ขั้นตอนวิธีประยุกต์สำหรับคัดกรองวัสดุนิวเคลียร์และกัมมันตรังสีที่มีผลต่อความมั่นคงปลอดภัย ทางนิวเคลียร์ จากสเปกตรัมที่มีการแยกแยะไม่ชัดเจน

นายเหงียน นิง จาง



ับทคัดย่อและแฟ้มข้อมูลฉบับเต็มของวิทยานิพนธ์ตั้งแต่ปีการศึกษา 2554 ที่ให้บริการในคลังปัญญาจุฬาฯ (CUIR)

เป็นแฟ้มข้อมูลของนิสิตเจ้าของวิทยานิพนธ์ ที่ส่งผ่านทางบัณฑิตวิทยาลัย

The abstract and full text อิทเกษโซนท์ขี้เป็นส่วนหนึ่งของอาร์ซื้อหาตามหลักสูตรุปริณญาวิทยาสุวศารแชาบัณ พืชpository (CUIR)

are the thesis authors files submitted through the University Graduate School.

คณะวิศวกรรมศาสตร์ จุ[ั]ฬาลงกรณ์มหาวิท[์]ยาลัย

ปีการศึกษา 2558

ลิบสิทธิ์ของจุฬาลงกรณ์มหาวิทยาลัย

APPLIED ALGORITHM FOR THE SCREENING OF NUCLEAR AND RADIOAC TIVE MATERIAL OF NUCLEAR SECURITY CONCERN FROM A POORLY RE SOLVED GAMMA-RAY SPECTRUM

Mr. Nguyen Ninh Giang



A Thesis Submitted in Partial Fulfillment of the Requirements for the Degree of Master of Science Program in Nuclear Technology Department of Nuclear Engineering Faculty of Engineering Chulalongkorn University Academic Year 2015 Copyright of Chulalongkorn University

Thesis Title	APPLIED ALGORITHM FOR THE SCREENING OF NUCLEAR AND RADIOACTIVE MATERIAL OF NUCLEAR SECURITY CONCERN FROM A POORLY RESOLVED GAMMA-RAY SPECTRUM
Ву	Mr. Nguyen Ninh Giang
Field of Study	Nuclear Technology
Thesis Advisor	Phongphaeth Pengvanich, Ph.D.
Thesis Co-Advisor	Associate Professor Nares Chankow, Ph.D.

Accepted by the Faculty of Engineering, Chulalongkorn University in Partial Fulfillment of the Requirements for the Master's Degree

> _____ Dean of the Faculty of Engineering (Professor Bundhit Eua-arporn, Ph.D.)

THESIS COMMITTEE _____Chairman (Associate Professor Somyot Srisatit, Ph.D.) _____Thesis Advisor (Phongphaeth Pengvanich, Ph.D.) Thesis Co-Advisor (Associate Professor Nares Chankow, Ph.D.) Examiner (Associate Professor Sunchai Nilsuwankosit, Ph.D.) _____External Examiner (Nguyen Hao Quang, Ph.D.)

เหงียน นิง จาง : ขั้นตอนวิธีประยุกต์สำหรับคัดกรองวัสดุนิวเคลียร์และกัมมันตรังสีที่มี ผลต่อความมั่นคงปลอดภัยทางนิวเคลียร์ จากสเปกตรัมที่มีการแยกแยะไม่ชัดเจน (APPLIED ALGORITHM FOR THE SCREENING OF NUCLEAR AND RADIOACTIVE MATERIAL OF NUCLEAR SECURITY CONCERN FROM A POORLY RESOLVED GAMMA-RAY SPECTRUM) อ.ที่ปรึกษา วิทยานิพนธ์หลัก: พงษ์แพทย์ เพ่งวาณิชย์, อ.ที่ปรึกษาวิทยานิพนธ์ร่วม: นเรศร์ จันทน์ ขาว, 70 หน้า.

สัญญาณจากรังสีแกมมาสามารถใช้ระบุชนิดของไอโซโทปรังสีได้ แต่การจะนำไป ประยุกต์ใช้ในงานด้านความมั่นคงปลอดภัย เช่นการตรวจหาวัสดุนิวเคลียร์และกัมมันตรังสีตามจุด ผ่านแดน ยังคงมีความท้าทายที่สำคัญต่างๆ ที่ต้องพิจารณา เช่น ระยะเวลาการตรวจวัคที่จำกัด เงื่อนไขการกำบังรังสี และสัญญาณรบกวนที่อาจมีอยู่ในบริเวณ การศึกษานี้ได้เสนอกระบวนการ ใหม่สำหรับระบุสัญญาณของไอโซโทปรังสีหนึ่งหรือหลายตัวจากสเปลตรัมที่แยกแยะได้ยาก โดย มีขั้นตอนวิธีตั้งแต่การลดสัญญาณรบกวนในสเปลตรัมด้วยวิธี Wavelet Decomposition การ กำจัดสัญญาณพื้นหลังที่ต่อเนื่องด้วยวิธี Baseline Determination ไปจนถึงการระบุไอโซโทป รังสีด้วยวิธี Matrix Linear Regression กระบวนการที่เสนอนี้ผ่านการตรวจสอบความถูกต้อง ด้วยการทำลองใช้กับสัญญาณรังสีแกมมาที่แยกแยะได้ยากภายใต้เงื่อนไขสถานการณ์ต่างๆ ซึ่ง รวมถึงการทดสอบกับต้นกำเนิดรังสีชนิดเดียว และการทดสอบการกำบังสัญญาณจากยูเรเนียม ธรรมชาติและยูเรเนียมเสริมสมรรถนะด้วยค้นกำเนิดรังสีต่างๆ พบว่า กระบวนการที่เสนอมี ประสิทธิภาพดีกว่าวิธีการที่ใช้ในซอฟท์แวร์สำเร็จรูปในปัจจุบัน

Chulalongkorn University

ภาควิชา วิศวกรรมนิวเคลียร์ สาขาวิชา เทคโนโลยีนิวเคลียร์ ปีการศึกษา 2558

ถายมือชื่อนิสิต
ลายมือชื่อ อ.ที่ปรึกษาหลัก
ลายมือชื่อ อ.ที่ปรึกษาร่วม

5670580021 : MAJOR NUCLEAR TECHNOLOGY

KEYWORDS: RADIOISOTOPE IDENTIFICATION / NUCLEAR MATERIALS MASKING DETECTION

NGUYEN NINH GIANG: APPLIED ALGORITHM FOR THE SCREENING OF NUCLEAR AND RADIOACTIVE MATERIAL OF NUCLEAR SECURITY CONCERN FROM A POORLY RESOLVED GAMMA-RAY SPECTRUM. ADVISOR: PHONGPHAETH PENGVANICH, Ph.D.. CO-ADVISOR: ASSOC. PROF. NARES CHANKOW, Ph.D., 70 pp.

Gamma-ray signal can be used as a fingerprint for radioisotope identification. In the context of radioactive and nuclear materials securing at the border control point, the detection task can present a significant challenge due to the limited measurement time, the shielding conditions and the noise interference. This study proposes a novel method to identify the signal of one or several radioisotopes from a poorly resolved spectrum. In this algorithm, the noise component in the raw spectrum is reduced by the wavelet decomposition approach, and the removal of continuum background is performed by the baseline determination algorithm. Finally, the identification of radioisotope is done using the matrix linear regression method. The proposed method has been verified by experiments using the poorly resolved gamma-ray signals from various scenarios, including single source, masking of natural uranium and enriched uranium (fresh fuel) with several radioactive sources. The novel method have better performance when compare with the commercial algorithm.

Department:	Nuclear Engineering	Student's Signature
Field of Study:	Nuclear Technology	Advisor's Signature
Academic Year:	2015	Co-Advisor's Signature

ACKNOWLEDGEMENTS

I am deeply indebted to my supervisor - Dr. Phongphaeth Pengvanich who stimulating motivation and invaluable supported me to complete this work.

I would like to thank to my co-advisor: Associate Professor Nares Chankow and Dr. Nguyen Hao Quang for the help and supervision during all of this research.

Special thanks to my family for keeping encourage me, without it, this work would become impossible for me.

The last but not least, I would like to thank to NSS' friend for all the fun we had in 2 years in Bangkok, Thailand.



CHULALONGKORN UNIVERSITY

CONTENTS

Page
THAI ABSTRACTiv
ENGLISH ABSTRACTv
ACKNOWLEDGEMENTSvi
CONTENTSvii
CHAPTER 1: INTRODUCTION
1.1. General introduction1
1.2. Problem statement
1.3. Motivation this work
1.4.Objectives
1.5. Scope of Study
1.6.Benefit of this research
1.7. Overview of the main section
CHAPTER 2: THEORY AND LITERATURE REVIEW
2.1.Physics theory
2.1.1. Interaction between gamma – ray radiation and detector
2.1.2. Scintillation detector characteristic7
2.1.3. Masking scenario of Nuclear with other radioactive materials9
2.1.4. Screening method at border control procedure11
2.2.Peak identification algorithm review13
2.2.1. Peak detection method14
2.2.2. Library (Gamma-M) Peak Locate16
2.2.3. Library correlation nuclide identification algorithm
2.3. Wavelet transform approach for poorly resolved signal
2.4. Theoretical background
2.5.Liner regression
CHAPTER 3: APPLIED PEAK DETECTION ALGORITHM AND SOFTWARE26
3.1.Noise Reduction
3.2. Background subtraction

	Page
3.3.Baseline Estimation	
3.4. Peak search	29
3.5. Linear Regression Equation	29
Gamma-ray emission modeling	29
3.6.Peak ratio comparison	34
CHAPTER 4: EQUIPMENT, MATERIALS AND	36
EXPERIMENT SET UP	36
4.1. Equipment and Materials	36
4.2.Experiments set-up	
CHAPTER 5: RESULTS AND DISCUSSION	41
5.1. Results and discussion	41
5.2. Conclusions and suggestion for future work	46
REFERENCES	51
APPENDIX 1. REFERENCE LIBRARY MATRIX X _n	52
APPENDIX 2: MATLAB CODE	63
VITA	70

จุฬาลงกรณ์มหาวิทยาลัย

viii

CHAPTER 1: INTRODUCTION

1.1. General introduction

The prime objective of international nuclear security program is to detect and interdict the potential illegal transfer and smuggling of nuclear or other radioactive materials that could be used in malicious acts. In company with laws and regulations, the development of effective methods for detection of radionuclides plays an important role in the first-line defense at border crossing and at actual or suspect nuclear sites.

For detection of fissile materials, there are two basic methods: "passive" detection of emitted radiation by materials itself, or several of "active" detection involving either radiographic imaging, or induced gammas when irradiating neutrons, high-energy photons to fissile materials. Each technique will have different advantages and its drawbacks, therefore no single method can work individually in the context of nuclear materials control at Port-of-Entry.

If the detector is "eyes and ears" of the radioisotope identification process, then algorithms are the "brain" to treat the information and produce the accuracy. There are many identification algorithms for low-resolution detectors like NaI(Tl) with different accuracy and processing capability. Consequently, assessing the performance of radioisotope identification algorithms for NaI(Tl) detector and finding improvements are important works.

The main objective of this research is to develop the methodology for determining the radio-isotope signatures and estimating its confidential level from gamma–ray spectrum by using of identification algorithm. The proposed method has been verified by experiments using the poorly resolved gamma-ray signals from various scenarios including single source, mixing of natural and enriched uranium with five of the most common industrial radioactive sources (⁵⁷Co, ⁶⁰Co, ¹³³Ba, ¹³⁷Cs, and ²⁴¹Am).

1.2. Problem statement

There are two central concepts of gamma-ray detector sensitivity [1]: detection efficiency and spectral resolution. The first element, efficiency, refers to the processing time and the fraction between the number of counts recorded by the detector and the actual number of gamma interaction with the detector. The second, spectral resolution, represents the sharpness of gamma-ray spectrum. At the seaport control, passive detection using NaI(Tl) detector for the analysis of gamma ray signal is the preferred technique for the investigation purposes because of its safety and simplicity. Nonetheless, the usage of NaI(Tl) detector for Nuclear materials can probably be evaded by high-density shielding materials and/or masked radio-isotope, like I-131, Cs-137; and this type of low resolution detector can cause difficult in the identification of radio-isotope because either the peaks from different gamma-ray energies may blur together or make inaccuracy of peaks information from high Compton effects of masked radio-isotope.

In most cases in the screening of the containers for nuclear security mission at seaport, the signals from nuclear materials are normally poor and weak, or its equivalent to a signal-to-noise ratio (SNR) of gamma–ray spectrum is low, because of several reasons (i) nuclear materials emit low energy gamma – ray, it is easy to attenuated (ii) the high variance of background at the field and (iii) by the shielding effects from the container, the cargos are near-by the source.

Peak detection algorithms is the popular method to analyze the gammaray spectrum when the signal-to-noise ratio is large and high-resolution detectors are available. In non-ideal conditions, like detection of nuclear material at the port, and/or under shielding conditions, the observed peaks from spectrum may not be obvious. Moreover, peak detection algorithms are highly sensitive to noise, so it is dominant to use in the laboratory rather than in the field measurements. Peak detection algorithms in Matlab-based can be found in [7]. There are several variations of linear regression methods ([5],[6],[8]) but all rely on the template library of reference gamma-ray spectrum from group of radio-nuclides composition and attempt to solve the identification problem by matching the relationship between the observed spectrum **Y** and the template library **X** via a mathematical model with parameter vector $\boldsymbol{\beta}$, i.e., **Y**=f(**X**, $\boldsymbol{\beta}$). The limitation of these methods is that, the reference library must contain a large number of different combination of expected isotopes, with variations of shielding and attenuating geometries conditions [3].

1.3. Motivation this work

In a global effort to ban and eliminate Nuclear Weapon, the investigation of using of Nuclear Materials, especially Special Nuclear Material (SNM, Plutonium and certain types of uranium) for non-peaceful purpose plays an important role. Nuclear Weapon contain SNM, which produce suspect signatures of radiation and a noticeable image on radiograph, which can be used to identify the presence of nuclear materials.

While a number of active detection methods have been proposed which can detect quite small amounts of nuclear material, they are ultimately deemed unsafe because of potentially lethal dose and complexity in manufacturing, it led passive sensing to the primary selected method.

Nuclear Materials emit gamma-ray and/or neutron which allowing the possibility for using of the passive detection. The passive neutron detection can only generates alarm of Nuclear Materials presence without the additional information of radio-isotope characteristics. The passive gamma ray method can identify which types of isotopes that generates alarm and its quantity within a detectable range of the detection system.

In the scene of seaport, the signal from Nuclear Materials to the detection system will be weak and poorly cause of a distance between truck and detector, shielding effects by intervene materials between the source and detector, atmosphere and the truck itself, so that the current techniques such as peak finding technique becomes more difficult task to evaluate the isotopes when the signals which are poorly resolved, in this case, there are some novel algorithms which are rely on linear regression technique and spectrum baseline determination will be considered. Moreover, the characteristics of expensive and heavy of the high resolution detectors like high-purity germanium (HPGe) are leaved the scintillation detectors like sodium iodine (NaI) became preference and popular in the seaports control. Therefore, the performance of identification algorithms in accordance with extracted gamma ray spectrum from scintillation detector is leading to an important activity and has not been sufficiently explored yet.

1.4. Objectives

The objectives of this research are following:

Develop software for the Screening of Nuclear and Radioactive Materials of Nuclear Security concern from a poorly resolved gamma–ray spectrum.

Evaluate the effects of shielding and distance in screening model for the control of nuclear materials.

1.5. Scope of Study

This research is limit by following scope:

- Study linear regression techniques such as peak finding, LASSO, etc.
- Use Matlab code for programming.
- Screening algorithms focus on reference library of nuclear materials.
- Verify the results using radio-isotopes which are locally available.

1.6. Benefit of this research

This research aims to develop the model of screening radioisotope by linear regression technique for nuclear security mission, when the template library spectra of nuclear materials compositions are not available. The outcome of the study are including:

• The applied algorithm for screening radioisotopes in a nuclear security mission, when the template library spectra of nuclear materials compositions are not available.

• The software used for screening radioisotopes in a nuclear security mission.

1.7. Overview of the main section

The remainder of this thesis is organized as follows. In Chapter 2, a significant published results in nuclear material detection were reviewed. The chapter started with physics concepts of the obtained spectra nature including detection of photoelectric effect, Compton effect in observed gamma-ray spectra, and detector characteristics. After that some masking scenarios of nuclear material with other interference radioactive source and the procedure of screening radioactive materials were reviewed. Finally, several current detection techniques have considered like Peak detection, Library Gamma-M method, LibCorNID were introduced. Most of my work was started in Chapter 3, a method for identification of nuclear materials was proposed and developed. The proposed method have been tested in Chapter 4 with varieties of experiments. Chapter 5 provides summaries and conclusions and future work studies. The obtained data and developed MATLAB code was published in Appendix 1 and Appendix 2, respectively. Finally, all the reference sources are listed.

จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

CHAPTER 2: THEORY AND LITERATURE REVIEW

2.1. Physics theory

2.1.1. Interaction between gamma – ray radiation and detector

There are several of possible interaction mechanisms between gammaray and matter, but only three mechanisms are considered as major types in radiation detection and measurements, including: (i) *photoelectric effect*, (ii) *Compton scattering*, and (iii) *pair production*.

Photoelectric effect: The effect in which a gamma photon interacts with and transfers its energy to an atomic electron, causing the ejection of that electron from the atom. The kinetic energy of the resulting photoelectron is equal to the energy of the incident gamma photon minus the energy that originally bound the electron to the atom (binding energy). The photoelectric effect is the dominant energy transfer mechanism for X-ray and gamma ray photons with energies below 100 keV but it is much less important at higher energies.

Compton scattering: This is an interaction in which an incident gamma photon loses enough energy to an atomic electron to cause its ejection, with the remainder of the original photon's energy emitted as a new, lower energy gamma photon whose emission direction is different from that of the incident gamma photon. The probability of Compton scattering decreases with the increasing of photon energy. Compton scattering is thought to be the principal absorption mechanism for gamma rays in the intermediate energy range 100 keV to 10 MeV. Compton scattering is relatively independent of the atomic number of the absorbing material, which is why very dense materials like lead are only modestly better shields, on a per weight basis, than are less dense materials.

Pair production: This becomes possible with gamma energies exceeding 1.022 MeV, and becomes important as an absorption mechanism at energies over 5 MeV. By interaction with the electric field of a nucleus, the energy of the incident photon is converted into the mass of an electron-positron pair. Any

gamma energy in excess of the equivalent rest mass of the two particles (totaling at least 1.022 MeV) appears as the kinetic energy of the pair and in the recoil of the emitting nucleus. At the end of the positron's range, it combines with a free electron, and the two annihilate, and the entire mass of these two is then converted into two gamma photons of at least 0.511 MeV energy each (or higher according to the kinetic energy of the annihilated particles).



FIGURE 1. Three major types of gamma-ray interaction (from the Glenn F.Knoll. Radiation Detection and Measurement – [10])

2.1.2. Scintillation detector characteristic

Thallium doped Sodium Iodide NaI(Tl) is the most widely used scintillation material. NaI(Tl)is used traditionally in nuclear medicine, environmental measurements, geophysics, medium-energy physics, etc. The fact of its great light output among scintillators, convenient emission range, the possibility of large-size crystals production, and their low prices compared to other scintillation materials compensate to a great extent for the main Nal(TI) disadvantage. Which is namely the hygroscopicity, on account of which NaI(Tl)can be used only in hermetically sealed assemblies. Varying of crystal growth conditions, dopant concentration, raw material quality, etc. makes it possible to improve specific parameters, e.g., to enhance the radiation resistance, to increase the transparency, and to reduce the afterglow. For specific applications, low-background crystals can be grown. NaI(Tl)crystals with increased dopant concentration are used to manufacture X-ray detectors of high spectrometric quality.



FIGURE 2. Comparison of background detector response functions from PVT, NaI, and HPGE detector of various sizes [Runkle, R.C.,L. E Smith, et al. (2009). The photon haystack and emerging radiation detection technology. Journal of Applied Physics 106(4)]

NaI(Tl)is produced in two forms: single - crystals and poly-crystals. The optical and scintillating characteristics of the material are the same in both states. In some cases of application, however, the use of the polycrystalline material allows coping with a number of additional problems. First, a press forging makes it possible to obtain crystals with linear dimensions exceeding significantly than those of grown single crystals. Second, the poly-crystals are ruggedized, which is important in some cases. Moreover, NaI(Tl) poly-crystals do not possess the

perfect cleavage, so the probability of their destruction in the course of the use is reduced. The use of extrusion in converting NaI(Tl) into the polycrystalline state makes it also possible to obtain complex-shaped parts without additional expensive machining.

2.1.3. Masking scenario of Nuclear with other radioactive materials

Countering the illicit trafficking at the border control points is one of the prime purposes of global nuclear security program. When the cargo container trigger an alarm to the primary detection - radiation portal monitor, then will be checked at the secondary inspection by spectroscopic portal monitor and handheld equipment.

The philosophy of masking scenario is to make confusing of the detection system by using of the declared radioactive materials such as radiopharmaceuticals or industrial used, to hide the signal from illegal shipments of nuclear and other radioactive materials. There is a study showed that four different features in a gamma-ray spectrum that can be used for masking scenario, including: (i) Backscatter peak, (ii) Compton scattering continuum, (iii) Compton edge, and (iv) full energy peak [13].



FIGURE 3. CZT measured gamma-ray spectrum of Cs-137 showing four regions of the spectra which can potentially be used for masking scenario [13].

Masking based on the backscatter peak: An emitted energy of Compton backscatter peak caused by the interaction by Compton scattering between the material surrounding the detector and the original emitted from the source is given by the equation 1:

$$E_{\gamma,(Bck,Sc)} = \frac{E_{\gamma}}{1 + \left[\frac{2 \times E_{\gamma}}{511}\right]}$$
(1)

In which: E_{γ} is a gamma-ray energy that emitted from the radiation source IN keV, E_{γ} ' is an energy of backscatter peak in keV.

The main emission energy of U^{235} is 186 keV with the abundance of 57.2 % which is the result of backscatter peak of energy of $E_{\gamma} = 684$ keV, that near the emitted gamma-ray energy of Cesium source - Cs^{137} . This phenomenon raised an issue that the detection system will be confused for determining the energy peak of 186 keV whether resulted from U^{235} or backscatter peak from Cs^{137} . The phenomenon can be accomplished by means of peak – ratio approach, further discussed in part 3.6, Chapter 3.

Masking based on the Compton scattering continuum and Compton edge: the philosophy behind is that the full peak energy of nuclear or other radioactive materials will be interfered by high continuum under the peak. Several algorithms such as Library Gamma – M, LibCoNid... are determine the peak by estimating the ratio between the full peak area and continuum under the peak that need to higher than certain level, so if the continuum under the peak is too high, the full energy peak has not be taken into account. This problem can be solved by changing the criterial for peak determining by comparing with the background continuum at the same channels, further discussion in Chapter 3.

Masking based on the full peak energy: in case U^{235} , the main emission energy is the same with Radium -226. There are several technique to differentiate

between these radioisotope such as X-Ray florescence or X-Ray Imaging, but out of the scope of this study.

2.1.4. Screening method at border control procedure

Standard operating for screening method using passive detection for nuclear and radioactive materials can be categorized into three processes as follows:

(i) – *Primary detection*: the first process to detect the presence of gamma and/or neutron radiation that emitted from radioactive materials by means of Radiation Portal monitor. The operator has legal authorize to search people and vehicle, check manifest to verify the alarm and perform further inspection as necessary.

(ii) – Secondary inspection: locate and identify the source of radiation to assess the level of threat according to verified alarm event whether innocent or non-innocent.

(iii) – *Expert support:* high level analysis by the senior expert team and handheld equipment to confirm illicit trafficking case and declared threat.

There are normally three types of alarm that can be defined in screening method for nuclear and radioactive material, including: (i) False alarm that results from a statistical count rate variation of the back ground and/or the change in the actual ambient background count rate, (ii) Innocent alarm which is a valid alarm due to the presence of naturally occurring radioactive materials - NORM or technologically enhanced naturally occurring radioactive materials -TENORM or legal medical or industrial radionuclides, and (iii) Real alarm results from the presence of illegally transported radioactive materials.



FIGURE 4. Example of Standard operating Procedure (SOP) [14]

Algorithm's performance can be expressed in various ways, but most simply how far the algorithm can detects all isotopes that are present in the set up experiment and does not detect any isotope that not in the source. To evaluate the performance of difference method, there are two terms of detection ability can be used, which are detection rate (DR) and false alarm rate (FA). The detection rate is defined as the percentage of the scenarios that all of the constituent radioisotopes have successfully been identified by the proposed method, i.e. when the corresponding peaks on the spectrum are higher than the peak threshold. The false alarm rate is defined as the percentage of the scenarios that at least one radioisotope has been identified even if it was not actually present in the experiment.

The alarm setting is defined as the peak threshold amplitude, that must be considered the statistical fluctuations either radiation source count rate or radiation background count rate and depending on the scan time and many other parameters. The alarm setting is inversely proportional with the false alarm rate but also affected to the detection rate especially in case of poorly resolved signal, for instance, if the peak threshold too low, the false alarm will be increased, if the peak threshold too high, the false alarm rate and the detection rate will be decreased.

The radiation alarm level of screening instruments should be set up so the false alarm rate is minimized as low as possible in considering to the detection rate, in this study, the alarm setting will be changed for estimating the algorithm performance.

2.2. Peak identification algorithm review

Most current *Radio-isotope identification algorithms (RIID) for NaI(Tl) detector* can be categorized into few distinct approaches including expert interactive, simple library comparisons, region of interest (ROI) method, peak finding and characterization, and template matching [2]. Library comparisons, ROI method and template matching are difficult due to the large size of the template spectrum library. In these cases, the library is reference source which used to determine the degree of similarity between some measured characteristics of the spectrum and reference radio-isotope information; Peak finding and characterization is a more popular method, that mainly relies on 4 steps: (i) differentiate to find the local extreme (ii) assess and identify the peaks (iii) fit the detected peak to a Gaussian function and determine the peak characteristics. Nevertheless, study shows that *current RIID algorithms for NaI(Tl)* detector is considerably *worse than desired* [2]. *Detection algorithms* like energy window technique are commonly used for the large size plastic scintillation detector at seaport. This technique only generates alarm in case of observed spectrum is different from background, but without providing additional information which radionuclides are presence. Detection algorithms can be formulated as the simplified version of identification algorithms, algorithms which can figure out the characteristics of suspected or hidden radionuclides.

Identification algorithms, in most cases, can be categorized into 3 types: (i) *Classification* – processing the spectrum into several classes, such as background, NORM, and SNM as opposed to an estimation of source strength and exact composition (ii) *Estimation* – determine the relationship between the observed data (**Y**) and the reference library (**X**) and (iii) *Hybrid approach* – combination of Classification and Estimation [3].

In *Identification – Estimation algorithms*, there are several available methods to perform, for instance: peak of interest method focus on sub-group of concerned radionuclides, like SNM, in observed spectrum; full spectrum method identify all the isotopes present in the spectrum when the signal in excess of background signal. In the context of Nuclear Security Mission in border control, the *Hybrid approach* which generates an alarm for either Industrial or Medical uses group or SNM and its quantities, is more preferred.

2.2.1. Peak detection method

Peak detection method using local maxima detection algorithm developed by T. O'Haver is one of the effective command-line function to locate and measure the positive peaks in a noisy data sets. It detects peaks by looking for downward zero-crossings in the smoothed first derivative that exceed slope threshold and peak amplitudes that exceed amplitude threshold, and determines the position, height, and approximate width of each peak by least-squares curvefitting the top part of the peak. Returns a list (in matrix **P**) containing the peak number and the estimated position, height, width, and area of each peak. This algorithm can find and measure over 1000 peaks per second in very large signals.

The data are passed to the findpeaksG function in the vectors x and y (x = independent variable, y = dependent variable). The other parameters are user-adjustable as follows:

SlopeThreshold - Slope of the smoothed first-derivative that is taken to indicate a peak. This discriminates on the basis of peak width. Larger values of this parameter will neglect broad features of the signal. A reasonable initial value for Gaussian peaks is 0.7*WidthPoints^-2, where WidthPoints is the number of data points in the half-width of the peak.

AmpThreshold - Discriminates on the basis of peak height. Any peaks with height less than this value are ignored. This is one of the drawback of this algorithm, when the optimum threshold is very difficult to determine, even it is not work in case of masking scenario where the signal from Nuclear materials were the peak to total ratio is small.

SmoothWidth - Width of the smooth function that is applied to data before the slope is measured. Larger values of SmoothWidth will neglect small, sharp features. A reasonable value is typically about equal to 1/2 of *the number of data points* in the half-width of the peaks.

FitWidth - The number of points around the "top part" of the (unsmoothed) peak that are taken to estimate the peak heights, positions, and widths. A reasonable value is typically about equal to 1/2 of *the number of data points* in the half-width of the peaks. The minimum value is 3.

Smoothtype: determines the smoothing algorithm, if smoothtype=1, rectangular (sliding-average or boxcar), if smoothtype=2, triangular (2 passes of sliding-average) if smoothtype=3, pseudo-Gaussian (3 passes of sliding-average). Basically, higher values iyield greater reduction in high-frequency noise, at the expense of slower execution.

2.2.2. Library (Gamma-M) Peak Locate

The Library (Gamma-M) Peak Locate is one of the algorithms that used in Genie 2000 software which is best suited for NaI(Tl) analysis with only specific nuclides are of interest. The algorithm includes several steps of determining the background continuum by means of erosion technique and determining the presented radionuclide is obtained spectrum.

The Library (Gamma-M) Peak Locate algorithm is consist of four main steps, including: (i) Peak erosion step to estimate a continuum area of the background of the spectrum by divided into energy bins depending on low or high channel, then grouped bins and modified eight times, finally reconstruct the continuum background and subtracted from original spectrum, (ii) Peak fitting step using of least square fit for Gaussian function, (iii) Unknown peak locate output is a quantity that is proportional to the ratio of peak area and detection limit as a results of averaging of the data over a region that is proportional to the detector resolution, and (iv) Peak rejection criterial compare the valid peak from the spectrum with the library location with tolerances for variations in the calibrations [16].

> จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

Gamma - 17, NU + Ba-13	3 20cm.5s.CNF	
		าคามุล
Idle Channel: 167 :	324.6 keV Counts: 115	Preset: 5/5.00
Acquire Start Stop		🖾 Gamma-M Library Peak Locate Setup 🛛 🔀 🕨
Expand On Clear		Search Region Start Channet 1 Stop Channet 1024
ROI Index:		Nuclide Library: C:\GENIE2K\CAMFILES\Stdlib.nlb Select
Datasource		Gain Shift Correction Use Stored Library
Prev Next		I Include Unknown Search
		Energy Tolerance: 1.50 FWHM 🔽 Generate Report
		Cancel More Help Evenue
		🔤 More Gamma-M Locate Setup
		Window Settings
		Area: 1 Variance: 1 Interference: 1.5 Background: 1
	÷	Overlap: 0.3 Nbr. Bkgnd Terms: 2
	a a constantino de la	Gain Correction Reject MDA
		Nax Nbr. Gain Passes: 10 Sigma: 2.5 Reject Gain Factor: 0.005 Constant: 3
		OK Cancel Help
	and the second	
Next	Acq. Start: 1/19/1998 4:07:03 AM	Elapsed Preset
Prev	Comp. Preset Region: Real (secs.):	5.710 0
	0 · 0 (channels) Total (cnts.):	0.00 0

FIGURE 5. Library (Gamma-M) Peak Locate algorithm user interface

The Library (Gamma-M) Peak Locate algorithm may be challenged in case of the measured signal is poorly resolved and if the Compton part under the interested peak is high.

2.2.3. Library correlation nuclide identification algorithm

Library correlation nuclide identification algorithm (LibCorNID) is the method that developed by William R. Russ that used in identiFINDER R400 radiation detector [15]. The algorithm including several key steps, as following: (i) using of peak erosion technique after background compensation for estimating the continuum baseline to isolate the valid peak from the original spectrum, (ii) performing nuclide filtering for interested peas which are constructed in accordance with energy, branching ratio, shape of peak and efficiency calibrations and reconstruct spectra as the data set of candidate nuclide peak spectra, (iii) applying tolerance for calibrations errors to fit constructed spectra to the isolated measured peak data, and (iv) Matching by normalized

correlation coefficient with a threshold for the reference shape library and the corrected measured shape. The block diagram of LibCorNID algorithm are showed in figure 6.



FIGURE 6. Block diagram of LibCorNID algorithm [15]

The performance of LibCorNID algorithm may decrease when the measured signal is poorly resolved and in case of Compton part under the interested peak is high.

2.3. Wavelet transform approach for poorly resolved signal

Wavelet methods are the most popular used approach for noise removal from poorly resolved signal. The application areas of wavelet transform are speed signal, biomedical signal analysis, producing and analyzing irregular signals or images, wavelet modulation in communication channels, in video coding and forecasting etc.

There are many types of noise in the signal from various source, such as from the detection system, measurement...etc. But all the noise source have the same characteristic of high frequency, so if the original signal can break into high and low frequency domain then eliminate the high frequency part, equivalent to the noise were removed.

There are two popular wavelet transform approaches that applying for denoised signal, including: (i) Continuous wavelet transform (CWT) used to break a continuous time function into time and frequency domain by contraction and dilatation of the wavelet functions, and (ii) Discrete wavelet transform (DWT) performed a filter banks with difference bandwidth for signal decomposition and reconstruction of the signal after thresholding [20]. In this study the DWT was employed for transformation and decomposition of radionuclide spectra in application of time series analysis. Figure 6 shows the two-level multi-resolution analysis.

In the first step of DWT - decomposition, the original signal was downsampled by 2 through high pass filter **H_filter** and low pass filter **L_filter**, and the results is two new coefficient named as **Signal_L1** and **Wn**, in which **Signal_L1** is approximation coefficient as a result of original signal passed through low pass filter, W_n is the detail coefficient as a result of original signal passed through high pass filter. The algorithm decomposed **Signal_L1** into **Signal_L2** and W_{n-1} . In the second step, reconstruction, wavelet thresholding is applied to the approximation coefficient (**Signal_L2**, **Signal_1'**) and detail coefficients (W_n , W_{n-1}) to up-sampling by 2 and again recombined for reconstructing the noise free signal **Signal'** as show in figure 7.



FIGURE 7. Two level of wavelet decomposition and reconstruction tree

2.4. Theoretical background

Detection efficiency: The number of received counts (background subtracted), denote as **C**, can be defined as follows [9]:

$$\mathbf{C} = (\mathbf{A}.\mathbf{f}).(\mathbf{G}.\ \boldsymbol{\varepsilon}).\mathbf{S} \qquad (2)$$

where:

A: Activity of the source (Bq),

f: Branching ratio,

G: Counting geometry,

 ϵ : Intrinsic efficiency of detector, and

S: Shielding factor.

In the context of field measurement and estimation of radioactive sources, the number of counts in each channels, **C**, is the values that can obtain from

system; activity of the source, \mathbf{A} , is the parameter that we need to find out. From the equation (5), the relationship between \mathbf{S} and \mathbf{A} are influenced by a several number of factors in the measurement – they are needed to evaluate by parameters or variances in calculation model.

The *branching ratio* is a term that used to characterize the probability of different decay modes, and in gamma-ray spectrum analysis, this parameter is used to estimate the amount of distribution of source to the spectrum in certain channels because each of radio-isotopes emitted different characteristic gamma-ray energies in accordance with branching ratio values.

The efficiency of a detector is a measure of how many pulses occur for a given of gamma-ray. There are many kinds of *detection efficiency* are in common use for gamma-ray detector [10] (i) *absolute efficiency* referred to the fraction between a number of pulses recorded and number of radiation emitted by source in all directions, which depends on either a detector response function and a counting geometry - a relative position of the source and detector; (ii) *intrinsic efficiency* presented the ratio of a number of pulse recorded and number of radiation striking the detector, consequently depends only on detector properties (iii) *Relative efficiency* is the efficiency of one detector to another and (iv) *full-energy peak efficiency* for producing full-energy peaks pulse rather than a pulse of any size for the gamma-ray. The *absolute* and *intrinsic* efficiencies have simply relationship is given as following equation [10]:

$$\boldsymbol{\varepsilon}_{abs} = \boldsymbol{\varepsilon}_{int}.\,\boldsymbol{G} = \boldsymbol{\varepsilon}_{int}.\,(\Omega/4\pi)$$
 (3)

In which, **G** is the *counting geometry*, Ω is the solid angle of the detector has subtended by the detector at the source position (in steradians). For the common case of a point source located along the axis of a right circular cylindrical detector, denoted **d** as source-detector distance, **a** as detector radius, Ω is shown as the sketch and given by equation as following [10]:

$$\Omega = 2\pi \left(1 - \frac{d}{\sqrt{d^2 + a^2}}\right) (4)$$

For **d** >>**a**, the solid angle reduces to [10]:

$$\Omega \cong \frac{\pi a^2}{d^2} \quad (5)$$

From (6) and (8), the geometry parameter, **G**, can be expressed by:

$$G = \frac{\pi a^2}{4\pi d^2} = \frac{A_d}{4\pi d^2} \quad (6)$$

Where A_d is the detector sensitivity surface area.

Signal-to-Noise ratio, generally represented by SNR or S/N is widely used for characterizing the response function of the detector. SNR is defined as the ratio between desired signal (useful information) and the level of background (noise). In decibels, the SNR is given by:

$$SNR = 10 \log_{10} \left(\frac{\text{signal}}{\text{background}} \right) \quad (7)$$

2.5. Liner regression

Gamma-ray emission of radioactive materials is a randomly process, and can be formulated using Poisson distribution. There are several studies on *gamma emission modeling* by linear equation [3-6].

The linear equation for gamma-ray emission can be written as:

$$Y = X. \beta^* + V \tag{8}$$

Where:

$$Y = \begin{pmatrix} y_1 \\ \vdots \\ y_n \end{pmatrix}; X = \begin{bmatrix} X_1, X_2, \dots, X_p \end{bmatrix} = \begin{bmatrix} x_{11} & \dots & x_{1p} \\ \vdots & \ddots & \vdots \\ x_{n1} & \dots & x_{np} \end{bmatrix}; \beta^* = \begin{pmatrix} \beta_1 \\ \vdots \\ \beta_p \end{pmatrix}; V = \begin{pmatrix} v_1 \\ \vdots \\ v_n \end{pmatrix}$$

In principle, an observed vector, \mathbf{Y} , is the number of counts in a spectrometer with n energy channels, which is given by $\mathbf{Y} = (y_1, y_2, ..., y_n)^T$, where the superscript T denotes the transpose of the vector, and the y_i 's are the counts in each channel.

A reference library matrix, \mathbf{X} , is the template spectra or a list of measured spectra from combination of expected isotope. \mathbf{X} is the matrix of (n x p), in which n is the energy channels, p is the total number of interested radionuclides (background is considered as a radionuclide). This modeling has two drawbacks: (i) it cannot be used if the template spectra are not available and (ii) it requires large number of template spectrum library with different combinations of isotopes.

 β^* is the contribution vector of p radioisotopes to the spectrum. Because the actual counts from the source are random and follow a Poisson distribution, which are different from observed mean counts in the system, therefore vector **V** is considered as the random noise in each channel.

Linear regression technique assuming that the reference library **X** is measured without error, the vector of noise **V** has zero mean and close to Gaussian distribution, one can solve the linear equation (8) to estimate the component β^* . Zero β_j is equivalent to the absence of radioisotope j^{th} .

Most of radionuclides have more than one gamma-ray characteristic peak. If the j^{th} isotope has m characteristic peaks, the reference library can be expressed as linear combination of the m sub-spectra. Then the reference library matrix **X** now will has the following form:

$$X = \begin{bmatrix} X_1, X_2, \dots & X_j & \dots, X_p \end{bmatrix}; X_j = \begin{bmatrix} x_{11} & x_{12} & \dots & x_{1m} \\ x_{21} & x_{22} & \dots & x_{2m} \\ \vdots & \vdots & \ddots & \vdots \\ x_{n1} & x_{n2} & \dots & x_{nm} \end{bmatrix}$$
(9)

Each column of X_j gives the mean gamma-ray counts per time unit and per unit of source material in n channel.

Effect of shielding thickness

Define t as the mass thickness of shielding material, μ_i is the attenuation coefficient of the *i*th energy level of isotope *j*th. Isotope *j*th has m characteristic gamma-ray peaks. For a known shielding material and mass thickness t, the matrix of attenuation is defined as follows [5]:

$$S = \begin{bmatrix} S_1 & 0 & \dots & 0 \\ 0 & S_2 & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & S_p \end{bmatrix}; S_j = \begin{bmatrix} e^{-\mu_1 t} & 0 & \dots & 0 \\ 0 & e^{-\mu_2 t} & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & e^{-\mu_m t} \end{bmatrix}$$
(10)

If each isotope has m characteristic peaks, thus the contribution vector $\boldsymbol{\beta}^*$ becomes:

$$\boldsymbol{\beta}^{*} = \begin{pmatrix} \boldsymbol{\beta}_{1} \\ \boldsymbol{\beta}_{2} \\ \vdots \\ \boldsymbol{\beta}_{p} \end{pmatrix}; \ \boldsymbol{\beta}_{j} = \begin{pmatrix} \boldsymbol{\beta}_{j1} \\ \boldsymbol{\beta}_{j2} \\ \vdots \\ \boldsymbol{\beta}_{jm} \end{pmatrix}; \quad (11)$$

Contribution vector under shielding condition, denoted as B_p , can be expressed by:

$$\boldsymbol{B}_{p} = \begin{pmatrix} \boldsymbol{B}_{1} \\ \boldsymbol{B}_{2} \\ \vdots \\ \boldsymbol{B}_{p} \end{pmatrix} = \boldsymbol{S} \cdot \boldsymbol{\beta}^{*} ; \boldsymbol{B}_{j} = \begin{pmatrix} \boldsymbol{B}_{j1} \\ \boldsymbol{B}_{j2} \\ \vdots \\ \boldsymbol{B}_{jm} \end{pmatrix} = \boldsymbol{S}_{j} \cdot \boldsymbol{\beta}_{j} \quad (12)$$

The linear equation (6) becomes:

$$Y = \sum_{j=1}^{p} X_j B_j + V \quad (13)$$

The shielding effects in the measurement can be estimated only when the information of the shielding materials and their thickness are available.



CHAPTER 3: APPLIED PEAK DETECTION ALGORITHM AND SOFTWARE

This chapter introduces to the proposed method that using for identification of radioactive materials for a given spectrum. For assumption of the measured spectrum is poorly resolved, low SNR, the proposed method will have ability to remove the noise, extract the useful and valid information and available to identify which radioisotope that present in the measured spectrum. The proposed method are programmed in Matlab, the script of the program can be found in Appendix 2.



FIGURE 8. Block diagram for proposed peak detection algorithm

The input for the proposed method are the measured spectrum, which may be poorly resolved and is taken under specific condition (scan time and distance), and the background spectrum which is taken in an absence of radiation source. After applying the Identification Algorithm, the output is a Radioisotope Identification results.

The proposed method includes five steps: noise reduction, background subtraction from the signal, determination of the baseline spectrum for isolate the valid peaks, application of peak search algorithm to identify the valid peaks from the signal, and application of the linear regression model for matching between the reference library and the recognized peak to verify whether the interested radionuclide that presence in the measured spectrum or not. Figure 8 shows the block diagram for the proposed method.

3.1. Noise Reduction

Most of the high-frequency components in the spectrum are noise, hence the spectrum can be decomposed by wavelet transform into high- and lowfrequency components in order to remove the high-frequency component [17]. In this study, the discrete-wavelet transform (DWT) based algorithm is used for spectrum de-noising.



FIGURE 9. Spectra of a natural Uranium and Cs-137 mixed source in accordance with each identification step: (a) measured spectrum under low-SNR condition (from a distance of 5cm, and 2s scan time), (b) de-noised spectrum after 3 levels of wavelet decomposition, (c) smoothed spectrum and the determined baseline, and (d) subtracted baseline spectrum

Figure 9(b) shows the result of noise reduction by 3-level DWT. At each level of decomposition, the signal is passed through the half-band high- and low- pass filters and subsampled by 2, giving the detail and approximation coefficients respectively. The approximation coefficients are then used as an input for the next level decomposition.

To de-noise the signal, the detail coefficients are set to 'zero' after applying the wavelet transform. Then the inverse wavelet transform is performed to recover the original signal which is free from noise components [17].

3.2. Background subtraction

The algorithm performs the subtraction of background at each channel of the measured spectrum. Due to the fluctuation of the background and the radiation sources itself, in case that the number of counts of the background in one channel is higher than the number of counts in the measured spectrum, the result of subtraction is set to zero.

3.3. Baseline Estimation

Figure 9(c) shows the baseline continuum estimation of the de-noised spectrum. The general concept underlying this method has already been described by Nikolaos Kourkoumelis [17]. The spectrum baseline determination algorithm proposed in this study is in essence analogous but has different processing and implementation. The proposed method consists of three steps, as following:

+ Firstly, the de-noised spectrum is smoothed by Pseudo-Gaussian smooth method to remove the rough part of the signal.

+ Secondly, the minimal point set of the spectrum is constructed with a valley check method by skipping the first minimal points in the first few channels [18].

+ Finally, the spectrum continuum baseline is constructed under two conditions: (i) for the channels that are located inside the minimal point set, the
baseline is drawn by linear interpolation between the two adjacent minimal points, (ii) for the channels that are located outside the minimal point set, the baseline is equal to the number of counts on the smoothed spectrum.

29

3.4. Peak search

This research use the local maxima detection method developed by T. O'Haver [7]. In the context of radioactive material detection at border control, especially in the case of nuclear material masking scenario, the local maxima detection method may have a drawback when identifying the low energy peak of nuclear material that lays on the high Compton continuum part. The peak height threshold applied in this study is three to ten sigma or square root of the background counts at the same channel. In this study, the peak search algorithm by local maxima method was performed after the spectrum baseline estimation and thresholding.

Figure 10 shows the comparison between peak threshold algorithms by applying the number of sigma with two difference methods: the spectrum background and the net counts under the peak which is determined by spectrum baseline algorithm. In case that the signal is poorly resolved and low, the detection rate of the proposed identification method can improve by applying the peak thresholding using of spectrum background method.

3.5. Linear Regression Equation

Gamma-ray emission modeling

Without shielding effect

Given a spectrometer with n = 1024 energy channels, the observed counts in each of energy bins is $\mathbf{Y} = (\mathbf{y_1}, \mathbf{y_2}, ..., \mathbf{y_n})^T$. Denoted \mathbf{p} , \mathbf{G} , $\boldsymbol{\varepsilon}$ is the number of radio-isotopes that need to be estimated activity whether they are presence in the observed spectrum, the counting geometry of the measurement, and the detection efficiency, respectively.



FIGURE 10. Example of peak thresholding by background and spectrum baseline method for natural Uranium spectrum

An operation library matrix $\mathbf{F} = (\mathbf{F_1}, \mathbf{F_2}, \dots, \mathbf{F_p})$; $\mathbf{F_j} = (\mathbf{f_{1-j}}, \mathbf{f_{2-j}}, \dots, \mathbf{f_{n-j}})^{\mathrm{T}}$, $j=(1, 2, ..., \mathbf{p})$ represents the value of branching ratio of characteristic gammaray energy of interested isotope j^{th} in n channel, $\mathbf{f_{n-j}}$ here means component of \mathbf{n}^{th} channel for j^{th} isotope. Matrix $\mathbf{F_j}$ played a role of establishing interested library of isotopes those that need to be determined in detection, such as SNM and some mixing radioactive sources. Matrix $\mathbf{F_j}$ can be developed based on the characteristic gamma-ray energy and branching ratio of the isotope, for example, Co-57 has two characteristic gamma-ray energy of 122 keV and 136 keV with the branching ratios are 85.6 and 10.7 respectively, so the vector $\mathbf{F_{Co-57}}$ is given by equation (14).

$$\mathbf{F_{Co-57}} = \begin{pmatrix} \mathbf{f_{11-1}} \\ \mathbf{f_{21-1}} \\ \vdots \\ \mathbf{f_{121-1}} \\ \mathbf{f_{122-1}} \\ \mathbf{f_{122-1}} \\ \mathbf{f_{123-1}} \\ \vdots \\ \mathbf{f_{135-1}} \\ \mathbf{f_{136-1}} \\ \mathbf{f_{136-1}} \\ \mathbf{f_{137-1}} \\ \vdots \\ \mathbf{f_{1024-1}} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ \vdots \\ 0 \\ 10.7 \\ 0 \\ \vdots \\ 0 \end{pmatrix}$$
(14)

The operation library matrix \mathbf{F} of group of the radioisotopes that used in this study can be found in Appendix 1.

Vector $\mathbf{Z} = (\mathbf{z_1}, \mathbf{z_2}, ..., \mathbf{z_n})^T$ is the background vector, and the $\mathbf{z_i}$'s is the counts of background in each channel.

A vector of contribution $\mathbf{A} = (\mathbf{A}_1, \mathbf{A}_2, ..., \mathbf{A}_p)^T$, where A_j 's is the activity of radioisotopes in expected library.

A noise vector $\mathbf{V} = (\mathbf{v}_1, \mathbf{v}_2, \dots, \mathbf{v}_n)^T$, which has several properties (i) zero mean, (ii) Gaussian distribution and (iii) each component is independent.

The linear equation for gamma-ray emission can be expressed as:

$$\boldsymbol{Y} = (\boldsymbol{G}.\boldsymbol{\varepsilon}).\,\boldsymbol{F}.\,\boldsymbol{A} + \boldsymbol{Z} + \boldsymbol{V} \qquad (15)$$

where:

$$Y = \begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{pmatrix}; F = \begin{bmatrix} f_{11} & f_{12} & \cdots & f_{1p} \\ f_{21} & f_{22} & \cdots & f_{2p} \\ \vdots & \ddots & \ddots & \vdots \\ f_{n1} & f_{n2} & \cdots & f_{np} \end{bmatrix}; A = \begin{pmatrix} A_1 \\ A_2 \\ \vdots \\ A_p \end{pmatrix};$$

$$\mathbf{Z} = \begin{pmatrix} \mathbf{Z}_{1bg} \\ \mathbf{Z}_{2bg} \\ \vdots \\ \mathbf{Z}_{nbg} \end{pmatrix}; \mathbf{V} = \begin{pmatrix} \mathbf{v}_1 \\ \mathbf{v}_2 \\ \vdots \\ \mathbf{v}_n \end{pmatrix}$$

For given type of detector, the counting geometry can be considered as parameter, which is only depended on the distance between detector and unknown radiation sources. In case of screening for $\mathbf{a} \times \mathbf{b} \times \mathbf{c}$, \mathbf{d} (m) containers at seaport, \mathbf{a} , \mathbf{b} , \mathbf{c} and \mathbf{d} is the length, height, width and thickness layer of the container, respectively. The size of the container shows as the sketch below.



Assuming that an inspector follows screening procedure such as (i) go a long with the length of container and (ii) stop at the highest received signal position, therefor the boundaries limitation for the source location inside the container is from $\mathbf{0}$ (m) to \mathbf{c} (m) in height and from \mathbf{d} (m) to \mathbf{b} (m) in width.

Shielding effect

There are two difference approaches to model the shielding effect, as follow:

Approach 1:

Defined a shielding matrix S has p components, it represented to the attenuation of p isotopes under shielding conditions and the last component showed that background counts could not be affected by shielding. The shielding matrix S can be expressed by:

$$S = \begin{bmatrix} S_1 & 0 & \dots & 0 \\ 0 & S_2 & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & S_p \end{bmatrix}; S_j = \begin{bmatrix} e^{-\mu_1 jt} & 0 & \dots & 0 \\ 0 & e^{-\mu_2 jt} & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & e^{-\mu_n jt} \end{bmatrix}$$
(16)

Denoted \mathbf{F}^* is the operation library under shielding conditions, matrix \mathbf{F}^* needs take into account the exponential attenuation for all the characteristic gamma-ray energies of the radioisotopes, thus matrix \mathbf{F}^* will be expressed as following form:

$$\mathbf{F}^{*} = \begin{bmatrix} F_{1}^{*}, F_{2}^{*}, \dots, F_{p}^{*} \end{bmatrix} = \begin{bmatrix} f_{11} \cdot e^{-\mu_{11}t} & \cdots & f_{1p} \cdot e^{-\mu_{1p}t} \\ f_{21} \cdot e^{-\mu_{21}t} & \cdots & f_{2p} \cdot e^{-\mu_{2p}t} \\ \vdots & \ddots & \cdots \\ f_{n1} \cdot e^{-\mu_{n1}t} & \cdots & f_{np} \cdot e^{-\mu_{np}t} \end{bmatrix}$$
(17)

Matrix \mathbf{F}^* can be also calculated from operation library \mathbf{F} and shielding matrix \mathbf{S} as following:

$$\mathbf{F}_{j}^{*} = \mathbf{S}_{j} \cdot \mathbf{F}_{j}$$
(18)
=> $\mathbf{F}^{*} = [(\mathbf{S} \cdot \mathbf{F})^{T}]^{T}$ (19)

The linear equation for gamma-ray emission (16) becomes:

$$\boldsymbol{Y} = \left((\boldsymbol{G}.\boldsymbol{\varepsilon}). \left[(\boldsymbol{S}.\boldsymbol{F})^T \right]^T \right) \cdot \boldsymbol{A} + \boldsymbol{V}$$
(20)

Approach 2:

Because the attenuation of gamma-ray both depended on energy levels and shielding characteristic (shielding material and thickness). Therefore, the shielding matrix S can be defined as follows:

33

$$\boldsymbol{S} = \begin{bmatrix} \boldsymbol{e}^{-\mu_{11}t} & \cdots & \boldsymbol{e}^{-\mu_{1p}t} \\ \boldsymbol{e}^{-\mu_{21}t} & \cdots & \boldsymbol{e}^{-\mu_{2p}t} \\ \vdots & \ddots & \cdots \\ \boldsymbol{e}^{-\mu_{n1}t} & \cdots & \boldsymbol{e}^{-\mu_{np}t} \end{bmatrix}$$
(21)

Consequently, the operation matrix under shielding conditions \mathbf{X}^* will have following form:

$$\mathbf{F}^{*} = \begin{bmatrix} F_{1}^{*}, F_{2}^{*}, \dots, F_{p}^{*} \end{bmatrix} = \begin{bmatrix} f_{11} \cdot e^{-\mu_{11}t} & \cdots & f_{1p} \cdot e^{-\mu_{1p}t} \\ f_{21} \cdot e^{-\mu_{21}t} & \cdots & f_{2p} \cdot e^{-\mu_{2p}t} \\ \vdots & \ddots & \cdots \\ f_{n1} \cdot e^{-\mu_{n1}t} & \cdots & f_{np} \cdot e^{-\mu_{np}t} \end{bmatrix} = F^{\circ}S \quad (22)$$

Where ° is the Hadamard product.

3.6. Peak ratio comparison





For the scenario of masking nuclear material by backscatter peak as mentioned in part 2.1.3, chapter 2, the problem is that the false alarm for energy peak of 186 keV of Uranium – 235 will be increased due to the backscatter peak from Cesium – 137 radiation source or the real alarm can dissemble.

To overcome this matter, the ratio comparison of a full peak energy of 662 keV from Cesium – 137 and the backscatter peak can be used. Denoted P_T and P_B is the net counts under the full energy peak and the backscatter peak of Cesium – 137, respectively. Depending on radiation source's activity and counting geometry, the full energy peak and the backscatter peak will be changed but proportional. To determine the optimum ratio of P_B/P_T for reduce the false alarm rate, several spectrum of Cesium – 137 was collected and tested.

CHAPTER 4: EQUIPMENT, MATERIALS AND EXPERIMENT SET UP

This chapter describes the equipment and materials that are used for estimating the performance of the proposed method.

4.1. Equipment and Materials

Detector: The 3 inches x 3 inches NaI(Tl) detector by BICRON shows in figure 12 is used for the measurement of gamma-ray spectra. The energy range and resolution of the detector are 10 to 30000 keV and 6.5% FWHM at 662 keV respectively. It has a proven record of long term reliability and stability [19].



FIGURE 12. The BICRON 3" x 3" NaI(Tl) detector

The 3" x 3" NaI(Tl) crystal is covered by aluminum housing and connected to photomultiplier tube, an internal magnetic/light shield, a high-voltage power supply, a preamplifier, an amplifier, a multichannel analyzer (MCA), and computer. Figure 13 shows the experiment set up diagram.

Nuclear material sources

This study used two types of nuclear material sources as following:

- 8.4 grams of natural Uranium (NU) formed in metal sheet by a size of 50 mm x 50 mm, in thickness of 0.175 mm, 0.71% U-235;

- Fresh TRIGA Mark III fuel of TRR/M-1 Thai research reactor: 20% U-235 enriched, diameter 3.63 cm (1.43 inches), length 55.88 cm (22 inches).



FIGURE 13. The diagram of detection system for NaI(Tl) detector

Radioactive mixing sources: The test mixing sources used to evaluate the performance of the proposed method consisted of five common industrial radioisotopes as showed in table 1, including: Co-57, Co-60, Ba-133, Cs-137, and Am-241. The activity and the characteristic peaks of these source showed in Table 1.

No.	Source's name	Activity (μCi)	Date	Half-life (years)	Energy (keV)	Branching ratio (%)
1	Cobalt–57	10.93	15/04/2015	0.74	122.06	85.60
2	Cobalt–60	10.32	15/04/2015	5.272	1173.22	99.85
					1332.49	99.98
3	Barium–133	10.86	15/04/2015	10.5	356.01	62.05
4	Cesium–137	10.28	15/04/2015	30.17	661.65	85.10
5	Americium– 241	1.132	1/11/1987	432	59.54	35.90

TABLE 1. The test mixing sources for the experiments

4.2. Experiments set-up

The detector was installed in a fixed location and various sets of radioisotopes were placed in front of the detector at various distances (5, 10, 15, 20, 35 and 60 cm). The scan times used for each location were varied (2, 5, 10, 20 and 30 seconds).



FIGURE 14. The diagram of experiment set-up for NaI(Tl) detector

Totally 480 spectrum were collected for testing the performance of the proposed method.

จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

Radioisotopes set up	Name of Radioisotope	Number of Spectrum
Single Source	Natural Uranium	1 - 30
	Cobalt–57	31 - 60
	Cobalt–60	61 - 90
	Barium–133	91 - 120
	Cesium–137	121 - 150
	Americium–241	151 - 180
Mixing source	Natural Uranium + Cobalt–57	181 - 210
	Natural Uranium + Cobalt–60	211 - 240
	Natural Uranium + Barium–133	241 - 270
	Natural Uranium + Cesium–137	271 - 300
	Natural Uranium + Americium–241	301 - 330
	Fresh Fuel + Cobalt–57	331 - 360
	Fresh Fuel + Cobalt–60	361 - 390
	Fresh Fuel + Barium–133	391 - 420
	Fresh Fuel + Cesium–137	421 - 450
	Fresh Fuel + Americium–241	451 - 480
Shielding	Cesium–137	481 - 483
	Cesium–137 + 1mm Al	484 - 486
	Cesium–137 + 2 mm Al	487 - 489
	Cesium–137 + 3 mm Al	490 - 492
	Cesium–137 + 5 mm Al	493 - 495

TABLE 2. The radioisotope set up the experiments

The diagram of setting for the experiment was showed in figure 14 and table 2.



FIGURE 15. The experiments with natural Uranium in the Laboratory



FIGURE 16. The experiments with fresh TRIGA Mark III fuel of a TRR-1/M1 Thai Research Reactor at TINT

CHAPTER 5: RESULTS AND DISCUSSION

For estimating the performance of the proposed method, 495 measured spectrum from chapter 4 are used as input for the Matlab – based program. This study used the available commercial algorithm, Library (Gamma – M) (introduced in part 2.2, chapter 2) to compare the performance's results.

5.1. Results and discussion

In this study, there are two concepts for evaluating of the proposed method's performance: detection rate (DR) and false alarm rate (FA). The detection rate is defined as the percentage of the scenarios that all of the constituent radioisotopes have successfully been identified by the proposed method, i.e. when the corresponding peaks on the spectrum are higher than the peak threshold. The false alarm rate is defined as the percentage of the scenarios that at least one radioisotope has been identified even if it was not actually present in the experiment.

Experiments with natural Uranium

Table 3 shows detection rate and false alarm rate for the proposed method, and comparison with the Library (Gamma – M) method available in the Genie 2000 Spectroscopy Software [16]. At 3 sigma, the detection rate has been improved from 65.04 % to 99.19 % but the false alarm rate has been increased from 15.45 % to 28.46 % for the single source scenario. For the mixing source scenario, the detection rate has been improved from 48.78 % to 81.12 % however the false alarm rate has raised from 15.45 % to 20.28 %.

The results for 5 and 10 sigma are also present in Table 3. Although the detection rate of the proposed method has reduced when the number of sigma changed from 3 to 5 and 10, but the detection rate still higher than the Library (Gamma – M) method. At 10 sigma of the proposed method in comparison to 3 sigma of Library (Gamma – M) method, the detection rate has been improved

from 65.04 % to 83.74 % and the false alarm rate has been reduced from 15.45 % to 11.38 % for the single source scenario. For the mixing source scenario, the detection rate has been improved from 48.78 % to 75.44 % and the false alarm rate has decreased from 15.45 % to 12.11 %.

The results showed that by using of background peak thresholding technique (mentioned in part 3.4, Chapter 3), the detection rate of proposed method has increased because of the algorithm take into account all the low and poorly signals in the spectrum. However, the false alarm rate has increased due to the algorithm could not differentiate between the low peak came from the background fluctuation or real signal from radioactive sources.

TABLE 3. Detection rate and false alarm rate for the radioisotope

 identification of the poorly resolved gamma-ray spectrum of natural uranium

			Pe	eak Height	t Thresho	ld	
		3 sig	gma	5 sig	gma	10 si	gma
		DR(%)	FA(%)	DR(%)	FA(%)	DR(%)	FA(%)
Single Source	Proposed method	99.19	28.46	92.68	20.33	83.74	11.38
	Library (Gamma-M)	65.04	15.45				
Mixing Source	Proposed method	81.12	20.28			75.44	12.11
	Library (Gamma-M)	48.78	15.45				

At three sigma peak thresholding level, in comparison between single source and mixing source scenario, the detection performance has more challenged because of the identification algorithm need to recognize all the source that present in the spectrum instead of single characteristic peak, the detection rate of proposed method and Library (Gamm – M) method has dropped in comparison between single source and mixing source scenario. Because criterial for determination peak of Library (Gamm – M) method has peak fitting step using Gaussian model, the false alarm rates are the same between two scenarios for Library (Gamm – M) method but it has changed for the proposed method.

Experiment with fresh fuel

TABLE 4. Detection rate and false alarm rate for the radioisotope

 identification of the poorly resolved gamma-ray spectrum of fresh fuel

			Peak Height Threshold					
		3 sigma		5 sigma		10 sigma		
		DR(%)	FA(%)	DR(%)	FA(%)	DR(%)	FA(%)	
Mixing Source	Proposed method	71.16	30.24	67.15	24.92	59.83	17.22	
	Library (Gamma- M)	27.64	18.33					

The tendency of performance's results is the same between the mixing source experiment with the fresh fuel and the natural uranium. At 3 sigma, the detection rate has been improved from 27.64 % to 71.16 % but the false alarm rate has been increased from 18.33 % to 30.24 %.



FIGURE 17. Spectrum of mixing fresh fuel and Cs-137 at distance of 15 cm, 30 seconds scan time



FIGURE 18. Spectrum of mixing natural uranium (8 gram) and Cs-137 at distance of 15 cm, 30 seconds scan time

From 3 sigma to 10 sigma of peak threshold setting, the detection rate and the false alarm rate have decreased. At 10 sigma of the proposed method in comparison to 3 sigma of Library (Gamma – M) method, the detection rate has been improved from 27.64 % to 59.83 % and the false alarm rate has been reduced from 18.33 % to 17.22 %. Table 4 shows the results of experiments with the fresh fuel.

The detection rate of the proposed method for the experiments of mixing fresh fuel has lower than the mixing natural uranium experiments because of the signal from the natural uranium is comparable with the signal of industrial mixing source but the signal from the fresh fuel is too high.

Two factors of high background environment and difference source strength can be used for explaining the high false alarm rate between single sources and mixing sources scenario. The nuclear material's source strength difference mixing with Cs-137 source at the same distance and scan time expressed in figure 17 and figure 18.

Peak ratio comparison technique for false alarm reduction:

TABLE 5. False alarm rate from Cs-137 spectrum with difference backscatter peak to full peak ratio

Peak ratio	False alarm rate
0	64 %
0.010	40%
0.020	12 %

By applying the peak ratio comparison technique, the false alarm rate of simulated masking scenario by backscatter peak from Cs-137 source has reduced from 64% to 40% when $P_B/P_T = 0.01$ and 64% to 12% when $P_B/P_T = 0.02$.

The false alarm has decreased by applying peak ratio comparison technique because of the contribution from backscatter peak of Cs- 137 was estimated and removed. The optimum ratio is changed depending on several conditions of conducting experiment, such as detector resolution, experiments set up and the detection criterial.

The preliminary results show that the peak ratio comparison technique is remarkable reduce the false alarm rate in the scenario of differentiate between backscatter peak and the signal from U-235 in Cs-137 spectrum analysis.

5.2. Conclusions and suggestion for future work

This study proposes a novel method to identify the signal of one or several radioisotopes from a poorly resolved gamma-ray spectrum. In this method, the noise component in the raw spectrum is reduced by the wavelet decomposition approach, and the removal of the continuum background is performed using the proposed baseline determination algorithm. Finally, the identification of radioisotope is completed using the matrix linear regression method.

The proposed method has been verified by experiments using the poorly resolved gamma-ray signals from various scenarios including single source, mixing of natural uranium, enriched uranium with five of the most common industrial radioactive sources (⁵⁷Co, ⁶⁰Co, ¹³³Ba, ¹³⁷Cs, and ²⁴¹Am). The results of the experiments showed that the proposed method has increase the detection rate and its drawback with the false alarm rate.

The developed Matlab program using of linear regression in this study was developed by single characteristic gamma-ray energy, if considering that one radioisotope has several characteristic peaks, the program need to be more completed.

This study using the radioactive sources that available at the Department of Nuclear Engineering Laboratory. The nuclear materials used are natural uranium and fresh fuel. Because of small amount of natural uranium, the signal is quietly low, almost cannot detect at distance of 20 cm from the detector. For the experiments with the fresh fuel at TINT, the strong signal from the fresh fuel made the experiments set up are not the same as expected.

The proposed method can be used at the secondary inspection at the border control site when the operating procedure are focusing on group of radioisotopes, such as SNM, industrial radiation group...etc. The shielding from the container, the detector resolution and sensitivity, background conditions are the main factors need to be considered.

The future work are on-going to reduce the false alarm rate of the algorithm by applying Gaussian fitting function and to test the performance under the gain shift and shielding conditions.



REFERENCES

- Jonathan Medalia. Detection of nuclear weapons and materials: Science, Technologies, Observations. Technical report, Congressional Research Service, 2010.
- [2]. Tom Burr and Michael Hamada. Radio-isotope Identification Algorithms for NaI(Tl)Spectra. Algorithms, 2009.
- [3]. Deborah K. Fagan, Sean M. Robinson, Robert C. Runkle. Statistic methods applied to gamma-ray spectroscopy algorithms in nuclear security missions. Apply Radiation and Isotope 70, 2012.
- [4]. Kung-Sik Chan, Jinzheng Li, William Eichinger, Erwei Bai. A new physics-based method for detecting weak nuclear signals via spectra decomposition. Nuclear Instruments and Methods in Physics Research A, 2012.
- [5]. Paul Kump, Erwei Bai, Kung-Sik Chan, William Eichinger. Detection of shielded radionuclides from weak and poorly resolved spectra using group positive RIVAL. Radiation Measurement 48, 2013.
- [6]. E. Bai, K. Chan, W. Eichinger, and P. Kump. Detection of radionuclides from weak and poorly resolved spectra using lasso and sub-sampling techniques. Radiation Measurements 46, 2011.
- [7]. T. O'Haver. Peak finding and measurement, version 2. http://terpconnect. umd.edu/toh/spectrum/PeakFindingandMeasurement.htm, 2014.
- [8]. Mitchell, D.J., Mattingly, J., 2008. A framework for the solution of inverse radiation transport problems. 2008 IEEE Nucl. Sci. Symp. Medical Imaging Conf. (2008 Nss/Mic) 1–9, 540–545.
- [9]. Syed Naeem Ahmed. Physics Engineering of Radiation Detection. First edition, 2007.
- [10]. Glenn F.Knoll. Radiation Detection and Measurement. Third Edition, 1999.

- [11]. S. Fetter, V. A. Frolov, M. Miller, R. Mozley, O. F. Prilutsky, S. N. Rodionov, and R. Z. Sagdeev, "Detecting Nuclear Warheads," Science & Global Security, Vol. 1, Nos. 3-4, pp. 225-253, 1990.
- [12]. Trevor Hastie, Robert Tibshirani, Jerome Friedman. The Elements of Statistical Learning, Data Mining, Inference, and Prediction. Second Edition, 2008.
- [13]. M.I. Reinhard, D. Prokopovich, H.Van der Gaat, and D.Hill. Detection of Illicit Nuclear Materials Masked with other Gamma-Ray Emitters. IEEE Nuclear Science Symposium Conference record, N14-21, 2006.

[14]. Nuclear Security and Safeguards Advanced Detection lecture, part 3, Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University.

[15]. William R. Russ. Library correlation nuclide identification algorithm. Nuclear Instruments and Methods in Physics Research A 579, 2007.

[16]. Genie 2000 Spectroscopy Software, Customization Tools.

[17]. Ratnakar Madan, Sunil Kr.Singh, and Aitisha Jain, "Signal Filtering Using Discrete Wavelet Transform", International Journal of Recent Trends in Engineering, Vol 2, No. 3, November 2009.

[18]. Nikolaos Kourkoumelis, "Continuum determination in spectroscopic data by means of topological concepts and Fourier filtering". Nuclear Instruments and Methods in Physics Research A 691 (2012) 1–4.

[19]. NaI(Tl) Detector, model 802 – 3x3, User Manual.

[20]. Prof. Dr. Ir. M. Steinbuch, Dr. Ir. M.J.G. van de Molengraft, June 7 (2005), Eindhoven University of Technology, Control Systems Technology Group Eindhoven, "Wavelet Theory and Applications", a literature study, R.J.E. Merry, DCT 2005.53.



จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

REFERENCES



จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

APPENDIX 1. REFERENCE LIBRARY MATRIX X_n

	Am-241	U-235	Cs-137	Co-57	Co-60	Ba-133
1	0	0	0	0	0	0
2	0	0	0	0	0	0
3	0	0	0	0	0	0
4	0	0	0	0	0	0
5	0	0	0	0	0	0
6	0	0	0	0	0	0
7	0	0	0	0	0	0
8	0	0	0	0	0	0
9	0	0	0	0	0	0
10	0	0	0	0	0	0
11	0	0	0	0	0	0
12	0	0	0	0	0	0
13	0	0	0	0	0	0
14	0	0	0	0	0	0
15	0	0	0	0	0	0
16	0	0	0	0	0	0
17	0	0	0	0	0	0
18	0	0	0	0	0	0
19	0	0	0	0	0	0
20	0	0	0	0	0	0
21	0	0	0	0	0	0
22	0	0	0	0	0	0
23	0	0	0	0	0	0
24	0	0	0	0	0	0
25	0	0	0	0	0	0
26	0	0	0	0	0	0
27	0	0	0	0	0	0
28	0	0	0	0	0	0
29	0	0	0	0	0	0
30	0	0	0	0	0	0

31	0	0	0	0	0	0	1
32	0	0	0	0	0	0	
33	0	0	0	0	0	0	
34	0	0	0	0	0	0	
35	0	0	0	0	0	0	
36	0	0	0	0	0	0	
37	0	0	0	0	0	0	
38	0	0	0	0	0	0	
39	0	0	0	0	0	0	
40	0	0	0	0	0	0	
41	0	0	0	0	0	0	
42	0	0	0	0	0	0	
43	35.9	0	0	0	0	0	
44	35.9	0	0	0	0	0	
45	35.9	0	0	0	0	0	
46	0	0	0	0	0	0	
47	0	0	0	θ	0	0	
48	0	0	0	0	0	0	
49	0	0	0	0	0	0	
50	0	0	0	0	0	0	
51	0	0	0	0	0	0	
52	0	0	0	0	0	0	
53	0	0	0	0	0	0	
54	0	0	0	0	0	0	
55	0	0	0	0	0	0	
56	0	0	0	0	0	0	
57	0	0	0	0	0	0	
58	0	0	0	0	0	0	
59	0	0	0	0	0	0	
60	0	0	0	0	0	0	
61	0	0	0	0	0	0	
62	0	0	0	0	0	0	
63	0	0	0	0	0	0	

64	0	0	0	0	0	0	
65	0	0	0	0	0	0	
66	0	0	0	0	0	0	
67	0	0	0	0	0	0	
69	0	0	0	0	0	0	
60	0	0	0	0	0	0	
70	0	0	0	0	0	0	
70	0	0	0	0	0	0	
71	0	0	0	0	0	0	
72	0	0	0	0	0	0	
73	0	0	0	U	U	U	
74	0	0	0	0	0	0	
75	0	0	0	0	0	0	
76	0	0	0	0	0	0	
77	0	0	0	0	0	0	
78	0	0	0	0	0	0	
79	0	0	0	0	0	0	
80	0	0	0	0	0	0	
81	0	0	0	0	0	0	
82	0	0	0	0	0	0	
83	0	0	0	85.6	0	0	
84	0	0	0	85.6	0	0	
85	0	0	0	85.6	0	0	
86	0	0	0	85.6	0	0	
87	0	0	0	85.6	0	0	
88	0	0	0	θ	0	0	
89	0	0	0	0	0	0	
90	0	0	0	0	0	0	
91	0	0	0	0	0	0	
92	0	0	0	0	0	0	
93	0	0	0	0	0	0	
94	0	0	0	0	0	0	
95	0	0	0	0	0	0	
96	0	0	0	0	0	0	

97	0	0	0	0	0	0
98	0	0	0	0	0	0
99	0	0	0	0	0	0
100	0	0	0	0	0	0
101	0	0	0	0	0	0
102	0	0	0	0	0	0
103	0	0	0	0	0	0
104	0	0	0	0	0	0
105	0	0	0	0	0	0
106	0	0	0	0	0	0
107	0	0	0	0	0	0
108	0	0	0	0	0	0
109	0	0	0	0	0	0
110	0	0	0	0	0	0
111	0	0	0	0	0	0
112	0	θ	0	0	0	0
113	0	0	0	0	0	0
114	0	0	0	0	0	0
115	0	0	0	0	0	0
116	0	0	0	0	0	0
117	0	θ	0	0	0	0
118	0	0	0	0	0	0
119	0	0	0	0	0	0
120	0	0	0	0	0	0
121	0	0	0	0	0	0
122	0	0	0	0	0	0
123	0	0	0	0	0	0
124	0	0	0	0	0	0
125	0	0	0	0	0	0
126	0	57.2	0	0	0	0
127	0	57.2	0	0	0	0
128	0	57.2	0	0	0	0
129	0	57.2	0	0	0	0

130	0	57.2	0	0	0	0
131	0	57.2	0	0	0	0
132	0	57.2	0	0	0	0
133	0	0	0	0	0	0
134	0	0	0	0	0	0
135	0	0	0	0	0	0
136	0	0	0	0	0	0
137	0	0	0	0	0	0
138	0	0	0	0	0	0
139	0	0	0	0	0	0
140	0	0	0	0	0	0
141	0	0	0	0	0	0
142	0	0	0	0	0	0
143	0	0	0	0	0	0
144	0	0	0	0	0	0
145	0	0	0	0	0	0
146	0	0	0	0	0	0
147	0	0	0	0	0	0
148	0	0	0	0	0	0
149	0	0	0	0	0	0
150	0	0	0	0	0	0
151	0	0	0	0	0	0
152	0	0	0	0	0	0
153	0	0	0	0	0	0
154	0	0	0	0	0	0
155	0	0	0	0	0	0
156	0	0	0	0	0	0
157	0	0	0	0	0	0
158	0	0	0	0	0	0
159	0	0	0	0	0	0
160	0	0	0	0	0	0
161	0	0	0	0	0	0
162	0	0	0	0	0	0

163	0	0	0	0	0	0
164	0	0	0	0	0	0
165	0	0	0	0	0	0
166	0	0	0	0	0	0
167	0	0	0	0	0	0
168	0	0	0	0	0	0
169	0	0	0	0	0	0
170	0	0	0	0	0	0
171	0	0	0	0	0	0
172	0	0	0	0	0	0
173	0	0	0	0	0	0
174	0	0	0	0	0	0
175	0	0	0	0	0	0
176	0	0	0	0	0	0
177	0	0	0	0	0	0
178	0	0	0	0	0	0
179	0	0	0	0	0	0
180	0	0	0	0	0	0
181	0	0	0	0	0	0
182	0	0	0	0	0	0
183	0	0	0	0	0	0
184	0	0	0	0	0	0
185	0	0	0	0	0	0
186	0	0	0	0	0	0
187	0	0	0	0	0	0
188	0	0	0	0	0	0
189	0	0	0	0	0	0
190	0	0	0	0	0	0
191	0	0	0	0	0	0
192	0	0	0	0	0	0
193	0	0	0	0	0	0
194	0	0	0	0	0	0
195	0	0	0	0	0	0

104	0	0	0	0	0	0
190	U	0	0	0	0	0
197	0	0	0	0	0	0
198	0	0	0	0	0	0
199	0	0	0	0	0	0
200	0	0	0	0	0	0
201	0	0	0	0	0	0
202	0	0	0	0	0	0
203	0	0	0	0	0	0
204	0	0	0	0	0	0
205	0	0	0	0	0	0
206	0	0	0	0	0	0
207	0	0	0	0	0	0
208	0	0	0	0	0	0
209	0	θ	0	0	0	0
210	0	0	0	0	0	0
211	0	0	0	0	0	0
212	0	0	0	0	0	0
213	0	0	0	0	0	0
214	0	0	0	0	0	0
215	0	0	0	0	0	0
216	0	0	0	0	0	0
217	0	0	0	0	0	0
218	0	0	0	0	0	0
219	0	0	0	0	0	0
220	0	0	0	0	0	0
221	0	0	0	0	0	0
222	0	0	0	0	0	0
223	0	0	0	0	0	0
224	0	0	0	0	0	0
225	0	0	0	0	0	0
226	0	0	0	0	0	0
227	0	0	0	0	0	0
228	0	0	0	0	0	0

229	0	0	0	0	0	0
230	0	0	0	0	0	0
231	0	0	0	0	0	0
232	0	0	0	0	0	0
233	0	0	0	0	0	0
234	0	0	0	0	0	0
235	0	0	0	0	0	0
236	0	0	0	0	0	0
237	0	0	0	0	0	0
238	0	0	0	0	0	0
239	0	0	0	0	0	0
240	0	0	0	0	0	0
241	0	0	0	0	0	62.5
242	0	0	0	0	0	62.5
243	0	0	0	0	0	62.5
244	0	0	0	0	0	62.5
245	0	0	0	0	0	62.5
246	0	0	0	0	0	62.5
247	0	0	0	0	0	62.5
248	0	0	0	0	0	0
249	0	0	0	0	0	0
250	0	0	0	0	0	0
251	0	0	0	0	0	0
252	0	0	0	0	0	0
253	0	0	0	0	0	0
254	0	0	0	0	0	0
255	0	0	0	0	0	0
256	0	0	0	0	0	0
257	0	0	0	0	0	0
258	0	0	0	0	0	0
259	0	0	0	0	0	0
260	0	0	0	0	0	0
261	0	0	0	0	0	0

262	0	0	0	0	0	0
263	0	0	0	0	0	0
264	0	0	0	0	0	0
265	0	0	0	0	0	0
266	0	0	0	0	0	0
267	0	0	0	0	0	0
268	0	0	0	0	0	0
269	0	0	0	0	0	0
270	0	0	0	0	0	0
271	0	0	0	0	0	0
272	0	0	0	0	0	0
273	0	0	0	0	0	0
274	0	0	0	0	0	0
275	0	0	0	0	0	0
276	0	0	0	0	0	0
277	0	0	0	0	0	0
278	0	0	0	0	0	0
279	0	0	0	0	0	0
280	0	0	0	0	0	0
281	0	0	0	0	0	0
282	0	0	0	0	0	0
283	0	0	0	0	0	0
284	0	0	0	0	0	0
285	0	0	0	0	0	0
286	0	0	0	0	0	0
287	0	0	0	0	0	0
288	0	0	0	0	0	0
289	0	0	0	0	0	0
290	0	0	0	0	0	0
291	0	0	0	0	0	0
292	0	0	0	0	0	0
293	0	0	0	0	0	0
294	0	0	0	0	0	0

295	0	0	0	0	0	0
206	0	0	0	0	0	0
207	0	0	0	0	0	0
209	0	0	0	0	0	0
290	0	0	0	0	0	0
	•		v	v	•	
	1			1		-
438	0	0	0	0	0	0
439	0	0	0	0	0	0
440	0	0	85.1	0	0	0
441	0	0	85.1	0	0	0
442	0	0	85.1	0	0	0
443	0	0	85.1	0	0	0
444	0	0	85.1	0	0	0
445	0	0	85.1	0	0	0
446	0	0	85.1	0	0	0
447	0	0	0	0	0	0
448	0	0	0	0	0	0
449	0	0	0	0	0	0
450	0	θ	0	0	0	0
451	0	0	0	0	0	0
452	0	0	0	0	0	0
453	0	0	0	0	0	0
454	0	0	0	0	0	0
455	0	0	0	0	0	0
456	0	0	0	0	0	0
457	0	0	0	0	0	0
458	0	0	0	0	0	0
459	0	0	0	0	0	0
460	0	0	0	0	0	0
461	0	0	0	0	0	0
462	0	0	0	0	0	0
463	0	0	0	0	0	0

GHULALONGKORN UNIVERSI I M

464	0	0	0	0	0	0
465	0	0	0	0	0	0
466	0	0	0	0	0	0
786	-	-	-	-		-
797	0	0	0	0	0	0
788	0	0	0	0	0	0
780	0	0	0	0	0	0
790	0	0	0	0	00.85	0
791	0	0	0	0	00.85	0
792	0	ů 0	0	0	99.85	0
793	0	ů.	0	0	99.85	0
794	0	0	0	Ő	99.85	0
795	0	0 0	0	Õ	99.85	0
796	0	0	0	0	99.85	0
797	0	0	0	0	99.85	0
798	0	0	0	0	99.85	0
799	0	0	0	0	99.85	0
800	0	0	0	0	99.85	0
801	0	0	0	0	0	0
802	0	0	0	0	0	0
803	0	0	0	0	0	0
804	0	0	0	0	0	0
805	0	0	0	0	0	0
806	0	0	0	0	0	0
807	0	0	0	0	0	0
808	0	0	0	0	0	0
809	0	0	0	0	0	0
810	0	0	0	0	0	0
811	0	0	0	0	0	0
812	0	0	0	0	0	0
813	0	0	0	0	0	0
814	0	0	0	0	0	0
815	0	0	0	0	0	0
1012	0	0	0	0	0	0
1013	0	0	0	0	0	0
1014	0	0	0	0	0	0
1015	0	0	0	0	0	0
1016	0	0	0	0	0	0
1017	0	0	0	0	0	0
1018	0	0	0	0	0	0
1019	0	0	0	0	0	0
1020	0	0	0	0	0	0
1021	0	0	0	0	0	0
1022	0	0	0	0	0	0
1023	0	0	0	0	0	0
1024	0	0	0	0	0	0

APPENDIX 2: MATLAB CODE

function P = Peakdetection(y,bg,xn)

% Function to determine the base line of gamma-ray spectrum, x is channel % number, y is the number of measured counts.

% P=[num2cell(En) num2cell(BR) cellstr(Name) cellstr(Type)]; cmt for % Library

% Step 1: Noise Reduction of the original spectrum by DWT 3 time.

```
y_bg = zeros(size(y));
y_bg1 = zeros(size(y));
y_bl = zeros(size(y));
y_bl0 = zeros(size(y));
y_bl1 = zeros(size(y));
k = y(1);
test = zeros(size(y));
```

```
for i = 1:1024
    y_bg(i) = y(i) - bg(i)*k/300; % suctract the backgound
    if y_bg(i) <0
        y_bg(i) = 0;
    end
end</pre>
```

[Lo_D,Hi_D] = wfilters('db1','d'); [A1,D1] = dwt (y_bg,Lo_D,Hi_D); [A2,D2] = dwt (A1,Lo_D,Hi_D); [A3,D3] = dwt (A2,Lo_D,Hi_D); X1 = idwt(A3,zeros(size(A3)),'db1'); X2 = idwt(X1,zeros(size(X1)),'db1');

```
%[A4,D4] = dwt (A3,Lo_D,Hi_D);
%[A5,D5] = dwt (A4,Lo_D,Hi_D);
%X1 = idwt(A5,zeros(size(A5)),'db1');
%X2 = idwt(A4,zeros(size(A4)),'db1');
```

% Step 2: Smooth DWT by using of P-G smooth function

```
ys = smoothPG(X3);
d = deriv(ys);
vectorlength = length(y);
point = 1;
point1 = 1;
P=[0 0 0];
P1=[0 0 0];
A = zeros(6,1);
x = 1:1024;
xk = zeros(size(x));
yk = zeros(size(x));
yk = zeros(size(x));
baseline = zeros(size(y));
point_index = 0;
shift = zeros(size(y));
```

for i = 3 : round(vectorlength-1)

%if sign(d(i-1))*sign(d(i))<0 % detect the extreme values(maxima and minima), eliminate the zero point

if sign(d(i-1))<sign(d(i)) % detect the minima points

% if $y_s(i) < 3*sqrt(bg(i)*k/300)$

% if d(i-2) + d(i-1) + d(i) + d(i+1) < 0 % check the trend of spectrum, or decrease the sensitivity of algorithm

xk(i) = x(i);
```
yk(i)=ys(i); % mark the channel and count of minima.
xkin(i) = 1; % to find the index of minima in the next step.
baseline(i) = ys(i);
P(point,:) = [point xk(i) yk(i)];
point = point+1; % Move on to next point
% end
end
%end
```

end

```
for j = 1 : vectorlength - 1
```

point_index = point_index + xkin(j); % find total minima point
end

```
for j = 1:point_index-1
    for i = 1 : vectorlength
        if P(j,2) < x(i)
            if x(i) < P(j+1,2)
        shift(i) = (P(j+1,3)-P(j,3))/(P(j+1,2)-P(j,2));
        baseline(i)= P(j,3)+ (x(i) - P(j,2))*shift(i);
        if baseline(i)>ys(i)
        baseline(i) = ys(i);
        end
        en
```

```
for i = P(end,2):vectorlength
  baseline(i) = ys(i);
end
m = baseline;
% plot(x,y,'green',x,ys,'blue',x,baseline,'red');
subplot(3,1,1);
plot(x,ys,'blue',x,baseline,'red')
```

```
% subtract the found baseline
```

```
for i = 1:1024
```

 $y_bg1(i) = 3*sqrt(k*bg(i)/300);$ % 3 sigma

 $y_bl(i) = ys(i) - baseline(i);$

end

```
subplot(3,1,2);
```

```
plot(x,y_bl,'blue',x,y_bg1,'red')
```

```
% determine the peak
```

for i = 1:1024

```
y_bl0(i) = y_bl(i) - y_bg1(i); % subtract also the 3 sigma - as 13/7/2015
```

```
if y_bl0(i) < 0
```

```
y_bl0(i) = 0;
```

end

```
end
```

```
for i = 4:1020
```

```
%if y_bl(i) > 3*sqrt(k*bg(i)/300
%if sign(d(i-1))>sign(d(i))
%if y_bl0(i)>y_bl0(i-1)
%if or(y_bl0(i)>y_bl0(i+1),y_bl0(i)==y_bl0(i+1))
%if y_bl0(i)>y_bl0(i+1)
if and(2*y_bl0(i)>y_bl0(i+1)+ y_bl0(i+2),2*y_bl0(i)>y_bl0(i-2)+y_bl0(i-1))
% find local maxima point
```

```
P1(point1,:) = [point1 x(i) y_bl(i)];
point1 = point1+1; % Move on to next point
end
%end
%end
```

end

% convert the spectrum vector to the idea case

```
for i = 1:1024
   for j = 1:point1-1
     if i == P1(j,2)
       y_{bl1(i)} = P1(j,3);
     end
   end
end
subplot(3,1,3);
plot(x,y_bl1)
test = y_{bl1};
A = xn y_b11;
if A(1) > 0
 fprintf(' Am-241 INDUSTRIAL')
end
if A(2) > 0
 fprintf(' U-235 NM')
end
if A(3) > 0
```

```
fprintf(' Cs-137 MEDICAL')
end
```

```
if A(4) > 0
    fprintf(' Co-57 INDUSTRIAL')
end
```

```
if A(5) > 0
fprintf(' Co-60 MEDICAL')
```

end

if A(6) > 0

```
fprintf(' Ba-133 IND')
```

end

end

```
function d=deriv(a)
```

```
% First derivative of vector using 2-point central difference.

n=length(a);

d(1)=a(2)-a(1);

d(n)=a(n)-a(n-1);

for j = 2:n-1

d(j)=(a(j+1)-a(j-1)) ./ 2;
```

end end

```
function ysmt = smoothPG(b)
```

% Smooth the spectrum by Pseudo-Gaussian smooth function n=length(b); for j = 1.6

```
ysmt(j) = b(j);
```

end

for j= n-6:n

```
ysmt(j) = b(j);
end
for j = 7 : n-7
ysmt(j)= (b(j-3)+3*b(j-2)+6*b(j-1)+7*b(j)+6*b(j+1)+3*b(j+2)+b(j+3))/27;
end
end
```



จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University

VITA

I graduated from Hanoi University of Science, Hanoi National University in 2007 in field of Nuclear Technology. I have worked for Vietnam Agency for Radiation and Nuclear Safety since June, 2007. In 2013, I got the scholarship from European Commission's CBRN Centers of Excellence for Master programme in Nuclear Security and Safeguards at Chulalongkorn University, Bangkok, Thailand.



จุฬาลงกรณมหาวทยาลย Chulalongkorn University



จุฬาลงกรณ์มหาวิทยาลัย Chulalongkorn University