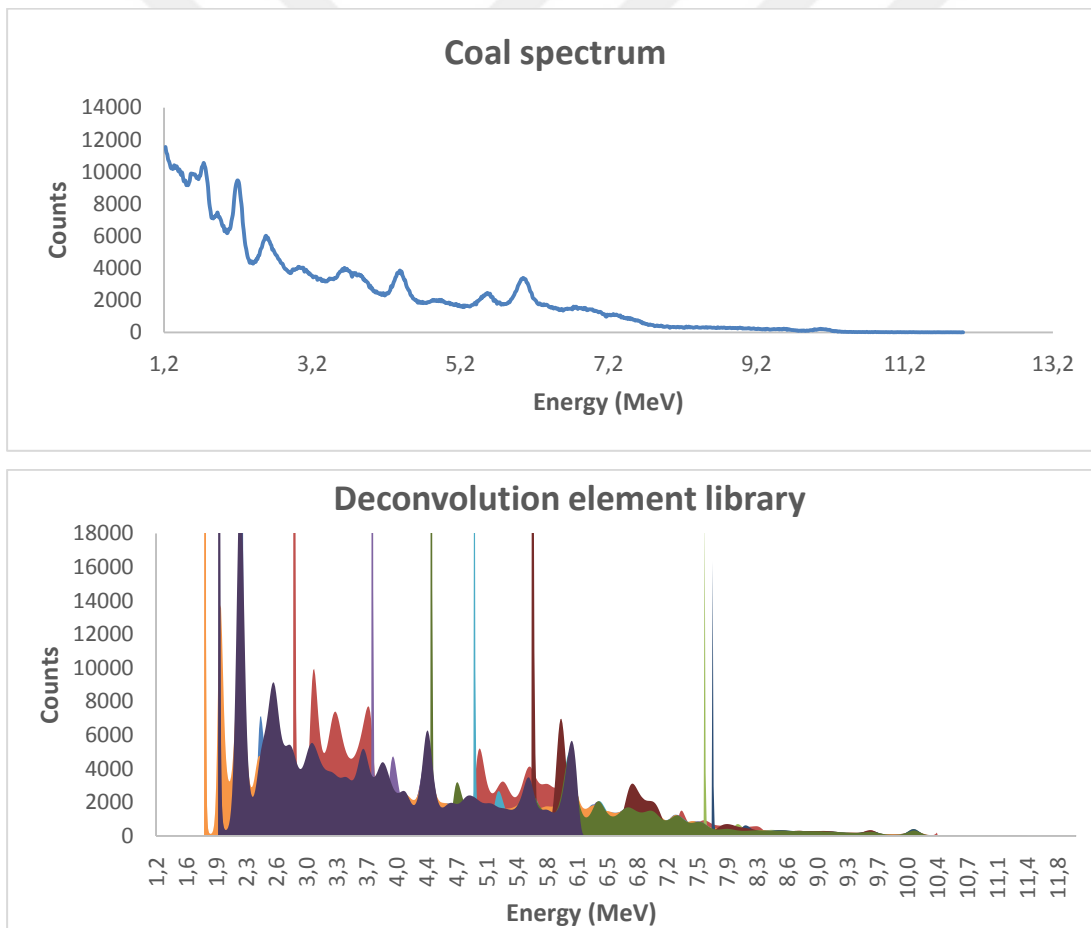


Figure 5. 46: The element library found by deconvolution

5.7.3 Element and coal parameter libraries by using experimental results

5.7.3.1 Element library with synthetic coal sample experiments

In order to analyse the gamma ray spectra created by the interaction of coal with neutrons, it can be used least square method as a third technique.

Analysis of PFTNA (Pulsed Fast Thermal Neutron Activation Analysis) experimental results are very hard since there are nonlinear structures in the process. Least square method is the most common technique used in such cases. These nonlinear effects are minimized by repetition processes and comparison of them with assumed results. LS method is important since it calculates the element weight in the coal sample. Ash, moisture, calorie and sulphur content of the coal material are easily calculated with the aid of LS method. It is obtained element library for the TNC/NIS reactions for the each element. These response functions are used for the determination of moisture, sulphur, ash and energy contents of coal sample which has unknown chemical composition.

Table 5.7: The element contents in coal prepared by synthetically

%	Set1	Set2	Set3	Set4	Set5	Set6	Set7	Set8	Set9	Set10
Element										
C	80	76	73	69	66	62	57	53	49	46
Si	3	3	3	4	5	5	6	6	7	8
Fe	2	2	2	2	3	3	4	4	5	5
Al	2	1	2	2	2	2	3	3	3	4
Mg	0.4	0.5	0.6	0.6	0.7	0.8	0.9	1	1	1
Ca	2	2	2	3	3	3	4	4	5	5
S	1	1	1	1	1	2	2	2	2	3
H	0.2	0.2	0.3	0.3	0.3	0.4	0.5	0.5	0.5	0.6
O	10	11	12	14	16	18	20	22	24	26

The main idea of LS method is that composition of an unknown sample is based on the summation of element responses. According to this approach, element library is created first, then it is used for the determination of element content of coal sample

under observation. It was obtained succesful results from this method in our experiments. In order to find successful results with LS method, the number of coal sets are at least one more than the number of elements in the sample (degrees of freedom). [2] For 9 individual elements it was prepared 10 coal sets. Table 5.7 shows the element contents in coal prepared by synthetically.

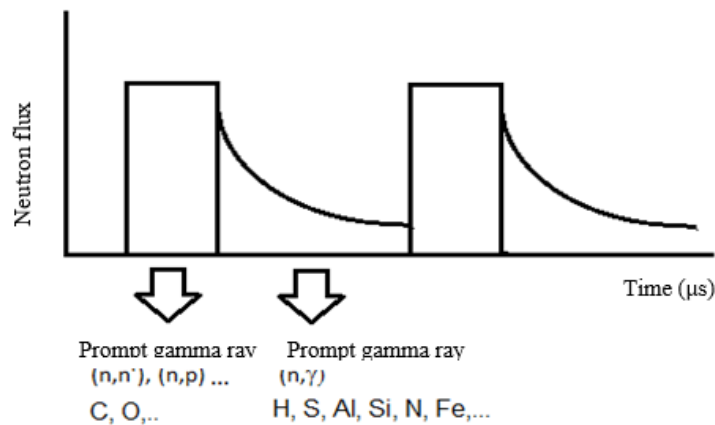


Figure 5. 47 : Carbon element photo peak

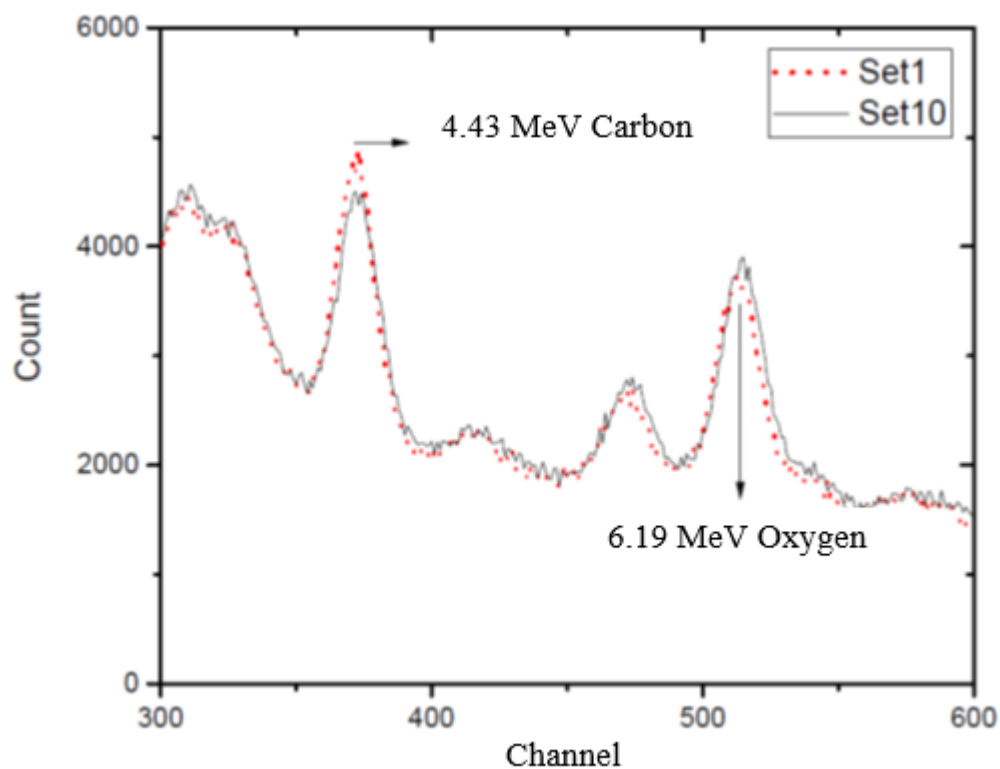


Figure 5. 48 : Created 9 element libraries from 10 coal sets

The coal sets written in the Table 5.7, were radiated with pulsed neutrons with duration and frequency of 10 min. and 2kHz respectively. Neutron generator produces neutrons for 100µs and stops for 400µs duration. In this way, thermal and fast neutrons are dissociated. In Figure – 5.46, carbon element photo peak shows a declining behaviour at 4.43MeV as the carbon ratio decreases through the Set1 to Set10 (from 80.45% to %46.08). In contrast to carbon peak, oxygen peak shows an increasing behaviour at 6.19MeV energy. There were created 9 element libraries from 10 coal sets. This library is represented in Figure – 5.47.

This shows the behaviour of the carbon and oxygen peaks for set1 and set10. The difference between them is expressed in terms of their counts.

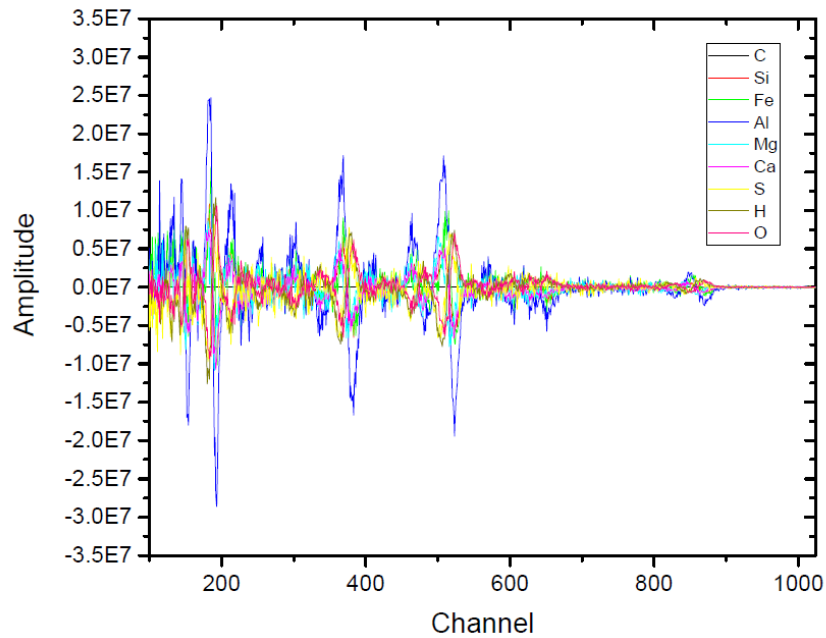


Figure 5. 49 : The element library for 9 elements

Figure 5.50 represents the element library for 9 elements. This data matrix is used for the determine element content of unknown bulk material.

For our experiment, LS method application is expressed by:

$$y_i = \sum_{k=1}^9 a_{ik}x_k, \quad i = 0, 1, 2, \dots, 1023 \quad (5.16)$$

By using equation 5.16, it is utilized least squares criteria to find most probable value.

$$M = \sum_{i=1}^{1024} (y_i - \sum_{k=1}^9 a_{ik} x_k)^2 \quad (5.17)$$

In this equation, it is assumed that the terms a_{ik} are constant. These values do not change with the element content. Therefore, it is calculated the minimum value of M by taking derivative with respect to x_k terms.

$$\frac{\partial M}{\partial x_k} = \sum_{i=1}^{1024} \left(y_i - \sum_{k=1}^9 a_{ik} x_k \right) a_{ik} = 0 \quad (5.18)$$

This is written in matrix form as follows:

$$X = (A^T A)^{-1} A^T Y \quad (5.19)$$

For LS method, element library should be known first, then A matrix is obtained after the derivation operation of equation 5.18 with respect to a_{ik} .

$$A = Y X^T (X X^T)^{-1} \quad (5.20)$$

Afterwards, by using matrix A , elemental weights of an unknown material are found. Table 5.9 shows the measured and calculated ratios.

Table 5. 8 : The measured and calculated ratios

Element	C	Si	Fe	Al	Mg	Ca	S	H	O
Measured	55.2	6.6	4.2	3.3	0.98	4.5	2.6	0.5	21.8
% Calculated	55.6	6.5	4.2	3.3	0.97	4.5	2.5	0.5	21.4

The coal samples having approximately 25kg weights were mixed so as to obtain elemental contents indicated in Table 5.8. Each sample was radiated for 10 minutes and emerging gamma rays are recorded by BGO detector. It was used MATLAB code for determining elemental ratios in coal sample with this algorithm.

5.7.3.2 Coal parameters library with real coal samples

It was needed coal samples which are analysed before in order to calibrate our system. It was worked with 750kg coal samples bought from Malkara. There are 3 different kinds of coal; “tüvenan”, “findık”, “toz”. They are classified according to

their physical dimensions and processing experience. These samples are analysed, their content is known and 7 parameters are taken into account. These values are represented in the Table 5.9.

Table 5.9 : Sample contents

	Tüvenan	Fındık	Toz
(%)Total moisture	28	35	37
(%)Ash	32	13	11
(%) Sulphur	1	2	1
(%) Volatile matter	21	26	26
(%) Fix carbon	16	24	24
(%) Carbon	27	37	38
Calorie (kcal/kg)	2391	3400	3437

According to these ratios, it was mixed three types of coal materials with different ratios to obtain coal sample having desired element content. There are 7 parameters belong to the coal sample and it is needed to calibrate the system in order to find these values. It must be used 8 coal sets so as to find these parameters. Table 5.10 show the content of these 8 sets.

Table 5.10 : Content of these 8 sets

	Set1	Set2	Set3	Set4	Set5	Set6	Set7	Set8
(%)Total moisture	28	33	33	35	36	37	37	37
(%) Ash	32	21	19	13	12	12	11	11
(%) Sulphur	1	1	2	2	1	1	1	1
(%) Volatile matter	21	24	24	26	26	26	26	26
(%) Fix carbon	16	20	21	24	24	24	24	24
(%) Carbon	27	33	34	37	37	37	38	38
Calorie (kcal/kg)	2391	2976	3107	3400	3410	3421	3431	3437

The system was calibrated by using equation 5.18 and 5.19 with values appearing in Table 5.10.

All of the samples were radiated with a duration of 10 minutes. Each of the samples were adjusted that they weigh 24kg. X matrix was found for each of the sets individually after writing a code in MATLAB. Additionally, at this stage parameter library is created by taking into account table 5.11 values. For this purpose A matrix was calculated once again. Figure 5.49 shows the parametric library of coal after all calculations are done. There was not found any study related with parametric library of coal in literature, therefore this calculation process has unique importance.

Table 5.11 shows the X matrix values after A matrix calculation.

These values refer to the ratios through set1 to set8 which are assumed as unknown samples. This calculation is in the form of verification and it is observed that these values are so close to the values written in Table 5.9. Table 5.12 shows the deviation values in terms of percentages.

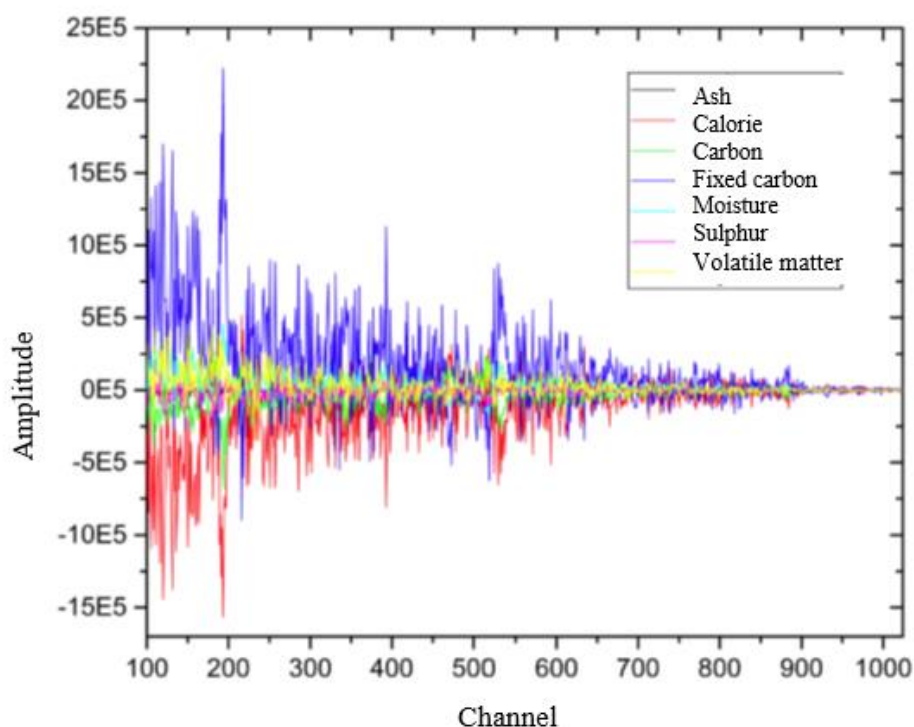


Figure 5. 50 : Parametric library of coal

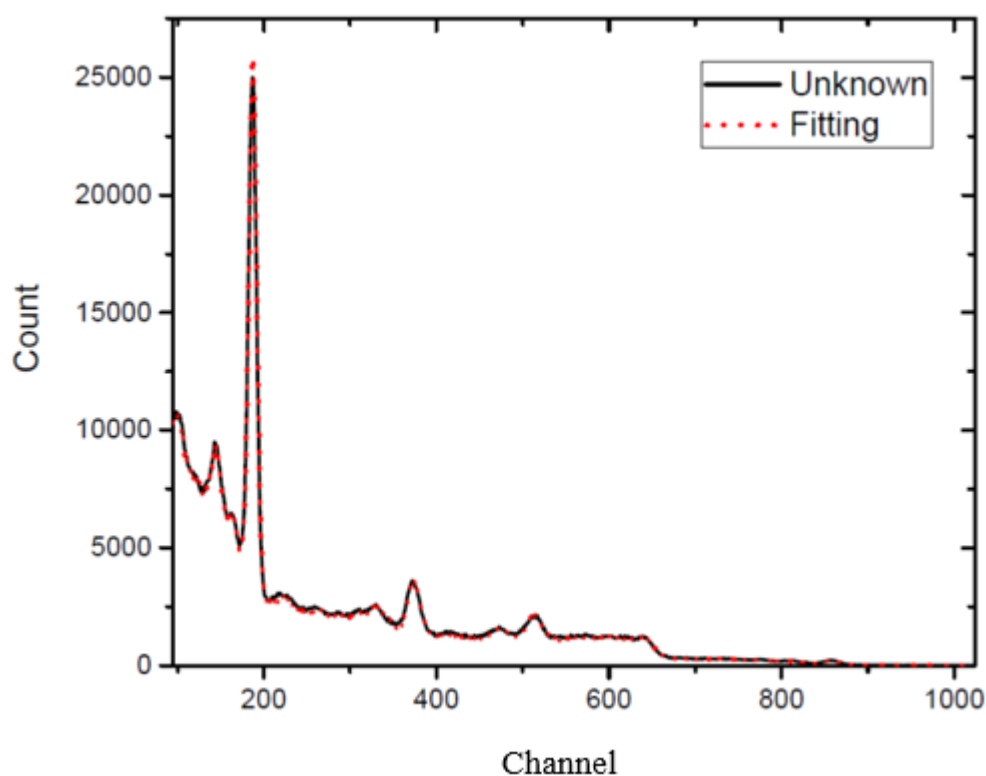
Table 5. 11 : The X matrix values after A matrix calculation

MATLAB calculated	Set1	Set2	Set3	Set4	Set5	Set6	Set7	Set8
(%) Moisture	28	32	34	35	36	36	36	38
(%) Ash	33	22	17	12	14	12	13	10
(%) Sulphur	1	1	2	2	1	1	1	1
(%) Volatile Matter	21	24	25	26	26	25	25	26
(%) Fixed Carbon	16	20	22	24	23	23	23	25
(%) Carbon	27	32	35	37	37	37	37	39
Calorie (kcal/kg)	2382	2921	3216	3422	3367	3365	3343	3555

In Figure 5.50, there is a fitting procedure between an unknown sample total gamma ray spectrum and parametric library. It can be observed that they are in good agreement with each other.

Table 5. 12 : The deviation values in terms of percentages

(%) Deviation	Set1	Set2	Set3	Set4	Set5	Set6	Set7	Set8
Total moisture	0.27	0.92	2.04	0.61	0.61	0.67	1.86	1.842
Ash	0.09	6.15	10.61	3.15	9.63	0.68	12.3	13.52
Sulphur	3.85	1.31	4.61	9.58	13.8	8.62	2.31	12.67
Volatile matter	0.36	0.93	2.09	0.38	0.41	1.59	1.62	2.17
Fixed C	0.43	2.22	4.06	0.99	1.93	1.97	2.83	3.76
C	0.47	1.57	3.06	0.29	0.84	1.45	2.30	3.14
Calorie(kcal/kg)	0.35	1.85	3.50	0.66	1.25	1.62	2.56	3.44

**Figure 5. 51 :** Fitting procedure between an unknown sample total gamma ray spectrum and parametric library

5.7.4 Calculation of element mass

In this part of the calculation, PFTNA technique is used for determination of elemental concentration as well as obtaining an empirical mass formula for elements. [33] Obtained spectra are separated by I and II numbers. Spectrum I refers to the summation of capture, inelastic and activation, spectrum II refers to capture and activation only.

In order to calculate the calorific value of samples, carbon oxygen, lead and hydrogen elements are necessary. Characteristic photo peak energies and energy window for each element in separate spectra are shown in Table 5.13.

Table 5. 13 : Characteristic photo peak energies and energy window for each element in separate spectra [33]

Element	Characteristic peak energy (MeV)	Identification	Energy window of characteristic peak (MeV)
C in spectrum I	4.43	N_{C_1}	4.20 – 4.67
O in spectrum I	6.13	N_{O_1}	5.89 – 6.36
Pb in spectrum I	7.36	N_{Pb}	6.78 – 7.59
C in spectrum II	4.44	N_{C_2}	4.20 – 4.67
O in spectrum II	6.13	N_{O_2}	5.89 – 6.36
H in spectrum II	2.22	N_H	2.00 – 2.47

Instability coming from the electrical circuit to the neutron generator causes neutron flux to be distorted. In order to normalize the neutron flux I_{All} and I_{Cap} are represented for spectrum I and spectrum II without subtracting the background from the total counts. After the measurements, multiple linear regression (MLR) was applied to find elemental mass formulation empirically. [33]

In this chapter it is needed coal sets that have known elemental concentrations. To create empirical mass formula for each element, it is a must to know mass of elements in the sample. After the formulation is done, empirical equations are used for different samples that are approximately the same weight with calibration samples. It was used 2.2kg synthetic coal sets for this goal. In Table 5.14, their concentrations are expressed for each element.

Table 5. 14 : Concentrations are expressed for each element in sample

% With water	Set1	Set2	Set3	Set4	Set5	Set6	Set7	Set8
C	81	75	69	63	56	50	44	38
Si	1	2	3	4	5	6	8	9
Fe	0	1	2	2	3	4	5	5
Al	0.5	1	1	2	2	3	4	4
Mg	0.1	0.3	0.5	0.6	0.8	1	1	1
Ca	0.7	1	2	3	3	4	5	6
S	3	3	3	3	3	3	3	3
H	1	1	1	1	1	1	1	1
O	10	13	15	18	21	23	26	28

These samples were used for creating linear equations to find the empirical mass formulation.

➔ Hydrogen mass formulation:

$$M_{H_2O} = a_1 \frac{N_H}{I_{cap}} + a_2 \frac{(N_{O_1} - N_{O_2})}{(I_{all} - I_{cap})} \quad (5.21)$$

Equation 5.21 is written for each sets of samples and the coefficients a_1 and a_2 are calculated by using matrix inversion process. Matrix representation of linear set of equations are expressed as:

$$\begin{pmatrix} M_{H_2O1} \\ M_{H_2O2} \\ \dots \\ \dots \\ \dots \\ \dots \\ \dots \\ M_{H_2O8} \end{pmatrix} = \begin{pmatrix} \text{ratio11} & \text{ratio21} \\ \text{ratio12} & \text{ratio22} \\ \text{ratio13} & \text{ratio23} \\ \dots & \dots \\ \dots & \dots \\ \dots & \dots \\ \dots & \dots \\ \text{ratio18} & \text{ratio28} \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} \quad (5.22)$$

$$\text{ratio1i} = \frac{N_H}{I_{cap}} \text{ for } i - \text{th set}$$

$$\text{ratio2i} = \frac{(N_{O_1} - N_{O_2})}{(I_{all} - I_{cap})} \text{ for } i - \text{th set}$$

According to this linear equations, it was found a_1 and a_2 coefficients.

➔ Carbon mass formulation:

$$M_C = a_3 \frac{(N_{C_1} - N_{C_2})}{(I_{all} - I_{cap})} + a_4 \frac{(N_{O_1} - N_{O_2})}{(I_{all} - I_{cap})} + a_5 \frac{N_{Pb}}{(I_{all} - I_{cap})} + a_6 \quad (5.23)$$

The same procedure is conducted for carbon and the matrix calculation is expressed as:

$$\begin{pmatrix} M_{C1} \\ M_{C2} \\ \dots \\ \dots \\ \dots \\ \dots \\ \dots \\ M_{C8} \end{pmatrix} = \begin{pmatrix} \text{ratio31} & \text{ratio21} & \text{ratio41} & 1 \\ \text{ratio32} & \text{ratio22} & \text{ratio42} & 1 \\ \text{ratio33} & \text{ratio23} & \text{ratio43} & 1 \\ \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & 1 \\ \text{ratio38} & \text{ratio28} & \text{ratio48} & 1 \end{pmatrix} \begin{pmatrix} a_3 \\ a_4 \\ a_5 \\ a_6 \end{pmatrix} \quad (5.24)$$

$$\text{ratio3i} = \frac{(N_{C1} - N_{C2})}{(I_{all} - I_{cap})} \text{ for } i - \text{th set}$$

$$\text{ratio4i} = \frac{N_{Pb}}{(I_{all} - I_{cap})} \text{ for } i - \text{th set}$$

According to these equations coefficients a_3 , a_4 , a_5 and a_6 are calculated.

➔ Oxygen mass formulation:

$$M_{O_2} = a_7 \frac{(N_{O1} - N_{O2})}{(I_{all} - I_{cap})} + a_8 \frac{N_{Pb}}{(I_{all} - I_{cap})} + a_9 \quad (5.25)$$

$$\begin{pmatrix} M_{O_21} \\ M_{O_22} \\ \dots \\ \dots \\ \dots \\ \dots \\ \dots \\ M_{O_28} \end{pmatrix} = \begin{pmatrix} \text{ratio21} & \text{ratio41} & 1 \\ \text{ratio22} & \text{ratio42} & 1 \\ \text{ratio23} & \text{ratio43} & 1 \\ \dots & \dots & 1 \\ \dots & \dots & 1 \\ \dots & \dots & 1 \\ \dots & \dots & 1 \\ \text{ratio28} & \text{ratio48} & 1 \end{pmatrix} \begin{pmatrix} a_7 \\ a_8 \\ a_9 \end{pmatrix} \quad (5.26)$$

According to these linear set of equations a_7 , a_8 and a_9 are calculated.

➔ Ash mass formulation:

$$M_{ash} = a_{10} \frac{(N_{Si1} - N_{Si2})}{(I_{all} - I_{cap})} + a_{11} \frac{(N_{O1} - N_{O2})}{(I_{all} - I_{cap})} + a_{12} \frac{(N_{Ca1} - N_{Ca2})}{(I_{all} - I_{cap})} + a_{13} \frac{N_{Pb}}{(I_{all} - I_{cap})} + a_{14} \quad (5.27)$$

$$\begin{pmatrix} M_{ash} \\ M_{ash} \\ \dots \\ \dots \\ \dots \\ \dots \\ \dots \\ M_{ash} \end{pmatrix} = \begin{pmatrix} \text{ratio51} & \text{ratio21} & \text{ratio61} & \text{ratio41} & 1 \\ \text{ratio52} & \text{ratio22} & \text{ratio62} & \text{ratio42} & 1 \\ \text{ratio53} & \text{ratio23} & \text{ratio63} & \text{ratio43} & 1 \\ \dots & \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & \dots & 1 \\ \dots & \dots & \dots & \dots & 1 \\ \text{ratio58} & \text{ratio28} & \text{ratio68} & \text{ratio48} & 1 \end{pmatrix} \begin{pmatrix} a_{10} \\ a_{11} \\ a_{12} \\ a_{13} \\ a_{14} \end{pmatrix} \quad (5.28)$$

$$\text{ratio5}_i = \frac{(N_{Si_1} - N_{Si_2})}{(I_{all} - I_{cap})} \text{ for } i - \text{th set}$$

$$\text{ratio6}_i = \frac{(N_{Ca_1} - N_{Ca_2})}{(I_{all} - I_{cap})} \text{ for } i - \text{th set}$$

After calculations, a_{10} , a_{11} , a_{12} , a_{13} and a_{14} coefficients are found. They are calculated with the aid of MLR method. The results belonging to ash, carbon, oxygen and water molecule are represented as an output of MLR in terms of regression statistics.

Table 5. 15 : Output of Ash

Summary output	
ASH	
Regression Statistics	
Multiple R	0.88
R Square	0.78
Adjusted R Square	0.49
Standard error	172.85
Observation	8

Table 5. 16 : Output of Carbon

Summary output	
CARBON	
Regression Statistics	
Multiple R	0.90
R Square	0.82
Adjusted R Square	0.62
Standard error	183.17
Observation	8

Table 5. 17 : Output of Water

Summary output	
H₂O	
Regression Statistics	
Multiple R	0.91
R Square	0.83
Adjusted R Square	0.76
Standard error	67.95
Observation	8

Table 5. 18 : Output of Oxygen

Summary output OXYGEN	
Regression Statistics	
Multiple R	0.87
R Square	0.75
Adjusted R Square	0.65
Standard error	172.84
Observation	8

According to these data sets, calculated and measured mass are compared. This is the verification process, obtained coefficients are substituted in the equations and known masses are calculated as if they are unknown values. Afterwards, they are compared as Table 5.19 indicates.

Table 5. 19: Compare of outputs

H2O (%)	C (%)	O2 (%)	Ash (%)
12.63787	0.747505	16.42442	29.94562
13.42221	2.717813	10.13933	30.7252
16.87966	0.159757	15.1441	8.431183
11.28213	7.776254	21.09069	24.23637
5.485235	14.20958	11.95118	12.53013
20.54309	3.07859	6.496901	13.07722
1.893426	6.153346	4.354894	3.058555
4.323858	26.33042	19.94167	17.5173

Table 5.19 shows the difference between measured mass and calculated mass. It is expressed in terms of percentages. This calculation mechanism can be further improved by adding other kinds of elements to ash calculation or prepared set mass amount is not sufficient for this type of formulation.

6. CONCLUSION

In this work elemental library and the concentration of elements appearing in coal are calculated by using experimental and mathematical methods. This work has a great importance since in coal industry the quality of the materials are examined and then they are classified according to the features of them. The moisture, calorific value, ash and sulphur content are determined by using nuclear techniques. Pulsed fast thermal neutron activation analysis (PFTNA) is a common method for this purpose. This technique gives accurate results even in a short period of time. Another outstanding feature of this study is storing the gamma ray data in list mode. In this way dead time is minimized and two separate spectra are examined accurately (Thermal neutron and fast neutron induced gamma rays). This offline analysis allow managing the data in a desired fashion. The data coming to the detector carries its information with 32-bit word and this word contains information about amplitude and time. In which channel gamma rays are collected at which time can be obtained by selecting desired time interval in 32-bit word. By separating two spectra as fast and thermal neutron induced gamma rays, elements present in the coal sample are identified more clearly.

Although list mode technique enhances the element identification, there are still ongoing problems in spectrum analysis. They can be caused by environmental conditions or/and data acquisition system. If so, it is observed a background information making the spectrum placed in a higher level. This baseline data is calculated by ROOT algorithm and then it is used in order to create element library. As an addition to this method, deconvolution algorithm with ROOT programming kit is utilized. It helps to identify the element peaks by separating the multiplets.

On the other hand, obtained elemental library is considered that they are in a linear summation relation and they form coal spectrum. After some verification procedures, it is obtained satisfactory results.

All in all, both experimental and mathematical methods are used for the determination of coal parameters such as sulphur content, calorific value, ash content

and moisture content. It is a powerful technique since it is utilized both experimental data and mathematical tools. As a result of the study, bulk coal sample quality which is an important factor for the coal classification is determined in a more accurate way.



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APENDICES

C++ code for decoding gamma ray spectrum obtained in list mode file format.

```
#include <iostream>
#include <fstream>
#include <cstdlib>
#include <iostream>
#include <assert.h>
#include <bitset>

using namespace std;
unsigned int Get_Data4Bytes();

ifstream inFILE("Si_60uA_80kV_20dk.Lis", ios::in | ios::binary);
int main()
{
    int data,PH;
    int SPEC_fast[1023];
    int SPEC_thermal[1023];
    int total_neutron[1023];

    for (int i=0;i<1024;i++) {
        SPEC_thermal[i] = 0;
        SPEC_fast[i] = 0;
        total_neutron[i] = 0;
    }
    inFILE.seekg(256,ios::beg); //Skip header
    while(!inFILE.eof()){

        data = Get_Data4Bytes();

        if ((data&0xE0000000) == 0xE0000000) {
            //fast neutron
            PH = (data&0x1FFE0000)>>17;
            SPEC_fast[PH]++;
        }
        if ((data&0xE0000000) == 0xC0000000) {
            //thermal neutron
            PH = (data&0x1FFE0000)>>17;
            SPEC_thermal[PH]++;
        }
    }
}
```

```

    total_neutron[1023] = 0;
    SPEC_fast[1023] = 0;
    SPEC_thermal[1023] = 0;
for(int i=0;i<1024;i++) {
    total_neutron[i] = SPEC_fast[i] + SPEC_thermal[i];
}
// Total neutron spectrum is written to the disk file
FILE * fpt;
fpt = fopen("Si-total.dat","w+");
for(int i=0;i<1024;i++) {
    fprintf(fpt,"%d\n",total_neutron[i]);
}
//Fast neutron spectrum is written to the disk file
FILE * fpf;
fpf = fopen("X-fast.dat","w+");
for(int i=0;i<1024;i++) {
    fprintf(fpf,"%d\n",SPEC_fast[i]);
}
//Thermal neutron spectrum is written to the disk file
FILE * fpth;
fpth = fopen("X-thermal.dat","w+");
for(int i=0;i<1024;i++) {
    fprintf(fpth,"%d\n",SPEC_thermal[i]);
}

//system("pause");
//return 0;
}

```

```

unsigned int Get_Data4Bytes(){
char ch;
int onebyte;
signed int data =0x00000000;
for(int i=0;i<4;i++){
    inFILE.get(ch);
    onebyte = ((int)ch )& 255;
    data = data | onebyte << 8*i;
}
return data;
}

```

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