The Pennsylvania State University

The Graduate School

Department of Mechanical and Nuclear Engineering

SPECIFICATIONS AND PERFORMANCE OF THE
COMPTON SUPPRESSION SPECTROMETER
AT THE PENNSYLVANIA STATE UNIVERSITY

A Thesis in

Nuclear Engineering

by

Nesrin Özgan Çetiner

© 2008 Nesrin Özgan Çetiner

Submitted in Partial Fulfillment
of the Requirements
for the Degree of

Master of Science

May 2008
The thesis of Nesrin Özgan Çetiner was reviewed and approved* by the following:

Kenan Ünlü  
Professor of Department of Mechanical and Nuclear Engineering  
Thesis Advisor

Kostadin Ivanov  
Professor of Department of Mechanical and Nuclear Engineering

Jack S. Brenizer  
J. ‘Lee’ Everett Professor of Department of Mechanical and Nuclear Engineering  
Chair of Nuclear Engineering Program

*Signatures are on file in the Graduate School
ABSTRACT

A Compton Suppression Spectrometer (CSS) consists of a primary high-purity germanium (HPGe) detector and surrounding secondary detectors with appropriate data acquisition and processing electronics. The CSS reduces the noise content from Compton scattered photons to the gamma energy spectrum. The CSS operates in anticoincidence mode, in which events that take place within a predetermined time window result in elimination of the event from the spectrum.

A custom-designed Compton Suppression Spectrometer was purchased from Canberra for Penn State Radiation Science and Engineering Center (RSEC). A detailed listing of the physical specifications of the Penn State Compton Suppression Spectrometer is presented. Various performance measurements were performed; among them are peak height-to-Compton ratio \((P/C)\) for suppressed and unsuppressed spectra, suppression factor \((SF)\) and reduction factor \((RF)\). Peak height-to-Compton ratio \((P/C)\) is defined as the ratio of the peak count in a photopeak to the average count per channel in a relatively flat region in the corresponding Compton continuum that avoids the Compton edge. This ratio is calculated for both the suppressed and unsuppressed spectra. Suppression factor is defined as the ratio of the suppressed peak height-to-Compton ratio to that of unsuppressed.

Preliminary measurements showed that a peak height-to-Compton ratio of \((P/C)_{\text{sup}} = 1001\) is obtained with the \(^{137}\text{Cs}\) source in the Compton suppressed mode, and \((P/C)_{\text{uns}} = 100.4\). This corresponds to a suppression factor of \(SF \approx 9.97\). Performance measurements cover a series of peak-to-Compton ratio calculations of various sources, the corresponding suppression factor and reduction factor calculations.

Natural environmental background spectra were collected with a long acquisition period in both the unsuppressed mode and the Compton suppressed mode.

A standard reference material NIST SRM-1571 Orchard Leaves was irradiated and counted. Peak height-to-Compton ratio and the suppression factor were calculated.
Another standard reference material NIST SRM-1570 Spinach sample was irradiated, and INAA calculations were performed. Among the identified elements are $^{24}Na$, $^{80}Br$, $^{27}Mg$, $^{52}V$ and $^{42}K$.

An example application of the Compton Suppression Spectrometer is the dendrochemistry study. The dendrochemistry research involves comprehensive conventional neutron activation analysis of dated tree rings collected from various regions of the world. It was shown that the Compton Suppression Spectrometer can determine certain peaks that correspond to elements such as gold, calcium, zinc and iron that cannot be easily identified and analyzed with the conventional NAA. Concentrations of these elements from each year were calculated, from which their chronologic variations were obtained and plotted.

For accurate determination of elemental concentrations, cascade corrections must be taken into account in concentration calculations. Therefore, peak-to-total calibration schemes must be incorporated into the analyses for future research.
# TABLE OF CONTENTS

LIST OF TABLES.........................................................................................................................x  

ACKNOWLEDGEMENTS..............................................................................................................xi  

DEDICATION ..........................................................................................................................xii  

CHAPTER 1 INTRODUCTION .................................................................................................1  

1.1 Objective ............................................................................................................................2  

CHAPTER 2 BACKGROUND AND THEORY ............................................................................4  

2.1 Theory of Compton Scattering .........................................................................................5  
2.2 Theory of Compton Suppression .......................................................................................9  
2.2.1 Adverse Effects of Suppression .................................................................................11  

CHAPTER 3 EXPERIMENTAL ....................................................................................................13  

3.1 Penn State Compton Suppression Spectrometer ..............................................................13  
3.1.1 Specifications of the Penn State Compton Suppression Spectrometer ..................16  
3.1.2 High-Purity Germanium Detector (HPGe) .............................................................16  
3.1.3 NaI(Tl) Guard Detector .........................................................................................19  
3.1.4 Shielding .................................................................................................................19  
3.1.5 Data Acquisition Electronics .................................................................................19  
3.1.6 Genie 2000 Basic Spectroscopy Software .........................................................20  
3.2 Breazeale Nuclear Reactor ..............................................................................................25  
3.2.1 Fast Neutron Irradiator and Fast Flux Tube .........................................................28  
3.2.2 Dry Tube Irradiation Facility ...............................................................................29  
3.2.3 Pneumatic Transfer System (PTS) Irradiation Facility .......................................29  

CHAPTER 4 CALIBRATION OF THE COMPTON SUPPRESSION SPECTROMETER ...........31  

4.1 Energy Calibration ............................................................................................................31  
4.2 Peak-to-Total Calibration ..............................................................................................33  
4.3 Efficiency Calibration ......................................................................................................33  
4.3.1 True Coincidence (Cascade) Summing ..................................................................34  

CHAPTER 5 PERFORMANCE MEASUREMENTS ..................................................................38
5.1 Definitions of Various Performance Metrics for Compton Suppression Spectrometers .................................................................38
  5.1.1 Peak Height-to-Compton Ratio, P/C .........................................................39
  5.1.2 Peak Area-to-Compton Area Ratio, A(P/C) ............................................39
  5.1.3 Peak Area-to-Total Area Ratio, A(P/T) ..................................................40
  5.1.4 Suppression Factor, (SF) .......................................................................40
  5.1.5 Reduction Factor, (RF) ..........................................................................41
  5.1.6 Area Suppression Factor, A(SF) ..............................................................41
5.2 Suppressed and Unsuppressed Measurements of the Natural Background Radiation with the Penn State Compton Suppression Spectrometer ........................................................................................................42
5.3 Performance Measurements with 137Cs ..................................................................................45
5.4 Performance Measurements with Dendrochronologically-Dated Tree Ring Sample ..................................................................................................................48
5.5 NIST SRM-1571 Orchard Leaves Sample ...........................................................................49

CHAPTER 6 INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS WITH THE COMPTON SUPPRESSION SPECTROMETER .................................................................51
6.1 Determination of Isotopic and Elemental Concentration with INAA .................52
  6.1.1 Neutron Activation ..................................................................................52
  6.1.2 Decay Equations ...................................................................................57
  6.1.3 Counting ..................................................................................................57
6.2 Instrumental Neutron Activation Analysis (INAA) Equations .............................................58
6.3 Neutron Flux Measurements ..................................................................................61
6.4 Standard Reference Material Measurements ......................................................................63
  6.4.1 NIST SRM 4275C-96 Mixed Gamma Source ........................................64
  6.4.2 NIST SRM-1570 Spinach Sample ................................................................65

CHAPTER 7 APPLICATIONS OF THE PENN STATE COMPTON SUPPRESSION SPECTROMETER .................................................................................................69
7.1 Application to Dendrochemistry Studies ..............................................................................69
7.2 Chronology of Elemental Concentrations ............................................................................75

CHAPTER 8 SUMMARY AND CONCLUSIONS ........................................................................84
8.1 Summary of the Research .................................................................................................84
8.2 Conclusions ......................................................................................................................85
8.3 Future Work ......................................................................................................................85

BIBLIOGRAPHY ..................................................................................................................87
LIST OF FIGURES

Figure 2.1: Sketch of the Compton scattering event..........................................................5

Figure 2.2: Variation of scattered gamma-ray energy with scattering angle ..................7

Figure 2.3: Polar plot of the probability of scattering through Compton event into a unit solid angle at the scattering angle $\vartheta$ for various energies. The photon is assumed to be incident from left.................................................................8

Figure 2.4: Geometric layout of the primary and the secondary detectors. .................10

Figure 3.1: The picture of the Penn State Compton Suppression Spectrometer showing the HPGe detector, the shield cave, measurement electronics, data acquisition electronics and the spectrometry computer running Canberra Genie-2000.................................................................14

Figure 3.2: Interior picture of the Penn State Compton Suppression Spectrometer....15

Figure 3.3: Picture of the NIM bin that has the spectroscopy instruments.................16

Figure 3.4: Schematic drawing of the Penn State Compton Suppression Spectrometer .........................................................................................................................17

Figure 3.5: Block diagram of the Penn State Compton Suppression Spectrometer....20

Figure 3.6: The Penn State Breazeale Reactor (PSBR) is seen during a pulse experiment.........................................................................................................................26

Figure 3.7. Drawing of the fast neutron irradiation facility for soft error rate measurements experimental setup..........................................................28

Figure 3.8. Locations of the dry tube irradiation facilities in the reactor core...........29

Figure 3.9: A simplified sketch of the dry tube and the pneumatic tube with respect to a fuel and control rod –drawing courtesy of Danielle K. H. Schwarz. ................................................................................................................30

Figure 4.1. An example energy calibration curve obtained with the Genie 2000 Gamma Analysis Software.................................................................32

Figure 4.2. Efficiency calibration curve obtained with the NIST standard reference material SRM4275C-109 $^{154}Eu$, $^{155}Eu$ and $^{125}Sb$ mixed gamma source....35
Figure 4.3. An example gamma decay scheme showing cascade transitions ..........36

Figure 5.1: The suppressed and unsuppressed background spectra obtained with the Penn State Compton Suppression Spectrometer. The acquisition times are 24 hours for both spectra. .................................................................42

Figure 5.2: Radioactive decay scheme of $^{137}$Cs; I.T. stands for isomeric or internal transition.................................................................45

Figure 5.3. (Main plot) Suppressed and unsuppressed spectra obtained with the $^{137}$Cs source with the reduction factors defined for multiple regions of the Compton continuum; (Inset plot) x-ray peaks originating from $^{137m}$Ba at 31.9 keV and 36.3 keV. ........................................................................................................46

Figure 5.4. (Main) Unsuppressed and suppressed spectra of the CTUPOR3-819 wood sample from Porsuk region, Turkey dated 819 Gordion year. (Inset) While the gold peak at 411.80 keV is not distinguishable in the unsuppressed spectrum, it is identified in the suppressed spectrum obtained with the Penn State Compton Suppression Spectrometer............................................49

Figure 5.5. Suppressed and unsuppressed spectra of the NIST standard reference material SRM-1571 acquired with the Penn State Compton Suppression Spectrometer. ........................................................................................................50

Figure 6.1. Prompt-gamma and delayed-gamma emission processes following activation by a neutron..........................................................51

Figure 6.2: The decay scheme of $^{198}$Au. Data obtained from Nuclear Data Evaluation Lab, Korea Atomic Energy Research Institute [8]. .........................56

Figure 6.3: Illustration of a simple background subtraction........................................59

Figure 6.4. The unsuppressed and the suppressed spectra obtained with the NIST SRM 4275C-96 mixed gamma source: ........................................64

Figure 6.5. Spectra of the NIST SRM-1570 Spinach sample obtained after the irradiation with and without Compton suppression..........................67

Figure 7.1: Unsuppressed spectrum of the dendrochronologically-dated tree ring sample CTUPOR3-819 collected from Porsuk, Turkey dated relative Gordion year 819. .................................................................................................71

Figure 7.2. Suppressed spectrum of the same tree ring sample CTUPOR3-819.
The unsuppressed spectrum of the same sample was shown in Figure 7.1........72
Figure 7.3: Suppressed spectrum of the dendrochronologically-dated tree ring sample CTUPOR3-824 from relative Gordion year 819.................................74

Figure 7.4: Chronologic variation of isotopic concentration of $^{82}\text{Br}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode. .................................................................75

Figure 7.5: Gamma ray decay scheme for $^{82}\text{Br}$. It is evident that the isotope is a strong cascade emitter (Data obtained from Nuclear Data Evaluation Lab, Korea Atomic Energy Research Institute). ........................................76

Figure 7.6: Chronologic variation of isotopic concentration of $^{47}\text{Ca}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode. ........................................................................78

Figure 7.7: Chronologic variation of isotopic concentration of $^{24}\text{Na}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode. ........................................................................80

Figure 7.8: Chronologic variation of isotopic concentration of $^{198}\text{Au}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode. ........................................................................82
LIST OF TABLES

Table 3.1. Physical and Electrical Characteristics of the HPGe Detector at the PSU-CSS.................................18

Table 3.2: List of irradiation facilities at the Penn State Breazeale Reactor (PSBR) .....27

Table 5.1: List of isotopes identified in the natural background and their decay series........................................44

Table 5.2: Suppression factors (SF) reported in the literature calculated for $^{137}$Cs........47

Table 6.1. Isotopes of interest in the NIST SRM-1570 Spinach sample with important physical parameters required to perform irradiation and decay calculations.........................................................65

Table 6.2. The irradiation parameters and the counting information for the NIST SRM-1570 Spinach sample.............................66

Table 6.3: Isotopic and elemental concentrations for the isotopes present in the SRM-1570 Spinach sample calculated by activation analysis.........................68

Table 7.1: Typical irradiation parameters for the Porsuk wood samples CTUPOR-xxx'.................................................................................................70

Table 7.2. Calculated $^{82}$Br concentrations and associated errors..................................77

Table 7.3. Calculated $^{47}$Ca concentrations and associated errors.................................79

Table 7.4. Calculated $^{24}$Na concentrations and associated errors.................................81

Table 7.5: Calculated $^{198}$Au concentrations and associated errors.................................83
ACKNOWLEDGEMENTS

First and foremost I would like to thank my spouse for his infinite support and love. His vision and knowledge always helped me get motivated and succeed in this field. He taught me how to be patient and organized to be a professional.

I would like to thank my adviser, Prof. Kenan Ünlü for his guidance and encouragement. He was very helpful and supportive during the completion of this research and through all my graduate studies. Prof. Jack Brenizer and Prof. Kostadin Ivanov provided support and feedback by reviewing the manuscript.

I would like to thank my mom and dad for giving their time and love for my comfort. They were always there to help me in every step of my life and I know that they will also be. Also I would like to thank my one and only sister Neslihan for her sincere support and for her glowing smile.

I would like to add my appreciation for the assistance of the following individuals from Radiation Science and Engineering Center in my experiments and my research: Danielle K. H. Schwarz, Thierry H. Daubenspeck, Mac Bryan, Ron Eaken and many others.
DEDICATION

To my purest new breath...

Balam,

Sarp Tarık Çetiner;

and To my endless love, Sacitim
CHAPTER 1

INTRODUCTION

Gamma rays interact with matter through three major interaction mechanisms: photoelectric absorption, Compton scattering and pair production. In all these interactions, the energy of the gamma ray photon is partially or completely transferred to an electron. In Compton scattering the incident gamma ray photon interacts with a valance electron and is deflected through an angle with respect to its original path. This interaction results in an emission of a gamma ray photon, which can then continue through the absorber and interact again or escape the absorber material.

The vast majority of scattered photons in Compton scattering escape the detector, hence the energy of the incident photon cannot be fully imparted to the detector. The partially deposited energy is registered as background in the gamma energy spectrum known as the Compton continuum. This continuum extends up to an energy that corresponds to the maximum transferrable energy known as the Compton edge.

Compton suppression is a commonly employed technique to reduce the contribution of Compton scattered photons to the gamma energy spectrum. Compton suppression technique relies on anticoincidence gating to eliminate gamma ray photons that are detected in more than one detector. A primary detector is used to acquire the actual spectrum of the source. The primary detector is surrounded by secondary detectors to catch the gamma ray photons that escape the primary detector after having undergone a Compton scattering. The secondary detectors are configured in a way to maximize the probability of detection of the escaped gamma ray photon.

The primary and the secondary detectors are operated in anticoincidence that eliminates a count event that takes place in the primary detector in case a gamma ray
photon is detected in the primary detector and any of the secondary detectors within a preset time window. The combination of the primary detector, surrounding secondary detectors and the measurement electronics is called the Compton Suppression Spectrometer (CSS).

The primary detector of the Penn State Compton Suppression Spectrometer consists of a central high-purity germanium (HPGe) detector surrounded by an assembly of NaI(Tl) scintillation detectors in annulus geometry as the secondary detectors. NaI(Tl) crystals have superior light yield and provide good timing resolution performance. Since gamma ray photons have a probability of interaction with every material, the amount of mass between the HPGe crystal and that of the NaI(Tl) scintillation crystal must be minimized to improve the performance of the Compton Suppression Spectrometer.

Compton Suppression Spectrometers improve the spectral data by reducing the Compton continua in the energy spectrum providing better identification of the peaks that otherwise would be buried in the Compton background. Compton Suppression Spectrometers also reduce the noise content in the energy spectrum caused by cosmic rays.

1.1 Objective

Compton Suppression Spectrometers offer unique capabilities that provide improved spectral quality at low count rates. Even for a commercial-grade system, performance characteristics of a Compton Suppression Spectrometer may vary considerably. The primary reason for discrepancy is possible variations in response characteristics of detector components. It is possible to say that each Compton Suppression Spectrometer has performance features that are unique to that particular system. Therefore, it is important to obtain a comprehensive collection of the performance specifications for the spectrometer.
The primary objective of this study is to introduce the components of the Penn State Compton Suppression Spectrometer and its characteristics and performance. An example application of the spectrometer will also be presented.

Chapter 2 gives brief background information on the theory of Compton scattering and the operation principles of the Compton Suppression Spectrometer. Chapter 3 gives information on the physical characteristics of the Compton Suppression Spectrometer at the Pennsylvania State University. The energy, peak-to-total and efficiency calibration procedures are introduced in Chapter 4, followed by a series of performance measurements given in Chapter 5. Chapter 6 presents a series of instrumental neutron activation analysis (INAA) measurements performed with the Compton Suppression Spectrometer. Chapter 7 introduces an application of the spectrometer in dendrochemistry study.

It is intended that this thesis serves as a reference material for future studies on the Compton Suppression Spectrometer at the Pennsylvania State University.
CHAPTER 2

BACKGROUND AND THEORY

Gamma ray quanta interact with matter through three mechanisms: (1) photoelectric absorption, (2) Compton scattering, and (3) pair production.

In the photoelectric absorption, a photon interacts with the atom as a whole and not with individual electrons. Upon interaction, the atom releases a photoelectron most likely from the most tightly bound shell, K shell, of the atom. The first mathematical description of the photoelectric process appeared in Einstein’s famous paper released in 1905 with the title “On a Heuristic Viewpoint Concerning the Production and Transformation of Light” [1]. The article offered a simple description of light quanta, or photons. The energy of the emitted electron is equal to the energy of the incident gamma ray photon less the binding energy of electron, given by the equation

\[ E_e = h\nu - E_b \]  

(2.1)

where \( E_e \) is the energy of the photoelectron, \( h\nu \) is the energy of the incident photon, and \( E_b \) is the binding energy of the electron in its original shell. Einstein’s explanation on the absorption of the gamma quanta and the subsequent release of electrons shed light on the phenomenon and the characteristic frequency, which won him the Nobel Prize in Physics in 1921 [2].

In Compton scattering, the incident photon interacts with an atomic orbital electron. Unlike the photoelectric absorption, the photon imparts only a fraction of its energy to the electron, and is deflected. A more comprehensive explanation of the Compton scattering process will be given in Section 2.1.

Pair production is creation of nuclear particles from a gamma quantum. The process takes place in the proximity of a strong Coulombic field of a nucleus. For the
creation of an electron-positron pair to be energetically possible, the gamma ray energy must exceed twice the rest mass energy of an electron, which is \( E_e = 0.511 \text{ MeV} \). The probability of pair production increases with the gamma ray energy above 1.022 \( \text{MeV} \), and is the predominant interaction mechanism for photon energies above 5 \( \text{MeV} \) [3].

This chapter will present further information on the theory of Compton scattering and the operation principles of the Compton Suppression Spectrometer (CSS).

### 2.1 Theory of Compton Scattering

Compton scattering takes place between an incident gamma-ray photon and an electron in the target material. In Compton scattering, the incoming gamma-ray photon is deflected through an angle \( \theta \) with respect to its angle of incidence. A portion of the photon energy is transferred to the electron, which is called the *recoil electron*. The energy transferred by the gamma-ray photon to the electron varies depending on the angle of scattering \( \theta \). A simplified sketch of the scattering process between the incident gamma-ray photon and the electron is shown in Figure 2.1.

![Figure 2.1: Sketch of the Compton scattering event.](image_url)
The deflected photon continues to travel in the medium until it completely transfers its energy through one of the mechanisms explained previously. The ejected electron loses its energy through cascade ionization.

The Compton scattering phenomenon was first explained by Arthur H. Compton. He discovered that x-ray photons scattered by thin foils underwent a wavelength shift [4]. The shift in wavelength of the scattered photon with respect to that of the incident photon was a function of the scattering angle $\theta$. He treated the x-ray as quanta according to the Einstein-Planck relation $E_\gamma = h\nu$, and the collision between the photon and the electron similar to billiard-ball collisions as illustrated in Figure 2.1. Compton derived the equation to describe the wavelength shift between the incident and scattered photon, and the scattering angle as

$$\lambda' - \lambda = \frac{h}{m_0c^2} (1 - \cos \theta) \quad (2.2)$$

where $\lambda$ and $\lambda'$ are the wavelengths of the incident and deflected photons, $h$ is Planck’s constant, $m_0$ is the rest mass of the electron, $c$ is the speed of light in vacuum, and $\theta$ is the scattering angle of the photon relative to its initial direction. The expression between the initial and final energy of the photon can be found by substituting $\lambda = \frac{hc}{E_\gamma}$ into Equation (2.2) as

$$E'_\gamma = \frac{E_\gamma}{1 + \frac{E_\gamma}{m_0c^2} (1 - \cos \theta)} \quad (2.3)$$

where $E'_\gamma$ and $E_\gamma$ are the final and initial energies of the photon, $m_0c^2$ is the rest mass energy of the electron, and $\theta$ is the scattering angle of the photon.

As shown by Compton, the angle of deflection of the photon and the energy imparted to the electron are correlated through Equation (2.3). The angle varies from just above $\theta = 0^\circ$ for low energies to a maximum of $\theta = 180^\circ$ for the highest Compton electron energy. Hence, Compton electrons are emitted with energies ranging between zero and the maximum energy, which is referred to as the Compton edge that corresponds
to photon scattering angle of $\theta = 180^\circ$. The variation of scattered gamma-ray photon energy with respect to the angle of scattering is shown in Figure 2.2.

![Figure 2.2: Variation of scattered gamma-ray energy with scattering angle.](image)

Compton edge can be easily found by letting $\theta = 180^\circ$ and substituting into Equation (2.3), which then becomes

$$E'_\gamma = \frac{E_\gamma}{1 + (E_\gamma/m_e c^2)(1 - \cos 180)} = \frac{E_\gamma}{1 + 2E_\gamma/m_e c^2}$$

(2.4)

As an example, the Compton edge of the gamma ray photons emitted from $^{137}\text{Cs}$ ($E_\gamma = 0.662\text{ MeV}$) is found as

$$E'_\gamma = \frac{0.662\text{ MeV}}{1 + \frac{2(0.662\text{ MeV})}{m_e c^2}} = 0.184\text{ MeV}$$

(2.5)
Spectra of Compton-scattered photon energies contain a peak known as the backscatter peak [3], which arises from Compton scattering into a gamma detector from the surrounding materials. The energy of the backscatter peak from the gamma-ray quantum is proportional to the incident photon energy for low energies, and asymptotically approaches a constant value of 0.25 MeV for higher energies.

The angular distribution of scattered gamma rays is predicted by the *Klein-Nishina formula* for the differential cross section $d\sigma/d\Omega$ [3]:

$$
\frac{d\sigma}{d\Omega} = Z\alpha r_0^2 \left( \frac{1}{1 + \alpha(1 - \cos \theta)} \right)^2 \left( \frac{1 + \cos^2 \theta}{2} \right) \left( 1 + \frac{\alpha^2(1 - \cos \theta)^2}{(1 + \cos^2 \theta)[1 + \alpha(1 - \cos \theta)]} \right) 
$$

(2.6)

where $\alpha \equiv h\nu/m_0c^2$ and $r_0$ is the classical electron radius. A polar plot of the Klein-Nishina formula is shown in Figure 2.3.

---

Figure 2.3: Polar plot of the probability of scattering through Compton event into a unit solid angle at the scattering angle $\theta$ for various energies. The photon is assumed to be incident from left.
The energy of the Compton electron $E_e$ can be expressed as

$$E_e = E_\gamma - E'_\gamma - E_b$$

(2.7)

where $E_b$ is the binding energy of the electron, which is relatively insignificant compared to the energy picked up by the electron. With the assumption $E_p \approx 0$, Eq. (2.7) becomes

$$E_e = E_\gamma - \frac{E_\gamma}{1 + (E_\gamma/m_e c^2)(1 - \cos \theta)} = \frac{E_\gamma^2}{m_e c^2 (1 - \cos \theta)}$$

(2.8)

The energy of the Compton electron scattered by an incident photon emitted by $^{137}$Cs ($E_\gamma = 0.662$ MeV) for photon scattering angle of $\theta = 180^\circ$ is found by Eq. (2.9):

$$E_e = 0.662 \text{ MeV} - \frac{0.662 \text{ MeV}}{1 + 2 (0.662 \text{ MeV} / 0.511 \text{ MeV})} = 0.478 \text{ MeV}$$

(2.9)

The probability of Compton scattering is a function of the number of electrons, therefore the Compton interaction probability increases linearly with $Z$.

### 2.2 Theory of Compton Suppression

The most probable channel of interaction for gamma rays with the germanium crystal is the Compton scattering for a wide range of energy of common interest. The effective energy range for Compton scattering of gamma rays in high-purity germanium crystal is $0.2 - 8.0$ MeV [6]. Reduction of Compton scattering in the spectrum improves the statistics of the spectrometer, and makes it possible that photopeaks that are buried in the Compton region of strong photopeaks can be analyzed with higher certainty.

Compton Suppression Spectrometers provide additional benefits besides the reduction of Compton noise: (1) excellent shielding capability for external radiation; and (2) reduction of the effect of the cosmic ray background.
The essential components of Compton Suppression Spectrometers are: (1) a primary gamma detector, (2) the secondary gamma detectors, and (3) an anticoincidence gate. The primary gamma detector is sought to have high resolution, whereas the secondary gamma detectors are expected to perform better in terms of detection efficiency. This configuration is quintessential since it provides the highest measurement resolution as well as highest suppression capability. Hence, semiconductor detectors such as lithium-drifted germanium Ge(Li), lithium-drifted silicon Si(Li) or high-purity germanium (HPGe) detectors are commonly used as the primary gamma ray detector to benefit from their excellent detection resolution. As the secondary detectors, usually thallium-drifted sodium iodide NaI(Tl) or bismuth germinate (BGO) detectors are used to benefit from their detection efficiency. The primary detector is located in the center of the detector network and is surrounded by the secondary detectors. The secondary detectors form an annulus around the active detection volume, and close off the top of the primary detector creating a geometric configuration similar to a Marinelli beaker. The secondary detectors are connected to multiple photomultiplier tubes. Figure 2.4 shows a simplified geometric layout of the primary, which is a high-purity germanium detector, and the secondary detectors, NaI(Tl) for the Compton Suppression Spectrometer (CSS) at Penn State University.

Figure 2.4: Geometric layout of the primary and the secondary detectors.
The signals from the primary and the secondary detectors are collected by respective preamplifier and amplifier units. The amplified and shaped signals are transferred to a coincidence unit that runs in anticoincidence mode. If a gamma ray is detected by the primary detector, this signal is also used as the start trigger on the coincidence unit, which effectively starts an internal stopwatch whose coincidence parameters are adjusted based on the geometry of the system. If the gamma ray deposits all of its energy into the crystal of the primary detector, it contributes to the full-energy peak. Consider that the gamma ray experiences a Compton scattering within the crystal of the primary detector and escapes the detector volume resulting in only a partial energy deposition. Depending on the angle of scattering, there is a high probability that the gamma ray will enter the active volume of the secondary detectors since they are close to a 2π-geometry. If the gamma ray undergoes any type of interaction in one of the secondary detectors, whether photoelectric event, Compton scattering or pair production resulting in generation of a signal of any amplitude, this signal is used as the stop trigger on the coincidence unit. Since the coincidence unit operates in anticoincidence mode, registration of both signals within the time window of the unit causes it to close the gate, hence not allowing the primary signal to reach the multichannel analyzer (MCA) and cannot contribute to the energy spectrum.

2.2.1 Adverse Effects of Suppression

Since the coincidence unit acts as a gate that opens by the start trigger for a certain period of time, detection of a particle by the secondary detectors will cause the gate to close resulting in the elimination of the primary signal. Although this scenario holds true for a Compton scattered photon, there is also a probability that the gamma rays that were detected by the primary and the secondary detectors are not the same gamma ray quantum as in the case of cascade emitters. An example of cascade emission process is shown in Figure 4.3. The presence of multiple coincident lines causes false
coincident signals in the gating unit, which results in elimination of these counts from the energy spectrum. The probability of false suppression is more pronounced at high count rates, especially when cascade emitter nuclides are involved in the measurement.
CHAPTER 3

EXPERIMENTAL

The Compton Suppression Spectrometer system consists of a high-purity germanium detector used as the primary detector, i.e. the central detector, NaI(Tl) detectors as the secondary detectors, i.e. the guard and the plug detectors, lead shield, measurement electronics, data acquisition device, a personal computer and the spectroscopy software Genie-2000 from Canberra Technologies, Inc.

Following sections will introduce the components of the Compton Suppression Spectrometer individually in detail.

3.1 Penn State Compton Suppression Spectrometer

A Compton Suppression System is used to reduce the contribution of scattered gamma rays that originate within the HPGe detector to the gamma ray spectrum. The HPGe detector is surrounded by an assembly of guard detectors, usually NaI(Tl). The HPGe and NaI(Tl) detectors are operated in anti-coincidence mode. The NaI(Tl) guard detector detects the photons that Compton scatter within, and subsequently escape from the HPGe detector. Since these photons are correlated with the partial energy deposition within the detector, much of the resulting Compton continuum can be subtracted from the spectrum reducing the unwanted background in gamma ray spectra. A commercially available Compton Suppression Spectrometer (CSS) was purchased from Canberra Industries and tested at the Radiation Science and Engineering Center at Penn State University. The PSU-CSS includes a reverse bias HPGe detector, four annulus NaI(Tl) detectors, a NaI(Tl) plug detector, detector shields,
data acquisition electronics, and a data processing computer with Genie-2000 Gamma Spectrum Analysis software. The HPGe detector is n-type with 54% efficiency. The guard detectors are on an annulus with 9-inch diameter and 9-inch height, plug detector that goes into/out of the annulus with the help of a special lift apparatus to raise/lower. The detector assembly is placed in a shielding cave. State-of-the-art electronics and software are used. Figure 3.1 shows a picture of the Penn State Compton Suppression Spectrometer with the measurement electronics.

Figure 3.1: The picture of the Penn State Compton Suppression Spectrometer showing the HPGe detector, the shield cave, measurement electronics, data acquisition electronics and the spectrometry computer running Canberra Genie-2000.
Four photomultiplier tubes that are connected to the NaI(Tl) crystals that form an annulus detection volume around the HPGe detector as well as the plug detector – also NaI(Tl) can be seen in Figure 3.2, which shows the interior of the spectrometer. The cylindrical enclosure has lead shields with copper lining.

Figure 3.2: Interior picture of the Penn State Compton Suppression Spectrometer.

The measurement electronics is shown in Figure 3.3. Two high-voltage power supplies are used to power the HPGe and NaI(Tl) detectors. The unit includes spectroscopy amplifiers and the coincidence unit. The output of the coincidence unit is sent to Canberra Multiport II multichannel analyzer (MCA).
3.1.1 Specifications of the Penn State Compton Suppression Spectrometer

Following sections introduce and give specifications of the components of the Penn State Compton Suppression Spectrometer.

3.1.2 High-Purity Germanium Detector (HPGe)

Germanium crystals of either p or n type can be grown at relatively larger sizes with very low impurity levels. The detectors fabricated from these crystals are called
intrinsic or high-purity detectors. A reverse electrode closed-end coaxial Ge (REGe) detector is used as central detector for PSU-CSS system. The outside of REGe is a \textit{p-type} while the inside is an \textit{n-type} electrode. This geometry is advantageous for radiation damage resistance since the holes trapped in germanium are collected by the p-type electrode.

The HPGe detector is combined with vertical slim line dipstick cryostat having 83-mm diameter end cap. Ultra-low background material was used for detector chamber, holder and internal hardware. A remote detector chamber is mounted on the cryostat with a neck of 10 cm in length. This provides a complete 360-degree shielding for the central detector crystal. The HPGe central detector crystal is 64 mm in diameter, and 71 mm in length. A diagram of the shield enclosure and the HPGe and NaI(Tl) detectors is given in Figure 3.4.

![Figure 3.4: Schematic drawing of the Penn State Compton Suppression Spectrometer.](image-url)
The HPGe detector used at PSU-CSS has a resolution of 2.2-keV FWHM at 1.33-MeV $^{60}\text{Co}$ energy line. The relative efficiency of the HPGe detector was measured to be 54% with a peak-to-Compton ratio of 58:1. Table 3.1 gives the physical and electrical characteristics of the HPGe detector used in the Penn State University Compton Suppression Spectrometer.

Table 3.1. Physical and Electrical Characteristics of the HPGe Detector at the PSU-CSS

<table>
<thead>
<tr>
<th>Physical Characteristics</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Geometry</td>
<td>Reverse electrode closed-end coaxial</td>
</tr>
<tr>
<td>Diameter</td>
<td>64 mm</td>
</tr>
<tr>
<td>Length</td>
<td>71 mm</td>
</tr>
<tr>
<td>Distance from Window</td>
<td>5 mm</td>
</tr>
<tr>
<td>Depletion Voltage</td>
<td>$-3000 , V$</td>
</tr>
<tr>
<td>Recommended Bias Voltage</td>
<td>$-4000 , V$</td>
</tr>
<tr>
<td>Capacitance</td>
<td>$\approx 45 , pF$</td>
</tr>
<tr>
<td>Peak-to-Compton Ratio</td>
<td>58:1</td>
</tr>
</tbody>
</table>

One major disadvantage of employing a well-type or annular with plug-type geometric configuration is that coincident gamma rays are subject to intense summation effect, which leads to significant deviation in the measured quantities. What is observed is reduction in individual peaks and increase in sum peaks. This requires that a sample specific efficiency calibration be done that employs a similar isotopic mixture in the calibrated standard.
3.1.3 NaI(Tl) Guard Detector

Suppression depends on the ability of the guard detector to detect the scattered photons leaving the central detector. The annulus detector system consists of four NaI(Tl) crystals with 23-cm diameter and 23-cm height. Each crystal is attached to a photomultiplier tube (PMT). A $76 \text{ mm} \times 76 \text{ mm}$ NaI(Tl) plug detector was added in order to enclose the upper section of the annulus to provide suppression of photons escaping upwards. Plug detector is supported by a guide tower allowing it to be raised for sample entry.

3.1.4 Shielding

A lead shield was used to reduce counts from external sources. The shield has 28-cm inner diameter and 61-cm height. The wall thickness of the shield is 10 cm. There is a cableway at top of the shield. Nitrogen purge port is also supplied on the lower side of the shield to flush the interior with $N_2$ gas from the Dewar. A nitrogen gas purge kit is used to provide positive pressure of nitrogen gas to purge natural sources of radioactivity. The 1-mm tin and 1.6-mm copper liner inside the shield helps prevent X-ray interferences. A lever actuated door with lifting mechanism helps open and firmly close the door on the top of the shield. A tall table with adjustable foot pads is used to hold the lead shield.

3.1.5 Data Acquisition Electronics

The block diagram of data acquisition electronics used for PSU-CSS is shown in Figure 3.5. A high voltage power supply is connected to four photomultiplier tubes (PMTs) on the annulus detectors and one PMT on the plug detector. A potential of $900 \text{ V}$ is applied to NaI(Tl) detectors. The output signal from PMTs is amplified and
shaped through preamplifier and amplifier. A bias of $4,000 \, V$ is applied to HPGe detector through the preamplifier. The signal from the preamplifier is fed into the amplifier. Both signals from the HPGe detector and the NaI(Tl) detectors are collected at the coincidence gate. The gate works in anti-coincidence mode where the signals produced at the detectors in coincidence cancel each other, leaving the non coincident signals to be counted. Signals are then analyzed by the multi-channel analyzer. The spectrum is displayed and data are collected with a personal computer.

![Block diagram of the Penn State Compton Suppression Spectrometer](image)

Figure 3.5: Block diagram of the Penn State Compton Suppression Spectrometer

### 3.1.6 Genie 2000 Basic Spectroscopy Software

Genie 2000 Basic Spectroscopy Software is a comprehensive environment for data acquisition, display and analysis on personal computers. It provides independent support for multiple detectors, networking capabilities, an intuitive graphical user interface and comprehensive scripting capabilities. Support for multiple detectors allows simultaneous detector sessions without interference.
Data acquisition and analysis capabilities are integrated within a single interface. Data analysis routines can be executed without affecting live detector sessions. Both data acquisition and data analysis can be carried out interactively or automatically using the scripting language capability. In the interactive mode, input provided by the user determines the evaluation of the data analysis. In the batch mode, scripts written the native Genie 2000 language control the process flow.

*Analysis Support*

The Genie 2000 Basic Spectroscopy Software includes a set of base spectroscopic analysis algorithms. Each analysis algorithm can be executed manually, as part of an analysis sequence or from within a batch script. These algorithms provide the ability to locate spectral peaks and calculate net peak areas.

Besides the aforementioned basic functions, a set of spectral manipulation algorithms are provided for spectral data smoothing and for stripping one spectrum from another.

*Library (Gamma-M) Peak Locate*

This algorithm provides library-driven peak search capability. The first step in the Gamma-M library locate phase is determination of the continuum background. This is done by an “erosion” process that effectively smooths the spectrum. At the end of this process, the resulting background spectrum is subtracted from the original spectrum to form a net spectrum.

Next, the algorithm builds a table of peak locations from the list of energies in the library. The system will then perform a linear least squares fit of the data, as well as a number of additional tests to determine if each peak in the table exists. If desired, the library peak locate can also be supplemented with an unknown peak search that will find peaks that are not in the library. Those peaks that are confirmed by this process are used to populate a second table that can be used for a later peak area analysis.
Library (Simple) Peak Locate

This peak locate algorithm will assign the peak locations based on the library energies as well. Unlike the Library (Gamma-M) algorithm, there is no erosion of the continuum, gain shift correction or MDA calculations to verify the existence of the peaks.

Peak Area Calculations

Peak area calculations use the results of the peak locate algorithms to compute net peak areas. Two algorithms are provided for area calculations: Sum/Non-Linear Least Squares Fit and Library (Gamma-M).

Sum/Non-Linear Least Squares Fit algorithm starts with the determination of peak region limits. Either the region of interest (ROI) limits specified by the user or automatically determined limits based on the results from peak locate algorithms.

Automatic ROI determination is followed by the determination whether the adjacent peaks are singlets or multiplets. Peak areas are calculated differently for singlet peaks and multiplet peaks. For singlets, the algorithm simply sums the gross counts in the peak ROI and subtracts the background continuum. For multiplets, the algorithm performs a non-linear least squares fit to the data, providing the best possible fit to the actual spectral data. The fit model uses both the peak width calibration and, optionally, a low tail shape calibration.

Calibration

Genie 2000 supports energy, width, low-tail and efficiency calibrations. Peak-to-total (P/T) calibration routine is also included as an initial step in the correction for cascade summing of coincident peaks in spectra.

The calibration process supports certificate files. Certificate files can be created with a graphical Certificate File Editor application. Certificate files store all the essential
information that comes with a calibration standard, such as nuclide, emission rates and uncertainties.

Calibration can also be performed in the batch mode or interactive mode. Efficiency calibration is carried out automatically. A peak search algorithm locates and quantifies peaks, then associates them with known (decay corrected) emission rates for each line to determine an efficiency curve and equation.

Interactive calibration plots are provided. The energy calibration curve may be a 1st, 2nd or 3rd order polynomial. The efficiency curves may be up to 9th order. Three efficiency calibration algorithms are available: dual polynomial, linear polynomial, empirical polynomial. It is also possible to choose an interpolated method for any of the calibration algorithms.

*Genie 2000 Gamma Analysis Software*

Genie 2000 Gamma Analysis Software package includes a set of advanced analysis algorithms for further processing of gamma spectra acquired with the Genie 2000 Basic Spectroscopy Software. These algorithms provide a complete analysis of gamma ray spectra obtained from any type of gamma detector and also are accessible from the Genie 2000 interactive environment. Analysis execution utilizes data resident in the Configuration Access Method (CAM) data structures. Results from each algorithm are also stored in CAM files, making the resultant data file a complete record of the entire analysis.

Genie 2000 features the following essential functions:

- Interactive Spectral Analysis allows visual inspection of a spectrum and modification by interactively adding, editing, and deleting peaks and peak regions. Calibrations and nuclide libraries can also be reviewed and updated.
- Efficiency correction involves calculating efficiency and associated uncertainty for each identified peaks.
• Nuclide Identification and Quantification algorithms take into account all energy lines of a nuclide entered into the analysis library with their proper branching ratios, as well as the half-life of the nuclide. For all identified nuclides, the algorithm calculates the value of the confidence index and a decay corrected activity per unit volume (or mass) for each energy with a matching peak in the spectrum.

• Library Correlation NID Peak Locate; analysis has the ability to locate minor peaks accurately, especially those hidden in multiplets, and correct calibration errors that might otherwise impair later nuclide identification analysis.

• Interference Correction and Weighted Mean Activity Calculations automatically finds the nuclides that have interferences, which is defined as two or more nuclides with at least one common energy. Energies for nuclides can be marked in the library for exclusion from weighted mean activity calculations.

• Parent/Daughter Decay Correction algorithm applies a correction to report proper nuclide activities when the parent nuclide and its daughters are not in equilibrium. Correction is based on sample time, acquisition start time, elapsed acquisition live time and nuclide parent/daughter information contained in the nuclide library.

• Background Subtraction and Reference Peak Correction: The background subtraction algorithm allows the subtraction of environmental background peaks from sample spectra. The purpose of a reference peak correction is to use a reference peak of a known count rate to normalize the areas of all other peaks in the spectrum.

• Minimum Detectable Activity (MDA) Calculations: Minimum detectable activity can be calculated for radionuclides. The MDA algorithms can
perform Currie MDA, KTA MDA and lower limit of detection (LLD) calculations.

- True Coincidence (Cascade) Summing Correction: This algorithm corrects cascade loss or gain of observable peak area as a function of nuclide decay scheme and geometry. Correction method uses LabSOCS technology to precisely describe the sample/detector.

3.2 Breazeale Nuclear Reactor

The Radiation Science and Engineering Center (RSEC) at the Pennsylvania State University houses the TRIGA Mark III research reactor known as the Breazeale Nuclear Reactor. TRIGA stands for Training, Research and Isotope Production built by General Atomic Company. The Penn State Breazeale Reactor (PSBR) core operates at a steady state power level of 1000 kW with pulsing capability up to 2000 MW in milliseconds. The peak flux for a maximum pulse is approximately $6 \times 10^{16} \text{ n cm}^{-2} \text{ sec}^{-1}$ with a pulse width of 15 milliseconds at half maximum. Penn State Breazeale Reactor core contains 3.4 kg of non-weapons form $^{235}\text{U}$, and operates at an approximate depth of 5.5 m in a pool of deionized water providing over 6.4 m of vertical shielding. The reactor core can be moved in the east-west as well as in the north-south direction. A picture of the Penn State Breazeale Reactor (PSBR) is shown in Figure 3.6 during a pulse.
Utilization of the PSBR can be described in four categories:

1. **Education**: laboratory classes for undergraduate, graduate students, and high school science groups. These classes are to bring in the experience of irradiation, sample analysis, non-destructive examinations of materials using neutrons or x-rays, transient behavior of the reactor, calibration of a reactor control rod for the undergraduate students in Radiation Detection and Measurement Laboratory and Nuclear Engineering Freshman Seminar. One of the educative activities is the workshop for Westinghouse Scholars. Also there are several tours set for public to inform them about the operation of the reactor and applications in Nuclear Science.

Figure 3.6: The Penn State Breazeale Reactor (PSBR) is seen during a pulse experiment.
2. **Research**: radionuclear applications, neutron dept profiling, neutron radiography, gamma irradiation, several research programs by faculty and graduate students throughout the University, and various applications by the industrial sector.

3. **Training**: programs for reactor operation staff.

4. **Service**: radionuclear applications, neutron transmission measurements, radioscopy, semiconductor irradiations, isotope production and other applications by industry.

The irradiation facilities are two beam ports, a pneumatic transfer system that is also called *rabbit*, and thermal and fast irradiation tubes. The beam ports are in use for the projects such as neutron depth profiling and neutron radiography. A list of irradiation facilities is given in Table 3.2 with their positions and measured neutron flux values. The irradiation facilities will be described in detail in the following sections.

Table 3.2: List of irradiation facilities at the Penn State Breazeale Reactor (PSBR)

<table>
<thead>
<tr>
<th>Irradiation Facilities</th>
<th>Neutron flux (n/cm²/s)</th>
<th>Position</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Dry Irradiation Tubes (DIT)</strong></td>
<td>1.8 x 10^13 thermal</td>
<td>E4 and E13 grid plate locations in the outermost core ring</td>
</tr>
<tr>
<td></td>
<td>8.1 x 10^11 epithermal</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6 x 10^12 fast</td>
<td></td>
</tr>
<tr>
<td><strong>Pneumatic Transfer System</strong></td>
<td>1.7 x 10^13 thermal</td>
<td>E6 grid plate location</td>
</tr>
<tr>
<td></td>
<td>9 x 10^11 epithermal</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6 x 10^12 fast</td>
<td></td>
</tr>
<tr>
<td><strong>Central Thimble</strong></td>
<td>3.3 x 10^13 thermal</td>
<td>H9 grid plate location</td>
</tr>
<tr>
<td></td>
<td>3 x 10^12 epithermal</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6 x 10^11 fast</td>
<td></td>
</tr>
<tr>
<td><strong>Fast flux tube</strong></td>
<td>3.8 x 10^11 thermal</td>
<td>Against the front core place for irradiations</td>
</tr>
<tr>
<td></td>
<td>3.7 x 10^11 epithermal</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6 x 10^12 fast</td>
<td></td>
</tr>
<tr>
<td><strong>Fast Neutron Irradiator</strong></td>
<td>3.8 x 10^11 thermal</td>
<td>Against the flat core face during irradiation</td>
</tr>
<tr>
<td></td>
<td>3.7 x 10^11 epithermal</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6 x 10^12 fast</td>
<td></td>
</tr>
</tbody>
</table>
3.2.1 Fast Neutron Irradiator and Fast Flux Tube

The Fast Neutron Irradiator (~23-cm diameter) and Fast Flux Tube (~15-cm diameter) use boron shielding to provide a hardened flux distribution. Thermal neutron irradiations utilize the so called 5 cm x 15 cm tube, the central thimble (~3-cm diameter) or the pneumatic transfer system (~3-cm diameter). The irradiation facility is shown in Figure 3.7.

---

Figure 3.7. Drawing of the fast neutron irradiation facility for soft error rate measurements experimental setup.
3.2.2 Dry Tube Irradiation Facility

Dry Irradiation Tubes (DIT) were designed and implemented specifically for the dendroanalysis project which is analyzing tree ring samples with Neutron Activation Analysis (NAA). DIT facilities consist of two closed bottom aluminum tubes which are placed in empty holes in the grid plate. The positions of the dry tubes correspond to grid plate locations E4 and E13 in the core as shown in Figure 3.8. One advantage of having the tubes semi-permanently installed in the grid plate is the freedom to perform an irradiation while the core is in any pool position.

---

![Figure 3.8. Locations of the dry tube irradiation facilities in the reactor core.](image)

3.2.3 Pneumatic Transfer System (PTS) Irradiation Facility

The pneumatic transfer system allows for samples to be transferred into the reactor core from a remote laboratory. The time between the release of the sample and arrival in the core is approximately 5 seconds. This relatively short transfer time is needed for analysis of short-lived isotopes. Samples are inserted into a pneumatic tube, which carries the sample from a radioisotope laboratory into the reactor core, where it
sits for a predetermined amount of time, and back to a fume hood in the laboratory. The position of the pneumatic transfer system is in grid plate location E-6, where thermal neutron flux is $1.7 \times 10^{13} \text{ cm}^{-2} \text{s}^{-1}$, epithermal neutron flux is $9 \times 10^{11} \text{ cm}^{-2} \text{s}^{-1}$ and fast neutron flux is $6 \times 10^{12} \text{ cm}^{-2} \text{s}^{-1}$ at 1000 kW of operation.

The dry tube and pneumatic transfer system irradiation facilities are shown in Figure 3.9.

---

Figure 3.9: A simplified sketch of the dry tube and the pneumatic tube with respect to a fuel and control rod – drawing courtesy of Danielle K. H. Schwarz.
CHAPTER 4

CALIBRATION OF THE COMPTON SUPPRESSION SPECTROMETER

In order to perform absolute quantification through instrumental neutron activation analysis (INAA) with any type of spectrometer, the spectroscopy system must be calibrated in terms of energy and efficiency. Energy calibration provides accurate peak identification while efficiency calibration makes it possible to perform concentration calculation of an isotope or element of interest. Besides energy and efficiency calibrations, Compton Suppression Spectrometers must be particularly calibrated for what is known as the peak-to-total calibration, which is defined as the ratio of the peak efficiency to total efficiency of the spectrum. Peak-to-total calibration provides accurate nuclide concentration calculation in a given sample, which otherwise might deviate from the actual value due to the presence of multiple cascade emitters in the sample of interest as well as the efficiency calibration sample.

In this chapter, energy, peak-to-total and efficiency calibration procedures will be briefly explained, and related references will be introduced.

4.1 Energy Calibration

Energy calibration is the technique that assigns each channel number with a unique energy value. Since the channel scale in a multichannel analyzer corresponds to the pulse-height scale at the output of the amplifier, the energy calibration is in fact a mapping methodology between the voltage height and the corresponding energy. In applications such as instrumental neutron activation analysis, a source of known gamma ray energies is used to obtain the calibration. With the a priori knowledge of a number of...
energies, unknown gamma ray peaks can be easily identified. Use of multiple calibration peaks is a common practice to eliminate undesired spectrometric effects such as nonlinearities.

A standard source that has at least two energy lines, each located at either end of the spectrum, is used to perform the energy calibration. A calibration curve is obtained from the correlation between the channel numbers and the corresponding energy values. Calibration curve provides a one-to-one mapping from the channel number of the multichannel analyzer to the energy, or vice versa. Commonly, a least square curve fitting is performed to obtain a calibration curve in the form of a polynomial as shown in Equation (4.1),

$$E_i = \sum_{n=0}^{N} a_n C_i^n$$  (4.1)

where $E_i$ is the energy value to the corresponding the $i$th channel number $C_i$. An example energy calibration curve created by Genie 2000 is shown in Figure 4.1.

---

**Figure 4.1.** An example energy calibration curve obtained with the Genie 2000 Gamma Analysis Software.
4.2 Peak-to-Total Calibration

Peak-to-total ratio, \( r \), is defined as the ratio of the peak efficiency to the total efficiency. Total efficiency is the total area under the entire spectrum, whereas the peak efficiency corresponds to the full energy deposition of the incident gamma rays. The definition of the peak-to-total calibration is given in Equation (4.2),

\[
r = \frac{\varepsilon_{\text{peak}}}{\varepsilon_{\text{total}}}
\]  

(4.2)

Single gamma emitting radio nuclides are good calibration sources for this purpose. The Genie-2000 software automatically calculates the peak-to-total ratios \( (P/T) \) at the main peak energies using chi-square minimization technique and fit it to a logarithmic function

\[
\ln \left( \frac{P}{T} \right) = a + b \ln E_\gamma + c (\ln E_\gamma)^2 + \frac{d}{E_\gamma^n}
\]  

(4.3)

where \( a, b, c \) and \( d \) parameters are obtained by chi-square minimization. Further information can be found in Genie 2000 Spectroscopy Software manual [7].

4.3 Efficiency Calibration

The intrinsic full-energy peak efficiency of semiconductor detectors is not constant for all gamma ray energies. In order to perform an absolute measurement of emission rates of gamma rays, the intrinsic detection efficiency of the detector as a function of the incident gamma-ray energy must be known.

Conventionally, the efficiency calibration is carried out using sources of known activity in a geometrically similar configuration as the actual measurement. This source is called calibration standard. Calibration standards are provided by National Institute of Standards and Technology (NIST) within a precision ranging from 0.5 to 2.0%. Any
errors in assumed detector dimensions will apply both to the calibration and actual measurement; hence they cancel out once the calibration is properly carried out. The calibration is done for a wide range of gamma-ray energies that covers the energy range of interest that the actual measurement will yield. This allows creating an empirical efficiency versus energy curve. One important point in achieving a valid and representative efficiency calibration is that the calibration standard and samples must be similar if not identical in size, geometry, density, spatial distribution of active material etc.

The most accurate way of calculating the efficiency of a detector is using a standard source of known initial activity that emits abundant gamma rays in a wide energy range. NIST certified europium-antimony mixed source SRM4275C-109 is used for efficiency calibration. Built-in functions of the Genie-2000 software is used to perform the efficiency calibration. An efficiency curve is obtained in the form as shown in Equation (4.4),

\[
\ln \epsilon = \sum_{i=1}^{N} a_i \left( \ln \frac{E}{E_0} \right)^{i-1}
\]

(4.4)

where \(E_0\) is a fixed reference energy and the values of \(a_i\) are the fitted parameters. A sample efficiency calibration curve obtained with SRM4275C-109 standard source is shown in Figure 4.2.

4.3.1 True Coincidence (Cascade) Summing

In most cases of a radioactive decay, a parent nuclide decays to an excited energy level of a daughter nuclide, which is subsequently followed by transition to ground state of the daughter nuclide by the emission of two or more gamma rays in a cascade.
Since the excited states have life times on the order of picoseconds, there is a probability that the successive emission of gamma rays is detected within the resolving time of the gamma ray spectrometer. This cascade detection is called true coincidence. The evident consequence of this detection scheme is the registration of the sum of the energy deposited by the cascade gammas from a nuclear decay. Events may be lost (summing-out) or gained (summing-in) from the full-energy peak of the gamma ray of interest. Activity determination based on the full-energy peak will be in error. Therefore, the spectrum must be corrected to take into account the true coincidence or cascade summing effects. An example decay scheme that involves cascade transitions is shown in Figure 4.3.

Figure 4.2. Efficiency calibration curve obtained with the NIST standard reference material SRM4275C-109$^{154}$Eu, $^{155}$Eu and $^{125}$Sb mixed gamma source.
As shown in Figure 4.3, de-excitation of the daughter nuclide $Z_X$ occurs via the emission of gamma rays $\gamma_1$ and $\gamma_2$, which are in cascade, or via the emission of gamma ray $\gamma_3$. If gamma rays $\gamma_1$ and $\gamma_2$ are detected in true coincidence, the full energy peak measurement of $\gamma_1$ and $\gamma_2$ suffers from cascade summing losses while the full energy peak measurement of $\gamma_3$ suffers from cascade summing gains. The cascade summing losses are not limited to the counts appearing in the sum peak. One of the cascade gammas, i.e. either $\gamma_1$ or $\gamma_2$ may deposit full energy while the other one may impart only a portion of its energy. The result is a continuum in the pulse height spectrum between the energy of $\gamma_1$ and the energy of the sum peak.

Cascade summing losses depend on the total efficiency of the detector for the gamma rays in the cascade. In the case of cascade summing gains, the two gamma rays $\gamma_1$ and $\gamma_2$ in the above example deposit their full energy in the detector. The resulting event appears at the same energy as that of $\gamma_3$. Cascade summing gains are dependent on the peak efficiencies of the detector at the gamma ray energies of interest. Besides the detection efficiencies, cascade summing losses or gains also depend on the gamma ray emission probabilities and any angular correlations involved in the gamma ray emission.
emission. It is important to note that the magnitude of cascade summing losses or gains is dependent on the counting geometry and not on the source activity. Cascade summing losses can be as high as 30 – 40% particularly for close-up sample geometries.
CHAPTER 5
PERFORMANCE MEASUREMENTS

Obtaining certain performance metrics of a Compton Suppression Spectrometer provides a rudimentary estimate of its response function under various circumstances. The performance measurements of the Compton Suppression Spectrometer at Penn State were performed with various samples, including a $^{137}$Cs source, an activated NIST SRM-1571 Orchard Leaves sample, and an activated dendrochronologically-dated tree ring sample. The peak height-to-Compton ratios for $^{137}$Cs source were found to be $(P/C)_{sup} = 1001$ for the Compton suppressed spectrum, and $(p/C)_{uns} = 1004$ for the unsuppressed spectrum. This gave a suppression factor of $SF = 9.97$. With the irradiated NIST SRM-1571 Orchard Leaves sample, the peak height-to-Compton ratios were calculated as $(P/C)_{sup} = 386.84$ for the Compton suppressed spectrum, and $(P/C)_{uns} = 101.77$ for the unsuppressed spectrum, from which the suppression factor was found as $SF = 3.80$. For the dendrochronologically dated tree sample CTUPOR3-819 from year 819, in which the gold concentration was significantly elevated, the gold peak was identified in the Compton suppressed spectrum using the Genie 2000 nuclide identification (NID) algorithm.

5.1 Definitions of Various Performance Metrics for Compton Suppression Spectrometers

There are numerous performance metrics defined to give an estimate of the response of a Compton Suppression Spectrometer. Although there are efforts to standardize certain metrics [10], the majority of the definitions are still limited to specific applications, hence their usage should be kept endemic only to the special case in the
definition. For instance, peak height-to-Compton ratio is typically calculated either for $^{137}$Cs or $^{60}$Co. Doing comparison between different performance evaluations is usually misleading. Therefore, care must be given to the type of evaluation when determining or comparing the performance of a system. The following sections introduce common definitions encountered in performance calculations used in Compton suppression spectroscopy.

### 5.1.1 Peak Height-to-Compton Ratio, P/C

The peak height-to-Compton ratio for a certain photopeak is defined as the ratio of the peak count to the average counts per channel in a relatively flat region in the corresponding Compton continuum that avoids the Compton edge. Let $C_p$ be the peak number of counts in the photopeak of interest, and $i$ and $j$ are the channel numbers at the beginning and at the end of the flat region in the associated continuum, and $C_x$ is the number of counts at the channel number $x$. The peak height-to-Compton ratio can be expressed by

$$\frac{C_p}{\frac{1}{j-i+1} \sum_{k=i}^{j} C_k}$$

The channels between 1040 keV and 1096 keV for $^{60}$Co, and 358 keV and 382 keV for $^{137}$Cs are the recommended regions for peak-to-Compton calculations [10].

### 5.1.2 Peak Area-to-Compton Area Ratio, A(P/C)

The peak area-to-Compton area ratio is defined as the ratio of the net photopeak area to the area of an equal spectral width in a relatively flat region in the associated
Compton continuum that avoids the Compton edge [11]. The quantity can be expressed mathematically by

\[
A(P/C) = \frac{\sum_{k=p_i}^{p_j} C_k - B}{\sum_{k=c_i}^{c_j} C_k}
\]  

(5.2)

where \(C_k\) is the number of counts in channel number \(k\), and indices \(p_i\) and \(p_j\) correspond to the channel numbers of the beginning and the end of the photopeak, \(c_i\) and \(c_j\) are the channel numbers of the beginning and the end of the flat region in the continuum, \(B\) is the area of the background under the photopeak. The flat region can be defined the same way as in the peak-height-to-Compton ratio.

5.1.3 Peak Area-to-Total Area Ratio, \(A(P/T)\)

The peak area-to-total area ratio is defined as the ratio of the net photopeak area to the total area in the spectrum including both the Compton continuum and the photopeak [11]. The mathematical expression for the quantity is

\[
A(P/T) = \frac{\sum_{k=p_i}^{p_j} C_k - B}{\sum_{k=1}^{M} C_k}
\]  

(5.3)

where \(M\) is the total number of channels of the multichannel analyzer (MCA).

5.1.4 Suppression Factor, (SF)

The suppression factor for a photopeak is defined as the ratio of the suppressed peak height-to-Compton ratio to that of unsuppressed peak height-to-Compton ratio [12], i.e.
5.1.5 Reduction Factor, (RF)

The reduction factor is used for nuclides emitting coincident gamma rays. It is defined as the ratio of average height of the unsuppressed Compton continuum to that of suppressed continuum defined in the same part of the spectrum [12], i.e.

\[
RF = \frac{(P/C)_{\text{uns}}}{(P/C)_{\text{sup}}}
\]

where \( i \) and \( j \) are the indices corresponding to the channel number of the beginning and the end of the region of interest.

5.1.6 Area Suppression Factor, A(SF)

Area suppression factor is defined as the ratio of the peak area-to-total area ratio in the suppressed spectrum to that of the unsuppressed spectrum, i.e.

\[
A(SF) = \frac{A(P/T)_{\text{sup}}}{A(P/T)_{\text{uns}}}
\]
5.2 Suppressed and Unsuppressed Measurements of the Natural Background Radiation with the Penn State Compton Suppression Spectrometer

In low-level counting practices, determination of the contribution of energy lines originating from the environmental background radiation to an acquired spectrum is crucial for performing a realistic identification of isotopic composition of the sample. Certain natural radioisotopes are hard to avoid even in high-end systems designed meticulously to minimize the background contamination in the collected energy spectrum. Suppressed and unsuppressed background spectra were collected for 24 hours. The acquired spectra are shown in Figure 5.1.

Figure 5.1: The suppressed and unsuppressed background spectra obtained with the Penn State Compton Suppression Spectrometer. The acquisition times are 24 hours for both spectra.
Among the noticeable peaks are $238.6$-keV $^{212}Pb$, $510.1$-keV $^{83}Se$, $583.2$-keV $^{208}Tl$, $727.3$-keV $^{212}Bi$ and $1460.8$-keV $^{40}K$, and especially $^{40}K$, $^{83}Se$ and $^{212}Pb$ stand out in the spectrum. For low-level sample analyses, background is one of the major limiting factors in accurate determination of elemental concentration. Therefore, the energy region that includes the photopeak of interest must be free of natural background contamination, i.e. specifically the Compton continua resulting from the major constituents of the natural background radiation.

The peak area-to-total area ratios $A(P/T)$ were used to determine the performance metric for a natural background spectrum. The peak area-to-total area ratio in the suppressed background spectrum was calculated as $A(P/T)_{sup} = 0.066$, and $A(P/T)_{uns} = 0.011$ in the unsuppressed background spectrum, from which the area suppression factor $A(SF)$ for the background spectra was calculated as

$$A(SF) = \frac{A(P/T)_{sup}}{A(P/T)_{uns}} = \frac{0.066}{0.011} = 6$$

(5.7)

Table 5.1 gives a detailed listing of the isotopes identified in the natural environmental radiation and the natural decay series they originate from. Compton Suppression Spectrometers perform an essential function by reducing the Compton continua that result from major photopeaks such as $1460$-keV $^{40}K$, $510$-keV $^{83}Se$ and $238.6$-keV $^{212}Pb$. Reduction of the Compton continua provides better peak identification, especially for weak peaks that fall into one the continuum as well as more accurate concentration determination.
Table 5.1: List of isotopes identified in the natural background and their decay series

<table>
<thead>
<tr>
<th>Decay Series</th>
<th>Isotope</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}U$ or $^{238}U$ Series</td>
<td>$^{169}Yb$</td>
<td>63.1</td>
</tr>
<tr>
<td></td>
<td>$^{197}Hg$</td>
<td>77.3</td>
</tr>
<tr>
<td></td>
<td>$^{67}Cu$</td>
<td>92.8</td>
</tr>
<tr>
<td></td>
<td>$^{186.2}Ra$</td>
<td>186.2</td>
</tr>
<tr>
<td></td>
<td>$^{169}Yb$</td>
<td>198.0</td>
</tr>
<tr>
<td></td>
<td>$^{212}Pb$</td>
<td>238.6</td>
</tr>
<tr>
<td></td>
<td>$^{234m}Pa$</td>
<td>1001.0</td>
</tr>
<tr>
<td></td>
<td>$^{214}Bi$</td>
<td>1120.3</td>
</tr>
<tr>
<td>$^{232}Th$ Series</td>
<td>$^{214}Pb$</td>
<td>295.2</td>
</tr>
<tr>
<td></td>
<td>$^{211}Bi$</td>
<td>351.1</td>
</tr>
<tr>
<td></td>
<td>$^{23}Ne$</td>
<td>439.9</td>
</tr>
<tr>
<td></td>
<td>$^{83}Se$</td>
<td>510.1</td>
</tr>
<tr>
<td></td>
<td>$^{208}Tl$</td>
<td>583.2</td>
</tr>
<tr>
<td></td>
<td>$^{214}Bi$</td>
<td>609.3</td>
</tr>
<tr>
<td></td>
<td>$^{212}Bi$</td>
<td>727.3</td>
</tr>
<tr>
<td></td>
<td>$^{228}Ac$</td>
<td>911.6</td>
</tr>
<tr>
<td></td>
<td>$^{228}Ac$</td>
<td>969.1</td>
</tr>
<tr>
<td>Non-Series</td>
<td>$^{99}Mo$</td>
<td>140.5</td>
</tr>
<tr>
<td></td>
<td>$^{60}Co$</td>
<td>1332.5</td>
</tr>
<tr>
<td></td>
<td>$^{40}K$</td>
<td>1460.8</td>
</tr>
</tbody>
</table>
5.3 Performance Measurements with $^{137}$Cs

Since most of the photopeaks used in activation analysis are located in the range below 2 MeV, Compton Suppression Spectrometer performance can be best evaluated with a $^{137}$Cs isotope because of its single gamma ray of 661.66 keV.

Figure 5.2 shows the decay scheme of Cs-137, one of the important long-lived fission products ($\tau_{1/2} = 30.17$ y) and a common environmental contaminant. It emits two beta rays of 1176 (6%) and 514 keV (94%). Transition to the first excited state through 514-keV beta emission is followed by de-excitation via isomeric transition with a single gamma ray of 661.66-keV energy with a 2.55-min half-life. De-excitation can take place through the emission of a gamma ray as well as by the ejection of an atomic electron (a conversion electron) with subsequent delayed emission of characteristic $^{137}$Ba x-rays.

![Radioactive decay scheme of $^{137}$Cs; I.T. stands for isomeric or internal transition.](image)

The suppressed and unsuppressed $^{137}$Cs spectra collected with the Penn State Compton Suppression Spectrometer are shown in Figure 5.3. The full energy peak at 661.66 keV carrying the full spectroscopic information can be easily identified. The x-rays of the daughter element $^{137m}$Ba doublet at 31.9 keV and 36.3 keV, are also clearly seen in the inset plot of Figure 5.3. For most other gamma transitions the intensity of the x-rays of the daughter element will be less pronounced since most gamma transitions
have much smaller conversion coefficient. The small peak at 693.3 keV corresponds to the random sum peak between the $^{137m}Ba$ x-rays and the 661.66-keV photopeak. Also shown in the main plot of Figure 5.3 are the reduction factors 4.60, 6.09 and 12.82 for the backscatter, lowest point and the Compton edge of the Compton continuum, respectively originating from the 661.66-keV photopeak of $^{137}Cs$. These reduction factors are found to be noteworthy compared to the reported reduction factors of 2.9, 4.6 and 7.9 for the same regions of the Compton continuum obtained with the same isotope [13].

Figure 5.3. (Main plot) Suppressed and unsuppressed spectra obtained with the $^{137}Cs$ source with the reduction factors defined for multiple regions of the Compton continuum; (Inset plot) x-ray peaks originating from $^{137m}Ba$ at 31.9 keV and 36.3 keV.
The peak height-to-Compton ratios of $(P/C)_{\text{sup}} = 1001.0$ and $(P/C)_{\text{uns}} = 100.4$ were calculated for the suppressed and unsuppressed spectra, respectively. From these values, the suppression factor $SF$ of the Penn State CSS is found to be

$$SF = \frac{(P/C)_{\text{sup}}}{(P/C)_{\text{uns}}} = \frac{1001.0}{100.4} = 9.97$$ (5.8)

Table 5.2 lists a few suppression factors reported in the literature, which demonstrates that the suppression factor obtained with the Penn State Compton Suppression Spectrometer is noticeable.

<table>
<thead>
<tr>
<th>Primary Detector Type</th>
<th>Primary Detector Efficiency</th>
<th>Suppression Factor (SF)</th>
<th>Cited by</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge(Li)</td>
<td>6.0%</td>
<td>6.7</td>
<td>Currie et al. [14]</td>
</tr>
<tr>
<td>Ge(Li)</td>
<td>11.0%</td>
<td>5.4</td>
<td>Pracoulis [15]</td>
</tr>
<tr>
<td>Ge(Li)</td>
<td>7.0%</td>
<td>7.7</td>
<td>Hoetzl et al. [16]</td>
</tr>
<tr>
<td>Ge(Li)</td>
<td>18.0%</td>
<td>5.0</td>
<td>Millard [17]</td>
</tr>
<tr>
<td>HPGe</td>
<td>25.0%</td>
<td>5.2</td>
<td>Chung et al. [18]</td>
</tr>
<tr>
<td>HPGe</td>
<td>18.0%</td>
<td>7.7</td>
<td>Landsberger [13]</td>
</tr>
<tr>
<td>HPGe</td>
<td>54.0%</td>
<td>9.97</td>
<td>Penn State</td>
</tr>
</tbody>
</table>

Besides the well-defined photopeak and two x-ray peaks in the spectrum shown in Figure 5.3, two broad peaks are observed: the Compton edge and the backscattered edge. The energy of the first peak is about 473.5 keV, which results from the Compton scattered photon originating from the incident 661.66-keV photon deflected with 180° angle inside the active volume of the detector with subsequent escape of the Compton gamma ray from the detector boundary. The second is due to 180° Compton scattering
in the detector surroundings with subsequent detection in the detector of the escaped Compton scattered gamma ray having an energy of about 186.3 keV. The broadness of these peaks is a result of the fact that the scattering angle of 180° is only approximately fulfilled.

5.4 Performance Measurements with Dendrochronologically-Dated Tree Ring Sample

Dendrochemistry project at Penn State Radiation Science and Engineering Center involves finding a correlation between the gold uptake in a tree and major global environmental effects such as volcanic eruptions. The main purpose is to search for the gold peak in various tree samples from around the world. These tree samples are dendrochronologically dated at Cornell University and sent for INAA analysis to Radiation Science and Engineering Center.

CTUPOR3 samples are from Porsuk, a town in central region in Turkey. CTUPOR samples were analyzed by conventional NAA, but no gold peak was found.

Same samples were also analyzed with the Compton Suppression Spectrometer at Penn State. It was found that the gold peak was observable in certain years at considerable concentration. Other elements such as zinc, calcium and iron were also identified with the Compton Suppression Spectrometer.

The CTUPOR3-819 wood sample was activated at 1 MW at the PSU Breazeale Nuclear Reactor and then counted in the CSS. Figure 5.4 shows the comparison of suppressed and unsuppressed spectra with determined gamma peaks. As demonstrated in the inset plot in Figure 5.4, the 411.8-keV gold peak is not identifiable in the unsuppressed spectrum, whereas it is possible to resolve the gold peak in the suppressed spectrum.
Another standard reference material, NIST SRM-1571 Orchard Leaves sample, was irradiated at 100 kW for 10 minutes at Penn State University Breazeale Nuclear Reactor. The sample was counted using the Compton Suppression Spectrometer in both the suppressed and unsuppressed modes. The acquired spectra are shown in Figure 5.5.
The most noticeable peak in the SRM-1571 sample is the 846.9-keV $^{56}$Mn peak, which also gives the most dominant Compton continuum in the spectrum. The peak height-to-Compton ratio for the 846.9-keV peak in the suppressed spectrum was calculated as $(P/C)_{sup} = 386.34$, and unsuppressed spectrum as $(P/C)_{uns} = 101.77$, from which the suppression factor SF can be found:

$$SF = \frac{(P/C)_{sup}}{(P/C)_{uns}} = \frac{386.34}{101.77} = 3.80 \quad (5.9)$$

---

![Figure 5.5. Suppressed and unsuppressed spectra of the NIST standard reference material SRM-1571 acquired with the Penn State Compton Suppression Spectrometer.](image)
CHAPTER 6

INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS WITH THE COMPTON SUPPRESSION SPECTROMETER

Neutron activation analysis can be considered in two categories: prompt-gamma and delayed-gamma neutron activation analyses. Irradiation and decay schemes and associated decay constants are shown in Figure 6.1.

![Diagram of neutron activation process](image)

Figure 6.1. Prompt-gamma and delayed-gamma emission processes following activation by a neutron.

Prompt-gamma neutron activation analysis is based on the detection of prompt gamma rays emitted by the compound nucleus. Since the decay constant for prompt gamma emission is very short ($<10^{-9}$ sec), data acquisition has to be carried out simultaneously with the irradiation of the sample.

In delayed-gamma neutron activation analysis, delayed gamma rays emitted by the daughter nucleus are detected and analyzed. Since the decay constant for delayed
gammas are much longer, i.e. minutes to days, data acquisition can be carried out once
the irradiation is completed.

6.1 Determination of Isotopic and Elemental Concentration with INAA

In a delayed-gamma neutron activation analysis, there are usually three stages in
the measurement: activation, decay and counting. During the activation stage, the
sample is irradiated with neutrons. The nuclides in the sample material are activated, i.e.
form compound nuclei that are highly unstable, based on their radiative neutron capture
cross sections. Simultaneously, the newly formed nuclei decay based on the decay
constant. In the decay and counting stages, the source term, i.e. activation, vanishes, and
only decay process continues.

In the following sections, rate equations that govern the activation, decay and
counting stages are derived for a simple case of activation/decay chain. The derivation
can be easily generalized to include irradiation schemes that are more complicated.

6.1.1 Neutron Activation

Given parent and daughter nuclides $X$ and $Y$ with isotopic concentrations $N_X$ and
$N_Y$, respectively. The rate equations for the isotopic concentrations $N_X$ and $N_Y$ can
be written as follows:

$$\frac{dN_X}{dt} = -N_X\sigma_X^{\gamma}\Phi$$

$$\frac{dN_Y}{dt} = N_X\sigma_X^{\gamma}\Phi - \lambda_Y N_Y$$

(6.1)

where $\sigma_X^{\gamma}$ is the radiative capture cross section for the parent nucleus $X$, $\Phi$ is the neutron
flux and $\lambda_Y$ is the decay constant for the daughter nucleus. In this equation system, the
parent nucleus $N_X$ was assumed to be stable. The system of equations can be written in matrix form as

$$
\frac{d}{dt} \begin{pmatrix} N_X \\ N_Y \end{pmatrix} = \begin{pmatrix} -\sigma_X^Y & 0 \\ \sigma_X^Y & -\lambda_Y \end{pmatrix} \begin{pmatrix} N_X \\ N_Y \end{pmatrix}
$$

(6.2)

$N_X$ and $N_Y$ can be found by solving $\dot{x} = Ax$.

$$
x(t) = x(0) \exp(At)
$$

(6.3)

where $x(0) = \begin{pmatrix} N_X(0) \\ N_Y(0) \end{pmatrix}$ represents the initial value of the vector $x(t) = \begin{pmatrix} N_X(t) \\ N_Y(t) \end{pmatrix}$ prior to the irradiation, and $\exp(At)$ is the matrix exponential. The matrix exponential $\exp(At)$ in Equation (6.3) can be shown to be identical to

$$
\exp(At) = \begin{pmatrix}
\exp(-\sigma_X^Y \Phi t) & 0 \\
\frac{\sigma_X^Y \Phi [\exp(-\sigma_X^Y \Phi t) - \exp(-\lambda_Y t)]}{\lambda_Y - \sigma_X^Y \Phi} & \exp(-\lambda_Y t)
\end{pmatrix}
$$

(6.4)

Equation (6.3) can then be rewritten as

$$
\begin{pmatrix} N_X(t) \\ N_Y(t) \end{pmatrix} = \begin{pmatrix}
\exp(-\sigma_X^Y \Phi t) & 0 \\
\frac{\sigma_X^Y \Phi [\exp(-\sigma_X^Y \Phi t) - \exp(-\lambda_Y t)]}{\lambda_Y - \sigma_X^Y \Phi} & \exp(-\lambda_Y t)
\end{pmatrix} \begin{pmatrix} N_X(0) \\ N_Y(0) \end{pmatrix}
$$

(6.5)

The rate equations can be expressed individually as

$$
N_X(t) = N_X(0) \exp(-\sigma_X^Y \Phi t)
$$

$$
N_Y(t) = \frac{\sigma_X^Y \Phi}{\lambda_Y - \sigma_X^Y \Phi} N_X(0) \left[ \exp(-\sigma_X^Y \Phi t) - \exp(-\lambda_Y t) \right] + N_Y(0) \exp(-\lambda_Y t)
$$

(6.6)
In most practical applications, the initial isotopic concentration of the daughter nuclide \( N_Y(0) \) can be assumed to be zero, i.e. \( N_Y(0) \approx 0 \). Then the concentrations given in Equation (6.6) becomes

\[
N_X(t) = N_X(0) \exp(-\sigma_X^Y \Phi t)
\]

\[
N_Y(t) = \frac{\sigma_X^Y \Phi}{\lambda_Y - \sigma_X^Y \Phi} N_X(0) \left[ \exp(-\sigma_X^Y \Phi t) - \exp(-\lambda_Y t) \right]
\]

(6.7)

If the concentration of the parent nuclide \( N_X(t) \) is assumed to be invariant, i.e. \( N_X(t) \equiv N_X \), which be expressed mathematically as \( \sigma_X^Y \Phi \ll \lambda_Y \), then the daughter nucleus concentration given in Equation (6.7) can be further simplified as

\[
N_Y(t) = \frac{\sigma_X^Y \Phi}{\lambda_Y} N_X \left[ 1 - \exp(-\lambda_Y t) \right]
\]

(6.8)

The activity of the daughter nuclide during the activation can be calculated by

\[
A_Y(t) = \lambda_Y N_Y(t) = N_X \sigma_X^Y \Phi [1 - \exp(-\lambda_Y t)]
\]

(6.9)

If the irradiation is carried out long enough, the system reaches an equilibrium, which can be found by letting \( t \to \infty \) in Equation (6.9)

\[
A_{Y\infty} = \lambda_Y N_{Y\infty} = N_X \sigma_X^Y \Phi
\]

(6.10)

\( A_{Y\infty} \) is called saturation activity, and is independent of the half-life of the activation product \( Y \), and depends solely on the value of the neutron flux and neutron cross section \([19]\). The implicit assumption in Equation (6.10) is that \( N_X \) does not vary significantly compared to \( N_Y \), as expressed in Equation (6.7).

The activation term \( \sigma_X^Y \Phi \) given in Equation (6.10) represents a single-energy group structure. However, a sample is exposed to a neutron flux with a certain spectrum, i.e. \( \Phi(E) \). The rate equation can then be represented as
In most cases, it is sufficient to represent the continuous energy spectrum in Equation (6.11) by three energy groups, i.e. thermal, epithermal and fast. The concentration of the daughter nuclide $N_Y$ in Equation (6.7) then becomes

$$N_Y(t) = \frac{\left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right)}{\lambda_Y - \left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right)} N_X \left(\exp\left[-\left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right) t\right] - \exp(-\lambda_Y t)\right)$$

(6.12)

where $\sigma_{x,th}^Y$, $\sigma_{x,epi}^Y$ and $\sigma_{x,f}^Y$ are the thermal, epithermal and fast radiative capture cross sections, and $\Phi_{th}$, $\Phi_{epi}$ and $\Phi_f$ are the thermal, epithermal and fast neutron fluxes. With the $\left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right) \ll \lambda_Y$ approximation, the daughter nuclide concentration in Equation (6.12) can be approximated by

$$N_Y(t) = \frac{\left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right)}{\lambda_Y} N_X \left[1 - \exp(-\lambda_Y t)\right]$$

(6.13)

and the daughter activity in Equation (6.9) can be represented by

$$A_Y(t) = \left(\sigma_{x,th}^Y \Phi_{th} + \sigma_{x,epi}^Y \Phi_{epi} + \sigma_{x,f}^Y \Phi_f\right) N_X \left[1 - \exp(-\lambda_Y t)\right]$$

(6.14)

In many practical applications, the contribution from fast flux irradiation, i.e. $\sigma_{x,f}^Y \Phi_f$, is usually insignificant in comparison to contribution from the thermal and epithermal components. Therefore fast flux contribution is commonly neglected.
A sample irradiation/decay scheme that matches the one given in Equation (6.4) is the neutron capture of $^{197}_{79}$Au.

$$^{197}_{79}Au + ^1_0n \rightarrow ^{198}_{79}Au^* \rightarrow ^{198}_{79}Au + \gamma_p$$

$$^{198}_{79}Au \rightarrow ^0_1\beta + ^{198}_{80}Hg^* \rightarrow ^{198}_{80}Hg + \gamma_d$$

(6.15)

where $\gamma_p$ is the prompt gamma ray emitted from the excited intermediate nucleus and $\gamma_d$ is the delayed gamma ray emitted from the daughter nucleus. The asterisks indicate excited nuclei, which de-excite through emission of gamma rays. The decay scheme is shown in Figure 6.2.

\[\]

Figure 6.2: The decay scheme of $^{198}_{79}$Au. Data obtained from Nuclear Data Evaluation Lab, Korea Atomic Energy Research Institute [8].

As the name implies, prompt gamma rays are emitted with a very small decay constant ranging between $10^{-12} - 10^{-9}$ sec following the irradiation. As seen in Eqs. (6.15), the prompt gamma ray originates from the excited intermediate nucleus $^{198}_{79}Au^*$, whereas the delayed gamma originates from the excited daughter nucleus $^{198}_{80}Hg^*$. The delayed gammas have much larger decay constants ranging from minutes to days.
6.1.2 Decay Equations

Once the irradiation is terminated, the source term in Equation (6.1) vanishes and the equation becomes a pure decay, i.e.

\[
\frac{dN_y}{dt} = -\lambda_y N_y \tag{6.16}
\]

whose solution is easily found as

\[
N_y(t) = N_y(0) \exp(-\lambda_y t) \tag{6.17}
\]

with \(N_y(0)\) as the initial value of the concentration of daughter nuclide \(N_y\) at the end of the irradiation process. Therefore, for an irradiation period of \(\tau_i\), the variation of the daughter nuclide concentration \(N_y(t)\) during the decay can be calculated by

\[
N_y(t) = \left(\frac{\sigma^Y_{X,th} \Phi_{th} + \sigma^Y_{X,epi} \Phi_{epi} + \sigma^Y_{X,f} \Phi_f}{\lambda_y} \right) N_X \left[1 - \exp(-\lambda_y \tau_i)\right] \exp(-\lambda_y t) \tag{6.18}
\]

and the variation of the nuclide activity \(A_Y(t)\) by

\[
A_Y(t) = \left(\frac{\sigma^Y_{X,th} \Phi_{th} + \sigma^Y_{X,epi} \Phi_{epi} + \sigma^Y_{X,f} \Phi_f}{\lambda_y} \right) N_X \left[1 - \exp(-\lambda_y \tau_i)\right] \exp(-\lambda_y t) \tag{6.19}
\]

6.1.3 Counting

During the counting process, the rate equations remain the same as given by Equation (6.16). The number of expected counts \(C_{\exp}\) can be calculated by

\[
C_{\exp} = \int_0^{\tau_c} dt \ A_Y(0) \exp(-\lambda_y t) = \int_0^{\tau_c} dt \ \frac{\lambda_y N_y(0)}{A_y(0)} \exp(-\lambda_y t) \tag{6.20}
\]
where $\tau_c$ is the counting time and $N_Y(0)$ is the nuclide concentration at the beginning of the counting process expressed by

$$N_Y(0) = \frac{\left(\sigma_{X,th}^{\gamma} \Phi_{th} + \sigma_{X,epi}^{\gamma} \Phi_{epi} + \sigma_{X,f}^{\gamma} \Phi_{f}\right)}{\lambda_Y} N_X \left[1 - \exp(-\lambda_Y \tau_i)\right] \exp(-\lambda_Y \tau_d) \quad (6.21)$$

The solution of Equation (6.20) can be shown to be

$$C_{\text{exp}} = \frac{\left(\sigma_{X,th}^{\gamma} \Phi_{th} + \sigma_{X,epi}^{\gamma} \Phi_{epi} + \sigma_{X,f}^{\gamma} \Phi_{f}\right)}{\lambda_Y} N_X \left[1 - \exp(-\lambda_Y \tau_i)\right] \exp(-\lambda_Y \tau_d) \left[1 - \exp(-\lambda_Y \tau_c)\right] \quad (6.22)$$

### 6.2 Instrumental Neutron Activation Analysis (INAA) Equations

Given a count distribution $C_i$ versus channel number $i$ as shown in Figure 6.3, the total peak area $A$ is calculated by

$$A = \sum_{k=l}^{j} C_k \quad (6.23)$$

The background $B$ can be calculated by

$$B = \frac{(C_i + C_j)}{2} (j - i + 1) \quad (6.24)$$

where $C_i$ and $C_j$ are the count values corresponding to the channel numbers $i$ and $j$. The net peak area $S$ can then be calculated by

$$S = A - B = \sum_{k=l}^{j} C_k - \frac{(C_i + C_j)}{2} (j - i + 1) \quad (6.25)$$

Given a peak with a net area $S$ corresponding to a gamma energy $E_\gamma$ with gamma abundance $\gamma$, the detector efficiency $\varepsilon$ corresponding to the gamma energy $E_\gamma$,
and the live and real counting times $\tau_{\text{live}}$ and $\tau_{\text{real}}$. The expected number of counts can be calculated by

$$C_{\text{exp}} = \frac{S}{\varepsilon y}$$  \hspace{1cm} (6.26)

For systems with considerable dead time, the expected number of counts can be corrected by scaling with $\tau_{\text{real}} / \tau_{\text{live}}$, i.e.

$$C_{\text{exp}} = \frac{S}{\varepsilon y} \frac{\tau_{\text{real}}}{\tau_{\text{live}}}$$  \hspace{1cm} (6.27)

---

Figure 6.3: Illustration of a simple background subtraction.
By equating Equations (6.22) and (6.27), one may calculate the isotopic concentration of the nuclide $X$, i.e.

$$\frac{S}{\varepsilon} \frac{\tau_{\text{real}}}{\tau_{\text{live}}} = \left( \frac{\sigma^Y_{X,\text{th}} \Phi_{\text{th}} + \sigma^Y_{X,\text{epi}} \Phi_{\text{epi}} + \sigma^Y_{X,f} \Phi_f}{\lambda_Y} \right) N_X$$

$$\frac{1}{[1 - \exp(-\lambda_Y \tau_i)] \exp(-\lambda_Y \tau_d) [1 - \exp(-\lambda_Y \tau_{\text{real}})]}$$

(6.28)

Solving Equation (6.28) for $N_X$, one finds the isotopic concentration $N_X$

$$N_X = \frac{S \lambda_Y \tau_{\text{real}}}{\varepsilon \gamma \tau_{\text{live}}} \left( \frac{\sigma^Y_{X,\text{th}} \Phi_{\text{th}} + \sigma^Y_{X,\text{epi}} \Phi_{\text{epi}} + \sigma^Y_{X,f} \Phi_f}{1} \right)$$

$$\frac{1}{[1 - \exp(-\lambda_Y \tau_i)] \exp(-\lambda_Y \tau_d) [1 - \exp(-\lambda_Y \tau_{\text{real}})]}$$

(6.29)

Let $m_X$, $M_A^X$ be the mass and atomic mass of the nuclide $X$, respectively, and $N_A$ be the Avogadro constant. The number of isotopes $n_X$ of the nuclide $X$ can be calculated by

$$n_X = \frac{m_X}{M_A^X} N_A$$

(6.30)

The isotopic concentration $C_X$ of the nuclide $X$ can be calculated by

$$C_X = \frac{N_X}{n_X} = \frac{N_X M_A^X}{m_X N_A}$$

(6.31)

with $N_X$ given by Equation (6.29). The elemental concentration $C_{\text{element}}$ can be calculated by dividing the isotopic concentration $C_X$ by the isotopic abundance $\gamma_{\text{isotope}}$, i.e.

$$C_{\text{element}} = \frac{C_X}{\gamma_{\text{isotope}}}$$

(6.32)

If the thermal, epithermal and fast components of the neutron flux, and sample mass are known, the isotopic and elemental concentrations can be calculated.
6.3 Neutron Flux Measurements

Accurate quantification of the neutron flux is an essential step in carrying out an absolute instrumental analysis. The neutron flux goes into the activation equation; hence it propagates into each and every calculation that is used to quantify the isotopic or elemental concentration of nuclides. Most commonly, two- or three-group energy scheme is used in activation calculations, i.e. thermal and epithermal, or thermal, epithermal and fast. Depending on the location and the composition of the sample, determination of the fast component of the flux might be important or insignificant. However, precise quantification of the thermal and epithermal components of the neutron flux is essential for the majority of the measurements.

In Sections 6.1 and 6.2, the equations for the determination of isotopic and elemental concentrations of nuclides in an irradiated sample were derived based on the \textit{a priori} quantities of the thermal, epithermal and fast components of the neutron flux. In the flux measurement calculations, a similar derivation is carried out; however, the equations are solved for the flux components instead of nuclide concentration.

Two samples that contain gold with known weight percentage are generally used to determine the thermal and epithermal components of the neutron flux. Commonly, the samples come in wire or foil form. For high-flux regions, $Al/Au$ wires are used, whereas for low-flux regions $Au$ foils are used. One of the samples is irradiated in bare form while the other wire is sheathed in a cadmium sleeve. A bare gold sample is activated by both thermal and epithermal neutrons. On the other hand, a cadmium-covered gold sample is only activated –to great extent– by epithermal neutrons since the cadmium cover absorbs a large portion of thermal neutrons due to its high thermal and low epithermal neutron capture cross section. The wire samples used in the flux measurements in this study are $Al/Au$ with $0.112^{W/o}$ gold.

Let the indices in $x_b$ and $x_{cd}$ designate the quantity $x$ in bare and cadmium covered forms. The number of gold atoms in the samples can be found by
where \( n_{Cd} \) and \( n_b \) are the total number of gold atoms, \( m_{Cd} \) and \( m_b \) are the sample mass, \( w_{Cd} \) and \( w_b \) are the weight percent of gold in the cadmium-covered and bare sample wires, and \( N_A \) is the Avogadro constant.

The expected count rates \( R_{Cd} \) and \( R_b \) from gold in the cadmium covered and bare sample wires can be calculated by

\[
R_{Cd} = n_{Cd} \sigma_{epi} \Phi_{epi} \\
R_b = n_b \left( \sigma_{th} \Phi_{th} + \sigma_{epi} \Phi_{epi} \right)
\]

The activities measured from the gold peak of the cadmium-covered and bare sample wires are

\[
R_{Cd} = \frac{S \lambda_y}{\varepsilon \gamma} \frac{1}{\left[1 - \exp(-\lambda_y \tau_i)\right] \exp(-\lambda_y \tau_d) \left[1 - \exp(-\lambda_y \tau_e)\right]}
\]

where \( S, \lambda_y, \varepsilon, \gamma, \tau_i, \tau_d \) and \( \tau_d \) were defined in the previous section for INAA calculations.

By equating Equation (6.34) and (6.35), the thermal and epithermal components of the neutron flux can be found:

\[
\begin{pmatrix}
0 & n_{Cd} \sigma_{epi} \\
n_b \sigma_{th} & n_b \sigma_{epi}
\end{pmatrix}
\begin{pmatrix}
\Phi_{th} \\
\Phi_{epi}
\end{pmatrix}
= 
\begin{pmatrix}
R_{Cd} \\
R_b
\end{pmatrix}
\]

The flux vector can be found by simple matrix algebra, i.e.

\[
\begin{pmatrix}
\Phi_{th} \\
\Phi_{epi}
\end{pmatrix} = 
\begin{pmatrix}
0 & n_{Cd} \sigma_{epi} \\
n_b \sigma_{th} & n_b \sigma_{epi}
\end{pmatrix}^{-1}
\begin{pmatrix}
R_{Cd} \\
R_b
\end{pmatrix}
= 
\begin{pmatrix}
\frac{1}{n_{Cd} \sigma_{th}} & \frac{1}{n_{Cd} \sigma_{epi}} \\
\frac{1}{n_b \sigma_{th}} & 0
\end{pmatrix}
\begin{pmatrix}
R_{Cd} \\
R_b
\end{pmatrix}
\]

(6.37)
from which the flux vector is easily found as

\[
\begin{pmatrix}
\Phi_{\text{th}} \\
\Phi_{\text{epi}}
\end{pmatrix}
= \begin{pmatrix}
\frac{R_b}{n_b\sigma_{\text{th}}} - \frac{R_{Cd}}{n_{Cd}\sigma_{\text{th}}} \\
\frac{R_{Cd}}{n_{Cd}\sigma_{\text{epi}}}
\end{pmatrix}
\] (6.38)

If written individually, the epithermal flux can be found by

\[
\Phi_{\text{epi}} = \frac{R_{Cd}}{n_{Cd}\sigma_{\text{epi}}}
\] (6.39)

and the thermal flux by

\[
\Phi_{\text{th}} = \frac{R_b}{n_b\sigma_{\text{th}}} - \frac{R_{Cd}}{n_{Cd}\sigma_{\text{th}}}
\] (6.40)

Once the thermal and epithermal components of the flux are determined with confidence, they can be used in the isotopic and elemental concentration calculations presented in the previous section.

The fast component of the flux can also be found in a similar fashion. In order to find a unique solution to the system of equations, an independent third measurement is needed that shows the effect of activation due to fast neutrons in comparable fractions.

### 6.4 Standard Reference Material Measurements

Instrumental analyses were performed to determine the elemental composition of standard reference materials (SRM) to do comparison between the measured quantity and the quantity given by the reference material. The SRM’s are sources prepared by National Institute of Standards and Technology (NIST) to use as a calibration standard.
6.4.1 NIST SRM 4275C-96 Mixed Gamma Source

NIST SRM 4275C-96 is a calibrated mixed gamma source that contains $^{125}$Sb, $^{154}$Eu, and $^{155}$Eu. This standard provides a wide range of gamma ray energies of various intensities. The abundance of energies makes the standard a very appropriate calibration material to obtain both the energy calibration and the efficiency calibration for conventional gamma spectrometry. However, as will be explained later in the thesis, the dominance of the multiple cascade transitions makes it impossible to achieve a reliable efficiency calibration in a Compton Suppression Spectrometer.

The unsuppressed and Compton suppressed energy spectra of the source obtained with the Penn State CSS are shown in Figure 6.4.

![Figure 6.4](image.png)

Figure 6.4. The unsuppressed and the suppressed spectra obtained with the NIST SRM 4275C-96 mixed gamma source.
6.4.2 NIST SRM-1570 Spinach Sample

One of the samples analyzed with the Compton Suppression Spectrometer was the Spinach sample obtained from NIST as a standard reference material with the code SRM-1570. The sample contains multiple isotopes of known quantity with associated uncertainty. The quantities and the associated uncertainties are provided by the NIST or available in the literature. The isotopic composition of the sample can be found in Glascock [9]. The isotopes of interest in the NIST SRM-1570 are listed in Table 6.1 with respective physical quantities required for calculations.

Table 6.1. Isotopes of interest in the NIST SRM-1570 Spinach sample with important physical parameters required to perform irradiation and decay calculations

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Isotope Abundance</th>
<th>Molecular Weight (g)</th>
<th>$T_{1/2}$ (min)</th>
<th>$\sigma_{th}$ (barns)</th>
<th>Resonance Integral (barns)</th>
<th>Gamma Peaks (keV)</th>
<th>Gamma Abundance</th>
<th>Gamma Factor ($\Gamma$)</th>
<th>$\lambda$ (1/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na-24</td>
<td>1</td>
<td>22.9898</td>
<td>897.6</td>
<td>0.51</td>
<td>0.303</td>
<td>1368.6</td>
<td>1</td>
<td>18.4</td>
<td>$1.29 \times 10^{-5}$</td>
</tr>
<tr>
<td>Br-80</td>
<td>0.5069</td>
<td>79.904</td>
<td>17.68</td>
<td>9.85</td>
<td>115.9</td>
<td>616.3</td>
<td>0.067</td>
<td>0.436</td>
<td>$6.53 \times 10^{-4}$</td>
</tr>
<tr>
<td>Mg-27</td>
<td>0.1101</td>
<td>24.305</td>
<td>9.46</td>
<td>0.04</td>
<td>0.024</td>
<td>1014.4</td>
<td>0.286</td>
<td>4.94</td>
<td>$1.22 \times 10^{-3}$</td>
</tr>
<tr>
<td>V-52</td>
<td>0.9975</td>
<td>50.94</td>
<td>3.75</td>
<td>4.79</td>
<td>2.63</td>
<td>1434.08</td>
<td>1</td>
<td>7.17</td>
<td>$3.08 \times 10^{-3}$</td>
</tr>
<tr>
<td>K-42</td>
<td>0.0673</td>
<td>39.0983</td>
<td>741.6</td>
<td>1.45</td>
<td>1.41</td>
<td>1524.58</td>
<td>0.188</td>
<td>1.4</td>
<td>$1.56 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

The NIST SRM-1570 Spinach sample was irradiated using the pneumatic transfer facility at the Breazeale Nuclear Reactor at the Pennsylvania State University. The facility was introduced earlier in Section 3.2. The irradiation parameters and counting information for both the suppressed and the unsuppressed spectra for the sample are given in Table 6.2.
Table 6.2. The irradiation parameters and the counting information for the NIST SRM-1570 Spinach sample

<table>
<thead>
<tr>
<th>IRRADIATION PARAMETER</th>
<th>QUANTITY</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation location</td>
<td>Pneumatic Transfer Facility</td>
</tr>
<tr>
<td>Date Performed</td>
<td>8/10/2006</td>
</tr>
<tr>
<td>Sample Name</td>
<td>SRM-1570 Spinach</td>
</tr>
<tr>
<td>Sample Weight (g)</td>
<td>0.1446</td>
</tr>
<tr>
<td>Reactor Power (kW)</td>
<td>10</td>
</tr>
<tr>
<td>Thermal Neutron Flux (n/cm^2sec)</td>
<td>(1.20 \times 10^{11})</td>
</tr>
<tr>
<td>Epithermal Neutron Flux (n/cm^2sec)</td>
<td>(3.52 \times 10^9)</td>
</tr>
<tr>
<td>Fast Neutron Flux (n/cm^2sec)</td>
<td>N/A</td>
</tr>
<tr>
<td>Irradiation Time (sec)</td>
<td>60</td>
</tr>
<tr>
<td>Decay Time for Unsuppressed Measurement (sec)</td>
<td>103</td>
</tr>
<tr>
<td>Counting Time for Unsuppressed Measurement (sec)</td>
<td>600</td>
</tr>
<tr>
<td>Real Time for Unsuppressed Measurement (sec)</td>
<td>616.455</td>
</tr>
<tr>
<td>Decay Time for Suppressed Measurement (sec)</td>
<td>794</td>
</tr>
<tr>
<td>Counting Time for Suppressed Measurement (sec)</td>
<td>600</td>
</tr>
<tr>
<td>Real Time for Suppressed Measurement (sec)</td>
<td>611.233</td>
</tr>
</tbody>
</table>
The suppressed and unsuppressed spectra of the NIST SRM-1570 Spinach sample acquired with the Penn State Compton Suppression Spectrometer are shown in Figure 6.5.

![Graph showing suppressed and unsuppressed spectra](image)

Figure 6.5. Spectra of the NIST SRM-1570 Spinach sample obtained after the irradiation with and without Compton suppression.

Table 6.3 gives a list of elemental concentrations for the isotopes identified in the SRM-1570 Spinach sample. The full energy peak used in the concentration calculations is also given along with the efficiency corresponding to that energy. Elemental concentrations were calculated using both the suppressed and the unsuppressed spectra. Also given in Table 6.3 is the elemental concentrations reported in the literature, and the measurement error between both concentrations and the literature value.
The largest measurement error was observed in the concentration of $^{24}$Na by $-50.8\%$ followed by the concentration of $^{42}$K by $-19.80\%$, both in the suppressed mode. The measurement error in $^{24}$Na concentration reduces significantly to $+5.26\%$ when the calculation is performed with the unsuppressed spectrum. This drastic improvement in measurement accuracy was attributed to deviation in the efficiency calibration in the suppressed mode since the true coincidence corrections (peak-to-total calibration) were not taken into account. It was mentioned earlier that, depending on the proximity of the sample to the detector, peak count losses due to true coincidences can be close to 40%.

Table 6.3: Isotopic and elemental concentrations for the isotopes present in the SRM-1570 Spinach sample calculated by activation analysis

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Peak Efficiency</th>
<th>Activity ($\mu$Ci/g)</th>
<th>Elemental Concentration (ppm)</th>
<th>Elemental Concentration (ppm)</th>
<th>Elemental Concentration (ppm)</th>
<th>Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>SUPPRESSED</td>
<td>UNSUPPRESSED</td>
<td>LITERATURE</td>
<td></td>
</tr>
<tr>
<td>Na-24</td>
<td>1368.6</td>
<td>$7.143 \times 10^{-3}$</td>
<td>305.632</td>
<td>6986</td>
<td>14947</td>
<td>14200</td>
<td>-50.8% (+5.26)</td>
</tr>
<tr>
<td>Br-80</td>
<td>616.7</td>
<td>$1.245 \times 10^{-2}$</td>
<td>8.863</td>
<td>54</td>
<td>57</td>
<td>48</td>
<td>+12.44 (+18.63)</td>
</tr>
<tr>
<td>Mg-27</td>
<td>1014.3</td>
<td>$8.447 \times 10^{-3}$</td>
<td>2.946</td>
<td>8809</td>
<td>10180</td>
<td>8650</td>
<td>+1.83 (+17.69)</td>
</tr>
<tr>
<td>V-52</td>
<td>1433.5</td>
<td>$7.054 \times 10^{-3}$</td>
<td>0.186</td>
<td>1.005</td>
<td>0.00</td>
<td>1.2</td>
<td>-16.25 (N/A)</td>
</tr>
<tr>
<td>K-42</td>
<td>1524.6</td>
<td>$6.997 \times 10^{-3}$</td>
<td>143.146</td>
<td>28551</td>
<td>30167</td>
<td>35600</td>
<td>-19.80 (-15.26)</td>
</tr>
</tbody>
</table>

* Suppressed (Unsuppressed)
 CHAPTER 7

APPLICATIONS OF THE PENN STATE COMPTON SUPPRESSION SPECTROMETER

7.1 Application to Dendrochemistry Studies

Another test was carried out using samples from the ongoing determination of trace element concentrations (in particular, trace amount of gold) in Dendrochronologically-dated tree ring samples using the neutron activation analysis (NAA) technique. The ultimate objective of this study is to come up with a correlation between annual uptake of gold and major environmental or climatologic changes, e.g. volcanic eruptions. Several thousand wood samples were analyzed by this technique [22]. All samples used in this study were independently dated by researchers at The Malcolm and Carolyn Wiener Laboratory for Aegean and Near Eastern Dendrochronology at Cornell University, where over 40,000 individually-dated wood samples with 4.5 million rings spanning the period from 7000 BC to present day are archived. Samples containing elevated levels of gold are being analyzed again for short and long half-life elements, e.g. silver, copper, etc., to investigate other elemental signatures of environmental changes using the Compton Suppression Spectrometer.

The measured rings span the relative Gordion years from 810 through 870. Table 7.1 gives typical irradiation and measurement parameters used for the Porsuk samples. The samples were irradiated in the pneumatic transfer facility.
The unsuppressed spectrum of the dendrochronologically-dated tree ring sample CTUPOR3-819 collected in Porsuk, Turkey from relative Gordion year 819 is shown in Figure 7.1. Identified nuclides are marked on the spectrum. The most noticeable peaks are $^{82}Br$, $^{24}Na$ and $^{47}Ca$.

The suppressed spectrum of the same tree ring sample is shown in Figure 7.2 with the nuclides identified by Genie-2000 spectroscopy software. In addition to the above nuclides easily identified in the unsuppressed spectrum, $^{65}Zn$, $^{60}Co$ and $^{198}Au$ are also identified.

Table 7.1: Typical irradiation parameters for the Porsuk wood samples CTUPOR-xxx *

<table>
<thead>
<tr>
<th>IRRADIATION PARAMETER</th>
<th>QUANTITY</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation location</td>
<td>Pneumatic Transfer Facility</td>
</tr>
<tr>
<td>Date Performed</td>
<td>3/14/2006</td>
</tr>
<tr>
<td>Sample Name</td>
<td>CTUPOR-810</td>
</tr>
<tr>
<td>Sample Weight (g)</td>
<td>0.0669</td>
</tr>
<tr>
<td>Reactor Power (kW)</td>
<td>1000</td>
</tr>
<tr>
<td>Thermal Neutron Flux $(n/cm^2/sec)$</td>
<td>$1.60 \times 10^{13}$</td>
</tr>
<tr>
<td>Epithermal Neutron Flux $(n/cm^2/sec)$</td>
<td>$7.0 \times 10^{11}$</td>
</tr>
<tr>
<td>Fast Neutron Flux $(n/cm^2/sec)$</td>
<td>N/A</td>
</tr>
<tr>
<td>Irradiation Time (sec)</td>
<td>14400</td>
</tr>
<tr>
<td>Decay Time (sec)</td>
<td>690000</td>
</tr>
<tr>
<td>Counting Time (sec)</td>
<td>21600</td>
</tr>
<tr>
<td>Real Time (sec)</td>
<td>21772.022</td>
</tr>
</tbody>
</table>

* Data provided in this table is for the sample from relative Gordion year 810.
Figure 7.1: Unsuppressed spectrum of the dendrochronologically-dated tree ring sample CTUPOR3-819 collected from Porsuk, Turkey dated relative Gordion year 819.
Figure 7.2. Suppressed spectrum of the same tree ring sample CTUPOR3-819. The unsuppressed spectrum of the same sample was shown in Figure 7.1.
It is important to note that the gold peak was visible in the suppressed spectrum while it was not identified in the unsuppressed spectrum as demonstrated in Figure 5.4. The improvement in identification of gold is of particular importance since gold was considered one of the possible signature elements that might unveil correlations between the uptake dynamics of certain elements by the tree and major environmental conditions [22].

CTUPOR3 samples that are dated from 800 through 870 are activated under similar irradiation conditions, and counted in the same geometric form for the same acquisition period after the same decay period. Nuclide concentrations were determined using the INAA formulations given in Section 6.1. It was observed that the concentrations of certain elements exhibited a stable behavior while some elements demonstrated sharp fluctuations. The chronologic analysis will be introduced in the following section. Another suppressed spectrum of CTUPOR3-824 sample from relative Gordion year 824 is shown in Figure 7.3 for visual comparison.
Figure 7.3. Suppressed spectrum of the dendrochronologically-dated tree ring sample CTUPOR3-824 from relative Gordion year 819.
7.2 Chronology of Elemental Concentrations

A series of spectra were obtained from the irradiated wood rings corresponding to a particular year. Concentration of each identifiable isotope in the spectra was individually determined thereby creating an array of concentrations with respect to years. Figure 7.4 shows the plot of chronologic variation of the isotopic concentration of $^{82}\text{Br}$. Calculated $^{82}\text{Br}$ concentration values are also listed in Table 7.2. It can be seen that the $^{82}\text{Br}$ concentration does not vary significantly over the span of the measurement. The average value of the concentration was calculated to be $\bar{C}_{\text{Br}} = 9.93 \text{ ppm}$, and the standard deviation of the distribution was found $\sigma_{\text{Br}} = 1.47 \text{ ppm}$.

---

Figure 7.4: Chronologic variation of isotopic concentration of $^{82}\text{Br}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode.
The decay scheme for $^{82}\text{Br}$ is shown in Figure 7.5. Although a large fraction of beta transitions go to the ground state of $^{82}\text{Kr}$, about 10% of the transitions go to the first and the second excited states of the nuclide. De-excitations from the second state consist of cascade transitions. Therefore, it is reasonable to state that certain degree of deviation is expected in the actual peak count due to coincidences in the Compton Suppression Spectrometer. These coincidences suppress not only the Compton continuum, but also legitimate counts for the photopeaks of interest. Consistency in the ratio of $^{82}\text{Br}$ concentrations can be considered as a validation to this hypothesis.

Figure 7.5: Gamma ray decay scheme for $^{82}\text{Br}$. It is evident that the isotope is a strong cascade emitter (Data obtained from Nuclear Data Evaluation Lab, Korea Atomic Energy Research Institute).
Table 7.2. Calculated $^{82}Br$ concentrations and associated errors.

<table>
<thead>
<tr>
<th>Year</th>
<th>$^{82}Br$ (ppm)</th>
<th>Year</th>
<th>$^{82}Br$ (ppm)</th>
<th>Year</th>
<th>$^{82}Br$ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>810</td>
<td>14.49 ± 0.18</td>
<td>831</td>
<td>9.38 ± 0.04</td>
<td>852</td>
<td>11.39 ± 0.09</td>
</tr>
<tr>
<td>811</td>
<td>11.87 ± 0.29</td>
<td>832</td>
<td>9.97 ± 0.05</td>
<td>853</td>
<td>11.14 ± 0.13</td>
</tr>
<tr>
<td>812</td>
<td>10.77 ± 0.17</td>
<td>833</td>
<td>9.28 ± 0.08</td>
<td>854</td>
<td>10.52 ± 0.07</td>
</tr>
<tr>
<td>813</td>
<td>8.12 ± 0.05</td>
<td>834</td>
<td>10.43 ± 0.18</td>
<td>855</td>
<td>11.49 ± 0.12</td>
</tr>
<tr>
<td>814</td>
<td>6.76 ± 0.06</td>
<td>835</td>
<td>10.52 ± 0.04</td>
<td>856</td>
<td>8.55 ± 0.04</td>
</tr>
<tr>
<td>815</td>
<td>10.99 ± 0.14</td>
<td>836</td>
<td>8.26 ± 0.04</td>
<td>857</td>
<td>11.43 ± 0.05</td>
</tr>
<tr>
<td>816</td>
<td>10.05 ± 0.1</td>
<td>837</td>
<td>10.54 ± 0.07</td>
<td>858</td>
<td>11.45 ± 0.06</td>
</tr>
<tr>
<td>817</td>
<td>9.51 ± 0.08</td>
<td>838</td>
<td>10.49 ± 0.06</td>
<td>859</td>
<td>12.14 ± 0.1</td>
</tr>
<tr>
<td>818</td>
<td>9.91 ± 0.1</td>
<td>839</td>
<td>7.61 ± 0.05</td>
<td>860</td>
<td>11.55 ± 0.04</td>
</tr>
<tr>
<td>819</td>
<td>9.66 ± 0.04</td>
<td>840</td>
<td>10.44 ± 0.03</td>
<td>861</td>
<td>10.46 ± 0.05</td>
</tr>
<tr>
<td>820</td>
<td>9.29 ± 0.03</td>
<td>841</td>
<td>8.05 ± 0.04</td>
<td>862</td>
<td>8.48 ± 0.05</td>
</tr>
<tr>
<td>821</td>
<td>9.55 ± 0.05</td>
<td>842</td>
<td>10.44 ± 0.04</td>
<td>863</td>
<td>10.69 ± 0.08</td>
</tr>
<tr>
<td>822</td>
<td>7.58 ± 0.04</td>
<td>843</td>
<td>10.56 ± 0.12</td>
<td>864</td>
<td>8.45 ± 0.07</td>
</tr>
<tr>
<td>823</td>
<td>9.68 ± 0.07</td>
<td>844</td>
<td>10.91 ± 0.12</td>
<td>865</td>
<td>11.24 ± 0.06</td>
</tr>
<tr>
<td>824</td>
<td>9.63 ± 0.09</td>
<td>845</td>
<td>10.89 ± 0.06</td>
<td>866</td>
<td>11.67 ± 0.06</td>
</tr>
<tr>
<td>825</td>
<td>9.67 ± 0.08</td>
<td>846</td>
<td>7.85 ± 0.03</td>
<td>867</td>
<td>11.42 ± 0.06</td>
</tr>
<tr>
<td>826</td>
<td>6.11 ± 0.04</td>
<td>847</td>
<td>8.35 ± 0.04</td>
<td>868</td>
<td>10.04 ± 0.09</td>
</tr>
<tr>
<td>827</td>
<td>9.68 ± 0.1</td>
<td>848</td>
<td>10.26 ± 0.06</td>
<td>869</td>
<td>8.32 ± 0.09</td>
</tr>
<tr>
<td>828</td>
<td>7.62 ± 0.05</td>
<td>849</td>
<td>10.36 ± 0.06</td>
<td>870</td>
<td>10.8 ± 0.13</td>
</tr>
<tr>
<td>829</td>
<td>8.11 ± 0.04</td>
<td>850</td>
<td>10.62 ± 0.04</td>
<td></td>
<td></td>
</tr>
<tr>
<td>830</td>
<td>9.34 ± 0.04</td>
<td>851</td>
<td>10.69 ± 0.11</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The concentration of $^{47}\text{Ca}$ was also stable throughout the span of the measured tree ring samples. Chronologic variation of $^{47}\text{Ca}$ elemental concentration is shown in Figure 7.6 and the complete listing of concentrations is given in Table 7.3. The average elemental concentration was calculated to be $\bar{C}_{\text{Ca}} = 386.89 \text{ ppb}$ and the standard deviation of the distribution was found to be $\sigma_{\text{Ca}} = 87.59 \text{ ppb}$.

Figure 7.6: Chronologic variation of isotopic concentration of $^{47}\text{Ca}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode.
Table 7.3. Calculated $^{47}\text{Ca}$ concentrations and associated errors.

<table>
<thead>
<tr>
<th>Year</th>
<th>$^{47}\text{Ca}$ (ppb)</th>
<th>Year</th>
<th>$^{47}\text{Ca}$ (ppb)</th>
<th>Year</th>
<th>$^{47}\text{Ca}$ (ppb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>810</td>
<td>0.89 ± 0.062</td>
<td>831</td>
<td>0.32 ± 0.020</td>
<td>852</td>
<td>0.37 ± 0.020</td>
</tr>
<tr>
<td>811</td>
<td>0.55 ± 0.136</td>
<td>832</td>
<td>0.33 ± 0.015</td>
<td>853</td>
<td>0.38 ± 0.018</td>
</tr>
<tr>
<td>812</td>
<td>0.49 ± 0.055</td>
<td>833</td>
<td>0.34 ± 0.016</td>
<td>854</td>
<td>0.37 ± 0.015</td>
</tr>
<tr>
<td>813</td>
<td>0.42 ± 0.037</td>
<td>834</td>
<td>0.38 ± 0.026</td>
<td>855</td>
<td>0.44 ± 0.019</td>
</tr>
<tr>
<td>814</td>
<td>0.41 ± 0.039</td>
<td>835</td>
<td>0.34 ± 0.025</td>
<td>856</td>
<td>0.37 ± 0.012</td>
</tr>
<tr>
<td>815</td>
<td>0.43 ± 0.030</td>
<td>836</td>
<td>0.35 ± 0.018</td>
<td>857</td>
<td>0.36 ± 0.018</td>
</tr>
<tr>
<td>816</td>
<td>0.37 ± 0.021</td>
<td>837</td>
<td>0.4 ± 0.027</td>
<td>858</td>
<td>0.38 ± 0.012</td>
</tr>
<tr>
<td>817</td>
<td>0.38 ± 0.026</td>
<td>838</td>
<td>0.37 ± 0.019</td>
<td>859</td>
<td>0.45 ± 0.018</td>
</tr>
<tr>
<td>818</td>
<td>0.41 ± 0.040</td>
<td>839</td>
<td>0.33 ± 0.011</td>
<td>860</td>
<td>0.37 ± 0.031</td>
</tr>
<tr>
<td>819</td>
<td>0.35 ± 0.022</td>
<td>840</td>
<td>0.29 ± 0.023</td>
<td>861</td>
<td>0.47 ± 0.035</td>
</tr>
<tr>
<td>820</td>
<td>0.29 ± 0.032</td>
<td>841</td>
<td>0.32 ± 0.029</td>
<td>862</td>
<td>0.44 ± 0.028</td>
</tr>
<tr>
<td>821</td>
<td>0.32 ± 0.026</td>
<td>842</td>
<td>0.3 ± 0.022</td>
<td>863</td>
<td>0.49 ± 0.030</td>
</tr>
<tr>
<td>822</td>
<td>0.34 ± 0.023</td>
<td>843</td>
<td>0.37 ± 0.023</td>
<td>864</td>
<td>0.45 ± 0.016</td>
</tr>
<tr>
<td>823</td>
<td>0.38 ± 0.026</td>
<td>844</td>
<td>0.36 ± 0.017</td>
<td>865</td>
<td>0.43 ± 0.023</td>
</tr>
<tr>
<td>824</td>
<td>0.41 ± 0.030</td>
<td>845</td>
<td>0.35 ± 0.047</td>
<td>866</td>
<td>0.44 ± 0.017</td>
</tr>
<tr>
<td>825</td>
<td>0.34 ± 0.061</td>
<td>846</td>
<td>0.31 ± 0.020</td>
<td>867</td>
<td>0.44 ± 0.018</td>
</tr>
<tr>
<td>826</td>
<td>0.34 ± 0.029</td>
<td>847</td>
<td>0.32 ± 0.024</td>
<td>868</td>
<td>0.44 ± 0.021</td>
</tr>
<tr>
<td>827</td>
<td>0.38 ± 0.037</td>
<td>848</td>
<td>0.34 ± 0.022</td>
<td>869</td>
<td>0.45 ± 0.020</td>
</tr>
<tr>
<td>828</td>
<td>0.33 ± 0.021</td>
<td>849</td>
<td>0.35 ± 0.017</td>
<td>870</td>
<td>0.51 ± 0.023</td>
</tr>
<tr>
<td>829</td>
<td>0.32 ± 0.034</td>
<td>850</td>
<td>0.3 ± 0.019</td>
<td></td>
<td></td>
</tr>
<tr>
<td>830</td>
<td>0.31 ± 0.018</td>
<td>851</td>
<td>0.32 ± 0.017</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Another easily identified nuclide in both the unsuppressed and suppressed spectra was $^{24}\text{Na}$. Chronologic variation of $^{24}\text{Na}$ is shown in Figure 7.7 and the complete listing of elemental concentrations with respect to years is given in Table 7.4. $^{24}\text{Na}$ concentration demonstrated an unsteady dynamic throughout the span of the measured samples. The average value of the measured concentration was found to be $\bar{C} = 218.70$ ppm and the standard deviation of the distribution was calculated as $\sigma_C = 85.78$. It must, however, be noted that the tree ring samples can be easily contaminated with sodium during the preparation process.

Figure 7.7: Chronologic variation of isotopic concentration of $^{24}\text{Na}$ calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode.
Table 7.4. Calculated $^{24}\text{Na}$ concentrations and associated errors.

<table>
<thead>
<tr>
<th>Year</th>
<th>$^{24}\text{Na}$ (ppm)</th>
<th>Year</th>
<th>$^{24}\text{Na}$ (ppm)</th>
<th>Year</th>
<th>$^{24}\text{Na}$ (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>810</td>
<td>432.00 ± 3.08</td>
<td>831</td>
<td>171.75 ± 0.40</td>
<td>852</td>
<td>238.39 ± 1.86</td>
</tr>
<tr>
<td>811</td>
<td>346.76 ± 6.28</td>
<td>832</td>
<td>172.89 ± 0.71</td>
<td>853</td>
<td>234.69 ± 5.01</td>
</tr>
<tr>
<td>812</td>
<td>313.69 ± 2.70</td>
<td>833</td>
<td>173.61 ± 1.51</td>
<td>854</td>
<td>235.82 ± 1.92</td>
</tr>
<tr>
<td>813</td>
<td>154.85 ± 0.33</td>
<td>834</td>
<td>207.75 ± 5.00</td>
<td>855</td>
<td>272.45 ± 5.04</td>
</tr>
<tr>
<td>814</td>
<td>87.35 ± 0.32</td>
<td>835</td>
<td>185.85 ± 0.29</td>
<td>856</td>
<td>153.21 ± 0.46</td>
</tr>
<tr>
<td>815</td>
<td>308.78 ± 4.68</td>
<td>836</td>
<td>105.00 ± 0.27</td>
<td>857</td>
<td>289.32 ± 0.63</td>
</tr>
<tr>
<td>816</td>
<td>266.33 ± 2.46</td>
<td>837</td>
<td>190.11 ± 0.82</td>
<td>858</td>
<td>292.60 ± 1.12</td>
</tr>
<tr>
<td>817</td>
<td>218.46 ± 1.24</td>
<td>838</td>
<td>182.33 ± 0.84</td>
<td>859</td>
<td>359.88 ± 2.51</td>
</tr>
<tr>
<td>818</td>
<td>223.06 ± 1.78</td>
<td>839</td>
<td>100.95 ± 0.69</td>
<td>860</td>
<td>290.25 ± 0.31</td>
</tr>
<tr>
<td>819</td>
<td>199.80 ± 0.43</td>
<td>840</td>
<td>173.28 ± 0.23</td>
<td>861</td>
<td>371.75 ± 0.60</td>
</tr>
<tr>
<td>820</td>
<td>184.36 ± 0.19</td>
<td>841</td>
<td>106.16 ± 0.27</td>
<td>862</td>
<td>216.08 ± 0.48</td>
</tr>
<tr>
<td>821</td>
<td>199.67 ± 0.35</td>
<td>842</td>
<td>195.38 ± 0.51</td>
<td>863</td>
<td>386.00 ± 1.24</td>
</tr>
<tr>
<td>822</td>
<td>112.46 ± 0.28</td>
<td>843</td>
<td>206.91 ± 2.78</td>
<td>864</td>
<td>206.40 ± 1.23</td>
</tr>
<tr>
<td>823</td>
<td>199.74 ± 0.84</td>
<td>844</td>
<td>196.15 ± 3.24</td>
<td>865</td>
<td>345.00 ± 0.88</td>
</tr>
<tr>
<td>824</td>
<td>197.77 ± 1.19</td>
<td>845</td>
<td>190.00 ± 0.31</td>
<td>866</td>
<td>335.02 ± 1.07</td>
</tr>
<tr>
<td>825</td>
<td>198.05 ± 0.64</td>
<td>846</td>
<td>92.19 ± 0.14</td>
<td>867</td>
<td>317.56 ± 1.33</td>
</tr>
<tr>
<td>826</td>
<td>64.91 ± 0.23</td>
<td>847</td>
<td>107.38 ± 0.25</td>
<td>868</td>
<td>363.84 ± 2.60</td>
</tr>
<tr>
<td>827</td>
<td>193.36 ± 1.02</td>
<td>848</td>
<td>188.88 ± 0.58</td>
<td>869</td>
<td>221.73 ± 2.59</td>
</tr>
<tr>
<td>828</td>
<td>103.48 ± 0.52</td>
<td>849</td>
<td>183.27 ± 0.83</td>
<td>870</td>
<td>397.53 ± 5.95</td>
</tr>
<tr>
<td>829</td>
<td>112.42 ± 0.22</td>
<td>850</td>
<td>197.49 ± 0.48</td>
<td></td>
<td></td>
</tr>
<tr>
<td>830</td>
<td>157.42 ± 0.25</td>
<td>851</td>
<td>210.78 ± 3.11</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Gold photopeak appears in a region of the spectrum where Compton noise is usually predominant. Therefore, it is possible that it cannot be identified with the conventional gamma spectroscopy, as was demonstrated in Sections 5.4 and 7.1.

Chronologic variation of elemental gold concentration is shown in Figure 7.8 and the complete listing of concentrations and associated errors is listed in Table 7.5. It can be seen that gold concentration exhibits sharp and rapid fluctuations throughout the span of the measured samples. Gold concentrations measured in the tree ring samples belonging to certain relative Gordian years such as 819, 824, 843 and 866 are elevated. As explained in the foregoing section, accurate determination of gold in the dendrochronologically-dated tree rings might give certain signature patterns that can be correlated with major environmental effects.

Figure 7.8: Chronologic variation of isotopic concentration of $^{198}$Au calculated from total activities measured with the Compton Suppression Spectrometer in suppressed mode.
Table 7.5: Calculated $^{198}\text{Au}$ concentrations and associated errors.

<table>
<thead>
<tr>
<th>Year</th>
<th>$^{198}\text{Au}$ (ppb)</th>
<th>Year</th>
<th>$^{198}\text{Au}$ (ppb)</th>
<th>Year</th>
<th>$^{198}\text{Au}$ (ppb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>810</td>
<td>0.000 ± 0.000</td>
<td>831</td>
<td>4.716 ± 0.277</td>
<td>852</td>
<td>2.745 ± 0.777</td>
</tr>
<tr>
<td>811</td>
<td>0.000 ± 0.000</td>
<td>832</td>
<td>4.095 ± 0.544</td>
<td>853</td>
<td>2.570 ± 1.119</td>
</tr>
<tr>
<td>812</td>
<td>0.000 ± 0.000</td>
<td>833</td>
<td>0.028 ± 0.342</td>
<td>854</td>
<td>0.028 ± 0.011</td>
</tr>
<tr>
<td>813</td>
<td>0.100 ± 0.019</td>
<td>834</td>
<td>0.070 ± 0.025</td>
<td>855</td>
<td>0.043 ± 0.017</td>
</tr>
<tr>
<td>814</td>
<td>0.073 ± 0.019</td>
<td>835</td>
<td>2.095 ± 0.567</td>
<td>856</td>
<td>0.597 ± 0.303</td>
</tr>
<tr>
<td>815</td>
<td>0.068 ± 0.018</td>
<td>836</td>
<td>0.053 ± 0.553</td>
<td>857</td>
<td>0.524 ± 0.430</td>
</tr>
<tr>
<td>816</td>
<td>4.306 ± 0.950</td>
<td>837</td>
<td>2.625 ± 0.865</td>
<td>858</td>
<td>0.051 ± 0.012</td>
</tr>
<tr>
<td>817</td>
<td>4.075 ± 0.863</td>
<td>838</td>
<td>0.452 ± 0.452</td>
<td>859</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>818</td>
<td>0.063 ± 0.022</td>
<td>839</td>
<td>0.043 ± 0.009</td>
<td>860</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>819</td>
<td>5.031 ± 0.352</td>
<td>840</td>
<td>2.206 ± 0.489</td>
<td>861</td>
<td>0.588 ± 0.023</td>
</tr>
<tr>
<td>820</td>
<td>0.000 ± 0.000</td>
<td>841</td>
<td>4.566 ± 0.507</td>
<td>862</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>821</td>
<td>0.000 ± 0.000</td>
<td>842</td>
<td>2.168 ± 0.493</td>
<td>863</td>
<td>1.502 ± 0.785</td>
</tr>
<tr>
<td>822</td>
<td>4.844 ± 0.552</td>
<td>843</td>
<td>4.823 ± 1.070</td>
<td>864</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>823</td>
<td>0.993 ± 0.583</td>
<td>844</td>
<td>1.663 ± 0.805</td>
<td>865</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>824</td>
<td>6.026 ± 0.481</td>
<td>845</td>
<td>0.000 ± 0.000</td>
<td>866</td>
<td>3.220 ± 0.279</td>
</tr>
<tr>
<td>825</td>
<td>4.121 ± 1.079</td>
<td>846</td>
<td>0.064 ± 0.015</td>
<td>867</td>
<td>0.031 ± 0.012</td>
</tr>
<tr>
<td>826</td>
<td>2.246 ± 0.706</td>
<td>847</td>
<td>3.171 ± 0.257</td>
<td>868</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>827</td>
<td>3.927 ± 1.153</td>
<td>848</td>
<td>0.000 ± 0.000</td>
<td>869</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>828</td>
<td>4.048 ± 0.335</td>
<td>849</td>
<td>0.000 ± 0.000</td>
<td>870</td>
<td>0.000 ± 0.000</td>
</tr>
<tr>
<td>829</td>
<td>3.724 ± 0.351</td>
<td>850</td>
<td>3.061 ± 0.500</td>
<td></td>
<td></td>
</tr>
<tr>
<td>830</td>
<td>2.183 ± 0.491</td>
<td>851</td>
<td>0.000 ± 0.000</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
CHAPTER 8

SUMMARY AND CONCLUSIONS

8.1 Summary of the Research

Compton Suppression Spectrometers are widely used for low-level counting applications as well as determining weak peaks in samples that contain mixed nuclides of various activities. They are effective in reducing the Compton continuum as well as other environmental background such as cosmic rays.

Theory of the Compton scattering process was given and the operating principles of Compton Suppression Spectrometers were introduced. The Compton Suppression Spectrometer at the Penn State Radiation Science and Engineering Center (RSEC) was introduced, and the specifications of the system components were given.

Energy, peak-to-total and efficiency calibration procedures were introduced; a sample energy and efficiency calibrations were performed.

Various performance metrics were introduced for Compton suppression spectrometry. Most of the performance metrics are not explicitly defined, therefore lack standardization. IEEE test procedures [10] were followed throughout this text for determination of peak height-to-Compton ratio where needed. Other performance measures introduced in this text are peak area-to-Compton area ratio $A(P/C)$, peak area-to-total area ratio $A(P/T)$, suppression factor $SF$, reduction factor $RF$ and area suppression factor $ASF$. Different performance measures were used depending on the source type. The peak height-to-Compton ratio was calculated to be $(P/C) = 1001$ in the suppressed mode, and $(P/C) = 100.4$ in the unsuppressed mode with a $^{137}\text{Cs}$ source, which gave a suppression factor of $SF = 9.97$. For natural background spectrum, peak area-to-total area $A(P/T)$ was used as the performance metric, which was calculated to
be $A(P/T) = 0.066$ in the suppressed spectrum, and $A(P/T) = 0.011$ in the unsuppressed spectrum, which gave an area suppression factor of $ASF = 6$.

8.2 Conclusions

The measurements that were presented in Chapter 5 demonstrate that the Penn State Compton Suppression Spectrometer has shown exceptional performance for a variety of standard samples. The spectrometer also demonstrated that certain peaks that are unidentifiable with the conventional gamma spectrometers due to excessive Compton noise can be identified in the Compton suppressed mode. One example is the gold peak in a dendrochronologically-dated wood sample CTUPOR3-819 collected from Porsuk, Turkey dating back to 819 relative Gordion year. The Penn State Compton Suppression Spectrometer was able to identify the gold peak at $E_\gamma = 411 \text{ keV}$ that cannot be identified with the conventional gamma spectrometer. The identified peak was also verified mathematically by running the nuclide identification script that is available in Genie-2000.

8.3 Future Work

Compton Suppression Spectrometers are very useful resources especially for low-activity nuclides. However, they also introduce considerable degree of measurement error when used for the determination of absolute quantity of a nuclide in a sample. It is well-known that true coincidence events can become significant sources of measurement error for any gamma spectrometer, particularly for nuclides with cascade emission lines. This measurement error is even more pronounced in Compton Suppression Spectrometers since the cascade emission, which is a true coincidence event, can be considered as a result of Compton coincidence, which results in
elimination of legitimate counts in the photopeak. The measurement error as a result of this process can be up to 40 – 50% depending on the geometry of the system.

For accurate determination of abscution isotopic or elemental concentrations, a procedure called peak-to-total calibration must be performed before the efficiency calibration. Both peak-to-total and efficiency calibrations must be carried out with standard radioactive sources that do not contain nuclides with strong cascade emission lines.

Genie-2000 Gamma Analysis spectroscopy software comes with a built-in module to perform corrections for true coincidence, and cascade summing losses and gains. Genie-2000 can calculate the true coincidence correction factors, for a wide variety of counting geometries and for an exhaustive list of nuclides and gamma ray lines. In order to compute the correction factors, the built-in algorithm requires a single intrinsic peak-to-total efficiency curve and a spatial response characterization or In-Situ Object Calibration Software (ISOCS) characterization for each detector. ISOCS is a powerful mathematical tool to calculate HPGe full energy peak efficiencies for practically any source geometry.

The Genie 2000 algorithm for the calculation of true coincidence correction factor for voluminous sources is based on the proposed methodology by Kolotov et al [21]. In this method, the source volume is divided into a large number of sub-sources of equal volume. A point is sampled in each sub-source using a pseudo-random sequence. The overall true coincidence factor is determined from the calculation of each of these points.
BIBLIOGRAPHY


