การบอกลักษณะของวัสดุที่มีธาตุยูเรเนียมและการวัดรังสีแกมมาโดยตรง โดยใช้เทคนิคการเรืองรังสีเอกซ์

นางสาวมาดิฮะห์ มูไจนี

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต
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Characterization of Uranium Bearing Material using X-Ray Fluorescence and Direct Gamma-Rays Measurement Techniques

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A Thesis Submitted in Partial Fulfillment of the Requirements
for the Degree of Master of Science Program in Nuclear Technology
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Thesis Title	Characterization of Uranium Bearing Material using X-Ray Fluorescence and Direct Gamma-Rays Measurement Techniques
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มาดิฮะห์ มูไจนี : การบอกลักษณะของวัสดุที่มีธาตุยูเรเนียมและการวัดรังสีแกมมาโดยตรง โดยใช้เทคนิคการเรืองรังสีเอกซ์ (Characterization of Uranium Bearing Material using X-Ray Fluorescence and Direct Gamma-Rays Measurement Techniques) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: รศ.นเรศร์ จันทน์ขาว, 88 หน้า.

แร่ยูเรเนียมสามารถตรวจวัดได้ง่าย เนื่องจากสามารถวัดพลังงานรังสีแกมมาที่ได้จากนิวไคลด์ลูกที่สลายตัวจากยูเรเนียม โดยเฉพาะอย่างยิ่งรังสีแกมมาที่มาจาก ^{238}U เช่น ^{214}Bi, ^{214}Pb และ ^{226}Ra ภายหลังการสกัดยูเรเนียมออกจากแร่แล้ว มีเพียงรังสีแกมมาพลังงานต่ำจาก ^{235}U เท่านั้น ซึ่งสามารถตรวจวัดได้เมื่อวางหัววัดรังสีอยู่ใกล้กับตัวอย่าง ในงานวิจัยนี้ได้ทดลองใช้วิธีวัดรังสีแกมมาจาก ^{235}U โดยตรง ร่วมกับเทคนิคการเรืองรังสีเอกซ์ เพื่อระบุและบอกลักษณะของวัสดุที่มีธาตุยูเรเนียม การตรวจวัดรังสีแกมมาสามารถตรวจวัดโดยใช้เจอร์เมเนียมความบริสุทธิ์สูง (HPGe) หรือหัววัดแคดเมียมเทลลูไรด์ (CdTe) ในส่วนของการวิเคราะห์ธาตุด้วยเครื่องวิเคราะห์ด้วยวิธีการเรืองรังสีเอกซ์ ได้ใช้หัววัดแคดเมียมเทลลูไรด์ โดยมีต้นกำเนิดรังสี ^{57}Co เป็นตัวกระตุ้น ได้ทำการทดสอบเทคนิคที่นำเสนอนี้กับวัสดุตัวอย่างทีมีธาตุยูเรเนียมต่าง ๆ กัน ได้แก่ ยูเรเนียมธรรมชาติ ธาตุยูเรเนียมด้อยสมรรถนะ และธาตุยูเรเนียมเสริมสมรรถนะ ในรูปแบบวัสดุตัวอย่างที่เป็นโลหะและตัวอย่างวัสดุที่เป็นชนิดผงผลการวิจัยพบว่าการใช้สองเทคนิคที่นำเสนอสามารถใช้ในการระบุและบอกลักษณะของวัสดุนิวเคลียร์ต้องสงสัยไม่เพียงแต่ที่มียูเรเนียม ยังสามารถใช้กับพลูโทเนียมและทอเลียมได้ด้วย

ภาควิชา วิศวกรรมนิวเคลียร์ ลายมือชื่อนิสิต .....

สาขาวิชา นิวเคลียร์เทคโนโลยี ลายมือชื่อ อ.ที่ปรึกษาหลัก .....

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Uranium ore can be easily detected due to various gamma-ray energies emitted from uranium daughters particularly from ^{238}U daughters such as ^{214}Bi, ^{214}Pb and ^{226}Ra. After uranium is extracted from uranium ore, only low energy gamma-rays emitted from ^{235}U may be detected if the detector is placed in close contact to the specimen. In this research, identification and characterization of uranium bearing materials is experimentally investigated using direct measurement of gamma-rays from ^{235}U in combination with the x-ray fluorescence (XRF) technique. Measurement of gamma-rays can be conducted by using high purity germanium (HPGe) detector or cadmium telluride (CdTe) detector while a ^{57}Co radioisotope-excited XRF spectrometer using CdTe detector is used for elemental analysis. The proposed technique was tested with various uranium bearing specimens containing natural, depleted and enriched uranium in both metallic and powder forms. The results indicated that combination of the two techniques can be used to identify and to characterize suspicious nuclear materials not only uranium bearing materials but also Plutonium (Pu) and Thorium (Th).

Department:	Nuclear Engineering	Student's Signature	.....
Field of Study:	Nuclear Technology	Advisor's Signature	.....
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CHAPTER 1

INTRODUCTION

1.1 Background

The characterization and identification of specimens using non-destructive analytical techniques is a part of the verification of nuclear materials and the detection of undeclared nuclear materials in international safeguards. The determination of ^{235}U using x-ray and gamma ray detector with high resolution cadmium telluride (CdTe) capable of detecting samples in the range from a few keV to several hundred keV. The isotope ratios in uranium bearing material samples with sufficient accuracy and measurement uncertainties to resolve differences between origins, identify clusters and patterns, and eventually attribute samples to or discriminate from known sources is one of the challenges in the field. The enrichment meter of the specimens will be determine on the basis of the measurement of the peak area of ^{235}U line. A good agreement predicted between the peak areas is measured the values of the ratios of ^{235}U with strong correlation to be found between the certified and the enrichments. The XRF technique measures from uranium and compares pulse heights of peaks to quantify elemental uranium. If unknown nuclear material needed to be identified rapidly, XRF could be used as a relatively portable measurement technique to obtain initial information about a sample, which could drastically reduce identification time by narrowing the range of further tests to be performed. The results obtain from the analysis would suggested the analytical method can be used to determine the uranium concentration, and the isotopic abundance, in the case of uranium bearing material depleted, natural and enriched uranium samples.

The safeguards mission of the IAEA is to provide assurance that no declared nuclear material (U, Pu, Th) is diverted to non-peaceful purpose and that no undeclared nuclear material or activities exist. To fulfill its mandate, the IAEA performs independent verification measurements of nuclear material using a variety of non-destructive assay

(NDA) instrumentation in attended or unattended mode. The nature of particular inspection activities required customization of the equipment and methods implemented, friendly and simple to use, short measurement time, and easily portable or transportable.

1.2 Objective of Study

(a) To characterize ^{235}U from uranium bearing materials using the XRF technique followed by direct gamma-ray measurement

(b) To determine concentration of ^{235}U from uranium bearing material

1.3 Significance of Study

The techniques can be used to identify suspicious metal and chemicals for security and safeguards. In a safeguards standpoint, monitoring usually sufficient sensitive if they detect small, say gram, amounts of ^{235}U concealed on person or vehicle.

The quality control for a common practice to pass such fuel rods through a rod scanning system detects ^{235}U content and spot any individual pellets that deviate from the specifications. This must be analyzed for manufacturing quality control reasons. Moreover, the same techniques can be used to identify Th and Pu bearing materials for the same purposes.

1.4 Scope of Study

(a) Investigate a technique for identifying uranium bearing materials by measurement of U K x-rays excited with gamma-rays from Co-57 source

(b) Investigate technique for determining uranium concentration by measurement of gamma-rays from ^{235}U and others

(c) Test the technique with known uranium bearing materials including depleted uranium and compound, natural uranium and compound, enriched uranium in fresh fuel elements of TRIGA III research reactor.

In this research, identification and characterization of uranium bearing materials is experimentally investigated using direct measurement of gamma-rays from ^{235}U in combination with the x-ray fluorescence (XRF) technique. Measurement of gamma-rays can be conducted by using high purity germanium (HPGe) detector or cadmium telluride (CdTe) detector while a ^{57}Co radioisotope-excited XRF spectrometer using CdTe detector is used for elemental analysis. The proposed technique was tested with various uranium bearing specimens containing natural, depleted and enriched uranium in both metallic and powder forms.

CHAPTER 2

PRINCIPLES AND LITERATURE REVIEW

2.1 Uranium Properties and Radioactive Series

Uranium is the heaviest element found in nature, with the atomic number Z = 92. However, the heaviest stable elements found in nature are several isotopes of lead with atomic number Z = 82 (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb found at 1.4%, 24.1%, 22.1%, and 52.1%, respectively) and one isotope of bismuth with atomic number Z = 83 (^{209}Bi). Except for the first natural isotope of lead (^{204}Pb), all of these isotopes are at least partially of radiogenic origin [1].

All natural elements with atomic numbers Z > 83 are radioactive. These elements decay either by emission of α-particles or by emission of negative β-particles. Both manners of radioactive decay might be followed by emission of γ-rays if the daughter nucleus is formed in an excited state.

Table 2-1: Natural Decay Series[1]

Series	First Isotope	Half-life (years)	Last Isotope
Uranium	^{238}U	4.49×10^{9}	^{206}Pb
Actinium	^{235}U	7.10×10^{8}	^{207}Pb
Thorium	^{232}Th	1.39×10^{10}	^{208}Pb
Neptunium	^{237}Np	2.14×10^{6}	^{209}Bi

Three of these radioactive decay series are named after the isotope with the longest half-life. The actinium series is named after one its members, in order to avoid having two series with identical names. Only three of these decay series are found in nature: uranium, actinium, and thorium. The isotope ^{237}Np has a half-life much shorter than the geological age of the Earth - about 5 billions of years. Virtually all neptunium decayed within the first 50 millions of years after the Earth was formed [1].

The radionuclides of the uranium-238 and uranium-235 decay series are shown in Figures 2.1 and 2.2 along with the major mode of radioactive decay for each. Radioactive decay occurs when an unstable isotope transforms to a more stable isotope, generally by emitting a subatomic particle such as an alpha or beta particle. Radionuclides that give rise to alpha and beta particles are shown in the figures, as are those that emit significant gamma radiation [1].

Table 2-2: Uranium Series[1]

Isotope	Half-life	Decay
^{238}U	$4.49x10^{9}$ years	α
^{234}Th	24.1 days	β-
^{234m}Pa	1.17 min	β-
^{234}U	$2.48x10^{5}$ years	α
^{230}Th	$7.7x10^{4}$ years	α
^{226}Ra	1600 years	α
^{222}Rn	3.82 days	α
^{218}Po	3.05 min	α
^{214}Pb	26.8 min	β-

Table 2-2: Uranium Series (Continue)[1]

Isotope	Half-life	Decay
^{214}Bi	19.8 min	β-
^{214}Po	162 μsec	α
^{210}Pb	22.3 years	β-
^{210}Bi	5.01 days	β-
^{210}Po	138.4 days	α
^{206}Pb	stable	

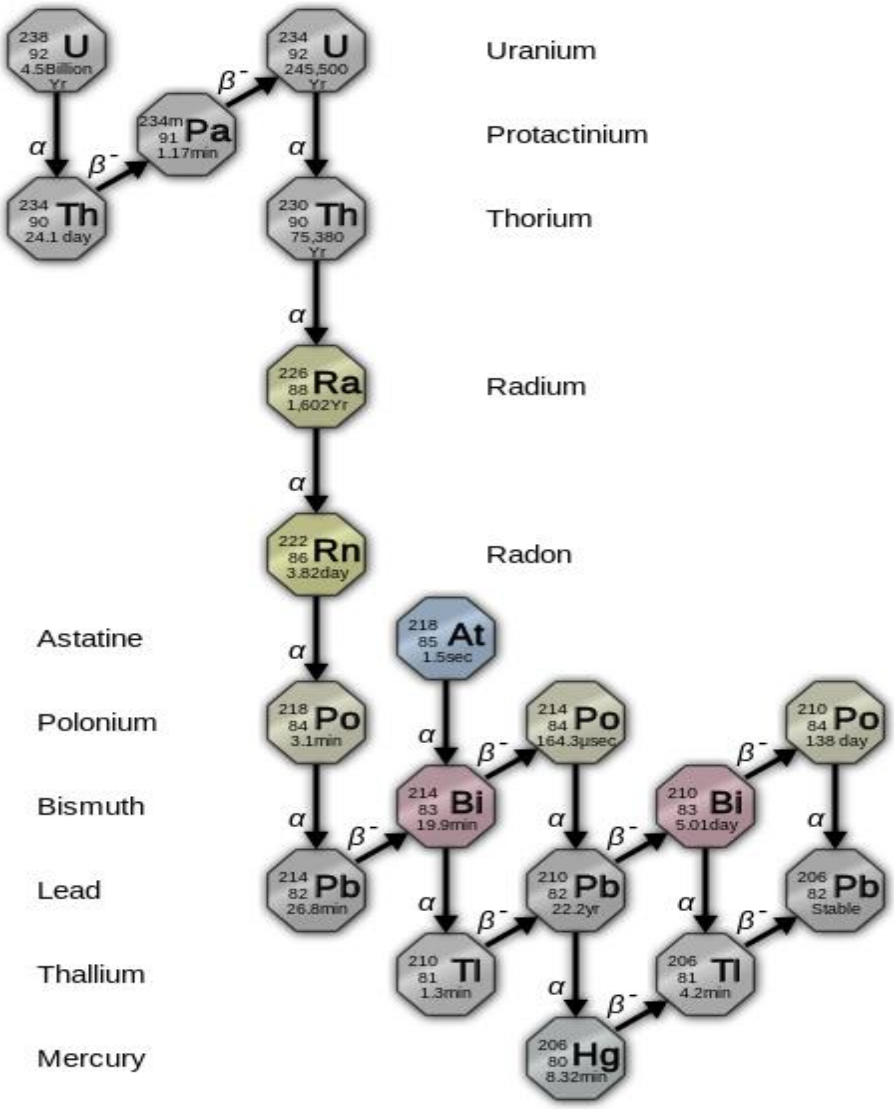


Figure 2.1: Natural Decay Series Uranium-238[2]

Table 2-3: Actinium Series Table [2]

Isotope	Half-life	Decay
^{235}U	$7.1x10^{8}$ years	α
^{231}Th	25.5 hours	β-
^{231}Pa	$3.25x10^{4}$ years	α
^{227}Ac	21.8 years	β-
^{227}Th	18.5 days	α
^{223}Ra	11.4 days	α
^{219}Rn	4.0 sec	α
^{215}Po	1.78 msec	α
^{211}Pb	36.1 min	β-
^{211}Bi	2.13 min	α
^{207}Tl	4.77 min	β-
^{207}Pb	stable	

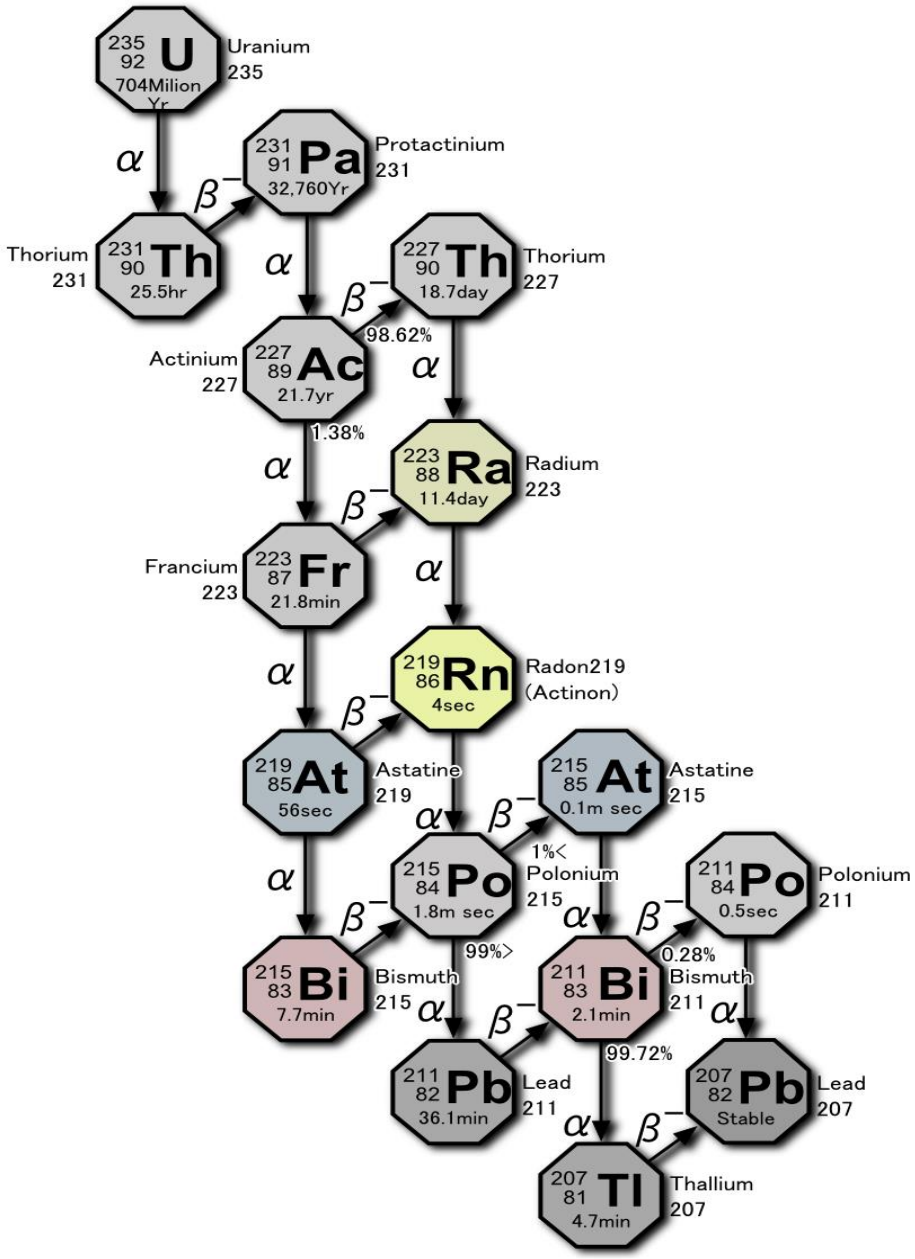


Figure 2.2: Natural Decay Series Uranium-235[2]

Natural Uranium in the process of uranium extraction from the ore, all radioactive daughter products in both radioactive decay series are eliminated (with the exception of the radiogenic isotope ^{234}U). A new secular equilibrium is established in a period of a few months between ^{238}U and two other radioactive daughter products (^{234}Th and ^{234m}Pa). Secular equilibrium between the uranium isotopes ^{238}U and ^{234}U is not disturbed by the extraction process. Similarly, a new secular equilibrium is established

in a period of a few days between the ^{235}U and one other radioactive daughter product, ^{231}Th.

Enriched and Depleted Uranium in order to sustain the chain reaction of nuclear fission, uranium has to be enriched by the fissile isotope ^{235}U to a reactor grade of 3-5% or weapon grade (90%+) uranium. This process not only produces the enriched product, but also a waste stream depleted in ^{235}U, typically to less than 0.3%. The Nuclear Regulatory Commission (NRC) defines depleted uranium (DU) as uranium in which the content of the ^{235}U isotope is less than 0.72%. The military specifications designate that DU used by the Department of Defense (DoD) contains less than 0.3% of ^{235}U. In actuality, DoD uses only DU that contains approximately 0.2% of ^{235}U. In other words, the ^{235}U content in depleted uranium is lowered to 28% of its content in natural uranium.

Nuclear Fuel Cycle

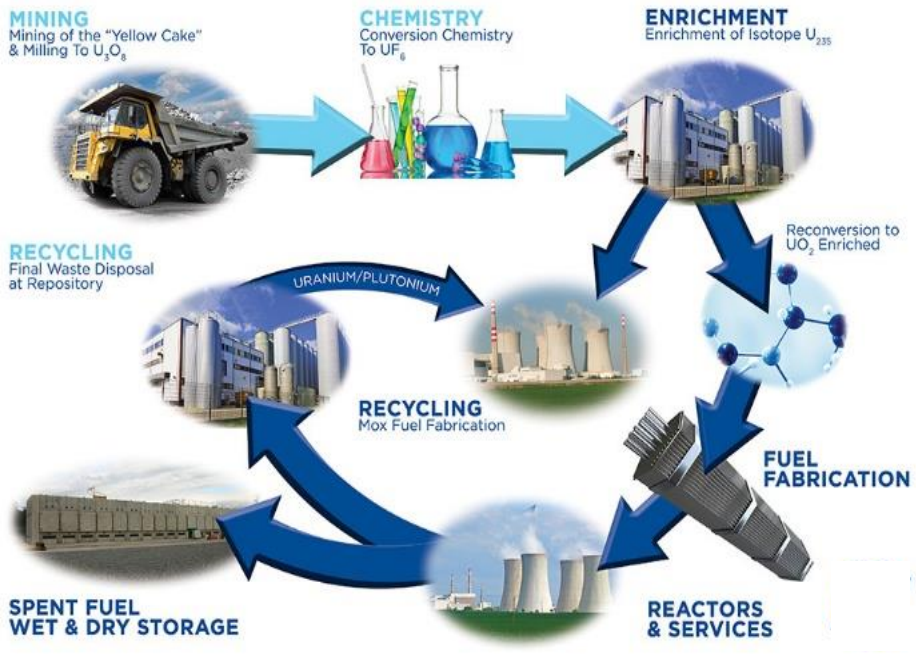


Figure 2.3: Nuclear Fuel Cycle Process[3]

[4]In the nuclear fuel cycle process the Uranium is an element that is widely distributed within the earth's crust. Its principal use is as the primary fuel for nuclear power reactors. Naturally occurring uranium is composed of about 99.3% ^{238}U, 0.7% ^{235}U and traces of ^{234}U. ^{235}U is the fissile isotope of uranium which its atoms have a high probability of undergoing fission after capture of a thermal neutron. In order to use uranium in the ground, it has to be extracted from the ore and converted into a compound which can be utilized in the further steps of the nuclear fuel cycle. The form of uranium to be used in next step is called uranium concentrate and known as yellowcake due to its color.

Then in the uranium is produced by conventional ore mines and ore processing plants. Uranium ores usually contain 0.1% to 0.5% of uranium although higher grades (up to several per cent) have been found in some cases. Uranium is extracted by several basic processes. In order to use the uranium for the nuclear fuel uranium concentrate has to be converted to other forms which is usable by the further steps in the fuel cycle, i.e. uranium hexafluoride (UF_6) in case of enriched uranium fuel, natural uranium oxide (UO_2) in case of PHWRs, metal uranium in case of fuel based on metallic uranium alloy.[4]

Next is the conversion to UF_6 is a two-stage process. In the first stage, the uranium is converted into uranium tetrafluoride (UF_4), green salt. The UF_4 is a solid with a melting point of 960 °C. This stage involves dissolving the uranium concentrates with acid and purifying and then calcining it to produce UO_3 powder. This product is then hydro fluorinated with hydrofluoric acid, which converts it into UF_4, which is granular and green. In the second stage, the UF_4 is converted into uranium hexafluoride (UF_6) through fluorination. One of the chemical characteristics of UF_6 is that it turns into a gas when heated at relatively low temperature. The fluorine used in this process is produced through electrolysis of hydrofluoric acid. [4]

The crucial part is the uranium enrichment process generates depleted UF_6, which might be converted into stable, insoluble and non-corrosive U_3O_8 that can be safely stored pending reuse.

The enrichment of uranium is a physical process used to increase the concentration of the ^{235}U isotope. Enrichment is the altering of isotope ratios in an element, and is usually done by isotope separation. Enrichment processes are made up of many stages, both in series and parallel, so it is usual to speak of separation factors per stage of process. When each process stage has only a small separation factor, many stages in series are needed to get the desired enrichment level.

The next step in the nuclear fuel cycle after enrichment (after conversion in the case of natural uranium fuel) is manufacturing the nuclear fuel in the form of an assembly in order to be utilized in the nuclear power reactors. The assembly has to be in a certain shape to meet the neurotic and thermal hydraulic design of the reactor and in a certain material form to provide first level of containment of radioactive material including fission products and other actinides which are produced during the irradiation of the nuclear fuel.

Usually, final product of fuel fabrication plant delivered to the electric utilities is a fuel assembly (FA). An LWR fuel assembly is made of cylindrical tubes called ‘fuel rods’ containing sintered uranium oxide pellets which is the fissile material held in place in a metal frame, or ‘skeleton’, usually made of zirconium alloy. An assembly can contain 200 to 500 kilograms of heavy metal, depending on the type of assembly.

[4]One of the important steps in the closed nuclear fuel cycle is the reprocessing of spent fuel. The spent nuclear fuel still consists of significant amount of fissile material that can be used to produce energy. The considerable amount of ^{235}U is still contained in the spent fuel and there are new fissile nuclides that were produced during the operation of nuclear power reactor such as ^{239}Pu. Closed nuclear fuel cycle considers taking out those fissile material from the spent fuel, refabricating it as fuel and burning in the reactor.

Then the reprocessing strategy considers spent fuel as an energy resource which is recovered through reprocessing. Spent fuel contains, (for 4% initial enrichment and 45 GWd/t discharge burn up), about 0.67% unburned ^{235}U, about 0.5% ^{236}U, about 93% ^{238}U, about 1% plutonium (0.67% is Fissile Pu), 0.1% Minor Actinides, 4% of fission products and small amounts of other actinides. After cooling in a pool for a few years, the fuel can be reprocessed. Reprocessing of irradiated nuclear fuel separates plutonium and uranium from the intensely radioactive fission products and other actinides.

2.2 Radiation from Uranium Sample

*H. A. Smith, J*r (1991) [5]stated that the primary radiation used in passive NDA of uranium samples is gamma radiation, which is usually dominated by emission from ^{235}U decay. However, in low enrichment uranium sample, the x-ray radiation is the most intense component of the emission spectrum. There are no common interferences except gamma ray from the ^{238}U daughter such as ^{214}Bi, ^{214}Pb and ^{226}Ra. Table 2.4 list the most intense gamma ray from uranium isotope of interest.

Table 2-4:Gamma radiation from Uranium Isotope[5]

Isotope	Gamma-Ray Energy (keV)	Specific Intensity (gamma/s-g of isotope)
^{232}U	129.1	6.5×10^{8}
	270.5	3.0×10^{7}
	327.8	2.7×10^{7}
^{233}U	119.0	3.9×10^{4}
	120.8	3.2×10^{4}
	146.4	6.6×10^{4}
	164.6	6.4×10^{4}
	245.3	3.8×10^{4}
	291.3	5.8×10^{4}
	317.2	8.3×10^{4}
^{234}U	120.9	5.4×10^{5}
^{235}U	143.8	7.8×10^{3}
	163.4	3.7×10^{3}
	185.7	4.3×10^{4}
	202.1	8.0×10^{2}
	205.3	4.0×10^{3}
^{238}U	742.8	7.1
In equilibrium	766.4	2.6×10^{1}
with ^{234m}Pa	786.3	4.3
	1001.0	7.5×10^{1}

The basic measurement viewing a uranium sample through a channel with a gamma ray detector. The enrichment is deduced from the intensity of the ^{235}U 185.7 keV gamma

ray. If the uranium sample is large enough, the 185.7 keV gamma rays from only a fraction of the total sample reach the detector because of the strong absorption of typical uranium bearing materials at this energy investigated by [6]*M.H. Naseef et al*. (2009).

J.L. Praker (1991)[7] in the studies observed, there are two reasons for the lack of the proportionality are the rate-related electronic processes of dead time and pulse pileup and self-attenuation of the samples.

Therefore in *E. Robu et al.* (2009)[8] studies, the need of gamma-ray self-attenuation correction factor F_{att}

$$F_{att} = - \ln (I_x/I_0)/ [1- (Ix/I_0)]$$

Where *I* and I_0 are the peak count rates for the actual sample and the standard air sample respectively.

The aim is to apply a simple practical to correct for self-attenuation effect. This is concern with low energy photon and the variations from samples to samples and sample to standards.

2.3 Method Non Destructive Analysis

[9]R. Sher et al. (1980), non-destructive methods advantages are all material is assayed and non-destructive methods are quick. They can handle large amount s of material and high throughputs therefore these methods are generally cheaper per sample. They also can analyse the material without any sample preparation, so they are not subject to the losses that sometimes plague chemical methods. On the other hand, non-destructive methods can influence by matrix effects due to other materials in the sample, which can require substantial corrections.

M.H. Naseef et al. (2009)[6], the studies suggested non-destructive technique in the determination of the uranium concentration and the isotopic abundance, especially in the case of depleted, natural and low enriched uranium samples.

[10]Y.Y. Ebaid (2009) recommended to use gamma-ray spectrometry as non-destructive and relative simple technique to assess the uranium isotopic ratio in

environmental samples in order to investigate their status regarding being normal, enriched or depleted.

P.D' Chunha et al. (2012)[11] in the studies concludes that XRF analysis method have simple lines, less interference and good stability and gamma ray spectrometry is useful non-destructive method that permits the simultaneous determination of more than one radionuclide without involving the radiochemical separation.

Gamma-ray spectrometry is an important technique for the measurement of quantities of nuclear material holdup in processing equipment. Gamma-ray spectrometry isotope specific in determination isotopic distribution. Spectrometry allow independent measurement of single or multiple isotopes. Gamma-ray detector can be collimated and shielded against background.

Good safeguards practice include unbiased measurement with the smallest quantities possible. Even a small relative's bias in the overall results for foldup in average facility can translate to a significant quantity. Unbiased measurements of holdup also contributed to reduced operation costs for safeguards, criticality safety, and management of waste.

Routine portable non-destructive analysis (NDA) measurements of in process deposits special nuclear material (SNM) use low and medium resolution gamma-ray spectrometry and methodologies that are implemented with generalized.

2.4 Selection of Detectors

Germanium detectors presently offer the best resolution but must be cooled by liquid nitrogen. They are capable of resolving complex gamma spectra and determining the isotopic composition of essentially all nuclear materials present in the nuclear fuel cycle. In Figure 2.4, High pure Germanium (HPGe) available with sufficient resolution to measure the composition of samples.

Then the room temperature superconductor detectors cadmium telluride (CdTe) has a proven record in safeguard verification measurements and related application. CdTe detector is providing medium resolution with reasonable efficiency and ideal for field measurement and for the design of small detection probe that can be operated in close proximity to the item to be verified even if there are space restrictions illustrate in Figure 2.5.

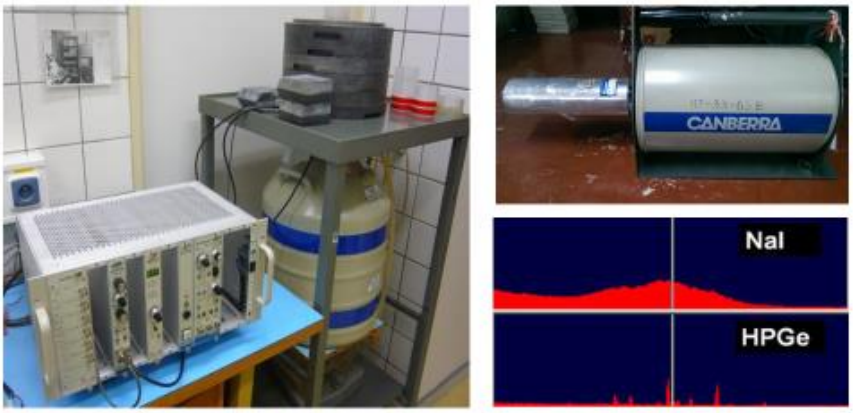


Figure 2.4: HPGe Detector

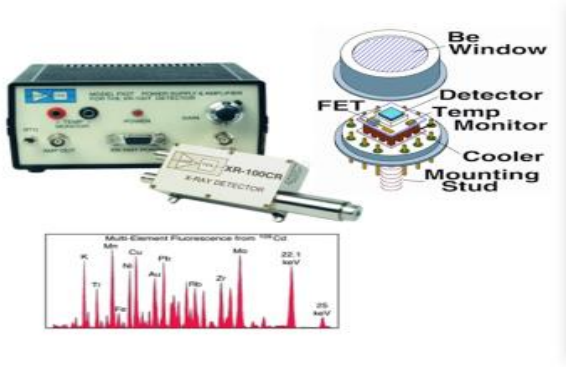


Figure 2.5: CdTe Detector

CHAPTER 3

METHODOLOGY

In this study a gamma ray spectrometer Canberra type, equipped with hyper-pure germanium (HPGe) detector and Cadmium Telluride (CdTe) model XR-100T-CdTe are used. The flow chart in Figure 5 shows the steps in the samples characterization measurement and the experimental procedure of the research study.

3.1 Characterization Techniques

The characterization samples of know uranium bearing material including natural and depleted uranium in yellow cake, natural uranium metal and enriched uranium in fresh fuel of TRIGA Mark III research reactor as shown in Table 3.1 for uranium bearing material samples measured by CdTe and HPGe.

Table 3-1: Uranium Bearing Material Samples Measured by CdTe and HPGe

Uranium Bearing	Sample
Natural Uranium	Metallic Uranium 0.7% ^{235}U
	Standard Uranium 0.527% U_3O_8
	Yellow cake
	Uranium ore (Euxenite)
Depleted Uranium	Depleted Uranium Block
	Radiography Source Projector with depleted uranium
	Uranium compound containing 0.4% ^{235}U
TRIGA Mark III (Fresh Fuel Rod) Enriched Uranium 20% ^{235}U	Control Rod (8268)
	Fresh Fuel Rod (9619)
	Fresh Fuel Rod (9621)

3.2 Gamma-Rays Detection System

HPGe system is used for measurement of energy spectrum of the emitted gamma rays in the energy range until 200 keV with an efficiency of approximately 10%.MCA function is controlled by Genie 2000 software. The energy calibration of the system is carried out before the analysis using the standard source ^{57}Co. Then energy transition of 185.7 keV is used to estimate the concentration of ^{235}U.

3.3 Elemental Analysis

Elemental concentration analysis is carried out with XRF spectrometry using CdTe detector. The detector model of XR-100T-CdTe is a high performance x-ray and gamma ray detector, preamplifier and cooler system. XRF analysis has been proven to be valuable tool for determining trace quantities of uranium bearing material in different type of samples and it can provide rapid, multi-element measurement with minimal sample preparation. In Table 3.2 shows the summarization of the detector selection with the purpose of use and the identification will be used in this research study.

Table 3-2: List of Detector Used for Measurement

Detector	**Purpose of Use**	**Comment**	**Study**
HPGe (Canberra)	Gamma-ray measurement and spectrometry	Excellent energy resolution	Allow a precise quantitative determination of the radioactive concentration of sample
CdTe (XR-100T-CdTe)	X-ray Spectrometry	Small size; Good energy resolution	Elemental Analysis

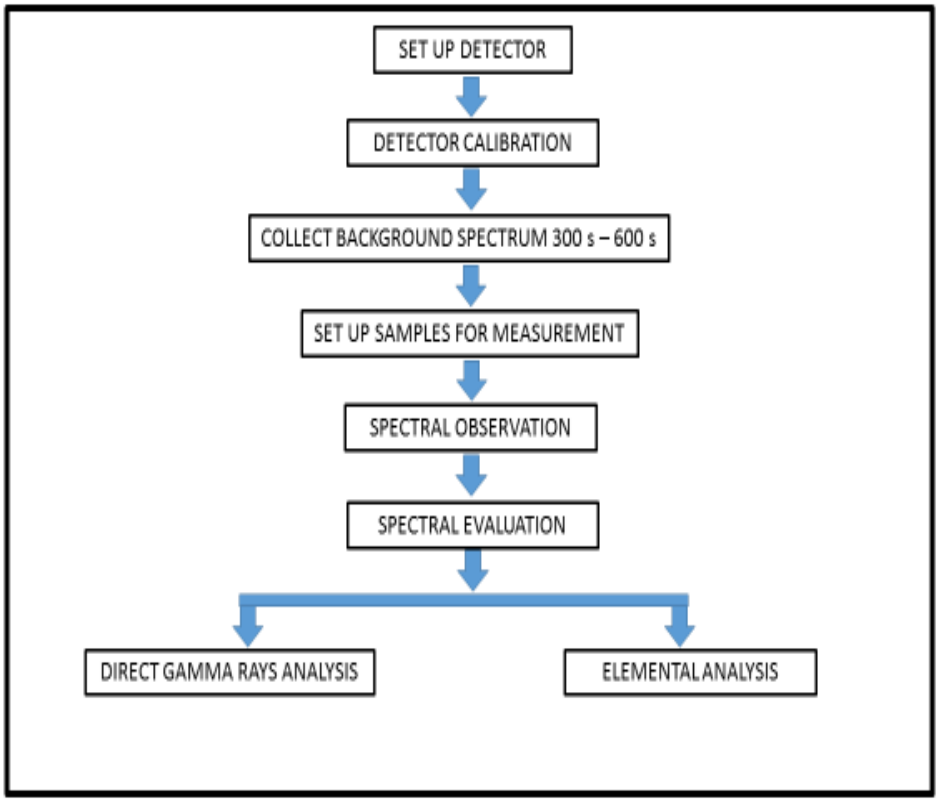


Figure 3.1: Flow chart of the sample preparation and identification techniques

Figure 3.1 shows the chart of the sample and the identification techniques. It is start with the set up the detector followed by the calibration by using the appropriate source. Before start the actual collection data, the measurement of background to ensure there low background count to ensure the accuracy of the measurement.

In this research study there are three part of experiment set up according to the segment below;

- **Part I**: Characterization of XRF and Direct Gamma Rays Measurement by CdTe and HPGe Detectors for Natural Uranium and Depleted Uranium Samples
- **Part II**: Characterization of Direct Gamma Rays Measurement by HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactor with Different Distances

- **Part III**: Characterization of XRF and Direct Gamma Rays Measurement by CdTe and HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactor

Table 3-3: Table Summary of TRIGA Mark III Fresh Fuel Element Specification

Fuel Moderator Material	**Nominal Value**
Uranium content	20 wt%
Uranium weight	~ 98 gm
H/Zr	1.6
Erbium content	0.5 wt%
Enrichment (U-235)	20%
Diameter	3.63 cm
Length	38.1
Cladding	
Material	Type 304 SS
Wall thickness	0.051 cm

The information above shows the TRIGA Mark III fresh fuel rod specification and the example of the fresh fuel rod as in Figure 3.2. Then in Figure 3.3 below shows the fresh fuel rod with the labeling of the segment.

Figure 3.2: TRIGA Mark III Fresh Fuel Rod with 20% ^{235}U

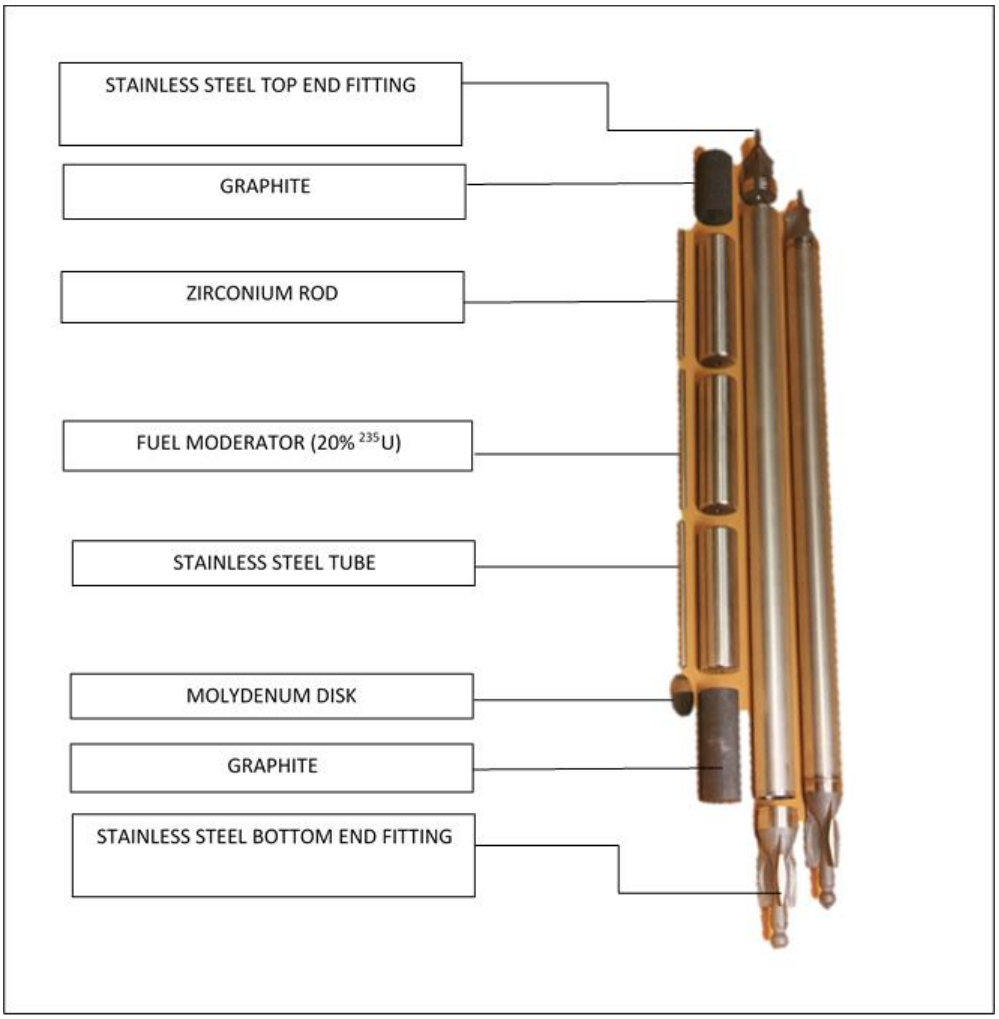


Figure 3.3: TRIGA Mark III Fresh Fuel Rod with 20% ^{235}U with the label

The diagram set up is shown in figure as below with the corresponding part of the experiment.

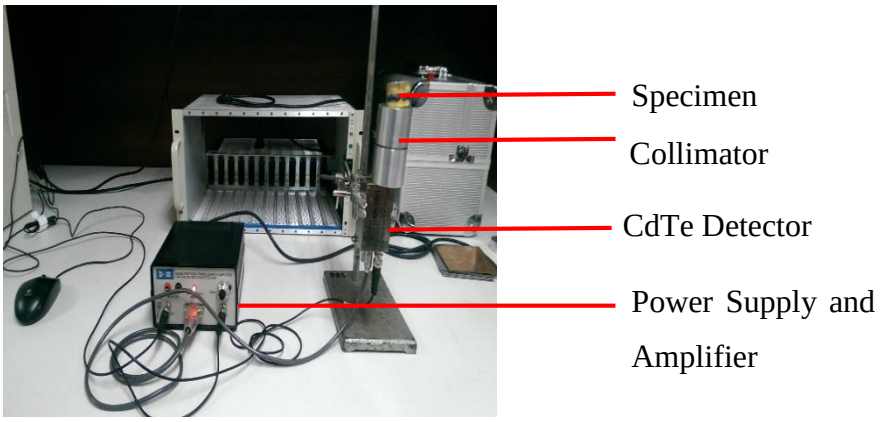


Figure 3.4: Experiment Set Up for Part I Samples with CdTe Detector

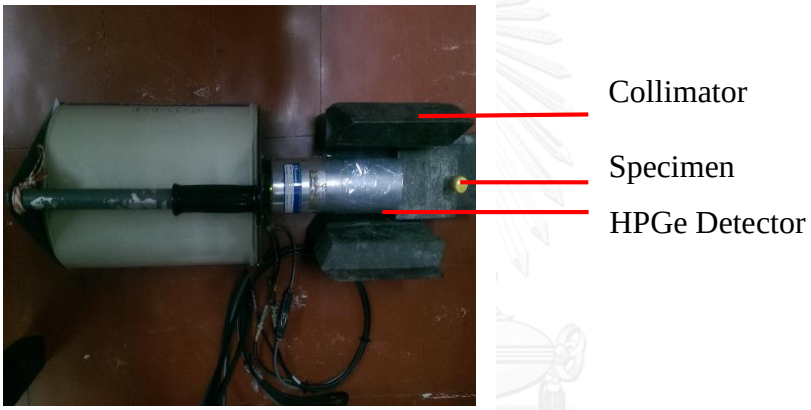


Figure 3.5: Experiment Set Up for Part I Samples with HPGe Detector

In the Figure 3.4 and Figure 3.5 shows the experiment set up for Part I, characterization of Uranium bearing material using x-ray fluorescence and direct gamma rays measurement for elemental and direct gamma ray analysis respectively. In part I there are two categories of uranium bearing material; natural uranium and depleted uranium. The specimens for natural are metallic uranium with 0.7% ^{235}U, metallic uranium 0.7% ^{235}U, standard uranium 0.527% U_3O_8, yellow cake and uranium ore (euxenite). Then the second category is depleted uranium with three specimens which are depleted uranium block, radiography source projector with depleted uranium and uranium compound containing 0.4% ^{235}U. Each of the specimens was measured in 300 second for both detectors CdTe and HPGe.

Figure 3.6: Experiment Set Up for Part II Samples with HPGe Detector

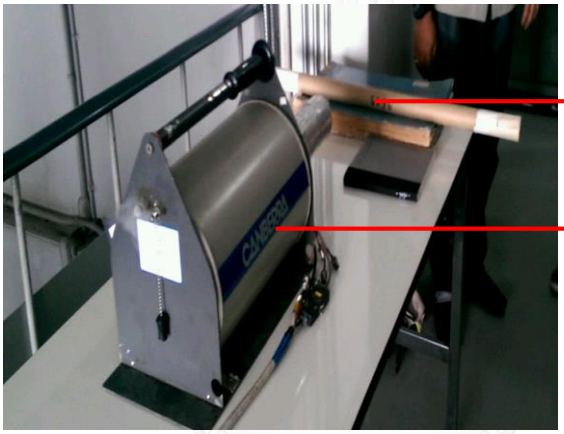


Figure 3.7: Experiment Set Up for Part II Samples with HPGe Detector front side

In the Figure 3.6 and Figure 3.7 shows the experiment set up for Part II, characterization of uranium bearing material of 20% ^{235}U fresh fuel rod of Thailand TRIGA Mark III research reactor using direct gamma rays measurement by HPGe. In part II there are three specimens with control rod (8268), 9619 and 9621 fresh fuel rod with 20% 235U. Each of the specimens were measured in 300 second for different distances started with 30 cm, 60 cm and 90 cm using HPGe detector.

In Part III the experiment set up shown in Figure 3.8 until Figure 3.13. In this part the measurement of specimen is same in Part II. The measurement with CdTe and HPGe detector are 60 seconds and 180 seconds respectively. The specimens are placed closed contact with the detector for both detector measurement.

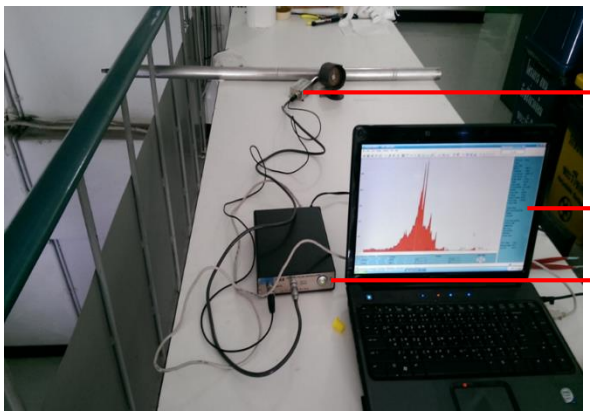


Figure 3.8: Experiment Set Up for Part III Samples with CdTe Detector

Figure 3.9: Experiment set up from the front side

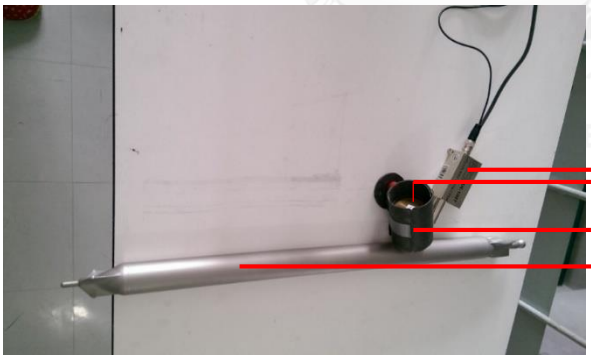


Figure 3.10: Experiment Set Up for Part III Samples with CdTe Detector up side

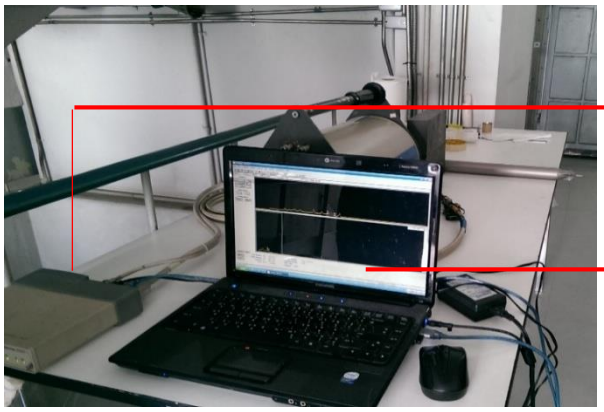


Figure 3.11: Experiment Set Up for Part III Samples with HPGe Detector

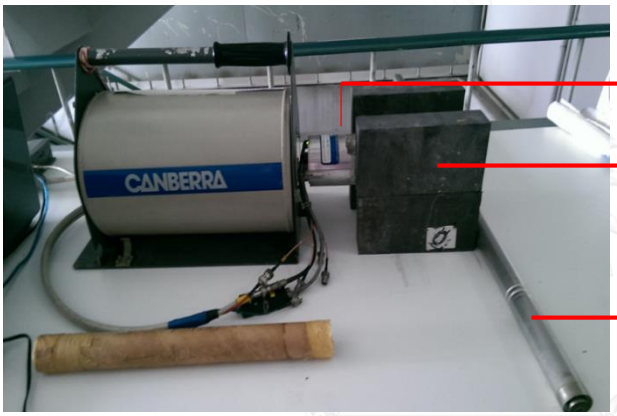


Figure 3.12: Experiment Set Up for Part III Samples with HPGe Detector

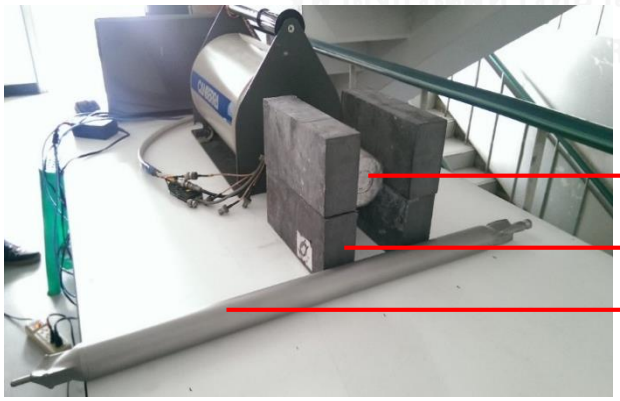


Figure 3.13: Experiment set up from the front side

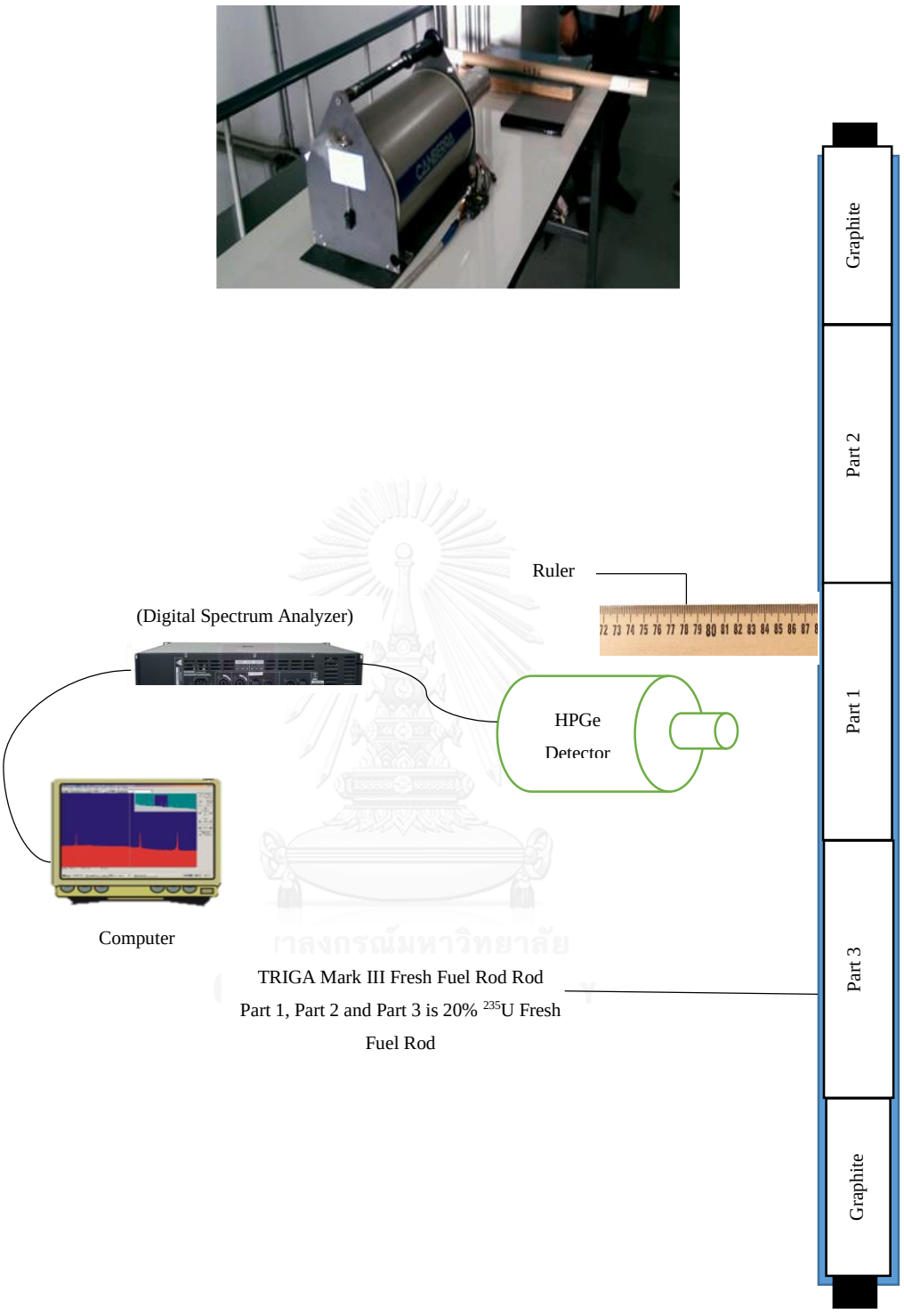


Figure 3.14: Experiment diagram set up with HPGe detector (Part II)

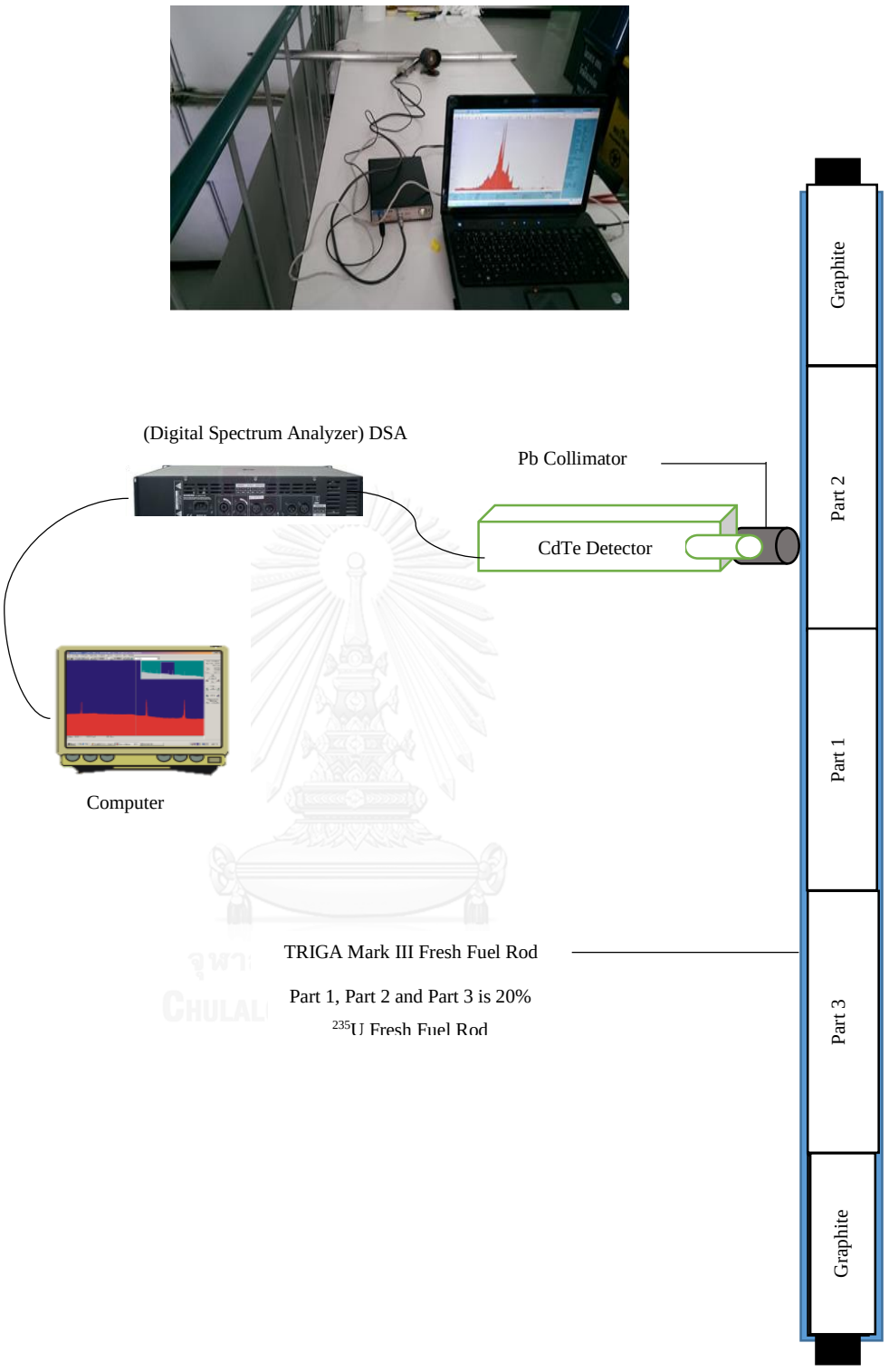


Figure 3.15: Experiment diagram set up with CdTe detector (Part III)

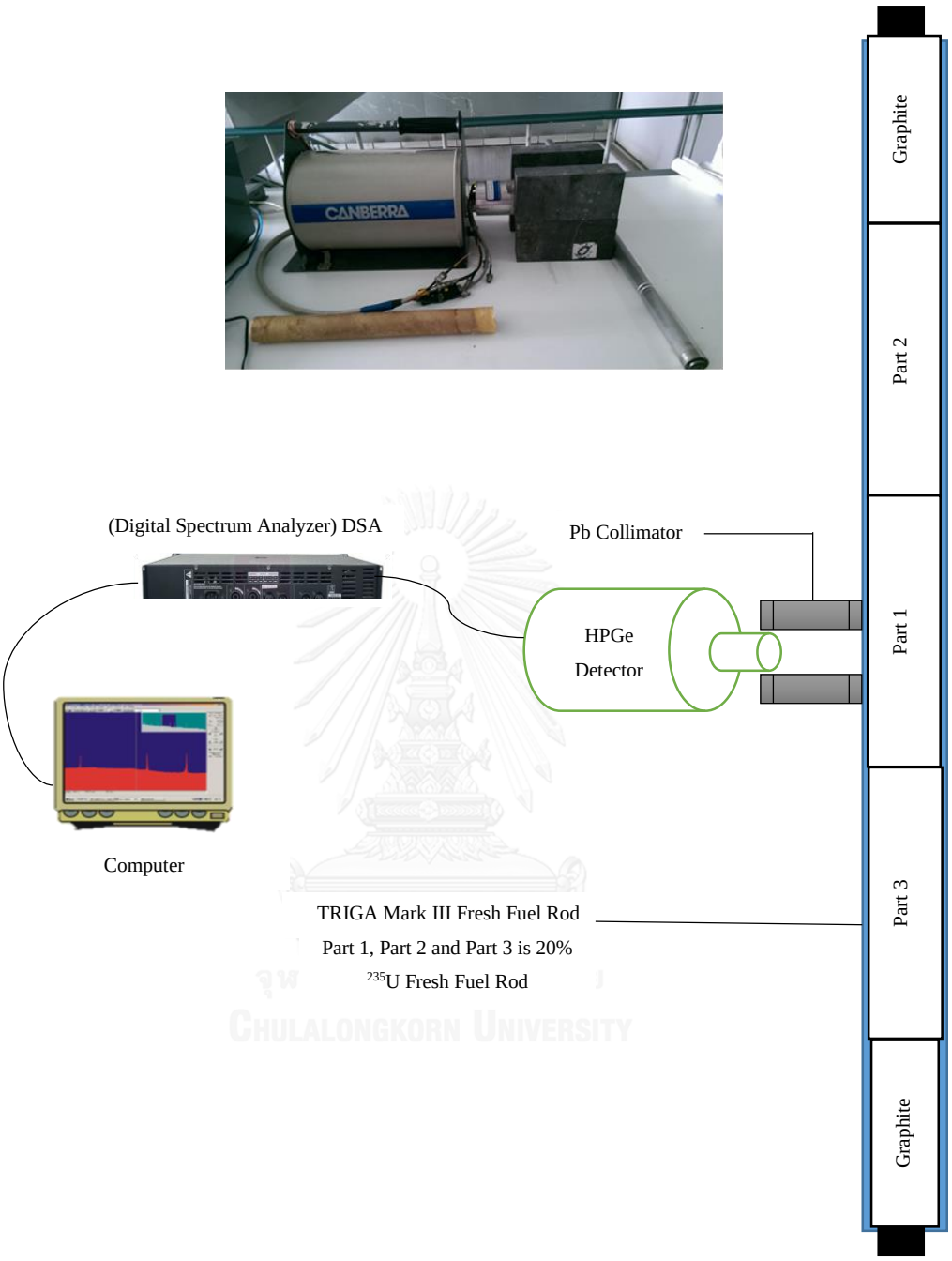


Figure 3.16: Experiment diagram set up with HPGe detector (Part III)

CHAPTER 4

RESULTS AND DISCUSSION

In this chapter discusses the results determined from the research. The results will be discussed in three parts;

- **Part I**: Characterization of XRF and Direct Gamma Ray by CdTe and HPGe Detectors for Natural Uranium and Depleted Uranium Samples

- **Part II**: Characterization of Direct Gamma Ray by HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactors with Different Distances

- **Part III**: Characterization of XRF and Direct Gamma Ray by CdTe and HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactors

The first part (**Part I**), there are seven samples for the characteristic of elemental analysis and direct gamma ray measurement by CdTe and HPGe detectors. The seven samples are categorized by two uranium bearing material which are natural uranium and depleted uranium. There are four sample for natural uranium, which are metallic uranium 0.7% ^{235}U, standard uranium 0.527% U_3O_8, yellow cake and uranium ore (Euxenite). Then the depleted uranium samples are depleted uranium block, a radiography projector source with depleted uranium and compound containing 0.4% ^{235}U. All samples are shown in Table 4.2.

Then, the second part (**Part II**) of the results and discussion are the results of gamma ray measurement for 20% ^{235}U of fresh fuel rod TRIGA Mark III research reactor of Thailand Institute of Nuclear Technology (TINT). There are three samples with series of 8268 the control rod and another two fresh fuel rod series of 9619 and 9621. These samples are measured in three different distance with HPGe detector.

Finally the third part (**Part III**) is the results of the x-ray fluorescence and direct gamma ray measurement with fresh fuel rod TRIGA Mark III TINT research reactor with both

CdTe and HPGe detectors. In this measurement and characterization of sample same as in **Part II**, which are three series of 20 % ^{235}U fresh fuel rod from TRIGA Mark III research reactor. The measurement of the XRF and direct gamma ray with closed contact with the specimen. The summarization of the experiment as shown in Table 4.1 below;

Table 4-1: Summarization Characterization of Uranium Bearing Material Samples Measured by CdTe and HPGe

Experiment	**Detector**	**Characterization**	**Specimens**
Part I	CdTe and HPGe	Elemental Analysis and Direct Gamma Rays Measurement	**Natural Uranium** Metallic Uranium 0.7% ^{235}U Standard Uranium 0.527% U_3O_8 Yellow cake Uranium ore (Euxenite) **Depleted Uranium** Depleted Uranium Block Radiography source Projector with Depleted Uranium Uranium compound containing 0.4% ^{235}U
Part II	HPGe	Direct Gamma Rays Measurement With Different Distances	**TRIGA Mark III Fresh Fuel Rod (20% ^{235}U)** Control Rod (8268) 9619 9621
Part III	CdTe and HPGe	Elemental Analysis and Direct Gamma Rays Measurement (Closed Contact with Detectors)	**TRIGA Mark III Fresh Fuel Rod (20% ^{235}U)** Control Rod (8268) 9619 9621

4.1 Part I: Characterization of XRF and Direct Gamma Rays Measurement by CdTe and HPGe Detectors for Natural Uranium and Depleted Uranium Series

There are seven samples of uranium bearing material, categorized by two, natural uranium and depleted uranium as below:

Table 4-2: Uranium Bearing Material Samples Measured by CdTe and HPGe

Uranium Bearing	**Sample**
Natural Uranium	Metallic Uranium 0.7% ^{235}U
	Standard Uranium 0.527% U_3O_8
	Yellow cake
	Uranium ore (Euxenite)
Depleted Uranium	Depleted Uranium Block
	Radiography source Projector with Depleted Uranium
	Uranium compound containing 0.4% ^{235}U

As the non-destructive method is known for its high accuracy, the XRF technique is extensively used in the elemental assay in the laboratories for all of the samples tested. In this study the main objective for the XRF assay is to detect the present of K x-ray from the specific energy from the uranium bearing material with the purpose being to test its rapid measurement and high accuracy.

X-ray is initially from the atomic electron transition and is elementally specific. The electron occupies specific energy levels that are designated. The exciting photon energy must be high enough to remove an electron from the orbit.

In Figure 4.1 shows characteristic of K x-ray of Metallic uranium 0.7% ^{235}U measured with CdTe for the x-ray fluorescence measurement. The results shows all the spectrums belong to ^{235}U with respective K line electrons. The exciting source use is ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are four major K line

are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.6 keV and $K\alpha_2$ at energy 94.6 keV. Then, $K_{\beta 1}$ and $K_{\beta 2}$ are at 111.3 keV and 114.7 keV respectively.

The Table 4.3 below shows the summarization of the Metallic uranium 0.7% ^{235}U with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of each energy is obtained. The peak area per 300 seconds for $K\alpha_1$ and $K\alpha_2$ are 18778±0.009 and 11126±0.015 respectively. Then, for $K_{\beta 1}$ and $K_{\beta 2}$ are 4075±0.029 and 1029±0.085 respectively.

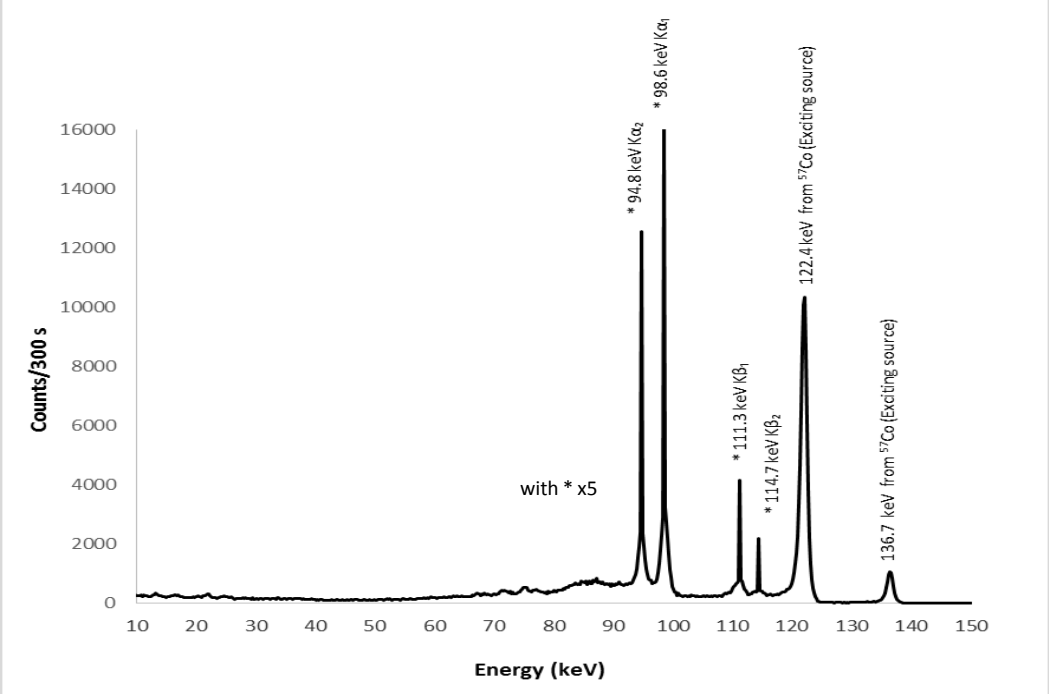


Figure 4.1 Characterization of K X-Ray of Metallic Uranium (0.7% ^{235}U) by CdTe Detector

Table 4-3: Summarization for Metallic Uranium (0.7% ^{235}U) by CdTe Detector

Sample	**Line**	**Energy (keV)**	**Peak Area/300 s**
Metallic	$K\alpha_1$	98.6	18778±0.009
Uranium	$K\alpha_2$	94.6	11126±0.015
(0.7% ^{235}U)	$K_{\beta 1}$	111.3	4075±0.029
	$K_{\beta 2}$	114.7	1029±0.085

Nevertheless, the x-ray fluorescence results based on the elemental analysis can only estimate the presence of uranium bearing material. Thus the presence of gamma ray

spectrometry is needed to confirm the existence of ^{235}U. The gamma ray measurement is tested by using HPGe to confirm the presence of ^{235}U based on the approach enrichment meter principle. Most prominent gamma ray was found from the decay of ^{235}U being 185.7 keV.

In Figure 4.2 referred to Metallic Natural Uranium 0.7% ^{235}U exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there are six major energy observed belong to ^{235}U are 98.9 keV, 111.7 keV, 143.9 keV, 163.4 keV, 185.7 keV and 205.2 keV.

Then Table 4.4 below, shows the summarization of the metallic uranium 0.7% ^{235}U with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each energy are obtained. The peak are per 300 seconds at 93.9 keV and 111.7 keV are 26859 ± 0.009 and 2360 ± 0.015 respectively. Then, at 143.7 keV and 163.4 keV are 2599 ± 0.042 and 1315 ± 0.074 respectively. Then the final two peaks at 185.7 keV with peak area 15523 ± 0.009 and at 205.2 keV is 1232 ± 0.066 peak area per 300 seconds.

As prediction, the prominent energy at 185.7 keV shows the highest photo peaks area from the decay of ^{235}U was determined. The photo peaks intensity of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample.

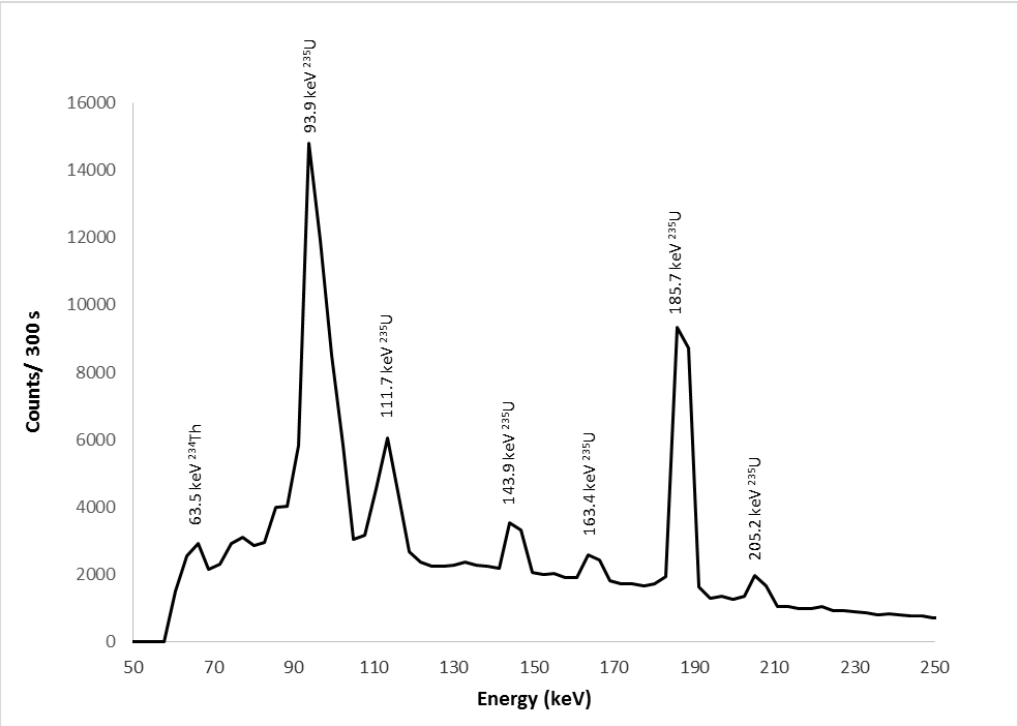


Figure 4.2: Characteristic of ^{235}U Gamma Ray for Metallic Uranium (0.7% ^{235}U) by HPGe Detector

Table 4-4: Summarization for Metallic Uranium (0.7% ^{235}U) by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Metallic Uranium	93.9	26859±0.009
(0.7% ^{235}U)	111.7	2360±0.015
	143.9	2599±0.042
	163.4	1315±0.074
	185.7	15523±0.009
	205.2	1232±0.066

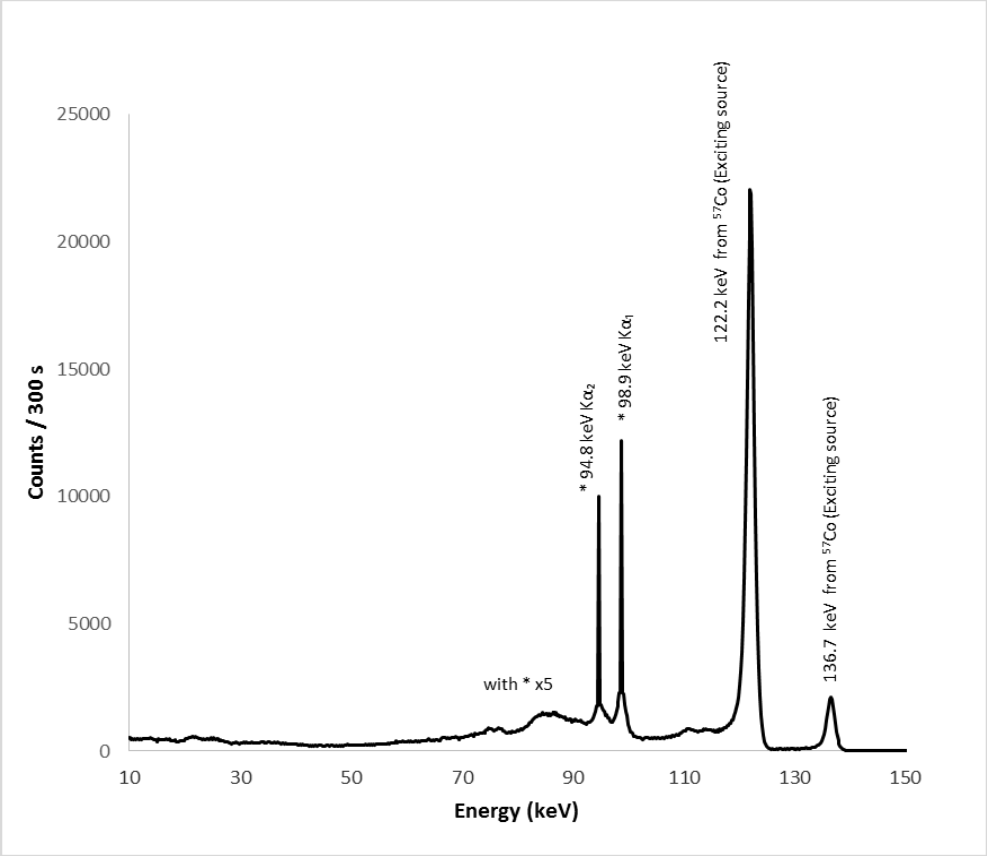


Figure 4.3: Characterization of K X-Ray of Yellow Cake by CdTe Detector

Table 4-5: Summarization for Yellow Cake by CdTe Detector

Sample	**Line**	**Energy (keV)**	**Peak Area/300 s**
Yellow Cake	$K\alpha_1$	98.9	12040±0.017
	$K\alpha_2$	94.8	6828±0.008

In Figure 4.3 shows characteristic of K x-ray of yellow cake measured with CdTe for the x-ray fluorescence measurement. The results shows all the spectrums belong to ^{235}U with respective two K line electrons. The same exciting source used ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are two major K line are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.9 keV and $K\alpha_2$ at energy 94.8 keV.

Then, Table 4.5 above, shows the summarization of yellow cake with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each

energy are obtained. The peak are per 300 seconds for $K\alpha_1$ and $K\alpha_2$ are 12040±0.017 and 6828±0.008 respectively. The elemental analysis from the x-ray fluorescence shows the existence of uranium element in the sample. These results may be explained by the fact that the strong absorption energy due to the relative intensities of the x-ray. X-ray originate from atomic electron transitions and are element specific.

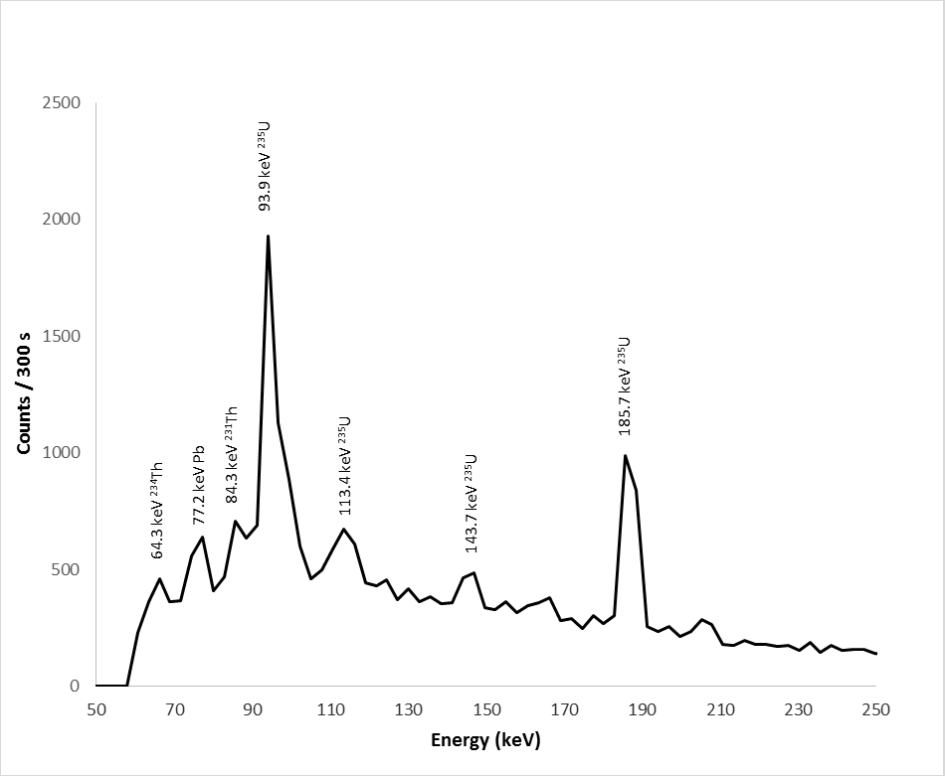


Figure 4.4: Characterization of Gamma Ray of Yellow Cake by HPGe Detector

Table 4-6: Summarization for Yellow Cake by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Yellow cake	93.9	2535±0.003
	113.4	254±0.019
	146.8	212±0.02
	185.7	1317±0.003

In Figure 4.4 referred to yellow cake sample exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there are four major energy observed belong to ^{235}U are 98.9 keV, 113.4 keV, 146.8 keV, 163.4 keV and 185.7 keV.

The Table 4.6, shows the summarization of the yellow cake with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each energy are determined. The peak are per 300 seconds at 93.9 keV and 113.4 keV are 2535±0.003 and 254±0.019 respectively. Then, at 146.8 keV and 185.7 keV are 212±0.02 and 1317±0.003 respectively. As prediction, the prominent energy at 185.7 keV shows the highest photo peaks area from the decay of ^{235}U was determined. The photo peaks intensity of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample.

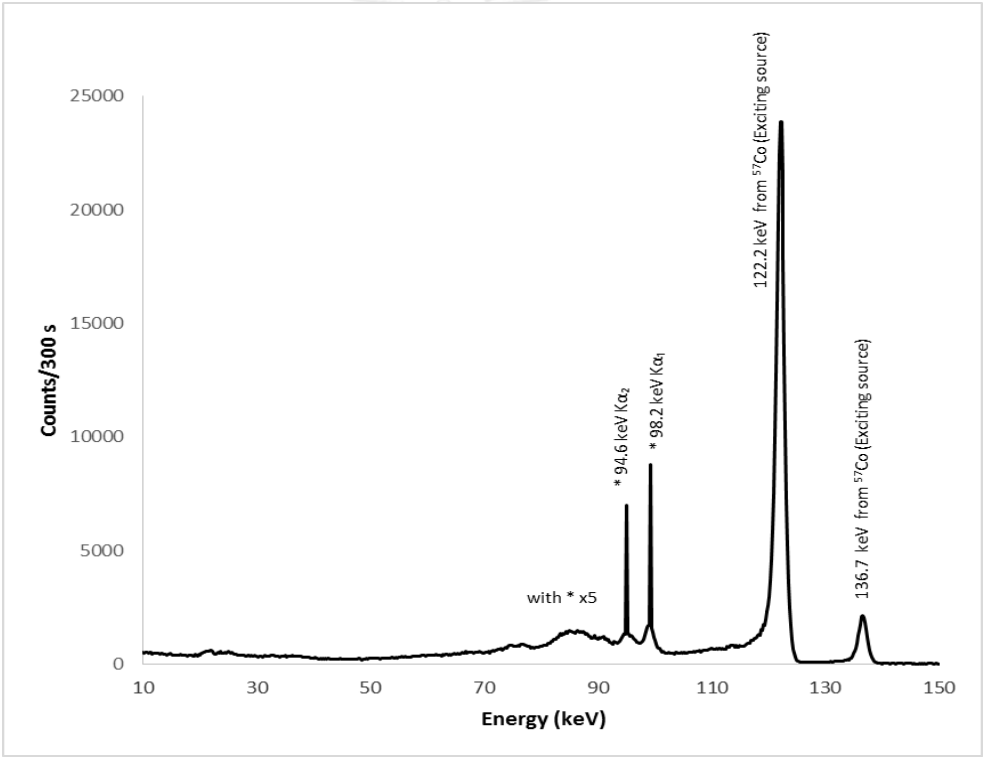


Figure 4.5 Characterization of K X-Ray of Standard Uranium (0.527% U_3O_8) by CdTe Detector

Table 4-7: Summarization for Standard Uranium (0.527% U_3O_8) by CdTe Detector

Sample	Line	Energy (keV)	Peak Area/300 s
Standard Uranium (0.527% U_3O_8)	$K\alpha_1$	98.2	7624±0.020
	$K\alpha_2$	94.6	4167±0.042

In Figure 4.5 shows characteristic of K x-ray of Standard Uranium (0.527% U_3O_8) measured with CdTe for the x-ray fluorescence measurement. The results shows all the spectrums belong to ^{235}U with respective two K line electrons. The same exciting source used ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are two major K line are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.4 keV and $K\alpha_2$ at energy 94.8 keV with the peak area are obtained 7624±0.02 and 4167±0.042 respectively. The elemental analysis from the x-ray fluorescence shows the existence of uranium element in the sample. These results may be explained by the fact that the strong absorption energy due to the relative intensities of the x-ray. X-ray originate from atomic electron transitions and are element specific.

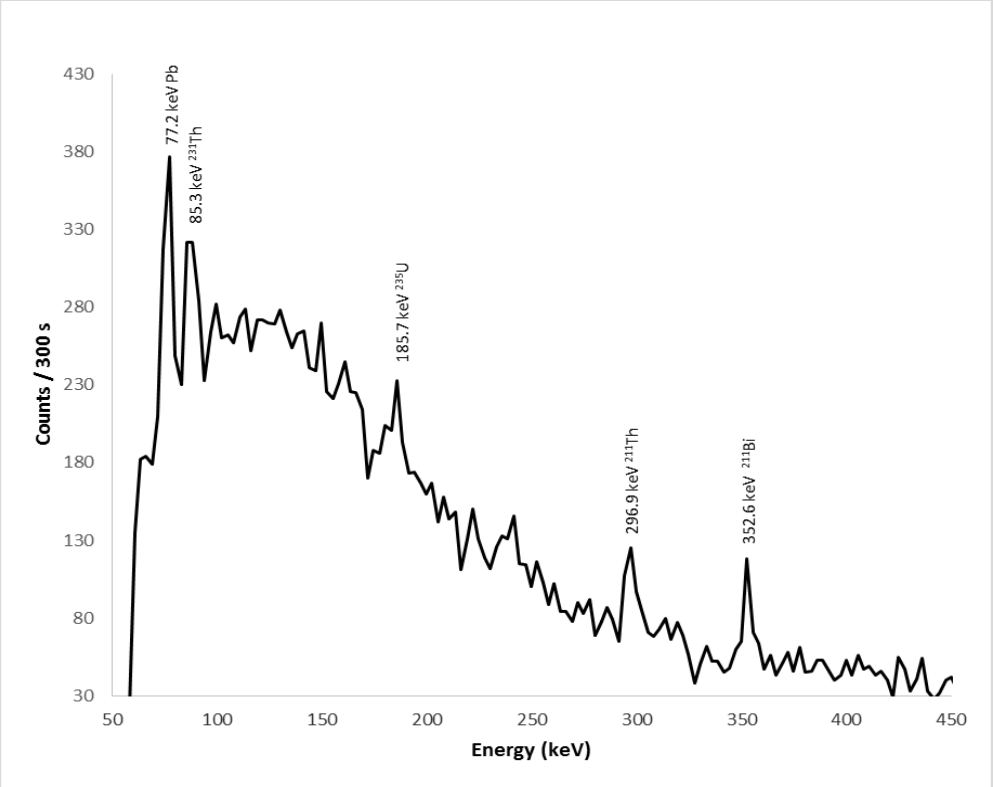


Figure 4.6 Characteristic of ^{235}UGamma Ray for Standard Uranium (0.527% U_3O_8) by HPGe Detector

Table 4-8: Summarization for Standard Uranium (0.527% U_3O_8) by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Standard Uranium	77.2	316±0.13
(0.527% U_3O_8)	85.3	125±0.32
	185.7	115±0.26
	296.9	85±0.31
	352.5	132±0.15

The x-ray fluorescence results based on the elemental analysis can only estimate the presence of uranium bearing material. Thus the presence of gamma ray spectrometry is needed to confirm the existence of ^{235}U. The gamma ray measurement is tested by using HPGe to confirm the presence of ^{235}U based on the approach enrichment meter principle. Most prominent gamma ray was found from the decay of ^{235}U being 185.7 keV.

In Figure 4.6 referred to Standard Uranium (0.527% U_3O_8) sample exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there is only one major energy observed belong to ^{235}U at 185.7 keV.

The Table 4.8, shows the summarization of the Standard Uranium (0.527% U_3O_8) with the energy and the peak area obtained from the experiment. The peak areas per 300 seconds of at each energy are determined. As prediction, the prominent energy at 185.7 keV shows the photo peaks area from the decay of ^{235}U was determined with 115±0.26 per 300 s. The photo peaks intensity of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample.

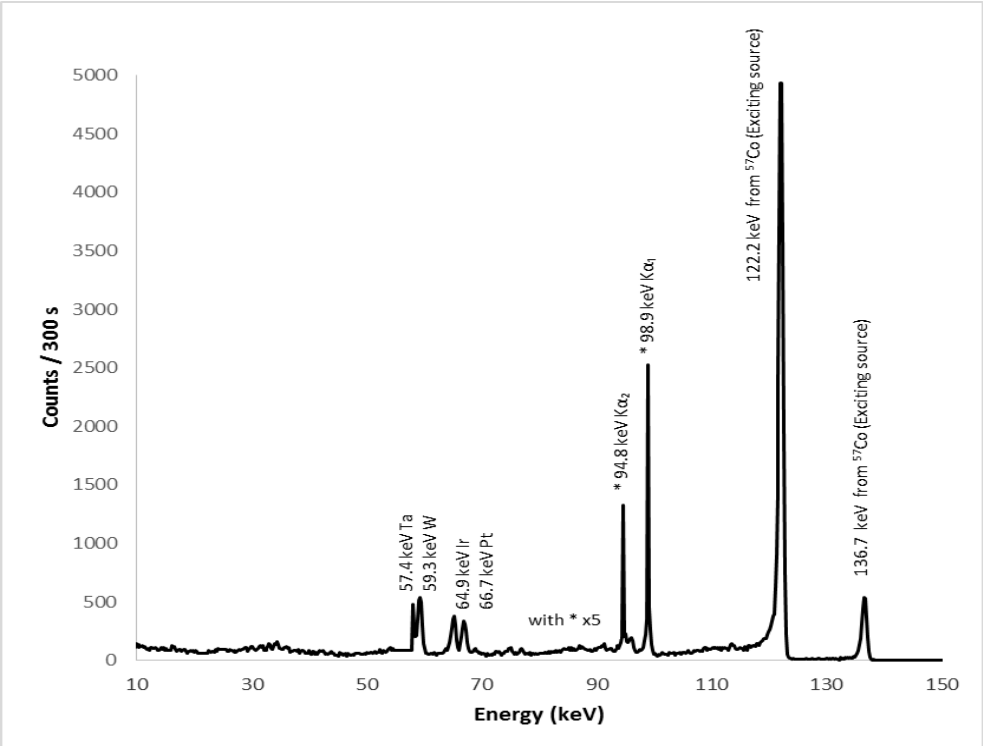


Figure 4.7 Characterization of K X-Ray of Euxenite (Uranium Ore) by CdTe Detector

Table 4-9: Summarization for Euxenite (Uranium Ore) by CdTe Detector

Sample	**Line**	**Energy (keV)**	**Peak Area/300 s**
Standard Uranium	$K\alpha_1$	98.9	2031±0.027
(0.527% U_3O_8)	$K\alpha_2$	94.8	354±0.110

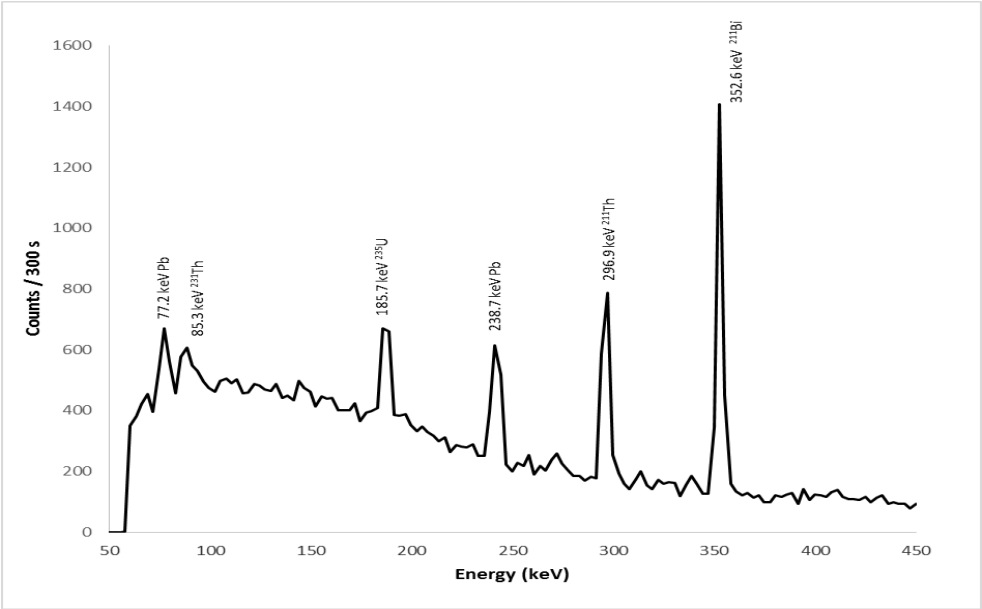


Figure 4.8 Characteristic of ^{235}U Gamma Ray for Euxenite (Uranium Ore) by HPGe Detector

Table 4-10: Summarization Euxenite (Uranium Ore) by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Euxenite	77.2	361±0.169
(Uranium Ore)	85.3	125±0.35
	185.7	670±0.09
	241.3	753±0.06
	296.9	1123±0.04
	352.6	1783±0.03

In this section, discussed the second categories of the samples which are depleted uranium bearing with three samples; depleted uranium block, compound contain 0.4% ^{235}U and radiography projector source with depleted uranium.

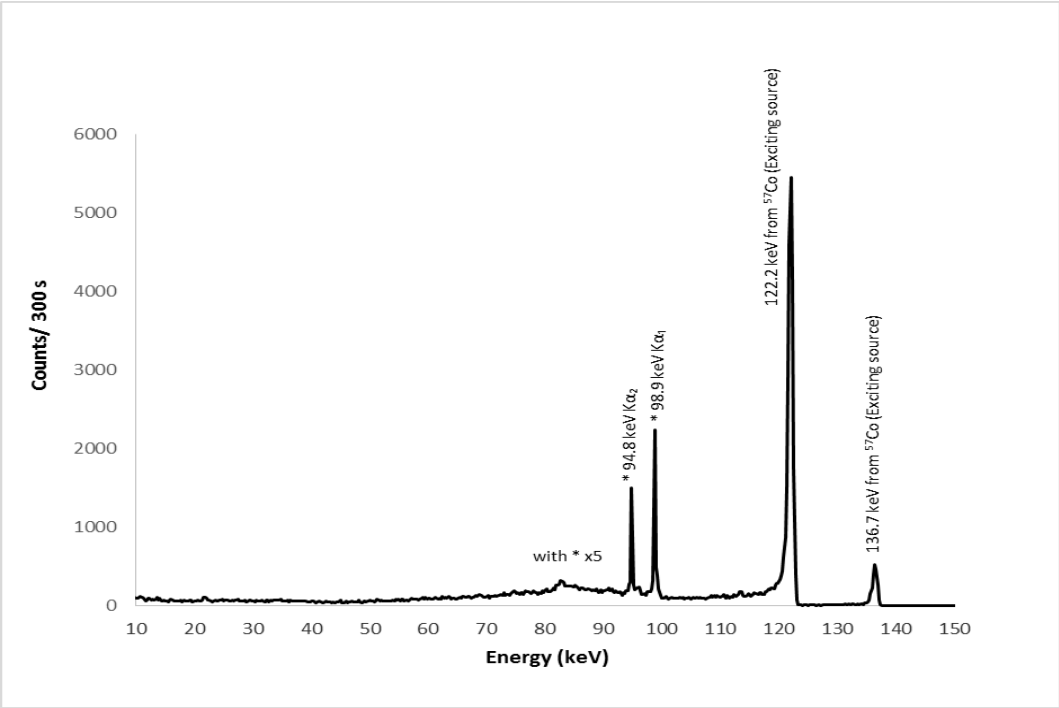


Figure 4.9 Characterization of K X-Ray of Depleted Uranium Block by CdTe Detector

Table 4-11: Summarization for Depleted Block by CdTe Detector

Sample	**Line**	**Energy (keV)**	**Peak Area/300 s**
Depleted Uranium	$K\alpha_1$	98.9	1289±0.041
Block	$K\alpha_2$	94.8	408±0.110

In Figure 4.9 shows characteristic of K x-ray of depleted uranium block measured with CdTe for the x-ray fluorescence measurement. The results shows all the spectrums belong to ^{235}U with respective two K line electrons. The same exciting source used ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are two major K line are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.9 keV and $K\alpha_2$ at energy 94.8 keV with the peak area are obtained 1289±0.041 and 408±0.11 respectively. The elemental analysis from the x-ray fluorescence shows the existence of uranium element in the sample. These results may be explained by the fact that the strong absorption energy due to the relative intensities of the x-ray. X-ray originate from atomic electron transitions and are element specific.

The x-ray fluorescence results based on the elemental analysis can only estimate the presence of uranium bearing material. Thus the presence of gamma ray spectrometry is needed to confirm the existence of ^{235}U. The gamma ray measurement is tested by using HPGe to confirm the presence of ^{235}U based on the approach enrichment meter principle. Most prominent gamma ray was found from the decay of ^{235}U being 185.7 keV.

In Figure 4.10 referred to depleted uranium block sample exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there are three major energy observed belong to ^{235}U at 93.9 keV, 111.7 keV, 163.4 keV and 185.7 keV.

The Table 4.12, shows the summarization of the depleted Uranium block with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each energy are determined. As prediction, the prominent energy at 185.7 keV shows the photo peaks area from the decay of ^{235}U was determined with 3322 ± 0.055 per 300 s. The photo peaks intensity of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample. Another three photo peaks are 93.9 keV, 111.7 keV, 163.4 keV are 28981 ± 0.057, 6436 ± 0.033 and 133 ± 0.18 respectively.

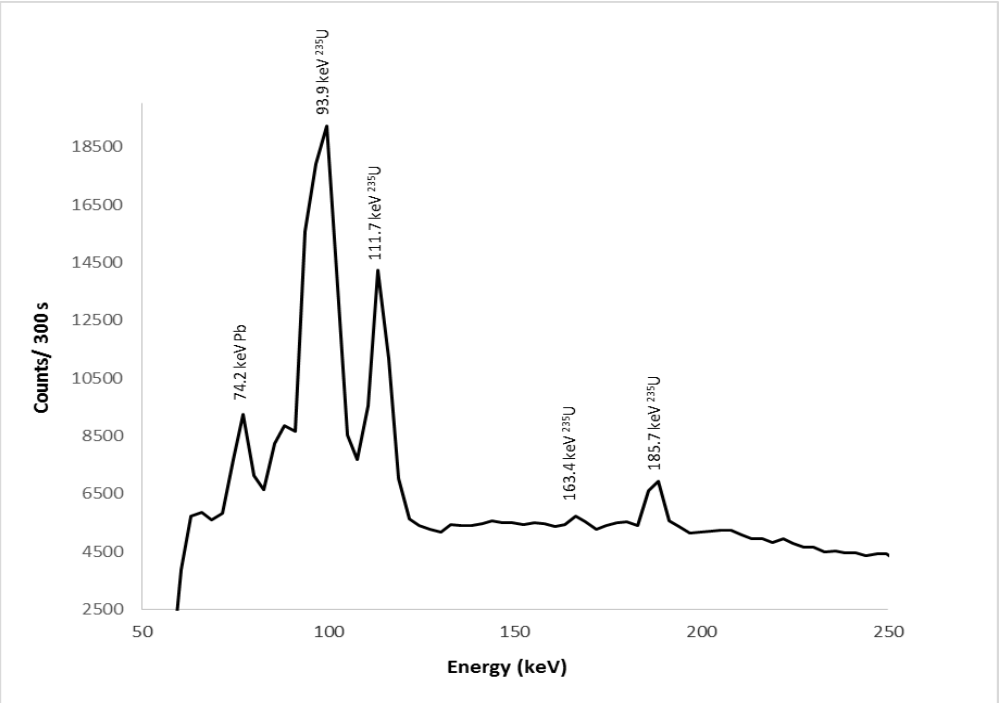


Figure 4.10 Characteristic of ^{235}U Gamma Ray for Depleted Block by HPGe Detector

Table 4-12: Summarization Depleted Block by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Depleted Uranium Block	74.2	3157±0.057
	93.9	28981±0.010
	111.7	6436±0.033
	163.4	133±0.18
	185.7	3322±0.055

In Figure 4.11 below shows the characteristic of K x-ray of 0.4% ^{235}U measured with CdTe for the x-ray fluorescence measurement. The results shows all the spectrums belong to ^{235}U with respective two K line electrons. The same exciting source used ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are two major K line are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.9 keV and $K\alpha_2$ at energy 94.8 keV with the peak area are obtained 6626±0.02 and 3872±0.034 respectively. The elemental analysis from the x-ray fluorescence shows the existence of uranium element in the sample. These results may be explained by the fact that the strong absorption

energy due to the relative intensities of the x-ray. X-ray originate from atomic electron transitions and are element specific.

The x-ray fluorescence results based on the elemental analysis can only estimate the presence of uranium bearing material. Thus the presence of gamma ray spectrometry is needed to confirm the existence of ^{235}U. The gamma ray measurement is tested by using HPGe to confirm the presence of ^{235}U based on the approach enrichment meter principle. Most prominent gamma ray was found from the decay of ^{235}U being 185.7 keV.

In Figure 4.12 referred to 0.4% ^{235}U sample exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there are three major energies observed belong to ^{235}U at 93.9 keV, 113.4 keV and 185.7 keV.

The Table 4.14, shows the summarization of the 0.4% ^{235}U with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each energy are determined. As prediction, the prominent energy at 185.7 keV shows the photo peaks area from the decay of ^{235}U was determined with 1605$\pm$0.037 per 300 s. The photo peak at 93.9 keV with 6500$\pm$0.021 of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample.

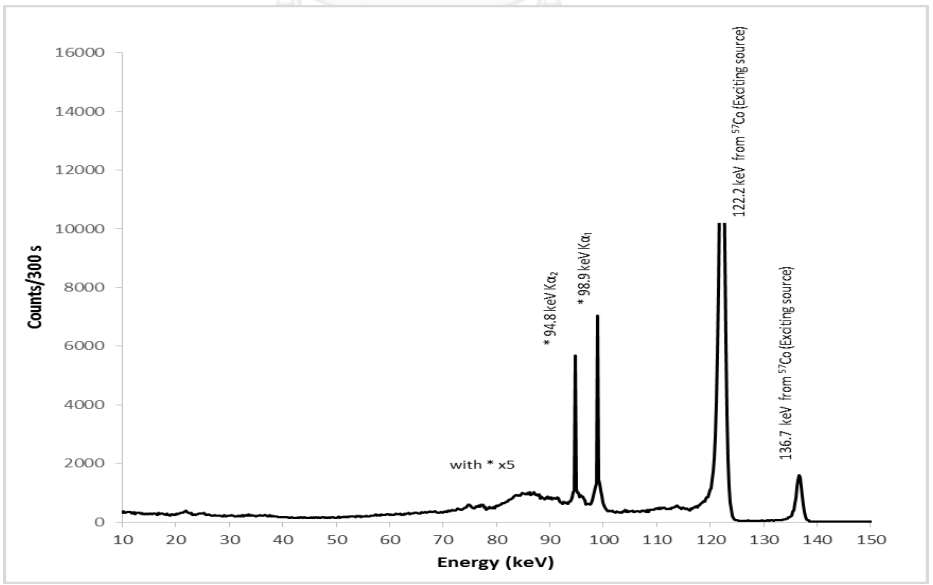


Figure 4.11 Characterization of K X-Ray of Compound containing 0.4% ^{235}U by CdTe Detector

Table 4-13: Summarization for compound containing 0.4% ^{235}U by CdTe Detector

Sample	Line	Energy (keV)	Peak Area/300 s
Compound containing 0.4% ^{235}U	$K\alpha_1$	98.9	6626±0.020
	$K\alpha_2$	94.8	3872±0.034

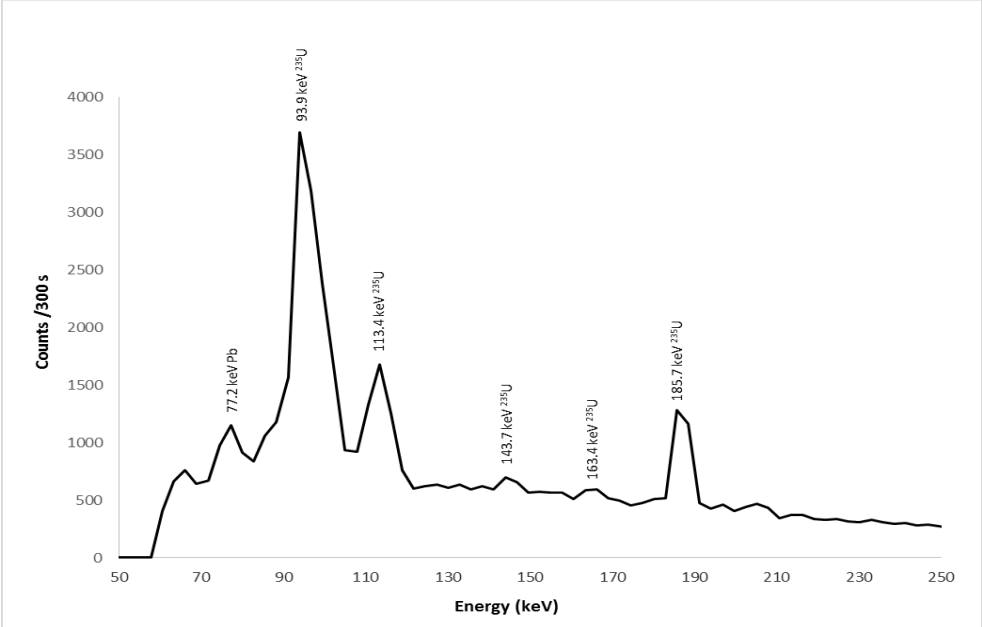


Figure 4.12 Characteristic of ^{235}U Gamma Ray for 0.4% ^{235}U by HPGe Detector

Table 4-14: Summarization 0.4% ^{235}U by HPGe Detector

Sample	Energy (keV)	Peak Area/300 s
0.4% ^{235}U	77.2	ND
	93.9	6500±0.021
	113.4	ND
	185.7	1605±0.037

ND = Not Detectable

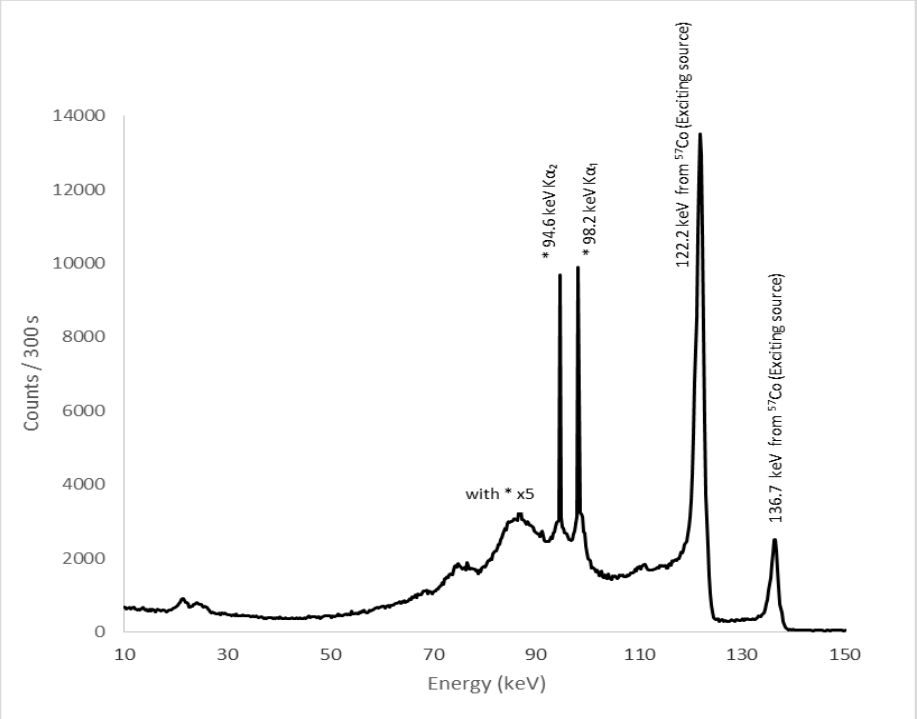


Figure 4.13 Characterization of K X-Ray of Radiography projector source with Depleted Uranium by CdTe Detector

Table 4-15: Summarization for Radiography projector source with Depleted Uranium by CdTe Detector

Sample	**Line**	**Energy (keV)**	**Peak Area/300 s**
Radiography projector source With Depleted Uranium	$K\alpha_1$	98.5	8417±0.020
	$K\alpha_2$	94.8	4198±0.034

Then, in Figure 4.13 above, shows characteristic of K x-ray of radiography projector source with depleted uranium measured with CdTe for the x-ray fluorescence measurement. The results show all the spectrums belong to ^{235}U with respective two K line electrons. The same exciting source used ^{57}Co as an efficient energy for K-shell ionization. Based in the figure there are two major K line are observed. $K\alpha_1$ at the energy of the emitted x-ray at 98.5 keV and $K\alpha_2$ at energy 94.8 keV with the peak area are obtained 8417±0.020 and 4198±0.034 respectively.

The x-ray fluorescence results based on the elemental analysis can only estimate the presence of uranium bearing material. Thus the presence of gamma ray spectrometry is needed to confirm the existence of ^{235}U. The gamma ray measurement is tested by using HPGe to confirm the presence of ^{235}U based on the approach enrichment meter principle. Most prominent gamma ray was found from the decay of ^{235}U being 185.7 keV.

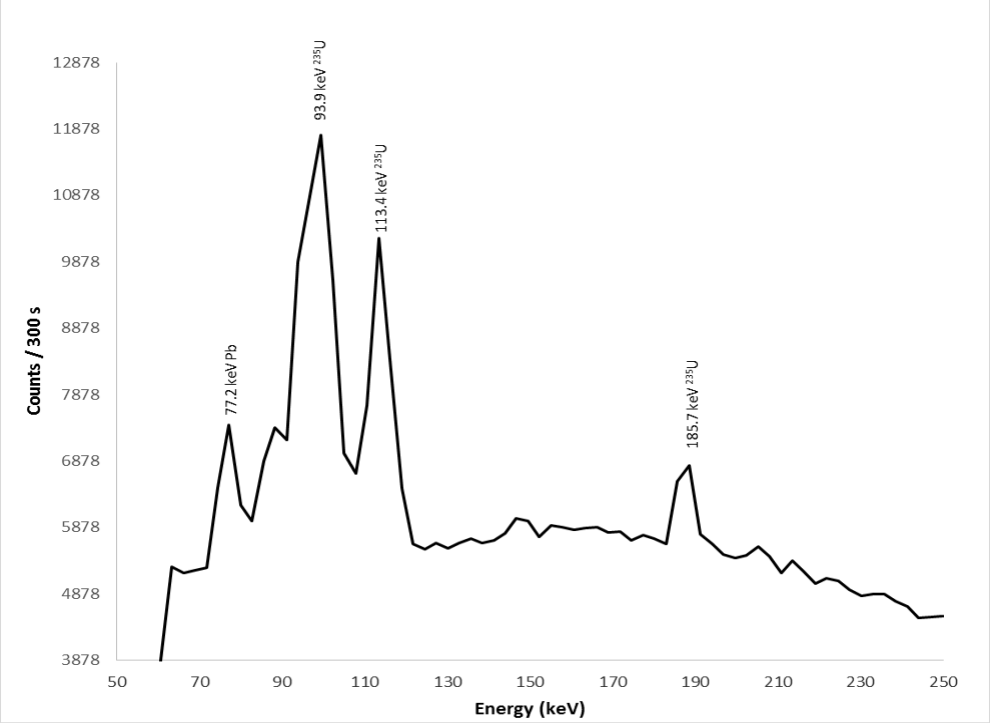


Figure 4.14 Characteristic of ^{235}U Gamma Ray for Radiography projector source with Depleted Uranium by HPGe Detector

Table 4-16: Summarization for Radiography projector source with Depleted Uranium by HPGe Detector

Sample	**Energy (keV)**	**Peak Area/300 s**
Radiography	77.2	1883±0.089
projector source	99.4	11697±0.021
with	113.4	981±0.217
Depleted Uranium	185.7	2302±0.072

In Figure 4.14 referred to a radiography projector source with depleted uranium sample exhibit the characteristic of gamma ray belong to ^{235}U measured by HPGe. The results shows all the spectrums belong to ^{235}U with respective energy. Based in the figure there is only one major energy observed belong to ^{235}U at 185.7 keV.

The Table 4.16 shows the summarization of the radiography projector source with depleted uranium with the energy and the peak area obtained from the experiment. The peak area per 300 seconds of at each energy are determined. As prediction, the prominent energy at 185.7 keV shows the photo peaks area from the decay of ^{235}U was determined with 2302 ± 0.072 per 300 s. There are another two with 93.9 keV and 113.4 keV belong to ^{235}U with peak area per 300 s, and 11697 ± 0.021 and 981 ± 0.217 respectively. This photo peaks intensity of the area helps to indicate the enrichment meter for the sample and the existence of ^{235}U in this sample.

In summary the measurement and characterization in **Part I,** result in significant results with same pattern belong to each uranium bearing material. The spectrum of samples from natural uranium and depleted uranium specimens shows the exact energy can be determine from the K x-ray results to indicate that there are uranium in the sample.
Further investigation on the determination of ^{235}U by using direct gamma ray measurement with HPGe. Based on the observation of the photo peak spectrum each sample shows the peak belongs to ^{235}U. This is as a complement standard for the uranium bearing measurement.

Therefore it is achieved the main of the of objectives to characterize the uranium bearing material by the elemental analysis and direct gamma ray measurement by identifying uranium K x-rays excited with gamma rays from ^{57}Co as exciting source.

Then the determination of the ^{235}U concentration based on enrichment meter which reference to peak of ^{235}U series especially the prominent photo peak 185.7 keV from the direct gamma ray measurement with HPGe detector. The enrichment meter used as an indicator for the technique to ensure the specimen does have the ^{235}U.

The peak area determination by the both spectrum analysis used to differentiate the specimen either in group of natural uranium or depleted uranium samples. From the spectrum results it is shows that the photo peak from the depleted uranium is lower than the natural uranium. Nevertheless it is also depends on the geometry of the specimens.

4.2 Part II: Characterization of Direct Gamma Rays Measurement by HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactor with Different Distances

In this section is the characterization of direct gamma ray by HPGe for TRIGA Mark III fresh fuel rod research reactors with different distances. There are three samples with the different series as shown in Table 4.16.

Table 4-17: TRIGA Mark III fresh fuel rod research reactors

Uranium Bearing	**Sample**
TRIGA Mark III (Fresh Fuel Rod) with Enriched Uranium 20% ^{235}U	Control Rod (8268) Fresh Fuel Rod (9619) Fresh Fuel Rod (9621)

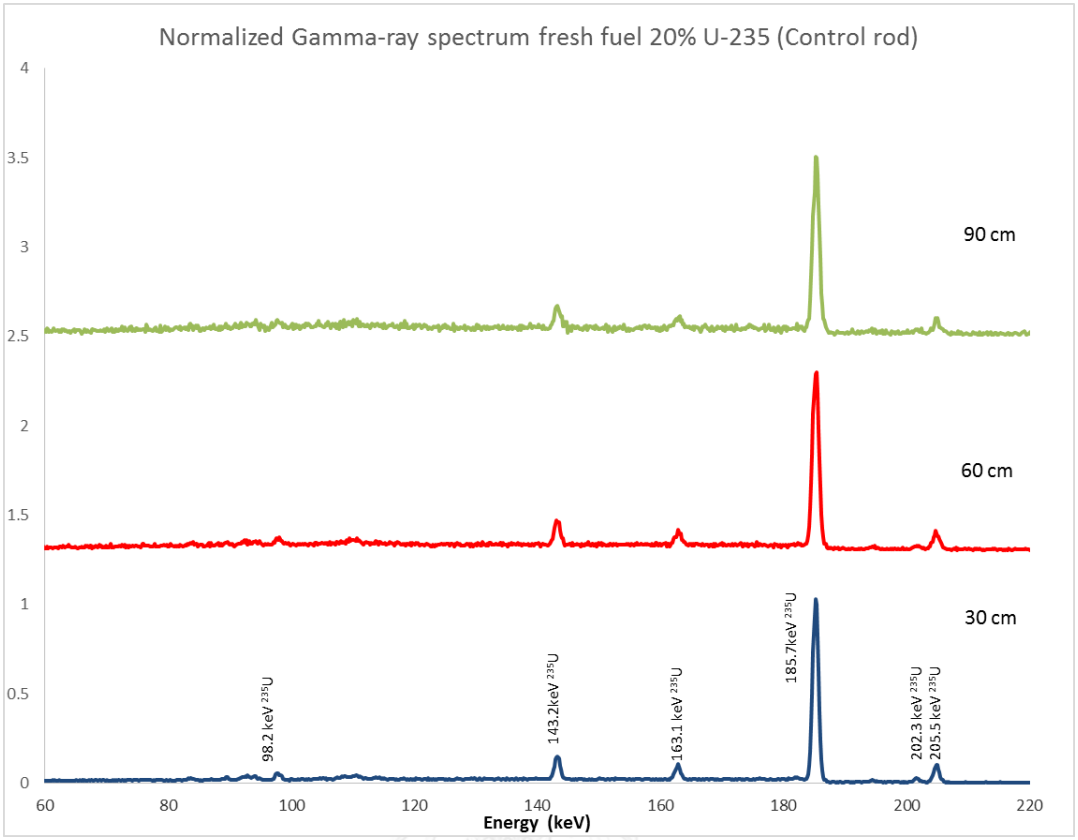


Figure 4.15 Normalized Gamma Ray Spectrum of Fresh Fuel Rod 20% ^{235}U (Control Rod)

The data analysis was performed to ensure the accuracy of the results and to explore the influence of the different peak shape from all the three fresh fuel rods. All data obtained with HPGe detector were analyzed with Genie 2000 for the peak identification and characterization, which also delivers peak area and full width of half-maximum (FWHM) value.

The results from the analysis are presented in Table 4.18 for control rod sample; 8268 with a varied distance starting at 30 cm, 60 cm and 90 cm. As shown, the characteristic of gamma rays energy at 144.1 keV, 163.7 keV, 185.7 keV, 202.3 keV and 205.5 keV belong to ^{235}U.

At a distance of 30 cm, in each spectrum energy shows a strong absorption of energy due to its internal conversions. Some gamma rays may be strongly converted that only a few percent of transition results in the emission of gamma ray. This leads to the observation of gamma ray intensities, which in turn is at a lower fraction. The highest

photo peak area for amongst the varied distance was 185.7 keV which is generally used for ^{235}U assay.

In the same table information, the findings of the ratio of energy over the highest photo peak area (E\E (185.7 keV)). From the findings of the table data, the differences in distance measurement correlates regularly with almost the same ratio for each area belong to the energy. This indicates that we can predict and estimate the area as we further the distance. As a result, 30 cm for the area reduction is around 12 % ratio from the nearest distance.

However the E (202.3 keV) over E (185.7 keV) spectrum cannot be characterize due to the low absorption of gamma energy. Therefore at this energy the ratio deduced as the distance of the detector to the sample is increasing.

Table 4-18: Summarization of Fresh Fuel Rod 20% ^{235}U (Control Rod) measured by HPGe Detector

Detector	**Series**	**Distance (cm)**	**Energy (keV)**	**Area/300s**	**Ratio/(185.7keV)**
HPGe	8268	30	98.2	1017±0.021	0.03
			143.2	3536±0.029	0.12
			163.1	2238±0.006	0.07
			185.7	29892±0.118	1.00
			202.3	602±0.052	0.02
			205.5	2899±0.020	0.10
		60	143.2	1082±0.043	0.12
			163.1	612±0.062	0.07
			185.7	8848±0.011	1.00
			202.3	ND	ND
			205.5	775±0.040	0.09

Table 4-18: Summarization of Fresh Fuel Rod 20% ^{235}U (Control Rod) measured by HPGe Detector (continue)

Detector	Series	Distance (cm)	Energy (keV)	Area/300s	Ratio/(185.7keV)
HPGe	8268	90	143.2	547±0.081	0.13
			163.1	309±0.132	0.07
			185.7	4261±0.016	1.00
			202.3	ND	ND
			205.5	366±0.076	0.09

The results from the analysis are presented in Figure 4.16 below, for the sample; 9619 with a varied distance starting at 30 cm, 60 cm and 90 cm. As shown, the characteristic of gamma rays energy at 98.2 keV, 144.1 keV, 163.7 keV, 185.7 keV, 202.3 keV and 205.5 keV belong to ^{235}U.

At a distance of 30 cm, each spectrum energy shows a strong absorption of energy due to its internal conversions. Some gamma rays may be strongly converted that only a few percent of transition results in the emission of gamma ray. This leads to the observation of gamma ray intensities, which in turn is at a lower fraction. The highest photo peak area for amongst the varied distance was 185.7 keV which is generally used for ^{235}U assay.

In Table 4.19, shows the results for the 9619 fresh fuel rod with 20 wt% ^{235}U enrichment. The data shows the spectrums which are the characteristic gamma rays of ^{235}U. The highest photo peak at 185.7 keV results and area of 58978/300 seconds due to ^{235}U assay. As predicted the ratio of area E\E (185.7 keV) is almost consistent for each energy area ratio to the highest photo peak.

Seen from the area at E (144.1 keV)\E (185.7 keV) around 10%-11% and the area at E (202.5 keV)\E (185.7 keV) reduces as the distance between the detector to the sample increases thus producing expected results seen for the third sample 9621 which justifies the same pattern.

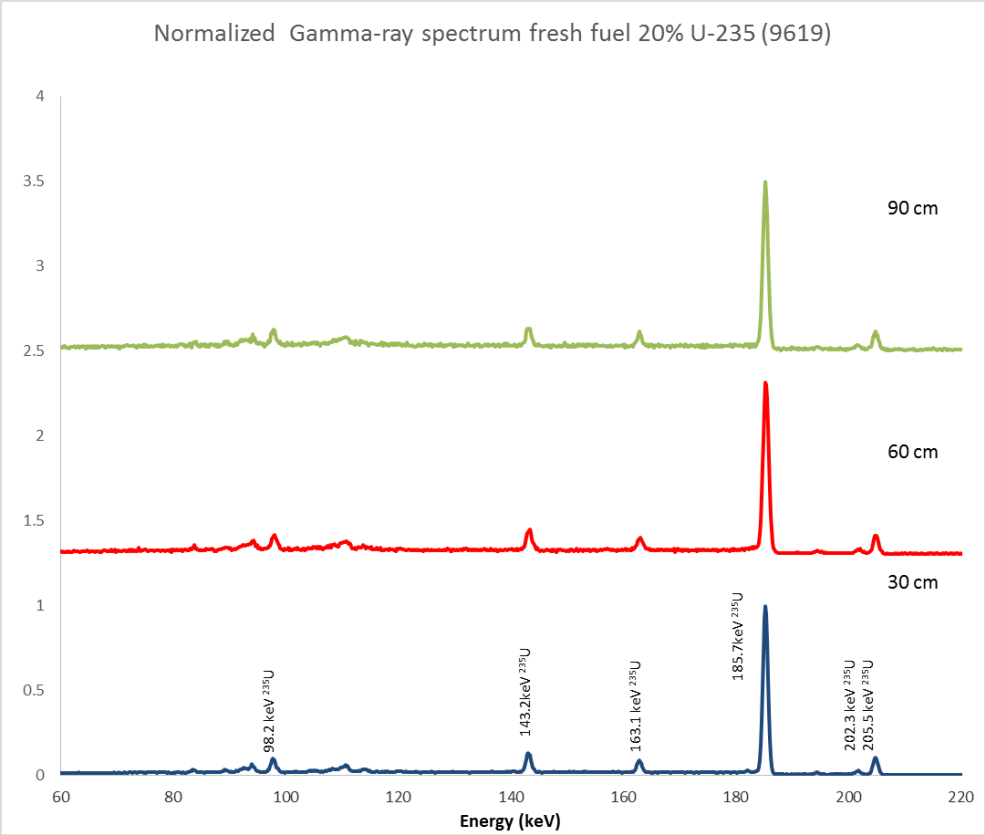


Figure 4.16 Normalized Gamma Ray Spectrum of Fresh Fuel Rod 20% ^{235}U (9619)

Table 4-19: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by HPGe Detector

Detector	Series	Distance (cm)	Energy (keV)	Area/300s	Ratio/(185.7keV)
HPGe	9619	30	94.4	1247±0.069	0.02
			98.2	4693±0.020	0.07
			111.2	1831±0.052	0.03
			143.2	6760±0.016	0.11
			163.1	3903±0.022	0.07
			185.7	58978±0.004	1.00
			202.3	888±0.046	0.02
			205.5	5803±0.014	0.10

Table 4-19: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by HPGe Detector (Continue)

Detector	Series	Distance (cm)	Energy (keV)	Area/300s	Ratio/(185.7keV)
HPGe	9619	60	94.4	341±0.058	0.02
			98.2	1267±0.047	0.07
			143.2	1838±0.031	0.11
			163.1	963±0.047	0.06
			185.7	17025±0.008	1.00
			202.3	ND	ND
			205.5	1688±0.029	0.10
		90	94.4	112±0.17	0.02
			98.2	547±0.073	0.07
			143.2	781±0.051	0.10
			163.1	499±0.070	0.06
			185.7	7810±0.001	1.00
			202.3	ND	ND
			205.5	830±0.043	0.11

The Table 4.20 below, shows the third fresh fuel rod with 20 wt% ^{235}U enrichment of TRIGA Mark III research reactor as predicted the data shows the same pattern as in Table 4.17 and Table 4.18. The most significant trait is the ratio of the area which reduces as the distance between the detector and sample increases, ranging between 10%-11% for the area at E (144.1 keV) and E (205.5 keV). The area at E (163.7 keV)\E (185.7 keV) on the other hand is in the range 6%-7% of the ratio. Finally the area at energy E (202.3 keV)\E (185.7 keV) is deduced as the distance between the detector and the sample are increases.

Therefore, it is obvious from this table data that the area of the photo peak can be estimated due to respective energy to the highest photo peak ratio. Moreover the significant pattern which proves beneficial in the safeguards approaches in the

verification and evaluation of nuclear material through independent measurement. The consistent results from the control rod results in it being a measurement for the other two fresh fuel rod as the standard comparison.

The results from the analysis are presented in Figure 4.17 below, for the sample; 9621 with a varied distance starting at 30 cm, 60 cm and 90 cm. As shown, the characteristic of gamma rays energy at 98.2 keV, 144.1 keV, 163.7 keV, 185.7 keV, 202.3 keV and 205.5 keV belong to ^{235}U.

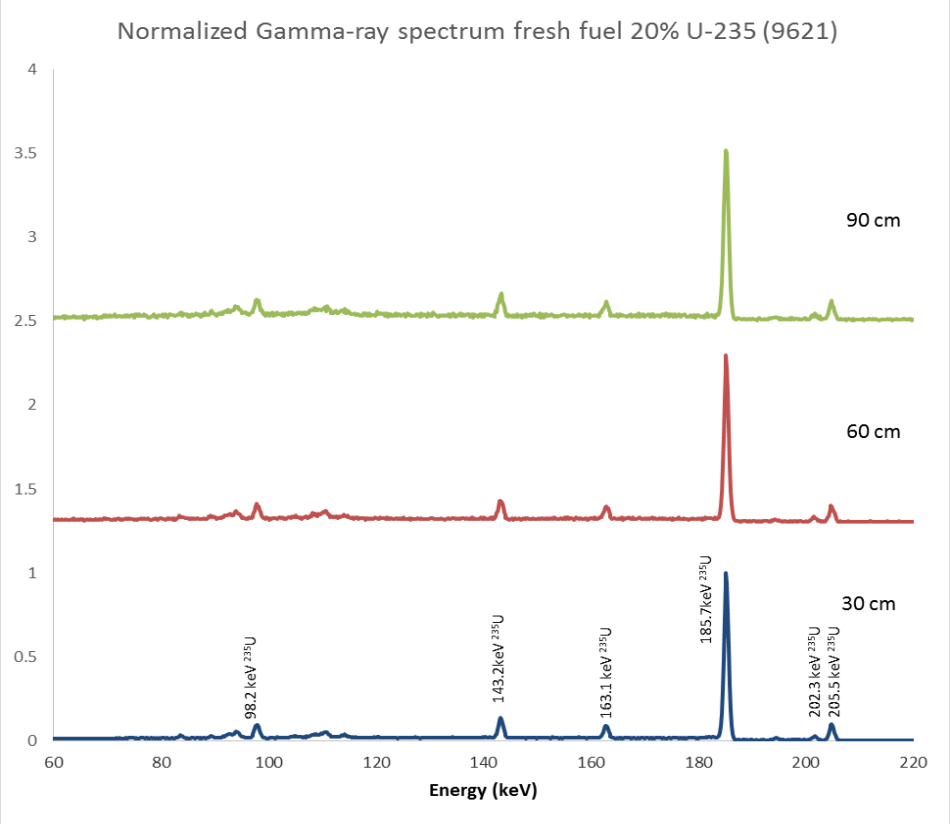


Figure 4.17: Normalized Gamma Ray Spectrum of Fresh Fuel Rod 20% ^{235}U (9621)

Table 4-20: Summarization of Fresh Fuel Rod 20% ^{235}U (9621) measured by HPGe Detector

Detector	**Series**	**Distance (cm)**	**Energy (keV)**	**Area/300s**	**Ratio/(185.7keV)**
HPGe	9621	30	94.4	1166±0.042	0.02
			98.2	4616±0.020	0.07
			143.2	6492±0.016	0.11
			163.1	4277±0.021	0.07
			185.7	59235±0.004	1.00
			202.3	1228±0.042	0.02
			205.5	5947±0.014	0.10
		60	98.2	1284±0.042	0.07
			143.2	1887±0.032	0.11
			163.1	1167±0.046	0.07
			185.7	17206±0.007	1.00
			202.3	ND	ND
			205.5	1594±0.028	0.09
		90	98.2	498±0.109	0.06
			143.2	850±0.052	0.11
			163.1	539±0.075	0.07
			185.7	7827±0.011	1.00
			202.3	ND	ND
			205.5	770±0.041	0.10

4.3 Part III: Characterization of XRF and Direct Gamma Rays Measurement for TRIGA Mark III Fresh Fuel Rod Research Reactor

In **Part III**, the characterization of XRF and direct gamma ray by CdTe and HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactors. The determination of full energy peak area; the gamma ray pulse height spectrum contains much useful information about gamma ray energies and intensities. Thus in this part of research the samples are set up closed contact with the detector in other to achieved the reasonable time of the sensitivity to ^{235}U by using the appropriate detector selection in order to analyse the elemental and gamma ray radiation to the energy in interest.

As the same sample measured in **Part II**, in this section will be examined with the same series but measurement are separated into four section of the TRIGA Mark III fresh fuel rod for series 9619 and 9621 which are top part of the graphite and the three part of uranium fuel.

Then, the control rod with series 8268 are measured divided into three part with top part of graphite, boron carbide and uranium fuel. The sample with the series as shown in Table 4.21.

Table 4-21: TRIGA Mark III (Fresh Fuel) Enriched Uranium 20% ^{235}U Series

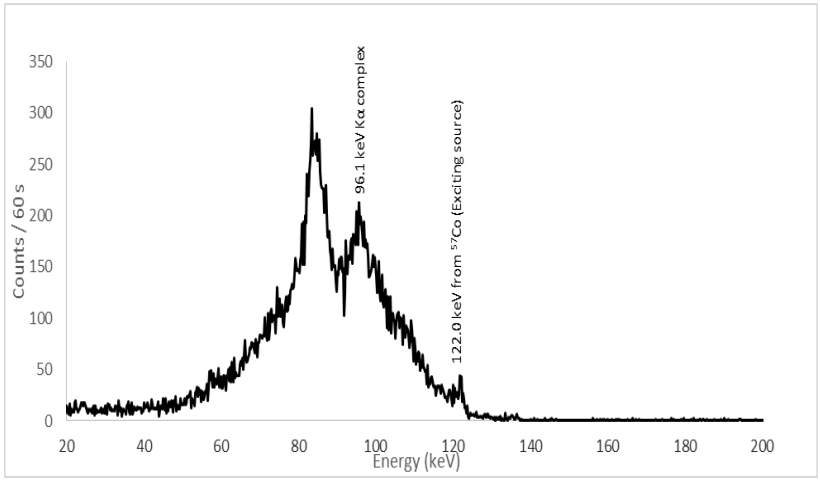
Uranium Bearing	**Sample**
TRIGA Mark III (Fresh Fuel)	Control Rod (8268)
Enriched Uranium 20% ^{235}U	Fresh Fuel Rod (9619)
	Fresh Fuel Rod (9621)

The basic measurement procedure involves viewing a uranium sample through a collimated channel with a gamma-ray detector. In Figure 4.18 (a) and (b) shows the spectrum results from the measurement of x-ray fluorescence by using CdTe for control rod sample (8268). The spectrum results shows a higher scattering belong to graphite and boron carbide. Figure 4.18 (a) shows the photo peak of $K\alpha_{complex}$ as a merging photo peak at 96.1 keV and the exciting source at 122 keV. Then at Figure 4.18 (b) also shows almost the same pattern of the spectrum.

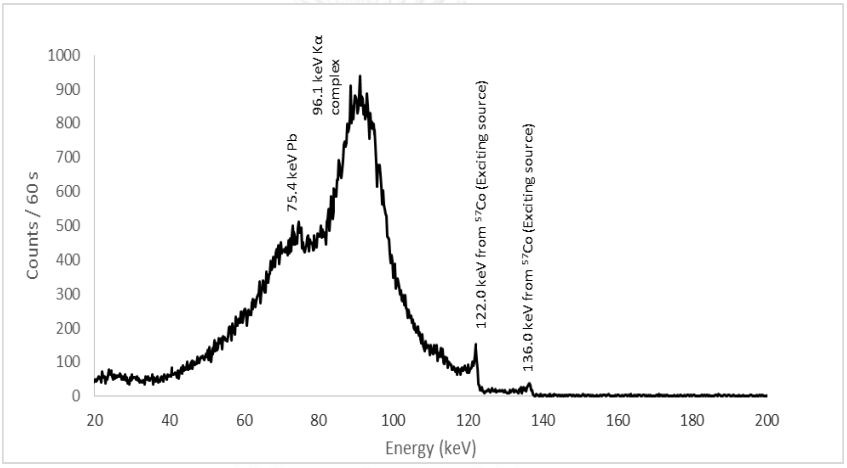
Then Figure 4.18 (c), results shows the measurement at the uranium fresh fuel part of the rod. From the observation we can see the K x-ray line belong to the uranium x-ray energy. Based on the figure there are three major K major line are observed which are 94.4 keV, 98. 3 keV and 111.0 keV belong to $K\alpha_2$, $K\alpha_1$ and $K_{\beta 1}$ respectively.

The most interesting the peak belong to ^{235}U at 185.7 keV is shown in the figure. The primary radiation used in passive NDA of uranium samples is gamma radiation which is usually dominated by emission of ^{235}U decay. However in low enriched uranium sample, the x radiation is the most intense component of the emission spectrum. The 185.7 keV gamma ray is the most frequently used signature to measure ^{235}U enrichment. It is the most prominent single gamma ray from any uranium sample enriched above natural ^{235}U level. There are no common interferences except in reprocessed fuel where the 236 kev from ^{232}Th daughter, ^{212}Pb, usually swamps the ^{235}U line. Gamma ray spectra from uranium sample varying degree of enrichment as an enrichment meter.

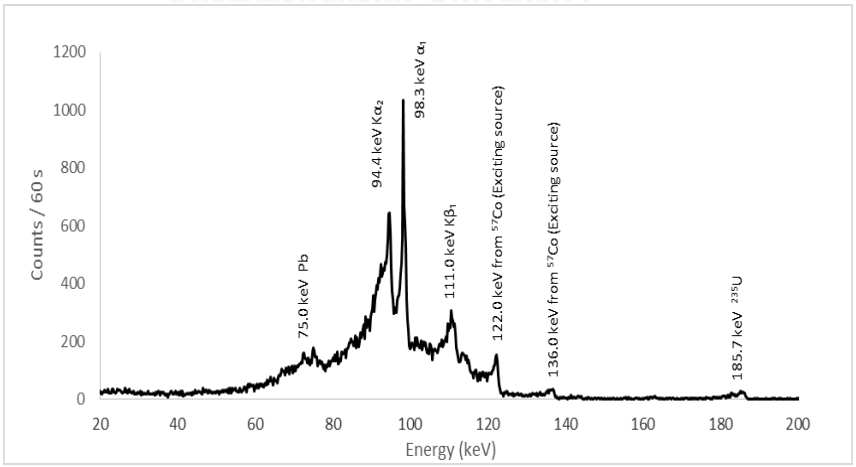
The enrichment is deduced from the intensity of the ^{235}U 185.7 keV gamma-ray. If the uranium large enough, the 185.7 keV gamma rays from only a fraction of the total sample reach the detector because of the strong absorption of typical uranium bearing material at this energy.



(a) Graphite



(b) Boron Carbide



(c) Fresh Fuel 20% ^{235}U

Figure 4.18: Characterization of K X-Ray of TRIGA Mark III Fresh Fuel rod (8268) by CdTe Detector

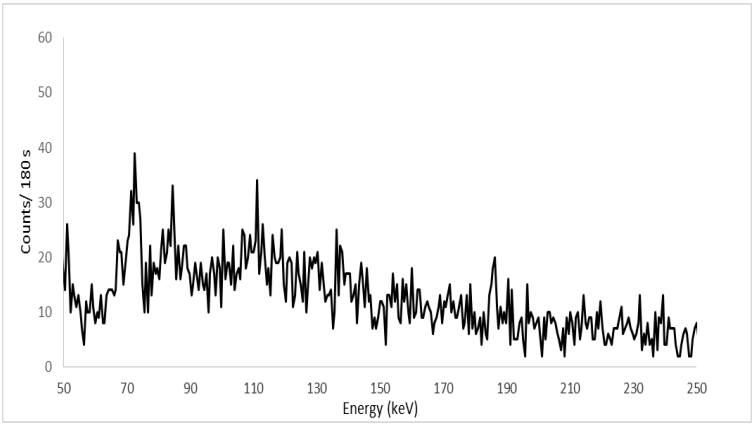
Table 4-22: Summarization of Fresh Fuel Rod 20% ^{235}U (8268) measured by CdTe Detector

Detector	Series	Part	Line	Energy (keV)	Area/60 s
CdTe	8268 (Control Rod)	Graphite	Kα complex	96.1	ND
			^{57}Co	122.0	ND
		Boron Carbide	Pb	75.4	ND
			Kα complex	96.1	ND
			^{57}Co	122.0	ND
				136.8	ND
		20% ^{235}U	Pb	75.0	121± 0.319
			$K\alpha_1$	94.4	1376 ± 0.074
			$K\alpha_2$	98.3	2190 ± 0.041
			$K_{\beta 1}$	111.0	260 ± 0.227
			^{57}Co	122.0	224 ± 0.161
				136.0	ND

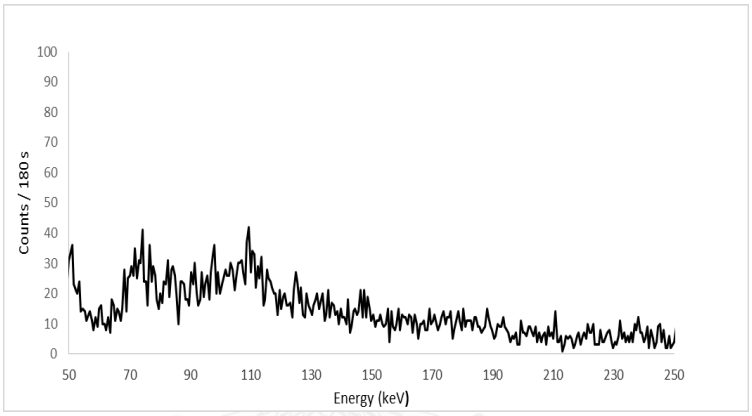
*ND = Not Detectable

Table 4-23: Summarization of Fresh Fuel Rod 20% ^{235}U (8268) measured by HPGe Detector

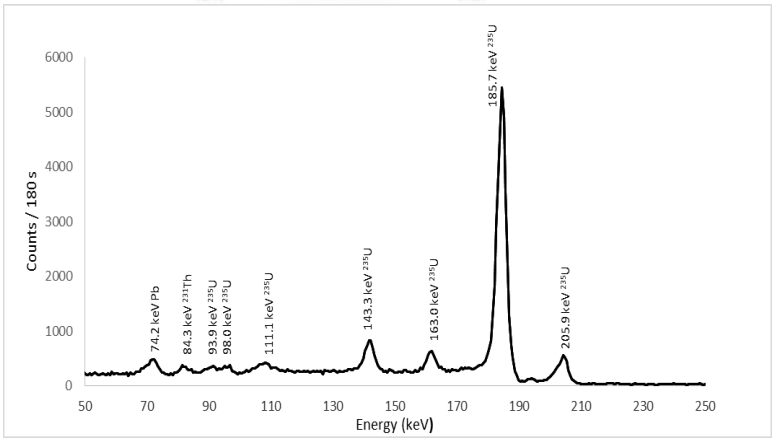
Detector	Series No.	Part(20% ^{235}U)	Energy (keV)	Area/180 s
HPGe	8268	1	74.2	2050 ± 0.040
			84.3	709 ± 0.086
			110.3	849 ± 0.187
			143.0	3305 ± 0.028
			163.0	2050 ± 0.039
			185.7	30839± 0.006
			205.9	3017 ± 0.022



(a) Graphite



(b) Boron Carbide



(c) Fresh Fuel 20% ^{235}U

Figure 4.19: Characteristic of ^{235}U Gamma Ray for TRIGA Mark III Fresh Fuel rod (8268) by HPGe Detector

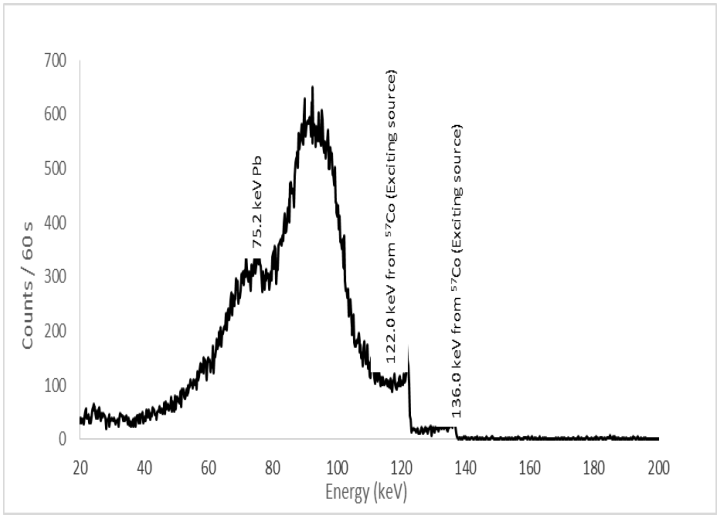
Then the Figure 4.19 (a), (b) and (c) shows the characterization of the sample for the control rod measured using the HPGe detector. Then, Table 4.23 shows the summarization of the 20% ^{235}U fresh fuel control rod with the energy and the peak area obtained from the experiment. The peak area per 180 seconds of at each energy are obtained. The peak is per 180 seconds at 111.1 keV and 143.3 keV are 849±0.187 and 3305±0.028 respectively. Then, at 163.0 keV and 185.7 keV are 2050±0.039 and 30839±0.006 respectively. Then the final peaks at at 205.2 keV is 3017±0.022 peak area per 180 seconds.

Table 4-24: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by CdTe Detector

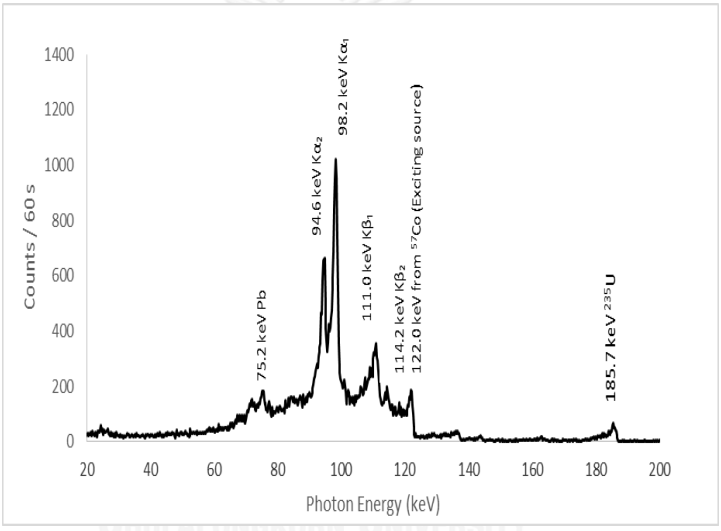
Detector	**Series**	**Part**	**Line**	**Energy (keV)**	**Area/ 60 s**
CdTe	9619	Graphite (Top)	Pb	74.0	ND
			$K\alpha_2$	92.0	ND
			^{57}Co	122.0	ND
				136.0	ND
		1 (20% ^{235}U)	Pb	75.2	253 ± 0.190
			$K\alpha_2$	94.6	2237 ± 0.044
			$K\alpha_1$	98.2	3586 ± 0.030
			$K_{\beta 1}$	111.1	ND
			$K_{\beta 2}$	114.6	892 ± 0.075
			^{57}Co	122.0	589 ± 0.074
			^{235}U	136.0	ND
				185.7	159 ± 0.147
		2 (20% ^{235}U)	Pb	75.2	ND
			$K\alpha_2$	94.6	779 ± 0.085
			$K\alpha_1$	98.2	2484 ± 0.036
			$K_{\beta 1}$	111.0	848 ± 0.087
			^{57}Co	122.0	266 ± 0.141
				136.0	ND

Table 4-24: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by CdTe Detector (continue)

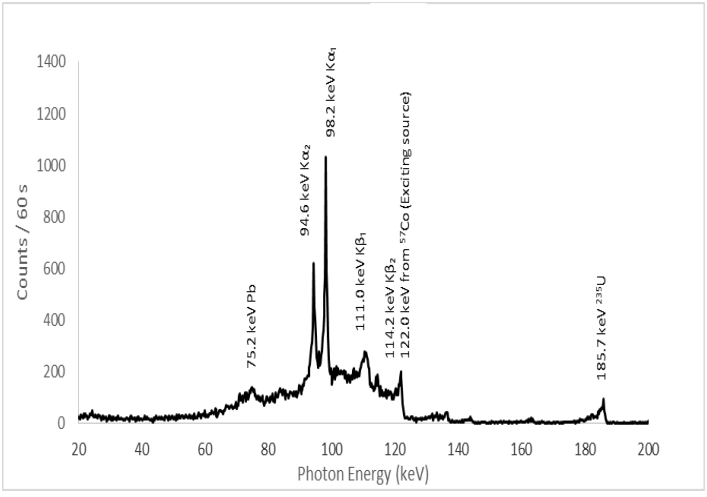
Detector	**Series**	**Part**	**Line**	**Energy (keV)**	**Area/ 60 s**
CdTe	9619	2 (20% ^{235}U)	^{235}U	143.9	ND
				163.8	ND
				185.7	117 ± 0.215
		3 (20% ^{235}U)	Pb	75.3	258 ± 0.148
			$K\alpha_2$	94.6	2015 ± 0.051
			$K\alpha_1$	98.2	2650 ± 0.042
			$K_{\beta 1}$	111.0	ND
			$K_{\beta 2}$	114.2	ND
			^{57}Co	122.0	ND
			^{235}U	185.7	ND



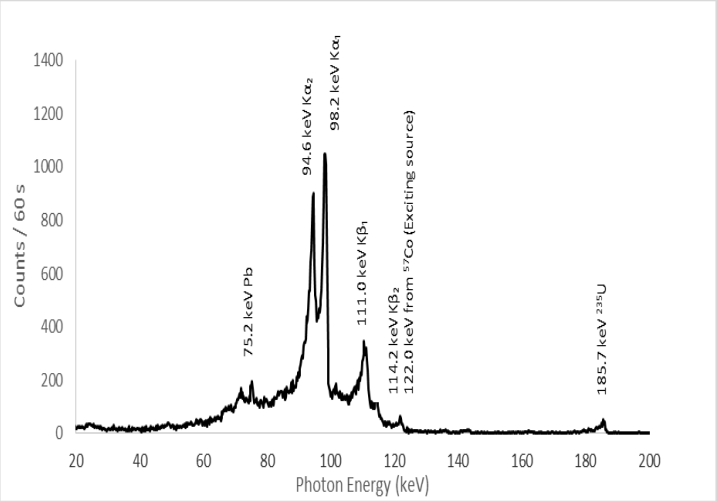
(a) Graphite



(b) Fresh Fuel 20% ^{235}U



(c) Fresh Fuel 20% ^{235}U



(d) Fresh Fuel 20% ^{235}U

Figure 4.20: Characterization of K X-Ray of TRIGA Mark III Fresh Fuel rod (9619) by CdTe Detector

In Figure 4.20 (a), (b), (c) and (d) shows the spectrum results from the measurement of x-ray fluorescence by using CdTe for TRIGA Mark III fresh fuel rod with series 9619 specimen. The measurement did in four part of the rod. The first part is the top part of graphite shown in Figure 4.20 (a). The spectrum results shows a higher scattering belong to graphite. Figure 4.18 (a) shows the photo peak of $K\alpha_{complex}$ as a merging photo peak but not detectable and the exciting source at 122.0 keV and 136.0 keV. All the part of the samples are measured within 60 seconds.

Then Figure 4.20 (b), (c) and (d) results shows the measurement at the uranium fresh fuel part of the rod. From the observation we can see the K x-ray line belong to the uranium x-ray energy. Based on the figure there are three major K major line are observed which are 98.2 keV, 94.6 keV belong to $K\alpha_1$ and $K\alpha_2$ respectively. Then at 111.0 keV and 114.2 belong to $K_{\beta 1}$ and $K_{\beta 2}$ respectively. The sample part all measured in 60 seconds

It is the same interesting the peak belong to ^{235}U at 185.7 keV is shown in the figure.. However in low enriched uranium sample, the x radiation is the most intense component of the emission spectrum. The 185.7 keV gamma ray is the most frequently

used signature to measure ^{235}U enrichment. It is the most prominent single gamma ray from any uranium sample enriched above natural ^{235}U level.

The main region of ^{235}U at 185.7 keV gamma-ray mainly used for the element of enrichment meter. If the uranium large enough, the 185.7 keV gamma rays from only a fraction of the total sample reach the detector because of the strong absorption of typical uranium bearing material at this energy.

Table 4-25: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by HPGe Detector

Detector	**Series No.**	**Part**	**Energy**	**Area/180 s**
HPGe	9619	Graphite	74.2	542 ± 0.084
		(Top)	84.3	304 ± 0.126
			94.0	ND
			98.5	98 ± 0.426
			111.1	858 ± 0.078
			143.3	624 ± 0.068
			163.0	373 ± 0.107
			185.7	7160 ± 0.012
			205.3	658 ± 0.050
			74.2	3768 ± 0.029
		1 (20% ^{235}U)	84.3	1270 ± 0.069
			94.0	641 ± 0.133
			98.5	1679 ± 0.059
			111.1	5199 ± 0.027
			143.3	6340 ± 0.020
			163.0	3859 ± 0.027
			185.7	60450±0.004
			205.3	6900 ± 0.014

*ND = Not Detectable

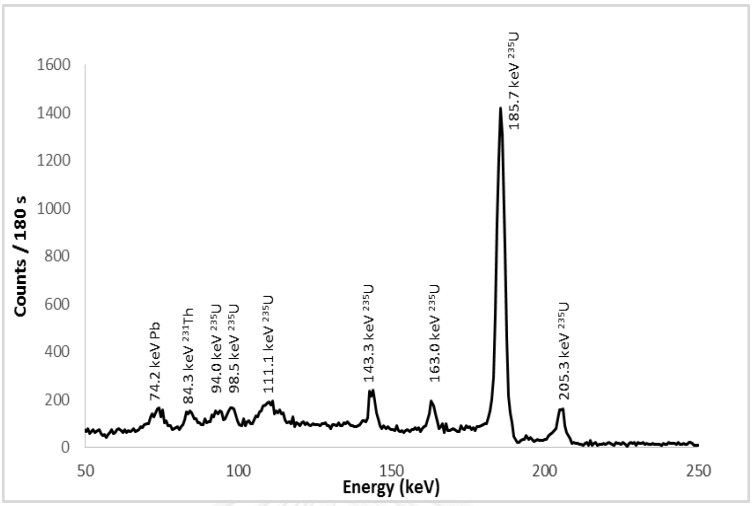
Table 4-25: Summarization of Fresh Fuel Rod 20% ^{235}U (9619) measured by HPGe Detector (Continue)

Detector	Series No.	Part	Energy	Area/180 s
HPGe	9619	2 (20% ^{235}U)	74.2	3439 ± 0.031
			84.3	1413 ± 0.062
			94.0	655 ± 0.130
			98.5	1692 ± 0.055
			111.1	2735 ± 0.041
			143.3	6158 ± 0.019
			163.0	3134 ± 0.030
			185.7	61017± 0.004
			205.3	6940 ± 0.014
		3 (20% ^{235}U)	74.2	3408 ± 0.032
			84.3	1485 ± 0.062
			94.0	483 ± 0.198
			98.5	1593 ± 0.056
			111.1	4715 ± 0.028
			143.3	5862 ± 0.019
			163.0	3950 ± 0.027
			185.7	59176± 0.004
			205.3	6876 ± 0.014

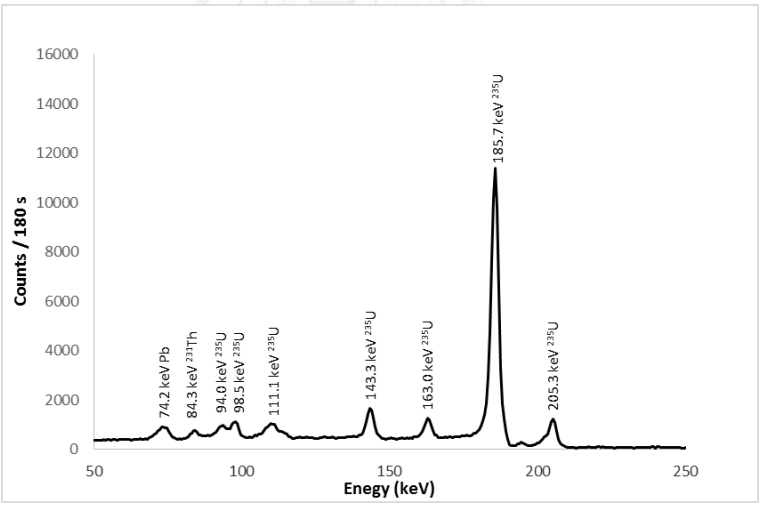
*ND = Not Detectable

Then the Figure 4.21 (a), (b) ,(c) and (d) shows the characterization of the sample for the 9619 series of the fresh fuel rod measured using the HPGe detector. Then, Table 4.26 shows the summarization of the 20% ^{235}U fresh fuel of 9619 rod with the energy and the peak area obtained from the experiment. The peak area per 180 seconds of at each energy are obtained. The peak are per 180 seconds at of the four segments of the rod are done with the results shows all the main peak energy belong to the ^{235}U series. Based on the observation it is clearly show that every part of the measurement shows

the almost the same ratio for the peak area determination. This lead to the conclusion that every part of the equally distributed and in the significant pattern.



(a) Graphite



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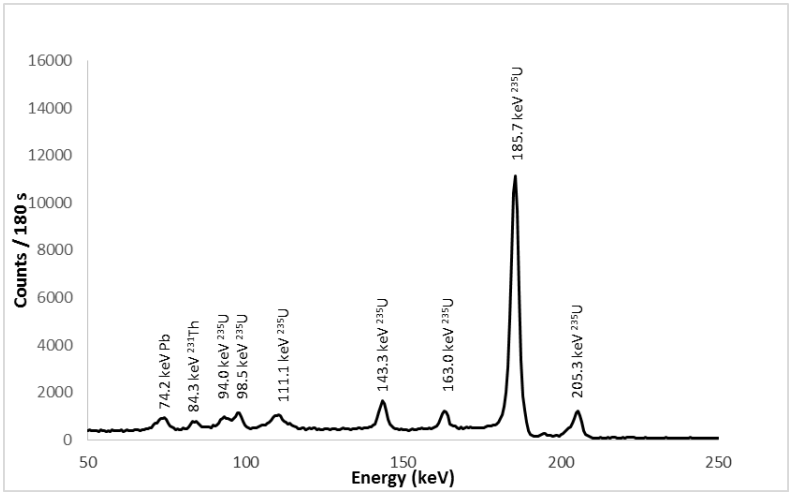
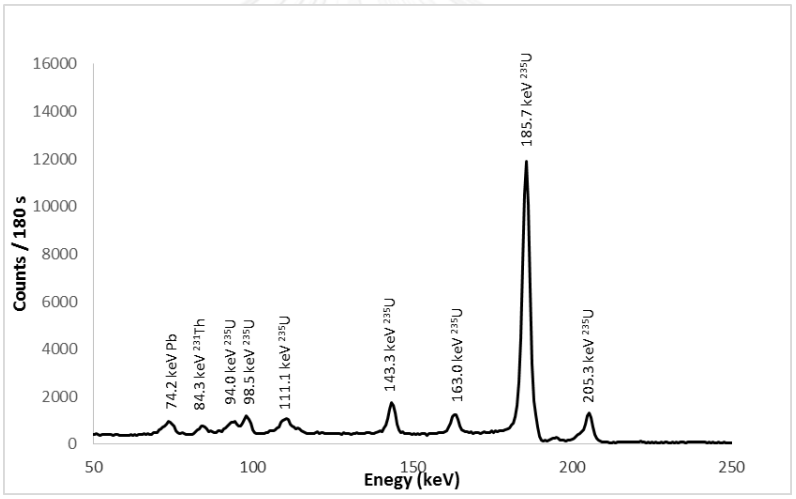


(c) Fresh Fuel 20% ^{235}U



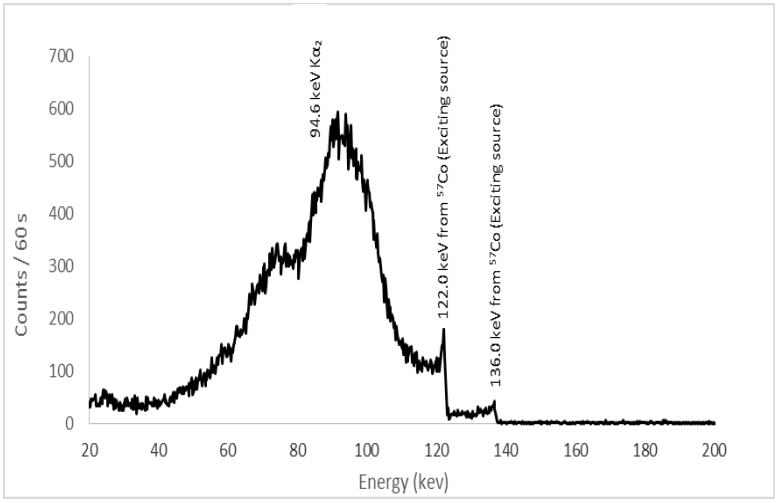
(d) Fresh Fuel 20% ^{235}U

Figure 4.21: Characteristic of ^{235}U Gamma Ray for TRIGA Mark III Fresh Fuel rod (9619) by HPGe Detector

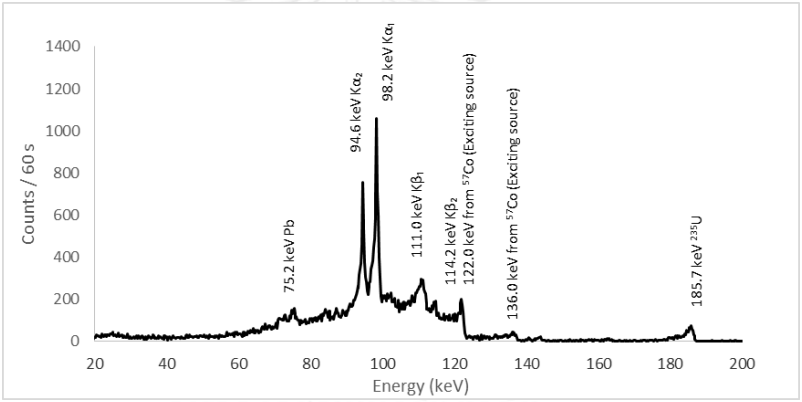
Table 4-26: Summarization of Fresh Fuel Rod 20% ^{235}U (9621) measured by CdTe Detector

Detector	Series	Part	Line	Energy (keV)	Area/ 60 s
CdTe	9621	Graphite (Top)	Pb	75.2	ND
			^{57}Co	122.1	ND
				136.6	ND
		1 (20% ^{235}U)	Pb	75.2	ND
			$K\alpha_2$	94.4	1378 ± 0.057
			$K\alpha_1$	98.3	3691 ± 0.027
			$K_{\beta 1}$	110.6	149 ± 0.313
			^{57}Co	122.0	266 ± 0.126
				185.7	145 ± 0.169
		2 (20% ^{235}U)	Pb	74.2	ND
			$K\alpha_2$	94.4	2174 ± 0.046
			$K\alpha_1$	98.1	4488 ± 0.025
			$K_{\beta 1}$	110.1	1047 ± 0.072
			^{57}Co	122.0	ND
				136.0	ND
			^{235}U	185.7	ND
		3 (20% ^{235}U)	Pb	75.2	127 ± 0.26
			$K\alpha_2$	94.6	1782 ± 0.048
			$K\alpha_1$	98.4	2236 ± 0.039
			$K_{\beta 1}$	111.0	456 ± 0.118
			^{57}Co	122.00	164 ± 0.210
			^{235}U	185.7	ND

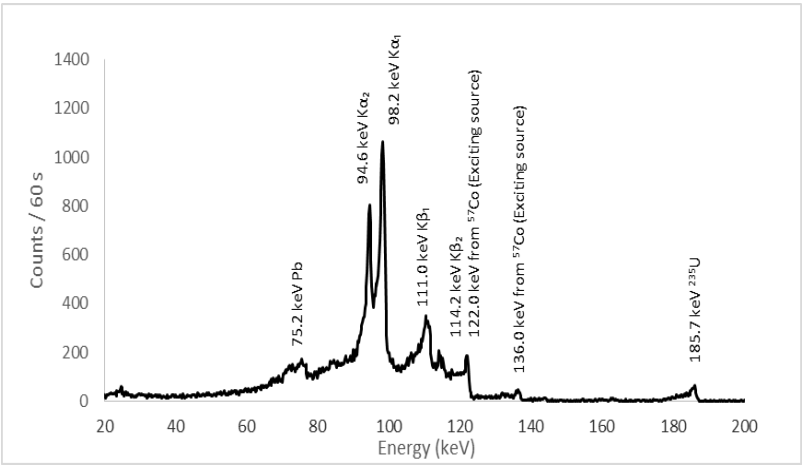
*ND = Not Detectable



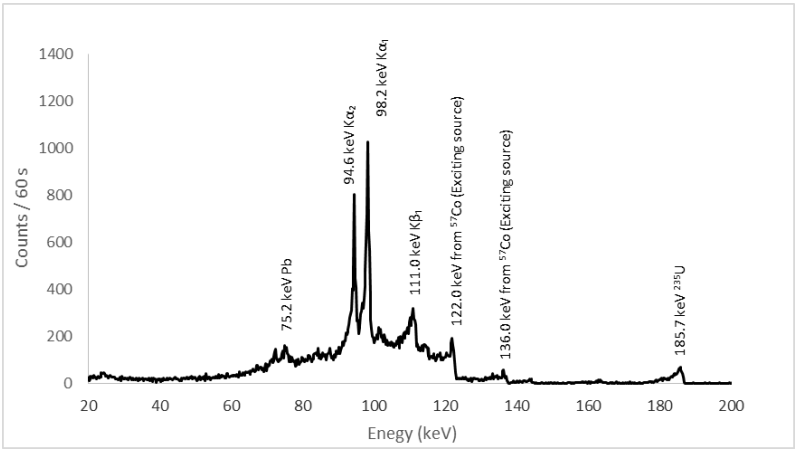
(a) Graphite



(b) Fresh Fuel 20% ^{235}U



(c) Fresh Fuel 20% ^{235}U



(d) Fresh Fuel 20% ^{235}U

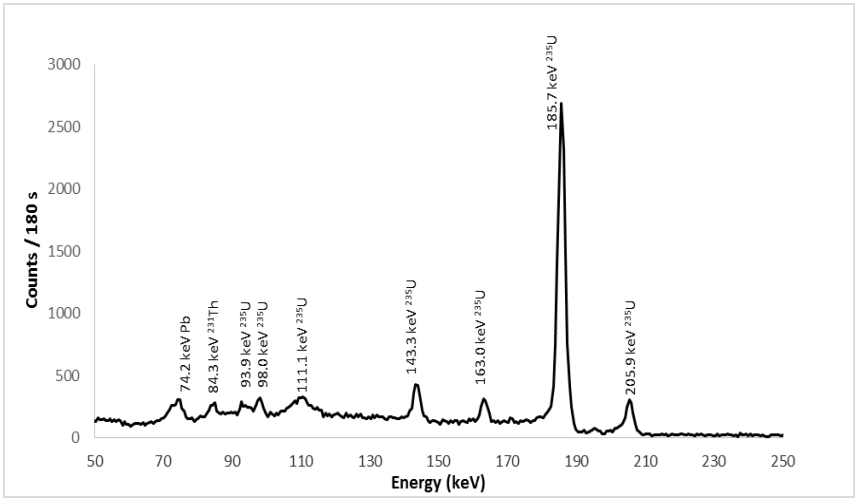
Figure 4.22: Characterization of K X-Ray of TRIGA Mark III Fresh Fuel rod (9621) by CdTe Detector

Table 4-27: Summarization of Fresh Fuel Rod 20% ^{235}U (9621) measured by HPGe Detector

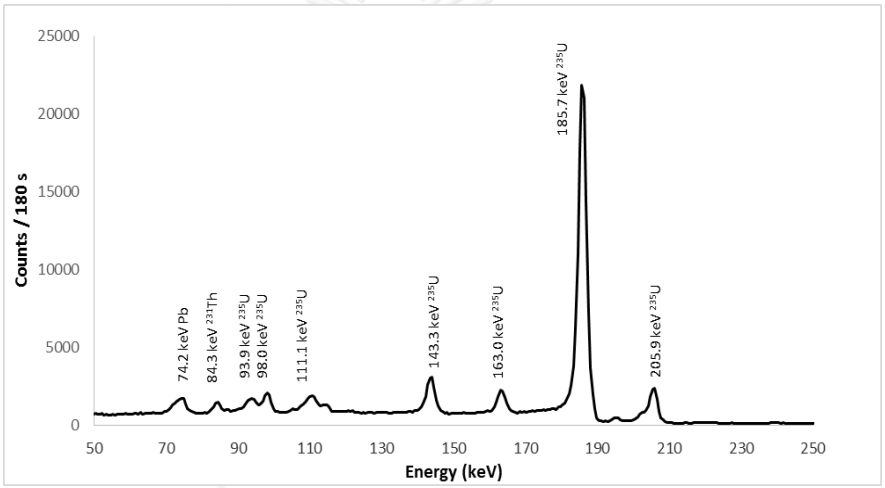
Detector	**Series No.**	**Part**	**Energy (keV)**	**Area/180 s**
HPGe	9621	Graphite (Top)	74.2	712 ± 0.058
			84.3	230 ± 0.130
			93.9	79 ± 0.318
			98.0	173 ± 0.16
			111.0	602± 0.075
			143.9	718± 0.045
			163.0	400 ± 0.071
			185.7	6894 ± 0.010
			205.9	659 ± 0.037
		1 (20% ^{235}U)	74.2	4014 ± 0.026
			84.3	1357± 0.049
			93.9	378± 0.204
			98.0	1880± 0.036
			111.0	2501± 0.036
			143.9	680± 0.014
			163.0	3946± 0.021
			185.7	63868±0.003
			205.9	5453± 0.013

Table 4-27: Summarization of Fresh Fuel Rod 20% ^{235}U (9621) measured by HPGe Detector (Continue)

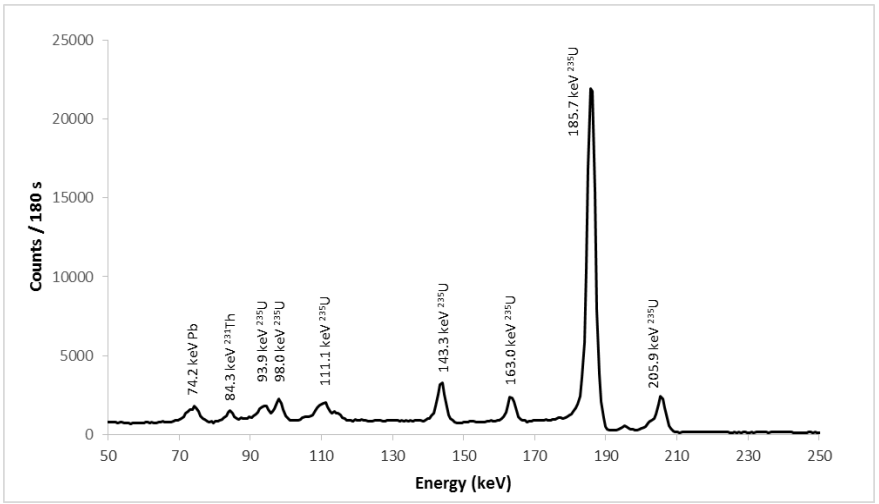
Detector	Series No.	Part	Energy (keV)	Area/180 s
HPGe	9621	2 (20% ^{235}U)	74.2	3417 ± 0.023
			84.3	1403 ± 0.048
			93.9	251± 0.332
			98.0	1811± 0.041
			111.0	2600 ± 0.0331
			143.9	697 ± 0.017
			163.0	4300 ± 0.019
			185.7	64300±0.003
			205.9	5609 ± 0.012
		3 (20% ^{235}U)	74.2	3209± 0.024
			84.3	1064 ± 0.056
			93.9	474± 0.140
			98.0	1436± 0.046
			111.0	2251± 0.035
			143.9	5511± 0.016
			163.0	3188± 0.023
			185.7	50691± 0.003
			205.9	3740± 0.016



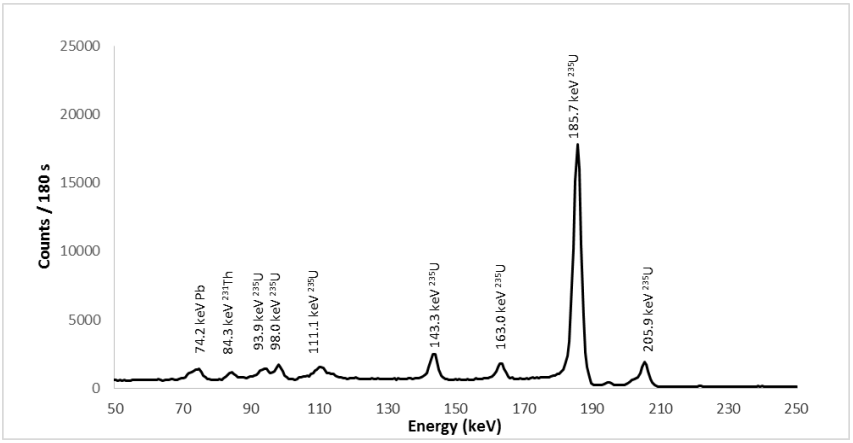
(a) Graphite



(b) Fresh Fuel 20% ^{235}U



(c) Fresh Fuel 20% ^{235}U



(d) Fresh Fuel 20% ^{235}U

Figure 4.23: Characteristic of ^{235}U Gamma Ray for TRIGA Mark III Fresh Fuel rod (9621) by HPGe Detector

Then finally is the third TRIGA Mark III fresh fuel rod of series 9621. This rod also under the same measurement as series 8268 and 9619 fresh fuel rod. In Figure 4.20 (a), (b), (c) and (d) shows the spectrum results from the measurement of x-ray fluorescence by using CdTe for TRIGA Mark III fresh fuel rod with series 9621 rod. The measurement done in four segment part of the rod. The first part is the top part of graphite shown in Figure 4.22 (a). The spectrum results show a higher scattering of the low atomic energy. In Figure 4.22 (a) shows the photo peak of $K\alpha_{complex}$ as a merging photo peak but not detectable and the exciting source at 122.0 keV and 136.0 keV. All the part of the samples are measured within 60 seconds.

Then Figure 4.20 (b), (c) and (d) results shows the measurement at the uranium fresh fuel of the three uranium fuel segment part of the rod. From the observation we can see the K x-ray line belong to the uranium x-ray energy. Based on the figure there are three major K major line are observed which are 98.2 keV, 94.6 keV belong to $K\alpha_1$ and $K\alpha_2$ respectively. Then at 111.0 keV and 114.2 belong to $K_{\beta 1}$ and $K_{\beta 2}$ respectively. The sample part all measured in 60 seconds. In table 4.26 shows the energy with the line of x-ray radiation and the peak area of each part per 60 seconds of measurement.

Fortunately the same interesting the peak belong to ^{235}U at 185.7 keV is shown in the figure. However in low enriched uranium sample, the x radiation is the most intense

component of the emission spectrum. The main region of ^{235}U at 185.7 keV gamma-ray mainly used for the element of enrichment meter. If the uranium large enough, the 185.7 keV gamma rays from only a fraction of the total sample reach the detector because of the strong absorption of typical uranium bearing material at this energy.

Then within the 60 seconds or 1 minute, it is can be estimated if the specimen is nearly closed enough and the energy of the interest are large to reach the material then the spectrum can be used as an scanning inspection method.

Nevertheless, it is not enough to verify that there are ^{235}U even the figure show the weak peak of 185.7 keV. Then the Figure 4.21 (a), (b) ,(c) and (d) shows the characterization of the sample for the 9621 series of the fresh fuel rod measured using the HPGe detector for the verification purpose.

Table 4.27 shows the summarization of the 20% ^{235}U fresh fuel of 9621 rod with the energy and the peak area obtained from the experiment. The peak area per 180 seconds of at each energy are obtained. The peak are per 180 seconds at of the four segments of the rod are done with the results shows all the main peak energy belong to the ^{235}U series. Based on the observation it is clearly show that every part of the measurement shows the almost the same ratio for the peak area determination. This lead to the conclusion that every part of the equally distributed and in the significant pattern.

In summary, in **Part III** characterization of XRF and direct gamma ray by CdTe and HPGe for TRIGA Mark III fresh fuel rod research reactors achieved the practical objective of the application of the reasonable time measurement with good resolution of spectrum with minimal interference of the spectrum. The specific energy of interest of enriched uranium of fresh fuel sample are determined from the K x-ray analysis with results show the major K lines of each segment part. Moreover the use of gamma ray measurement by HPGe verify of the existence of the ^{235}U indicate as enrichment meter. This is a positive results cooperate with the IAEA safeguards inspection to at least to have a scanning inspection through any sample that possible to be detect in a reasonable time with the minimal preparation of the procedure.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Overview of Study

The nuclear technology urges the parallel development of the necessary human resource potential. In this sophisticated nuclear sector with the same high level standard of safety, security and safeguards required highly skilled staff for design, operations and inspection.

Therefore in this research study, perhaps there are specifically stated principle consistency, completeness and timeliness. These all required for inspectors to have transparency of operations, with reasonable time measurement and minimal preparation of sample and the proper selection of the detector in the field involved.

For safeguards detection of diversion effectiveness of the internal control:

- to detect and correct biases, detecting mistake, abnormal conditions and trends;
- to maintain data authenticity, especially during any manipulation, processing or manual intervention

The objectives of safeguard are simple. The aim is to be able to detect a loss of significant amount of nuclear material in reasonably short detection time. Verification seeks out inconsistences in the accountancy, the measurement or plant layout.

The IAEA safeguard criteria are expressed in terms of the concepts of significant quantity, detection probability and timeless. Giving values to these three elements, for each specific category of the material, specifies a requirement for detection sensitivity is relevance for this research study.

5.2 Restatement of Objectives

As mention in Chapter 1, there are two objectives of the study for this research which are;

a) To characterize ^{235}U from uranium bearing materials using the XRF technique followed by direct gamma-ray measurement

b) To determine concentration of ^{235}U from uranium bearing material

Both of the objectives are achieved with the three part of the results and discussion. The sample are being investigate with the technique as a purpose of elemental analysis done by the XRF technique then the interest energy respect to the uranium bearing material are investigated. Managed by the scope of the study, the investigation with a technique for identifying uranium bearing materials by measurement of U K x-rays excited with gamma-rays from Co-57 source as the exciting source.

Moreover the investigation technique for determining uranium concentration by measurement of gamma-rays from ^{235}U and others as the enrichment meter. This measured by the intensity of the peak area of the sample of each the part of the study. The determination of the peak area gives an advantages for the verification of the measurement.

As an advantage, the technique tested with known uranium bearing materials including depleted uranium Metallic ic and compound, natural uranium Metallic ic and compound, enriched uranium in fresh fuel elements of TRIGA Mark III research reactor.

5.3 Review of the Finding

As discussed in Chapter 4, the finding is significant with the purpose of safeguard scanning in section. The result shows that the measurement can be handling in reasonable time of the sensitivity of the detector. Moreover with the proper selection of the detector helps the investigation lead to achieve the purposes and objectives.

As in **Part I**: Characterization of Elemental Analysis and Direct Gamma Ray by CdTe and HPGe Detectors for Natural Uranium and Depleted Uranium Samples, the measurement of seven samples with the elemental analysis and the gamma ray. All the samples shows the major K x-ray lines for the uranium line. Natural uranium samples shows the spectrum belong to ^{235}U which as the interested photo peak with the determination from the experiment. The depleted uranium sample shows lower intensities due to the determination of the peak area comparing with natural uranium bearing material. All the samples are measured in 300 seconds for both of the detectors.

Then in **Part II**: Characterization of Direct Gamma Ray by HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactors with Different Distances. The results demonstrate the characterization of 20 wt% fresh fuel of TRIGA reactor by directing gamma rays measurement as ^{235}U bearing material. Three fresh fuel samples; 8268 (control rod), 9619 rod and 9621 rod were investigated. The data shows the energy 185.7 keV results as the highest peak area in every sample due to the strong absorption of typical uranium bearing material at this energy level.

The analysis of data also shows the E\E (185.7 keV) have the consistent ratio to the highest peak area. This indicate that, we can make an estimation of the peak area where the measurements of special nuclear material due to the energy over the highest peak area is done.

Lastly in **Part III**: Characterization of Elemental Analysis and Direct Gamma Ray by CdTe and HPGe for TRIGA Mark III Fresh Fuel Rod Research Reactors. In this part of the investigation it is highly beneficial to the objective of safeguards to detect in a reasonably short amount of time to verify for seek out inconsistency in regards to the

amount of nuclear material as results of the existence of uranium K x-ray line in 1 minutes and the verification of ^{235}U in 3 minutes as the scanning inspection results.

As the conclusion, the x-ray fluorescence and direct gamma rays measurement have been proven to be a valuable tool for the trace element in the different types of samples such as uranium. It can provide rapid, multi element measurement with minimal preparation consisting of simple lines, less interference and good stability. By using the gamma ray spectrometry as a complement to trace element measurement by enriching the meter photo peak at 185.7 keV for special nuclear material ^{235}U. These analytical method has been playing an important role in safeguard inspection.

5.4 Recommendation based on the Finding and the Future Research

From the dealing with the detailed implementation of safeguards, a suspicious approach coupled with highly automated facilities has led to significant complexity in safeguards approaches. A comprehensive and all-embracing safeguards approach is increasingly costly and in sensitive bulk handling requires significant capital investment by the operator on NM and by the inspectorate on installing in the line independent monitoring equipment.

As in the future this research study should be further to the measurement with the standard in determination of the significant grams in the samples. Thus we can investigate the actual quantity of the uranium inside the sample.

As an advantages it is suggested that the measurement of uranium bearing material with one effective detector instead of using two detectors in the process of the safeguard scanning inspection. For this study it is recommended to use HPGe detector which is can analyze the characterization of direct gamma rays and x-ray fluorescence. In order to do this study it is suggested to do the measurement of the direct gamma ray from the specimen then as the exciting source is place, the elemental analysis can be determine with the same detector.

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APPENDIX

Specification of CdTe Detector – Model XR-100T-CdTe

SPECIFICATIONS

MODEL XR-100T-CdTe X-RAY and GAMMA RAY DETECTOR	
GENERAL	
Detector Type	Cadmium Telluride (CdTe) Diode
Detector Areas	3 x 3 mm (9 mm²) 5 x 5 mm (25 mm²)
Detector Thickness	1 mm
Energy Resolution @ 122 keV, ^{57}Co	9 mm²: <1.2 keV FWHM, typical 25 mm²: <1.5 keV FWHM, typical
Dark Counts	<5 x 10^{-3} counts/sec @ 10 keV <E <1 MeV
Be Window	4 mil thick (100 µm)
Preamplifier	Charge Sensitive, with Current Divider Feedback
Case Size	3.00 x 1.75 x 1.13 in 7.7 x 4.4 x 2.9 cm
Case Weight	4.4 ounces / 125 g
Total Power	Less than 1 Watt
Operation Conditions	0°C to +40°C
Storage and Shipping Typical Long-term storage	-20°C to +50°C, 10 to 90% humidity non condensing 10+ years in dry environment
TÜVRheinland	TUV Certification Certificate #: CU 72072412 01 Tested to: UL 61010-1: 2004 R7 .05 CAN/CSA-C22.2 61010-1: 2004
INPUTS	
Preamp Power	± 8 Volts @ 25 mA
Detector Power	+ 500 Volts @ 1 µA
Cooler Power	Current = 350 mA maximum Voltage = 4 V maximum
OUTPUTS	
Preamplifier Sensitivity Polarity	0.82 mV/keV Negative signal out 1 kΩ max. load
Temperature Monitor Sensitivity	PX5: direct reading in K through software
CONNECTORS	
Preamp Output	BNC coaxial connector
Power and Signal	6-Pin LEMO connector (Part# ERA.1S.306.CLL)
Interconnect Cable	6-Pin, LEMO (Part# FFA.1S.306. CLAC57) to 6-Pin LEMO (5 ft length)
6-PIN LEMO CONNECTOR ON THE XR-100T-CdTe	
Pin 1	Temperature monitor diode
Pin 2	+ H.V. detector bias, +500 V
Pin 3:	-8 Volt preamp power
Pin 4	+8 Volt preamp power
Pin 5	Cooler power return
Pin 6	Cooler power (0 to +4 Volt @ 0.350 A max.)
Case	Ground and shield
OPTIONS	
Other detector sizes (5x5x1mm) available on special orders.	
Other Be window thicknesses available on special orders.	
Components for vacuum applications.	
Collimator kit for high flux applications.	
See also XR-100CR specifications using Si-PIN for detection of low energy X-Rays with high resolution (149 eV FWHM @ 5.9 keV, ^{55}Fe).	
Available in X-123CdTe configuration (see Figure 1).	

MODEL PX5 DIGITAL PULSE PROCESSOR, MCA and POWER SUPPLY

Power to the XR-100T-CdTe is provided by the PX5. The PX5 is DC powered by an AC adaptor and provides both a variable Digital Pulse Shaping Amplifier (0.2 µs to 100 µs shaping time), the MCA function, and all necessary power supplies for the detector and preamplifier. The PX5 connects via USB, RS232, ot Ethernet to a PC. Please see PX5 specifications at http://www.amptek.com/px5-.html.

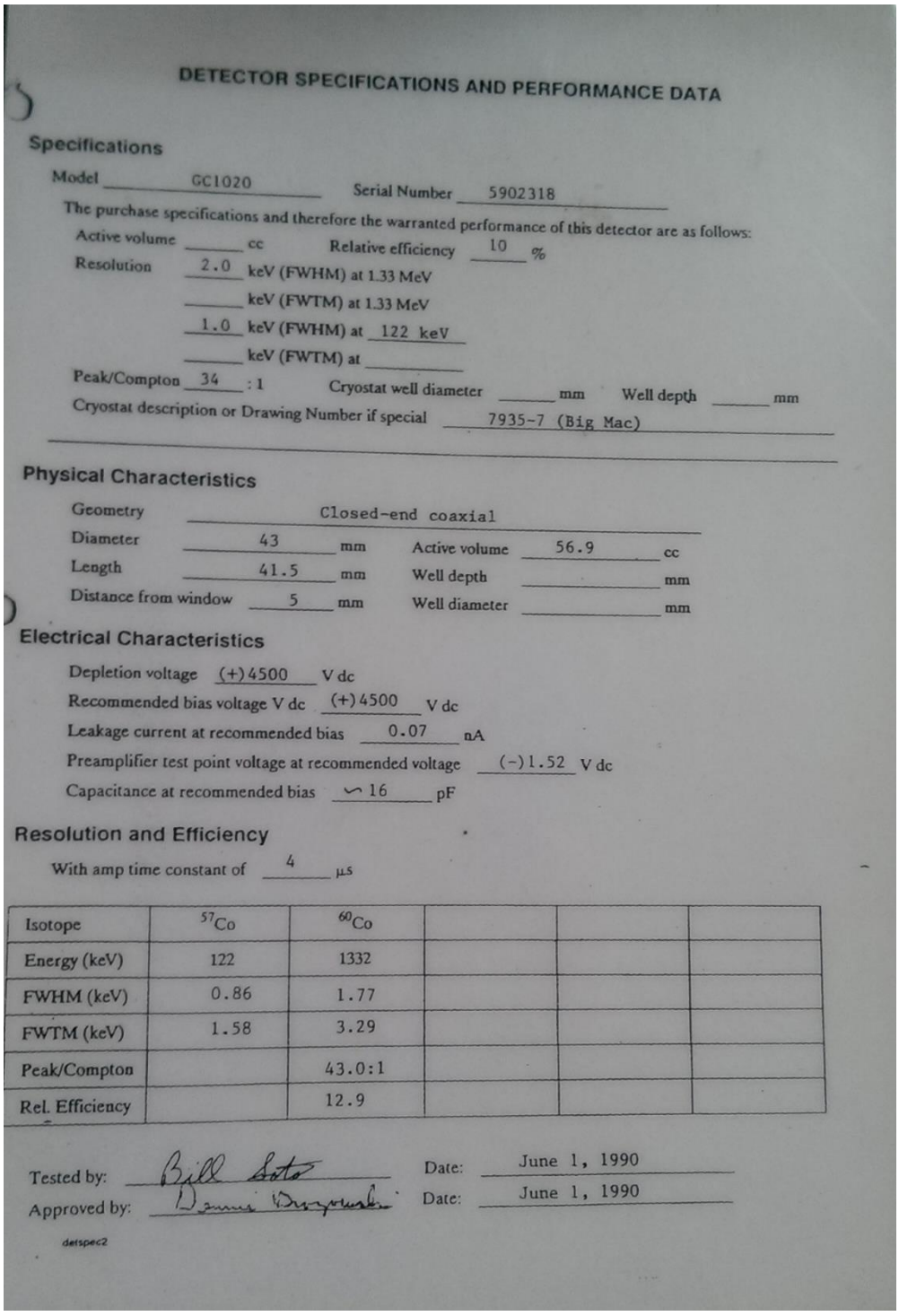
VACUUM OPERATION

The XR-100T-CdTe can be operated in air or in vacuum down to 10-8 Torr. There are two ways the XR-100T-CdTe can be operated in vacuum:

1) The entire XR-100T-CdTe detector and preamplifier box can be placed inside the chamber. In order to avoid overheating and dissipate the 1 Watt of power needed to operate the XR-100T-CdTe, good heat conduction to the chamber walls should be provided by using the four mounting holes. An optional Model 9DVF 9-Pin D vacuum feedthrough connector on a Conflat is available to connect the XR-100T-CdTe to the PX5 outside the vacuum chamber.
2) The XR-100T-CdTe can be located outside the vacuum chamber to detect X-Rays inside the chamber through a standard Conflat compression O-ring port. Optional Model EXV9 (9 inch) vacuum detector extender is available for this application.

FIGURE 1. The CdTe detector in the X-123CdTe configuration

Specification of HPGe Detector – Model GC1020



DETECTOR SPECIFICATIONS AND PERFORMANCE DATA

Specifications

Model GC1020 Serial Number 5902318

The purchase specifications and therefore the warranted performance of this detector are as follows:

Active volume _____ cc Relative efficiency 10 %

Resolution 2.0 keV (FWHM) at 1.33 MeV

_____ keV (FWTM) at 1.33 MeV

1.0 keV (FWHM) at 122 keV

_____ keV (FWTM) at _____

Peak/Compton 34 : 1 Cryostat well diameter _____ mm Well depth _____ mm

Cryostat description or Drawing Number if special 7935-7 (Big Mac)

Physical Characteristics

Geometry Closed-end coaxial

Diameter 43 mm Active volume 56.9 cc

Length 41.5 mm Well depth _____ mm

Distance from window 5 mm Well diameter _____ mm

Electrical Characteristics

Depletion voltage (+)4500 V dc

Recommended bias voltage V dc (+)4500 V dc

Leakage current at recommended bias 0.07 nA

Preamplifier test point voltage at recommended voltage (-)1.52 V dc

Capacitance at recommended bias ~16 pF

Resolution and Efficiency

With amp time constant of 4 μs

Isotope	^{57}Co	^{60}Co			
Energy (keV)	122	1332			
FWHM (keV)	0.86	1.77			
FWTM (keV)	1.58	3.29			
Peak/Compton		43.0:1			
Rel. Efficiency		12.9			

Tested by: Bill Soto Date: June 1, 1990

Approved by: Dennis Breymeier Date: June 1, 1990

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VITA

Madihah Binti Mujaini

Currently, Madihah Binti Mujaini is a MSc. student in Chulalongkorn University under Nuclear Security and Safeguard Program at Nuclear Engineering Department. Madihah is a Physics lecturer, having graduated from the University Technology of Mara (UiTM) with a B.Sc. in Physics. Madihah worked for UNITEN (Universiti Tenaga Nasional) for five years. From 2007 to 2009 Madihah worked as a research assistant at (UiTM).