การพัฒนาเทคนิคในการตรวจยูเรเนียมด้อยสมรรถนะในกำบังรังสีสำหรับต้นกำเนิดรังสีความแรงสูง

นายนีล เรมอน กิลเลียร์โม

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Development of a Technique to detect the presence of Depleted Uranium in the radiation shielding of highly radioactive sources

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นิล เรมอน กิลเลียร์โม : การพัฒนาเทคนิคในการตรวจยูเรเนียมด้อยสมรรถนะในกำบังรังสีสำหรับต้นกำเนิดรังสีความแรงสูง (Development of a Technique to detect the presence of Depleted Uranium in the radiation shielding of highly radioactive sources) อ.ที่ปรึกษาวิทยานิพนธ์หลัก: นเรศร์ จันทน์ขาว, อ.ที่ปรึกษาวิทยานิพนธ์ร่วม: สุวิทย์ ปุณณชัยยะ, 67 หน้า.

ยูเรเนียมด้อยสมรรถนะในรูปของโลหะมักถูกใช้เป็นวัสดุกำบังรังสีอุปกรณ์บำบัดโรคทางการแพทย์และการถ่ายภาพทางรังสีเนื่องจากมีเลขอะตอมและความหนาแน่นสูง ยูเรเนียมด้อยสมรรถนะได้ถูกจัดเป็นวัสดุนิวเคลียร์โดยทบวงการพลังงานปรมาณูระหว่างประเทศและต้องมีการขึ้นบัญชีเพื่อการตรวจสอบยืนยัน ซึ่งในปัจจุบันมีการใช้กำบังรังสีชนิดที่ไม่ได้ใช้ยูเรเนียมด้อยสมรรถนะ เช่น ตะกั่ว และทังสะเตน มาแทน หน่วยงานที่รับผิดชอบและควบคุมจึงต้องมีการตรวจสอบ แต่ยังอาจขาดอุปกรณ์และเทคนิคที่เหมาะสมในการตรวจสอบว่ามียูเรเนียมด้อยสมรรถนะอยู่หรือไม่

โดยเฉพาะอย่างยิ่งขณะที่มีต้นกำเนิดรังสีที่มีความแรงสูงมากอยู่ภายใน การวิจัยนี้ได้ทดลองใช้วิธีวัดสเปกตรัมรังสีแกมมาพลังงานต่ำด้วยหัววัดรังสีชนิดแคดเมียมเทลลูไรด์และเจอร์มาเนียมบริสุทธิ์สูง

เพื่อหลีกเลี่ยงการรบกวนจากรังสีแกมมาพลังงานสูงจากต้นกำเนิดรังสีที่อยู่ภายในกำบังรังสี โดยได้ทดสอบวิธีการนี้ในห้องปฏิบัติการกับตัวอย่างยูเรเนียมด้อยสมรรถนะต่างๆ กัน และทำการตรวจสอบจริงกับโปรเจ็กเตอร์สำหรับต้นกำเนิดรังสีแกมมาที่ใช้ในงานถ่ายภาพด้วยรังสีและเครื่องฉายรังสีแกมมาจากต้นกำเนิดรังสีโคบอลต์-60 ซึ่งพบว่าสามารถตรวจสอบโปรเจ็กเตอร์ที่บรรจุต้นกำเนิดรังสีอิริเดียม-192 ได้ทุกกรณี แม้ขณะมีอิริเดียมที่มีความแรงรังสีสูงถึง 60 คูรีบรรจุอยู่ ส่วนเครื่องฉายรังสีแกมมาจากต้นกำเนิดรังสีโคบอลต์-60 ได้เปลี่ยนไปใช้ตะกั่วและทังสะเตนแล้ว จึงไม่จำเป็นต้องตรวจสอบ

ภาควิชา	วิศวกรรมนิวเคลียร์	ลายมือชื่อนิสิต .....
สาขาวิชา	เทคโนโลยีนิวเคลียร์	ลายมือชื่อ อ.ที่ปรึกษาหลัก .....
ปีการศึกษา	2558	ลายมือชื่อ อ.ที่ปรึกษาร่วม .....

5670567021 : MAJOR NUCLEAR TECHNOLOGY
KEYWORDS: DEPLETED URANIUM / GAMMA-RAY SPECTROSCOPY / CADMIUM TELLURIDE / HIGH PURITY GERMANIUM / RADIATION SHIELDING

NEIL RAYMUND GUILLERMO: Development of a Technique to detect the presence of Depleted Uranium in the radiation shielding of highly radioactive sources. ADVISOR: ASSOC. PROF. NARES CHANKOW, CO-ADVISOR: ASST. PROF. SUVIT PUNNACHAIYA, 67 pp.

Depleted Uranium (DU) metal is commonly used as shielding in medical radiation therapy and industrial radiography equipment because of its high atomic number and density. DU though is classified as a nuclear material by the International Atomic Energy Agency (IAEA) and must be accounted for and subject to IAEA verification measures. Nowadays shielding containers may be replaced by lead and tungsten, thus Nuclear Regulating Authorities should be able to check the presence of DU. They may however lack the proper equipment to detect low energy gamma from DU particularly while having the high activity source present. In this research, low energy gamma-ray spectrometry using Cadmium Telluride (CdTe) and High Purity Germanium (HPGe) detectors was experimentally tested in laboratory and field measurements with various DU samples as well as with industrial radiography source projectors and Co-60 cancer treatment equipment. The proposed method was applied successfully with all gamma radiography source projectors containing Ir-192 up to 85 Ci. However, it was deemed unnecessary to inspect the Co-60 cancer treatment equipment since DU was no longer used as the shielding material.

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

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Chapter I

INTRODUCTION

1.1 Background on problems of interest

Depleted uranium (DU) is a by-product of the uranium enrichment process. It is mildly radioactive and has the same chemical toxicity and radiological properties as natural uranium. Uranium which has an atomic number Z = 92, is much denser than lead, so it has much better shielding properties. DU metal is commonly used as shielding in medical radiation therapy and industrial radiography equipment. It is used to make storage flasks for nuclear waste and flasks for the safe transport of highly active radiation sources that are used in the medical and engineering sectors.

Safeguards is a system of international inspections and other verification activities, undertaken by the International Atomic Energy Agency (IAEA) in order to evaluate, on an annual basis, a State's compliance with its obligations regarding the peaceful use of nuclear material. The IAEA refers nuclear material to the metals uranium, plutonium, and thorium, in any form. This is differentiated further into "source material", consisting of natural and depleted uranium, and "special fissionable material", consisting of enriched uranium U-235, uranium-233, and plutonium-239. Uranium ore concentrates are considered to be a "source material", although these are not subject to safeguards under the Nuclear Non-Proliferation Treaty.

According to the IAEA's Comprehensive Safeguards Agreement (CSA), DU must be accounted for and is subject to IAEA verification measures to confirm the reported quantities and peaceful use of the material. This is because DU stills contain some U-235, although in smaller concentration than natural uranium. The import and export of all DU, regardless of quantity, requires a license issued under the *Nuclear Non-Proliferation Import and Export Control Regulations*. Containers made of DU metal utilized to transport radioactive material, like tritium gas, are also subject to licensing. A State therefore must have an independent and competent regulating authority to effectively protect, regulate and control nuclear materials, facilities and nuclear-related activities. A State regulating authority is needed in order for the State

to establish and maintain its system of accounting and control of nuclear material (SSAC), which is an obligation each State accepts when it concludes a CSA.

The problem arises when these State regulating authorities lack or simply do not have the proper equipment to verify the presence of nuclear materials in particular depleted uranium. Many activities involving the use of nuclear material are related to the shielding of radioactive sources. Therefore, some safeguards activities such as establishing and verifying the inventory of nuclear material and supervising nuclear material control procedures, might be discharged by staff members responsible for radiation protection or security. Without the proper equipment, the regulating authority will not be able to verify and prove that the information given by an applicant for a license is true and valid. Much more if an existing license holder withholds pertinent information such as the declaration of lead shielding rather than the actual depleted uranium shielding. Without the proper equipment, the regulating authority cannot ensure that licensed holders of nuclear material are complying with their license conditions, including those related to accounting for and controlling nuclear material, when they conduct periodical audits and inspections of their inventories.

1.2 Thesis Objective

To develop a technique to detect the presence of depleted uranium in the shielding materials of containers with highly radioactive sources.

1.3 Scope of the study

The scope of this study will cover the following:

1. Developing a technique to detect the presence of depleted uranium in the shielding materials of containers with highly radioactive sources by using low energy gamma-ray detectors such as Cadmium Telluride (CdTe) and High Purity Germanium (HPGe).
2. Determining the sensitivity of the developed technique.
3. Investigating the factors that affect the sensitivity such as the energy and activity of gamma-ray source, age of depleted uranium, type and thickness of the container's cover material.

4. Testing the developed technique with industrial radiography projectors containing Iridium-192 and/or Selenium-75 sources and medical Cobalt-60 source used for cancer treatment in hospitals.

1.4 Expected benefits

This research could offer a simple method to detect the presence of depleted uranium in shielding materials for highly radioactive sources. This study could help regulating authorities in the inspection and verification of nuclear materials in particular depleted uranium. The development of such a technique will improve the accounting and control of nuclear materials within a State and an effective regulatory control builds confidence within the international community and demonstrates a strong commitment to the responsible use of nuclear materials.

1.5 Research Methodology

For this research work, the experimental part will be composed of the:

a. Conduct of gamma-ray spectroscopy on various gamma-ray sources and materials of different energy and activity, by CdTe and HPGe detectors.
b. Analysis of spectroscopy results
c. Develop procedures for gamma detection system to detect depleted uranium .
d. Determine the optimum settings of detection system by varying parameters such as distance, time and shielding material.
e. Test the detection system with the obtained optimum settings, on industrial projectors with Iridium-192 and/or Selenium-75 sources and medical Cobalt-60 sources used for cancer treatments in hospitals.
f. Evaluate test results

Chapter II

THEORY AND LITERATURE REVIEW

2.1 Properties and occurrence of Uranium

Uranium is a heavy, silvery-white, ductile and slightly paramagnetic metal, which is pyrophoric when finely divided. It is slightly softer than steel and reacts with cold water when present in a finely divided state. In air it easily oxidizes and becomes coated with a layer of oxide. Thus in nature uranium mainly occurs in oxidized form. Uranium is the heaviest naturally occurring element and is found at an average concentration of 0.0003% (3 mg/kg) in the earth's crust. In seawater the concentration is about 3.0 μg/l. Due to its presence in soil, rocks, surface and underground water, air, plants, and animals it occurs also in trace amounts in many foods and in drinking water. The daily intake of uranium is estimated to be 1–2 μg in food and 1.5 μg in water consumed[1]. The human body contains approximately 56 μg of uranium, 32 μg (56%) are in the skeleton, 11 μg in muscle tissue, 9 μg in fat, 2 μg in blood and less than 1 μg in lung, liver and kidneys[2]. The uranium in the human body is derived mostly from uranium in food, especially from vegetables, cereals, and table salt[3].

Natural uranium consists of a mixture of three radioactive isotopes which are identified by the mass numbers ^{238}U (99.27% by mass), ^{235}U (0.72%) and ^{234}U (0.0054%)[4]. Uranium isotopes undergo radioactive decay to stable Lead (Pb) isotopes via a series of radioactive elements called progeny or daughter radionuclides. 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 (radioactive) 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 these figures, as are those that emit significant gamma radiation.

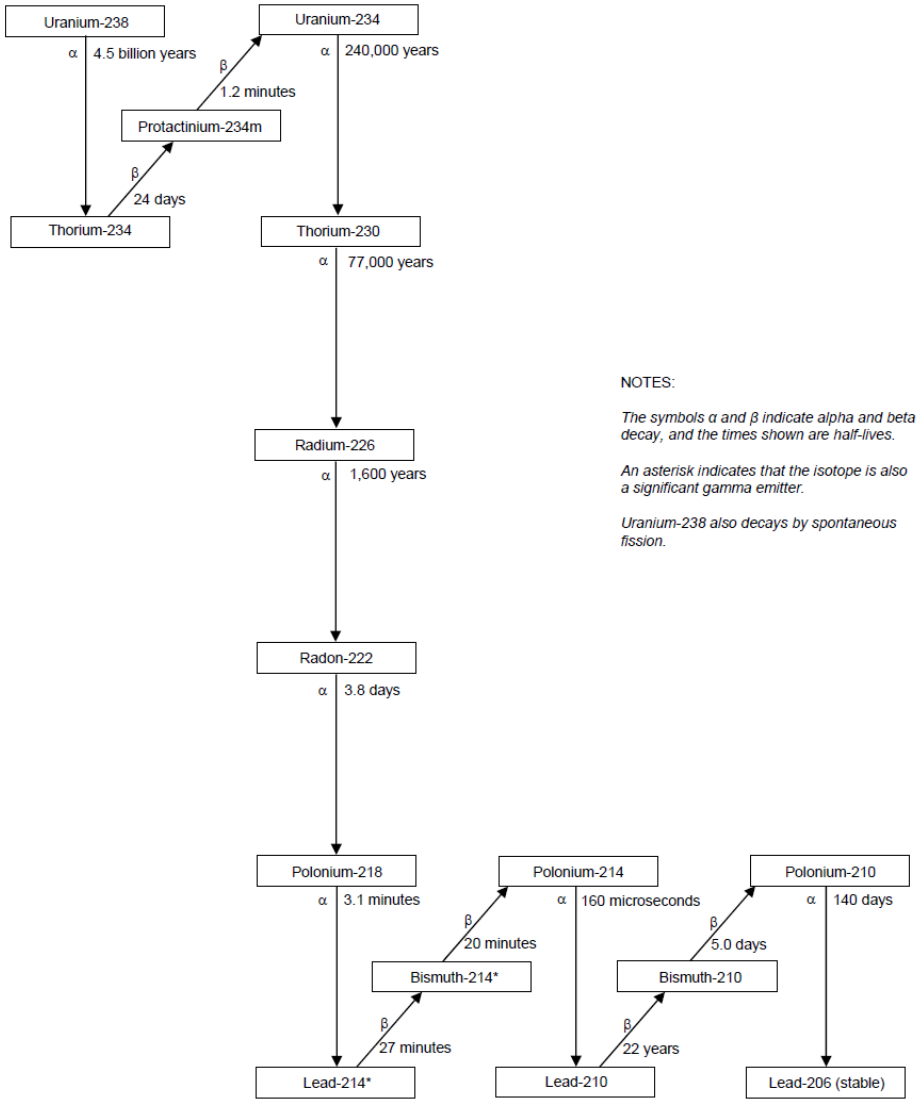

Figure 2-1 Natural decay series: Uranium-238[5]

Gamma radiation is not a mode of radioactive decay (such as alpha and beta decay). Rather, it is a mechanism by which excess energy is emitted from certain radionuclides, i.e., as highly energetic electromagnetic radiation emitted from the nucleus of the atom. For simplicity, only significant gamma emissions associated with the major decay modes are shown in Figures 2-1 and 2-2; that is, radionuclides listed are those for which the radiation dose associated with gamma rays may pose a health concern. The gamma component is not shown for those radionuclides whose gamma emissions do not generally represent a concern.

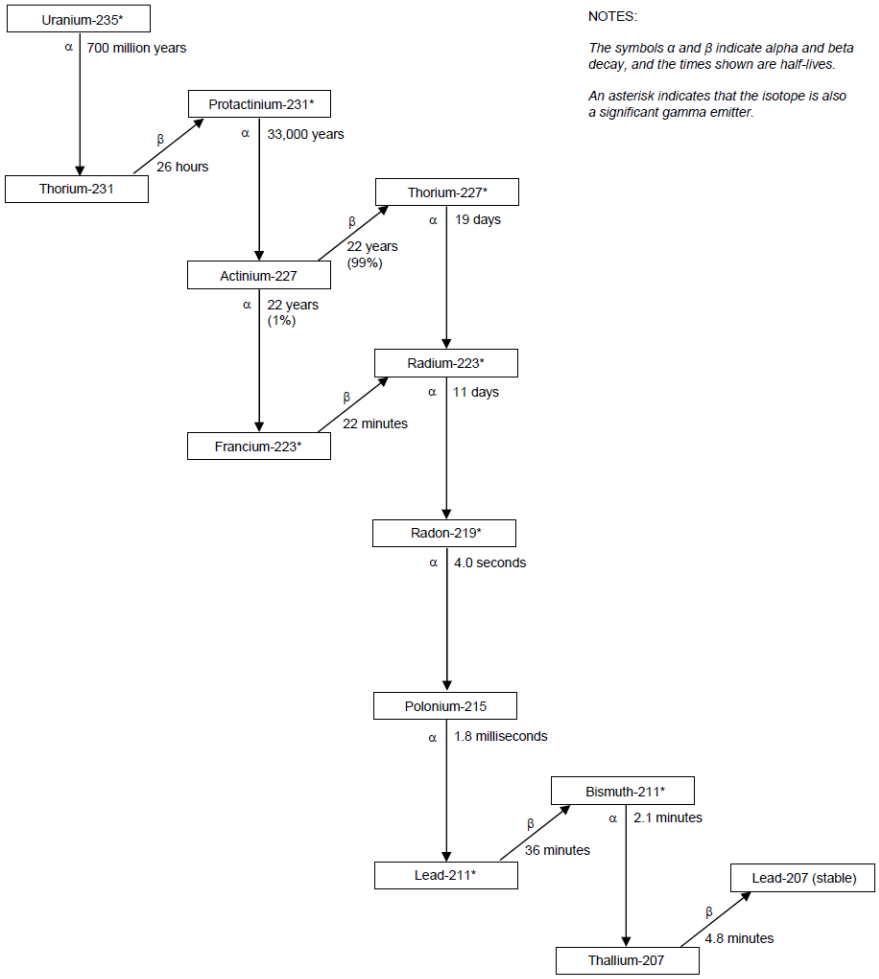

Figure 2-2 Natural decay series: Uranium-235[5]

In nature, the radionuclides in these two series are approximately in a state of secular equilibrium, in which the activities of all radionuclides within each series are nearly equal. Two conditions are necessary for secular equilibrium. First, the parent radionuclide must have a half-life much longer than that of any other radionuclide in the series. Second, a sufficiently long period of time must have elapsed, for example ten half-lives of the decay product having the longest half-life, to allow for ingrowth of the decay products. Under secular equilibrium, the activity of the parent radionuclide undergoes no appreciable changes during many half-lives of its decay products.

Of the two conditions noted above for secular equilibrium, the first is generally met for the uranium-238 and uranium-235 decay series in naturally occurring ores.

While the second condition may not be met for all ores or other deposits of uranium (given the extremely long half-lives for the radionuclides involved and the geological changes that occur over similar time scales), it is reasonable to assume secular equilibrium for naturally occurring ores to estimate the concentrations of the various daughter radionuclides that accompany the parent. The state of secular equilibrium in natural uranium ores is significantly altered when they are processed to extract specific radionuclides.

After processing, radionuclides with half-lives less than one year will reestablish equilibrium conditions with their longer-lived parent radionuclides within several years. For this reason, at processing sites what was once a single, long decay series (for example the series for uranium-238) may be present as several smaller decay series headed by the longer-lived decay products of the original series (that is, headed by uranium-238, uranium-234, thorium-230, radium-226, and lead-210 in the case of uranium-238). Each of these sub-series can be considered to represent a new, separate decay series. Understanding the physical and chemical processes associated with materials containing uranium, thorium, and radium is important when addressing associated radiological risks.

Natural uranium together with its daughters yield four–five times as many decays per second as pure uranium. Radon (^{222}Rn), a radioactive noble gas in the decay chain of ^{238}U, can easily escape from soil or rock and, contrary to uranium, is a major contributor to the radiation exposure of the world population. When uranium is separated from its ores, the decay chain is broken. Only thorium (^{234}Th) and protactinium (^{234}Pa) reach equilibrium with ^{238}U within about 1 year and are the major contributors to the radioactivity of the purified uranium. The remaining members of the decay chain following ^{234}U take thousands of years to reach equilibrium and can be neglected. ^{235}U follows the same pattern and only thorium (^{231}Th) reaches equilibrium rapidly. The decay products of ^{238}U (^{234}Th and ^{234}Pa) and ^{234}U (^{231}Th) are responsible for the presence of beta and gamma radiation in purified natural uranium.

2.2 Depleted Uranium

Uranium is used primarily in nuclear power plants; most reactors require uranium in which the ^{235}U content is enriched from 0.72% to about 3%. The uranium

remaining after removal of the enriched fraction is referred to as depleted uranium. Depleted uranium typically contains about 99.8% ^{238}U, 0.2% ^{235}U and 0.0006% ^{234}U by mass. In typical DU the content of ^{235}U is about one-third of its original value (0.2–0.3%). Consequently, the activity of DU is about 60% the activity of natural uranium. Metallic uranium (including DU) is 65% more dense than lead (density of 19 g/cm3), has a high melting point (1132 °C), is highly pyrophoric, and has a tensile strength comparable to most steels. These properties, as well as the relative high availability and low cost, has led to various civilian and military applications of DU.

DU metal is commonly used as shielding in medical radiation therapy and industrial radiography equipment and it is used to make storage flasks for nuclear waste and flasks for the safe transport of highly active radiation sources that are used in the medical and engineering sectors. DU has often been used as a shield for radioactive sources in tele-therapy units used in the treatment of cancer and in linear accelerators. Typical quantities of DU used in such equipment range from tens to hundreds of kilograms.

Current developments in waste management have also employed DU as a shielding material. For example, casks used for holding spent fuel in the nuclear power industry have been constructed by combining DU with concrete (e.g. DUCRETE™[6]). This achieves a significant increase in gamma-radiation shielding with thinner shield walls and much lighter weight casks than traditional storage casks.

2.3 Nuclear Safeguards of depleted uranium

Nuclear Safeguards are measures to verify that States comply with their international obligations not to use nuclear materials (plutonium, uranium and thorium) for nuclear explosives purposes. Global recognition of the need for such verification is reflected in the requirements of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) for the application of safeguards by the International Atomic Energy Agency (IAEA). Organizations such as the IAEA and EURATOM and others are tasked with ensuring declared amounts of material are indeed present in the facilities in which they are stored and that such material is not being diverted to other uses.

As part of the global nuclear non-proliferation regime, the State Regulating Authority (SRA) participates in domestic and international activities to account for and control nuclear material assigned to commercial and other peaceful purposes. In order to comply with its Government non-proliferation commitments, the SRA participates in many different activities such as implementing international safeguards treaties, reviewing import and export licenses, and collecting data through the national system of accounting for source and special nuclear materials. The technical measures used to provide credible assurance vary, however some practices are fairly commonplace such as nuclear material accountancy, containment (i.e., tamper indicating seals), surveillance, and inspections. Using all of the available expertise, equipment, and data, the IAEA generates and implements a safeguards approach at both the facility level, and the State level. At the end of the year, the IAEA will draw conclusions about a particular State, and publish these findings in its annual report.

Most of the world's non-nuclear-weapon States (NNWSs) have concluded comprehensive safeguards agreements (CSAs) with the IAEA, pursuant to the (NPT). The IAEA and States are required to cooperate in the implementation of such agreements. Effective cooperation demonstrates a State's commitment to the peaceful use of nuclear energy and furthers the State's national interests by reducing the risk of unauthorized use of nuclear material.

Over 100 NNWSs party to the NPT have very limited quantities of nuclear material and have concluded protocols to their CSAs which hold in abeyance many procedures in Part II of a CSA. These protocols are referred to as 'small quantities protocols' or 'SQPs' and remain in effect as long as the State meets certain eligibility criteria. The purpose of an SQP is to reduce the burden of safeguards implementation for States with little or no nuclear activities, while retaining the integrity of the safeguards system.

Nuclear material as defined in Article XX of the IAEA Statute[7]. The term source material does not apply to ore or ore residue. The definitions of special fissionable material and source material are provided below.

"The term 'special fissionable material' means plutonium-239, uranium-233, uranium enriched in the isotopes 235 or 233; any material containing one or more of the foregoing, and such other fissionable material as the Board of Governors shall from

time to time determine; but the term ‘special fissionable material’ does not include source material.”

“The term ‘source material’ means uranium containing the mixture of isotopes occurring in nature (e.g. 99.3% uranium-238, 0.7% uranium-235); uranium depleted in the isotope 235; thorium; any of the foregoing in the form of metal, alloy, chemical compound, or concentrate; any other material containing one or more of the foregoing in such concentration as the Board of Governors shall from time to time determine; and other such material as the Board of Governors shall from time to time determine.”

Depleted uranium, plutonium and enriched uranium are 34(c) nuclear material (referring to paragraph 34(c) of INFCIRC/153 Corr.)[8], regardless of form, purity, use or quantity. Ore and ore deposits are not 34(c) nuclear material. The SRA should make all reasonable efforts to identify and locate all nuclear material in the State. Nuclear material in SQP States is often used in medical, industrial, academic and research applications. One of the most common uses of 34(c) material in SQP States is the use of depleted uranium as radiation shielding in containers or instruments that contain high-activity radioactive sources. Therefore, the SRA could use its radioactive source registry to begin identifying locations which use high-activity sources (such as Co-60) as they may also have depleted uranium shielding[9].

Equipment that is imported and that contains depleted uranium should be accompanied by certification or documentation that specifies the quantity, composition, form and number of items that contain nuclear material, such as collimators or source removal/replacement machines. Containers with depleted uranium shielding should also be labelled with the quantity of depleted uranium indicated either on the label or on the documentation that accompanied the container. The shipper’s address and contact information should also be indicated on the document or certificate, and the SRA can contact the shipper for additional information regarding the nuclear material as necessary.

Appendix 1 of the SQP Guide[9] provides information about common applications of nuclear material in each sector and a list of various equipment models in medical and industrial applications and the quantity of depleted uranium shielding in each instrument, based on manufacturer specifications. This information is provided to help the SRA to locate, control and report all nuclear material in the State.

2.4 Gamma-ray spectrometry

Gamma-ray spectrometry is an analytical method that allows the identification and quantification of gamma emitting isotopes in a variety of matrices. In one single measurement and with little sample preparation, gamma-ray spectrometry allows you to detect several gamma emitting radionuclides in the sample. The measurement gives a spectrum of lines, the amplitude of which is proportional to the activity of the radionuclide and its position on the horizontal axis gives an idea on its energy.

2.4.1 Gamma emission and detection of nuclear materials

Most nuclear materials under IAEA safeguards emit gamma-rays that can be used for Non-destructive analysis (NDA) of the materials. Gamma-rays are electromagnetic radiation produced by nuclear interactions. It is generally characterized as high energy radiation and short wavelengths within the electromagnetic spectrum. Gamma-rays have well defined energies that are characteristic of the isotopes emitting them. Determination of the gamma-ray energies and their relative intensities serves to identify the isotopic composition of the materials. When combined with a measurement of absolute intensities, the gamma-ray energies can provide quantitative information on the amount of material that is present. Enriched uranium fuel, for example, has a strong 186 keV gamma-ray associated with the alpha decay of ^{235}U, and the ^{235}U enrichment can be verified by measuring this gamma-ray. To detect gamma-rays, the radiation must interact with a detector to give up all or part of the photon energy. The basis of all spectroscopic gamma ray detector systems is the collection of this liberated electrical charge to produce a voltage pulse whose amplitude is proportional to the energy deposited by a gamma-ray in a detector. These pulses are then sorted according to amplitude (energy) and counted using appropriate electronics, such as a single or multichannel analyzer. With a multichannel analyzer (MCA), the gamma-rays of different energies can be displayed or plotted to produce a gamma-ray energy spectrum that provides detailed information on the measured material.

2.4.2 Gamma-ray detectors

The detector is the center piece of the gamma spectroscopy system. The gamma photons interact with the detection material and transfer their energies to electrons or to positrons in the case of annihilation. These produced particles lose their energy

within the detector, creating ionized atoms and ion pairs. These secondary entities form the basis of the detector signal. The detector choice must be evaluated in the light of the technical requirements of a proposed application and the non-technical but often over-riding matter of budgetary constraints. The first and most important detector parameter to consider is resolution. Resolution is a measure of the width (full width half max, FWHM) of a single energy peak at a specific energy, either expressed in absolute keV (as with Germanium Detectors), or as a percentage of the energy at that point (Sodium Iodide Detectors). Better (lower FWHM value) resolution enables the system to more clearly separate the peaks within a spectrum.

The various types of detector that are now used for inspection purposes namely are, high purity germanium (HPGe), cadmium zinc telluride (CdZnTe)/cadmium telluride (CdTe) and sodium iodide (NaI) detectors — allowing high, medium and low resolution spectrometry. Germanium detectors have energy resolution far superior to that of NaI detectors and are better suited to the task of resolving complex gamma ray spectra and providing information about the isotopic content of materials. A disadvantage of these detectors is that they must be operated at a very low temperature, which is usually achieved by cooling with liquid nitrogen. Recently, electric cooling systems have become available, mitigating this disadvantage with almost no effect on detector performance characteristics. Standard CdZnTe detectors (and cadmium telluride (CdTe) detectors) do not need cooling, and currently in widespread use since they have the highest intrinsic detection efficiency. Recent progress in fabrication techniques has substantially improved CdZnTe resolution. Detectors with volumes ranging from 5 to 1500 mm^3 are available. The portability and small size of CdZnTe and CdTe detectors have made them especially suitable for use in a wide range of applications, including in confined spaces such as for in situ verification of fresh fuel assemblies whose design permits insertion of only a small detector probe into the assembly interior, and of spent fuel bundles stored underwater in closely packed stacks. The NaI detectors can be made with large volumes and generally have higher gamma-ray detection efficiencies than do germanium detectors. Their safeguards applications include the verification of both ^{235}U enrichment in fresh fuel and the presence of spent fuel through detection of fission product gamma-radiation. Their ability to distinguish between gamma-rays of different energies, however, is relatively poor, and of the

detector types mentioned here they have the worst energy resolution. Figure 2-3 illustrates the capabilities of various types of detector with low, medium and high resolution.

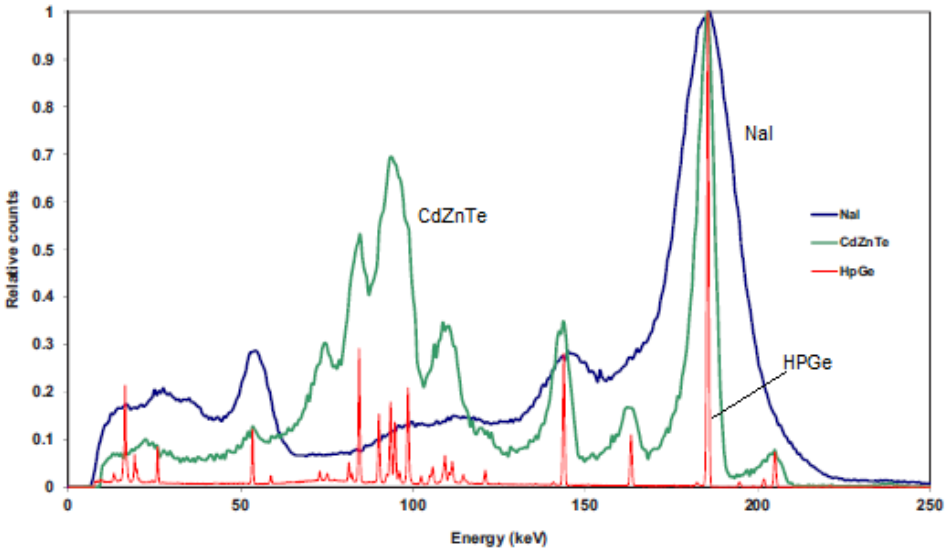

Figure 2-3 Comparison of gamma ray spectrometric performance of various types of detector with low, medium and high resolution [10]

Low and medium resolution gamma spectrometry applications in safeguards range from quantitative verification of enrichment levels to the purely qualitative detection of plutonium and uranium in fresh and spent fuel, and of the presence of nuclear material in general. When coupled to a germanium detector, the Miniature Multichannel Analyzer (MMCA) or MCA becomes a high resolution gamma ray spectrometer. This type of spectrometer is often used to determine the ^{235}U enrichment of uranium hexafluoride (UF_6) in shipping cylinders.

2.4.2.1 Cadmium (Zinc) Telluride Detectors

Cadmium telluride and cadmium zinc telluride have been regarded as promising semiconductor materials for hard X-ray and gamma-ray detection. The high atomic number of the materials (Z_{Cd} =48, Z_{Te}=52) gives a high quantum efficiency in comparison with Si. The large band-gap energy (E_g = 1.5 eV) allows us to operate the detector at room temperature. However, a considerable amount of charge loss in these detectors produces a reduced energy resolution. This problem arises due to the low

mobility and short lifetime of holes. Recently, significant improvements have been achieved to improve the spectral properties based on the advances in the production of crystals and in the design of electrodes. Silicon (Si) and germanium (Ge) are traditional semiconductors used for radiation detectors that offer good performance in a wide range of applications[11]. The growing field of applications has stimulated the development of detectors based on compound semiconductors [11-13] . They were first investigated as radiation detectors in 1945 by Van Heerden[14], who used AgCl crystals for detection of alpha particles and gamma rays. A great advantage of compound semiconductors is the possibility to grow materials with a wide range of physical properties (band gap, atomic number, density) making them suitable to almost any application. Interests in radiation detectors operating at room temperature gave rise to development of compound semiconductors with wide band gaps, in comparison to Si and Ge. Moreover, for X-ray and gamma ray detection, compound semiconductors with high atomic number were preferred in order to emphasize photoelectric interaction. Compound semiconductors are generally derived from elements of groups III and V (e.g. GaAs) and groups II and VI (e.g. CdTe) of the periodic table. Besides binary compounds, ternary materials have been also produced, e.g. CdZnTe and CdMnTe. Table 2-1 reports the physical properties of the most common compound semiconductors typically used for radiation detection.

Table 2-1Physical properties of the principal compound semiconductors [15]

Material	Si	Ge	GaAs	CdTe	$Cd_{0.9}Zn_{0.1}Te$	HgI_2	TlBr
Crystal structure	Cubic	Cubic	Cubic (ZB)	Cubic (ZB)	Cubic (ZB)	Tetragonal	Cubic (CsCl)
Growth method*	C	C	CVD	THM	HPB, THM	VAM	BM
Atomic number	14	32	31, 33	48, 52	48, 30, 52	80, 53	81, 35
Density (g/cm^3)	2.33	5.33	5.32	6.20	5.78	6.4	7.56
Band gap (eV)	1.12	0.67	1.43	1.44	1.57	2.13	2.68
Pair creation energy (eV)	3.62	2.96	4.2	4.43	4.6	4.2	6.5
Resistivity (Ω cm)	10^4	50	10^7	10^9	10^{10}	10^{13}	10^{12}
$\mu_e\tau_e$ (cm^2/V)	> 1	> 1	10^{-5}	10^{-3}	10^{-3} - 10^{-2}	10^{-4}	10^{-5}
$\mu_h\tau_h$ (cm^2/V)	~ 1	> 1	10^{-6}	10^{-4}	10^{-5}	10^{-5}	10^{-6}

* The more common growth methods: C = Czochralski, CVD = chemical vapor deposition, THM = traveler heater method, BM = Bridgman method, HPB = high-pressure Bridgman and VAM = vertical ampoule method

Among the compound semiconductors, CdTe and CdZnTe have attracted growing interests in the development of X-ray and gamma ray detectors[16]. Due to the high atomic number, the high density and the wide band gap, CdTe and CdZnTe detectors ensure high detection efficiency, good room temperature performance and are very attractive for X-ray and gamma ray applications. Difficulties in producing detector-grade materials and in growing chemically pure and structurally perfect crystals are the critical issues of CdTe and CdZnTe detectors. In fact, the great potential of these compounds has not been exploited for many decades due mainly to the limited commercial availability of high-quality crystals. This situation has changed dramatically during the mid-nineties with the emergence of a few companies committed to the advancement and commercialization of these materials.

The typical operation of semiconductor detectors is based on collection of the charges, created by photon interactions, through the application of an external electric field. The choice of the proper semiconductor material for a radiation detector is mainly influenced by the energy range of interest. Among the various interaction mechanisms of X-rays and gamma rays with matter, three effects play an important role in radiation measurements: photoelectric absorption, Compton scattering and pair production. In photoelectric absorption the photon transfers all its energy to an atomic electron, while a photon interacting through Compton process transfers only a fraction of its energy to an outer electron, producing a hot electron and a degraded photon; in pair production a photon with energy above a threshold energy of 1.02 MeV interacts within the Coulomb field of the nucleus producing an electron and positron pair. Neglecting the escape of

characteristic X-rays from the detector volume (the so called fluorescent lines), only the photoelectric effect results in the total absorption of the incident energy and thus gives useful information about the photon energy[15].

2.4.2.2 High Purity Germanium Detectors

High-Purity Germanium (HPGe) detector is a semiconductor particle detector. These detectors are mainly used in gamma ray spectroscopy. Germanium detectors are semiconductor diodes having a p-i-n structure in which the intrinsic (i) region is sensitive to ionizing radiation, particularly X-rays and Gamma rays. These detectors directly collect the charges produced when the semiconductor material is ionized by the ionizing radiations. Electron-hole pairs are produced by ionization. These pairs drift under an external electric field to the p and n electrodes where they generate the pulse. In the fabrication of high purity Ge crystals, if the remaining low level impurities are acceptors, the electrical properties of the Ge crystal is mildly p type and if donor impurities are present, the crystal is n type. But still the impurities are so low that we can easily call it intrinsic Germanium. So in HPGe, Germanium is used as intrinsic semiconductor. There is a wide region of intrinsic Ge in the middle surrounded by p-type and n-type semiconductor contacts. The PIN diode is reverse biased. A photon entering the intrinsic region creates an electron-hole pair. The reverse bias filed sweeps the carrier out of the region and creates a current.

When a gamma-ray interacts in the active volume, it creates electrons which are collected on the n+ contact, while the holes are collected on the p+ contact, and the current that occurs as the electrons and holes move toward the electrodes, once integrated, constitutes the gamma-ray energy signal. Generally the n type material is made by diffusing lithium over the germanium crystal and the p type material is made by implantation of boron.

Simple junction and surface barrier detectors can easily detect alpha particles and other short range radiations but are not easily adaptable for more penetrating radiations. The major limitation is the maximum depletion depth or the active volume that can be created. Using Silicon or Germanium of normal semiconductor purity, depletion depths of only 2-3mm can be achieved. Much greater thickness is required for the detectors intended for gamma ray spectroscopy.

The thickness of depletion region is inversely related to the net impurity concentration in bulk semiconductor material. So we use further refining techniques capable of reducing impurity concentration to approximately 10^{10} atoms/cm^3. Techniques have been developed to achieve this goal in germanium, but not in silicon because of much higher melting point of Si as compared to Ge which makes the exclusion of impurities in there fining process more difficult. High purity Germanium detectors are manufactured using this ultrapure Germanium and are now available with depletion depths of several centimeters. High Purity Germanium (HPGe) is the only radiation detection technology that provides sufficient information to accurately and reliably identify radionuclides from their passive gamma ray emissions. HPGe detectors have a 20-30 times improvement in resolution as compared to that of Sodium Iodide (NaI) detectors. So the major characteristics of the HPGe detector are high atomic number, low impurity concentration (large depletion depth), low ionizing energy required to produce an electron-hole pair, high conductivity, compact size, first time response, high resolution and relative simplicity of operation[17].

2.5 Related Research Work on determination of Natural and depleted uranium

Shoji, et al [18] developed a method for discriminating between natural and depleted uranium of reagent grade by measuring and analyzing the gamma ray spectrum of the sample without any standard reference sources. The method is based on the principle that the counting efficiency of photoelectric peaks of gamma rays can be expressed as a function of gamma ray energy under specified measuring conditions. The discrimination between natural and depleted uranium is important for the proper management of nuclear fuel materials. One way to achieve this is a comparison of the gamma ray spectrum of an unknown sample with that of a natural uranium sample. This method needs a reference uranium standard that should be authenticated to be of natural isotopic composition. It should also be of reagent grade because natural uranium minerals are not suitable for the purpose. The natural uranium minerals contain ^{226}Ra and its daughter nuclides in the ^{238}U decay chain and ^{231}Pa and its daughter nuclides in the ^{235}U decay chain. The gamma rays from ^{226}Ra (186.10 keV) in the ^{238}U decay chain obstruct those from ^{235}U (185.72 keV) (Firestone and Shirley, 1996) in the case of uranium minerals. Another disadvantage of the method is that the influence of

absorption of gamma rays by samples themselves and their containers should be corrected because the samples and their containers are of different shapes and materials. Three among 16 samples of uranium of reagent grade were determined to be natural in our laboratory. Using this method, depleted uranium of which the isotopic composition is close to that of natural uranium maybe identified as natural. However, this method appears to be a useful and a convenient method for discriminating between natural and depleted uranium of reagent grade.

Saleh and Abdel-Halim [19] used a high-resolution gamma-ray spectrometer based on a hyper-pure germanium detector to determine the amounts of depleted uranium in ground features subjected to military operations during the Gulf War of 1991 and in beach sediment samples collected from the northern side of the Arabian Gulf. The determination of $^{235}U/^{238}U$ was evaluated using spiked samples with a series of depleted uranium solutions. According to this method, the levels of depleted uranium were found to exceed 6.5% of the total natural uranium required to achieve reasonable levels for detection. Soil results indicated that the average of the total radioactivity of ^{238}U is 50.59 Bq/kg, with approximately 41.41% of this being represented by depleted uranium. ^{235}U radioactivity was determined using the gamma energy lines of 143.76 keV and 163.36 keV. However, the most intensive gamma line of 185.71 keV was excluded to avoid its interference with the 186.21 keV gamma line of ^{226}Ra. ^{238}U radioactivity was determined using its direct daughter ^{234}Th(24d) at the gamma lines 63.39 keV, and both 92.38 keV and 92.80 keV were taken as one line, assuming that ^{238}U and ^{234}Th are in secular equilibrium.

The Nuclear Engineering Section of the National Technical University of Athens (NES-NTUA)[20] uses high-resolution Low energy Germanium (LEGe) detectors and especially developed gamma spectroscopic analysis techniques for the determination of ^{238}U and ^{235}U and subsequently for the isotopic abundance of the uranium isotopes in the sample. ^{235}U is determined from the analysis of the multiplet photopeak at 186 keV. The efficiency calibration curves of the Ge detectors of NES-NTUA for the sample geometry used are presented in Figure 2-4.

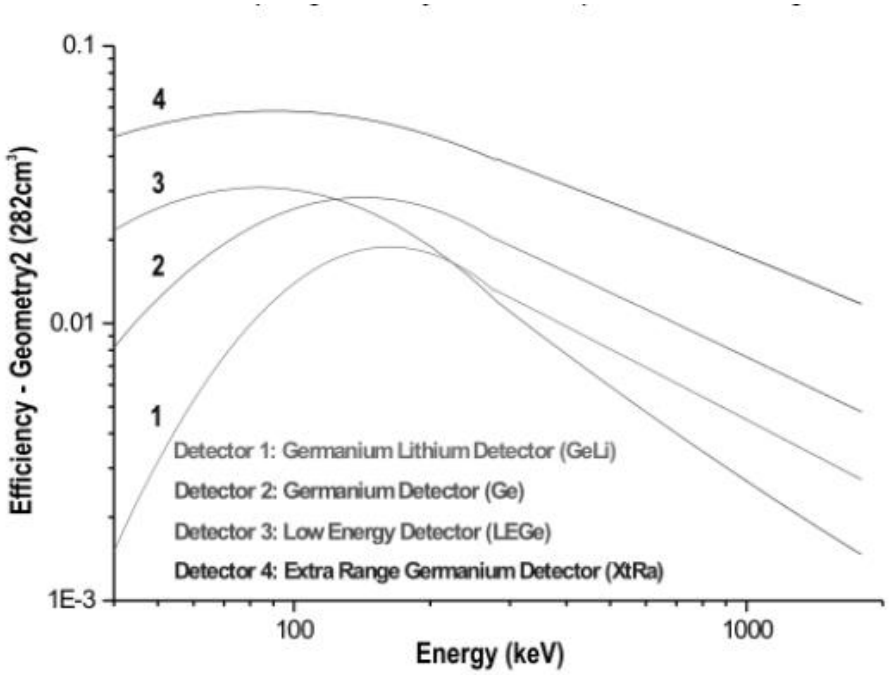

Figure 2-4 Full energy peak efficiency curves for sample geometry of NES-NTUA

The technique used in this work for the determination of ^{235}U is based on the analysis of the multiplet photopeak at ~186 keV using the LEGe detector calibrated in the low energy region 0-200keV, which allows for sufficient number of channels per keV (e.g. ~20 channels / keV). The results obtained found that the multiplet photopeak at ~186 keV may be analyzed into its two components, at 185.75 and 186.25 keV, leading to the direct determination of ^{235}U and ^{226}Ra respectively, in the cases where the statistics of the multiplet photopeak at ~186 keV was satisfactory. The spectrum of a soil sample in the energy region 0-200 keV is presented in Figure 2-5.

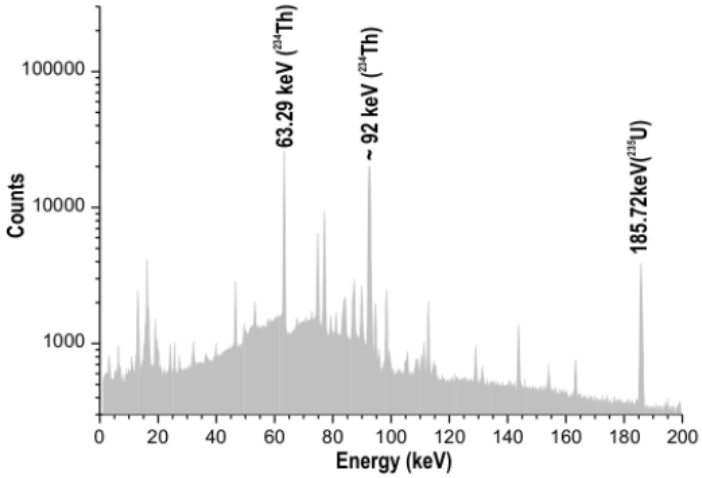

Figure 2-5 Gamma spectrum of DU contaminated soil sample in the energy region 0-200keV

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2.6 Summary of Literature Review

From the collection of the literatures, common focus of the study were established. The properties and occurence of uranium and its isotopes have well been documented. For this study, the application of depleted uranium as radiation shielding and its classification as a nuclear source material justifies the importance of its accounting and safeguarding. No research work on exactly the detection of DU in radiation shielding has been published but there are more than enough research on the determination of DU by gamma spectroscopy which will help support the objective and scope of this study. Literature might overlook the importance of this specific topic but the significance and the technology is there to make this research study a novel and very useful innovation.

Chapter III

MATERIALS AND METHODOLOGY

3.1 Materials

In this study the various materials investigated, analyzed and used are composed of standards, calibration and radionuclide sources, radiation shielding materials, projectors and detectors.

3.1.1 Samples and standards

3.1.1.1 Natural Uranium Metal Standard (NUMS)

The NUMS shown in Figure 3-1 is from Koch-light Limited which has the dimensions of 50 mm x 50 mm x 0.175 mm. The composition of which is 99.3% U-238 and 0.7% U-235.

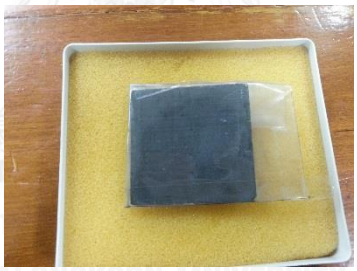

Figure 3-1 Natural Uranium Metal Standard

3.1.1.2 Depleted Uranium Block

In Figure 3-2 is the Depleted Uranium block (8cm x 6.7cm x 15.6 cm) that will serve as the DU standard.

Figure 3-2 Depleted Uranium block

3.1.2 Calibration and Radionuclide sources

Calibration of the detecting systems was done by using ^{238}Pu and ^{241}Am sources whose energies are in Table 3-1.

Table 3-1 Calibration and Radionuclide sources

Isotope	Half-life	Activity	Photon energies (keV)
Pu-238	87.7 years	100 μCi	X-rays
		January 01, 1988	13.613
			17.218
			20.163
Am-241	433 years	0.977 μCi	Gamma
		January 04, 1988	59.543
Co-60	5.3 years	10.32μCi	Gamma
		January 01, 2015	1173.2
			1332.5

3.1.3 Radiation Shielding Materials

Radiation shielding materials such as Aluminum (Al), Iron (Fe) and Lead (Pb) will be used to test the penetration capabilities of the gamma-rays emitting from the NUMS

and DU block. These materials are in the form of square (10 cm x 10 cm) sheets with thicknesses of about 1 mm. The densities of these materials are shown in Table 3-2.

Table 2-2 Density of some common Shielding materials

Element	Density, ρ (g/cc)
Aluminum	2.70
Iron	7.87
Lead	11.3
Uranium	19.1
Gold	19.3
Tungsten	19.3

3.1.4 Industrial Radiography Projectors

The industrial radiography projectors measured for this study are listed in Table 3-3.

Table 3-3 Industrial radiography projectors

Model	Serial Number	Source	Activity (Curies, Ci)
Sentinel 880 Delta	D1229	Iridium-192	0Ci
Sentinel 660B	B3132	Iridium-192	2Ci
Sentinel 880 Delta	D9693	Iridium-192	40Ci
Sentinel 880 Delta	AD623	Iridium-192	86Ci
Sentinel 680B	B256	Cobalt-60	8 Ci

Sentinel 880 Delta Sentinel 660B

Figure 3-3 Two types of Industrial Radiography projectors

Figure 3-4 Cobalt-60 radiography projector (Sentinel 680B) at NDT Division of the Nuclear Technology Service Center of Thailand Institute of Nuclear Technology

3.1.5 Medical Cobalt-60 Tele-therapy

The medical cobalt-60 tele-therapy machine at the Ramathibodi Hospital of the Mahidol University, Bangkok, Thailand is shown in Figure 3-5. It is a Theratron Model 80 Elite.

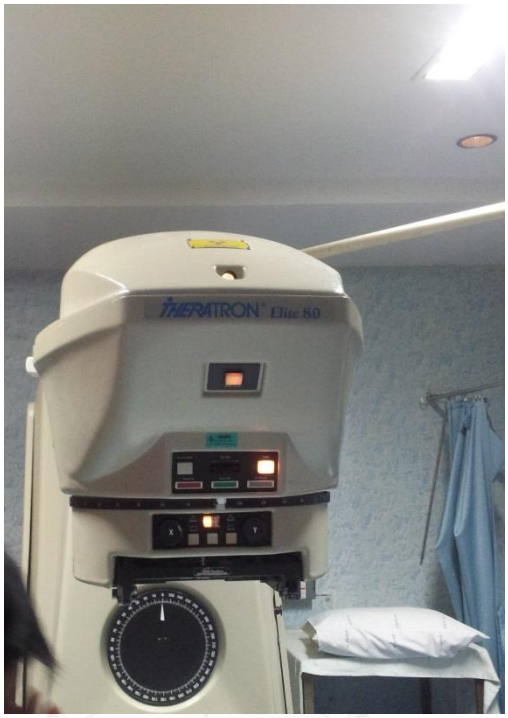

Figure 3-5 Theratron 80 Elite at the Ramathibodi Hospital

For information references and especially as a guide to a Cobalt-60 tele-therapy machine, are the schematic diagrams of the Theratron 780 shown in Figure 3-6.

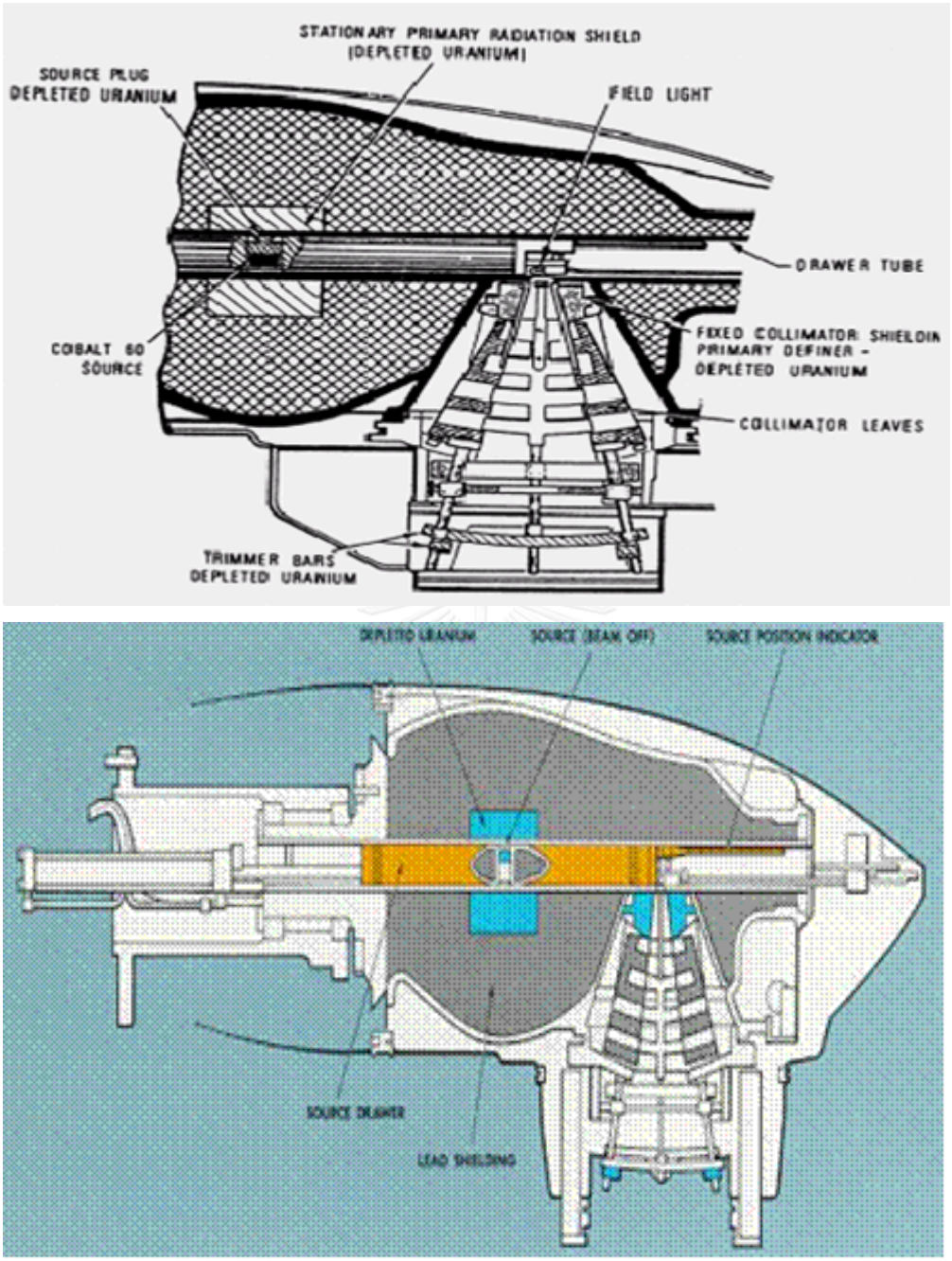

Figure 3-6 Schematic diagrams of the Theratron 780 Co-60 tele-therapy machine [21]

3.1.6 Detectors

3.1.6.1 CdTe

The Amptek XR-100T-CdTe is a high performance x-ray and gamma-ray detector, preamplifier, and cooler system using a 3 x 3 x 1 mm Cadmium Telluride (CdTe) diode detector mounted on a two-stage thermoelectric cooler. The XR-100T-CdTe is capable of detecting energies from a few keV to several hundreds of keV, with

an efficiency that peaks from 10 to 100 keV. The traditional -T option provides very good energy resolution but is limited to count rates of < 10 kcps. A resistive feedback option permits operation at even higher rates but at slightly lower resolution.

Power to the XR-100T-CdTe is best provided by the PX5. The PX5 is DC powered by an AC adapter and provides a variable Digital Pulse Shaping Amplifier (0.2 µs to 100 µs peaking time), the MCA function, and all necessary power supplies for the detector and preamplifier. The PX5 connects via USB, RS-232, or Ethernet to a PC. The XR-100T-CdTe/PX5 system ensures stable operation in less than one minute from power turn-on. The features of the XR-100T-CdTe are shown in Figure 3-7.

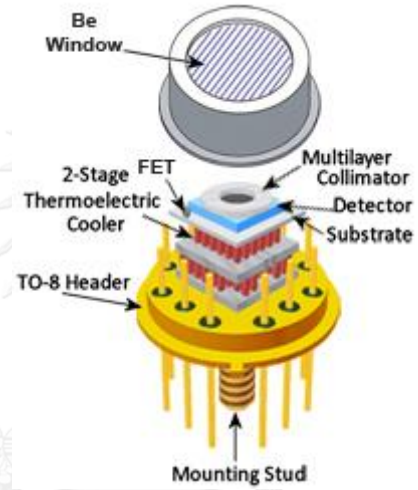

Figure 3-7 Diagram of the features of the Amptek XR-100T-CdTe [22]

The Amptek XR-100T CdTe detector and laboratory measurement setup is shown in Figure 3-8.

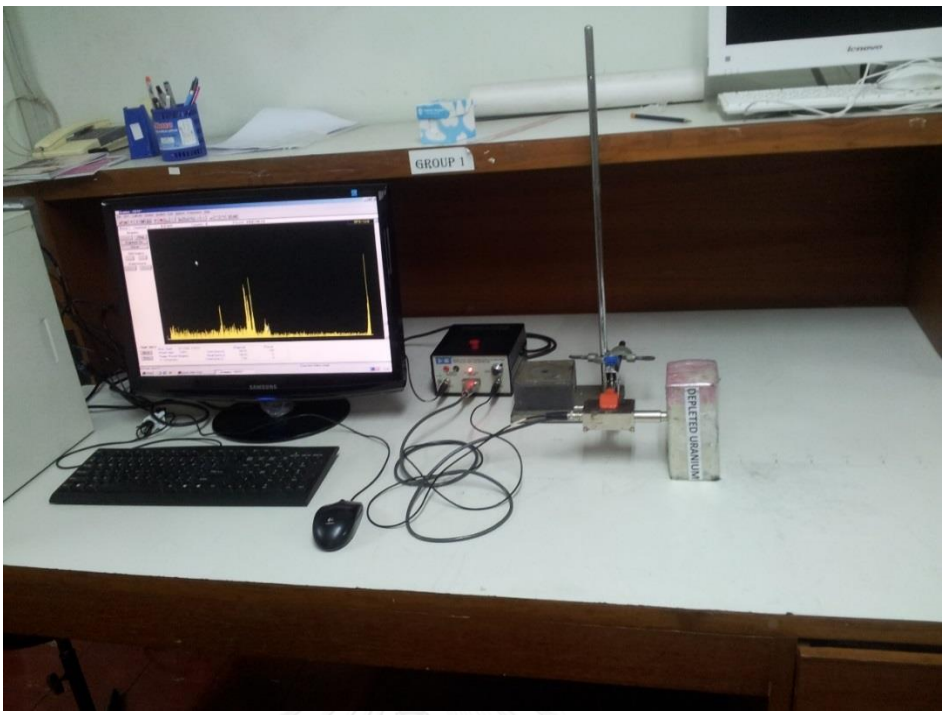

Figure 3-8 Laboratory measurement set-up for the Amptek XR-100T-CdTe

3.1.6.2 HPGe

The Ortec GLP-06165/05 is a GLP Series Planar Low Energy Photon Spectrometer (LEPS). It is equipped with a high purity germanium crystal (diameter 6 mm, length 5 mm) and a 70mm diameter endcap all mounted on a vertical cryostat/30-liter dewar model. Figures 3-9 and 3-10 show the actual Ortec GLP-06165/05 LEPS-HPGe detector and measurement set-up, respectively.

Figure 3-9 Ortec GLP-06165/05 LEPS-HPGe

Figure 3-10 Measurement set-up for the Ortec GLP-06165/05 LEPS-HPGe detector

3.2 Methodology

To accomplish the objective of this study, experiments must be performed by using the materials and equipment mentioned at the beginning of this chapter.

3.2.1 Experimental Procedures

The experimental procedures to be carried out are as follows:

1. Calibration of the detecting system

   After sufficient warm up of the equipment (electronics and computer), acquire the gamma-ray energy emissions from either of the following radionuclides, ^{238}Pu, ^{241}Am or both. In the obtained spectrum, identify the respective peaks and set as calibration peaks.
2. Acquire the gamma-ray spectrum of the natural uranium metal standard and the depleted uranium block.
3. Analyze the spectra.
4. Vary set-up and acquisition parameters such as (a) distance between sample and detector, (b) acquisition time, (c) introduction of radionuclide sources between and around sample and detector and (d) the use of different radiation shielding materials between sample and detector.
5. Analyze results, identify pattern, significant details and information in the gamma-ray spectra to establish basis and criteria in determining the presence of DU in the material.
6. Develop procedure to test the technique on industrial radiography projectors.

3.2.2 Application of technique in the field

1. Set up and perform Gamma-ray spectroscopy on Industrial radiography projectors.
2. Analyze results and compare performance of both detecting systems.
3. Optimize parameters, reevaluate technique and modify if necessary.

Chapter IV

RESULTS AND DISCUSSION

The experimental procedures and field tests were carried out as described in Chapter III, with the results and analyses to be presented in this chapter.

4.1 Gamma spectroscopy of NUMS and DU block

Gamma-ray spectroscopy was conducted on the Natural Uranium metal standard and a Depleted Uranium block by the AmpTek XR-100T-CdTe and the Ortec LEPS-HPGe, The LEPS-HPGe was able to obtain the gamma-ray spectrum of the NUMS as shown in Figure 4-1. This was acquired in 300 seconds with the NUMS placed on top of the endcap. The uranium x-ray peaks, ($U_{K\alpha's}$, $U_{K\beta's}$ and even the $U_{L's}$) have been detected and identified in the spectrum. The low gamma energies of the U-238 daughter (Th-234) have also been detected (63.3 keV and 92.3 keV). The signature peak of U-235 (185.7 keV) is also very distinguishable by the LEPS-HPGe.

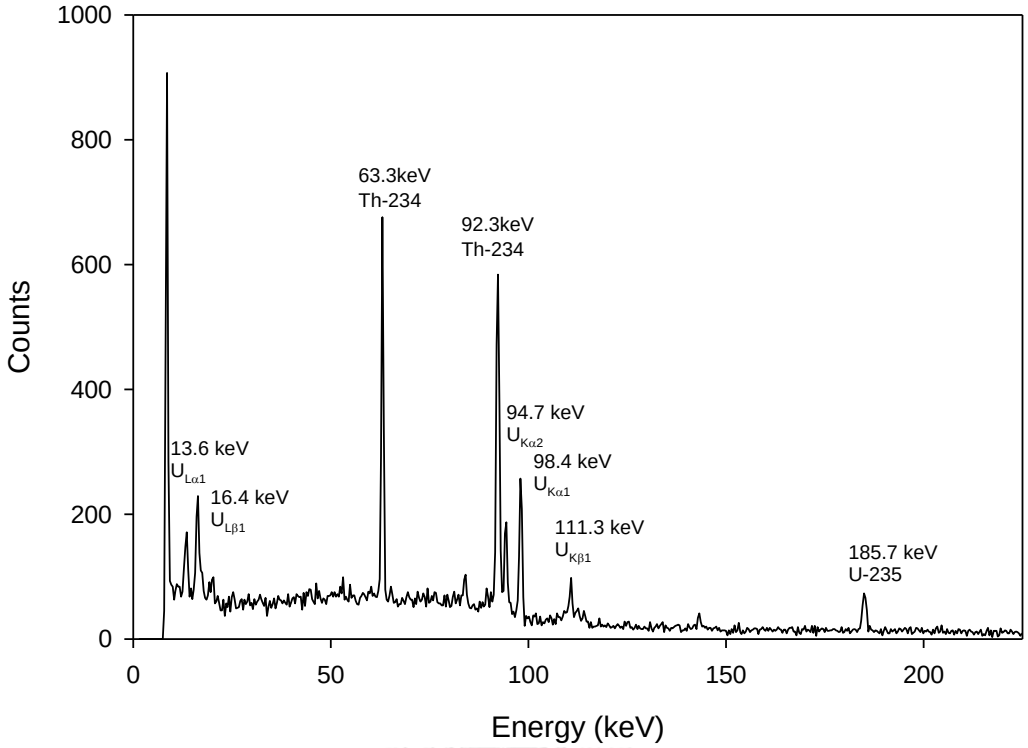

Figure 4-1 Gamma-ray spectrum of Natural uranium metal standard by the Ortec LEPS-HPGe

Figure 4-2 is the gamma-ray spectrum of the DU block by the LEPS-HPGe. The peaks observed for the NUMS can also be observed in this spectrum but with major differences. These differences are:

1. For NUMS the most intensed peak is the Th-234 gamma 63.6 keV while for the DU it is the Uranium x-ray $U_{K\alpha 1}$ = 98.4 keV.
2. The U-235 gamma-ray with 185.7 keV has decreased in the DU spectrum which is the appropriate result since it is the U-235 that is lacking or depleted in DU thus the lower counts due to lower concentration of U-235.

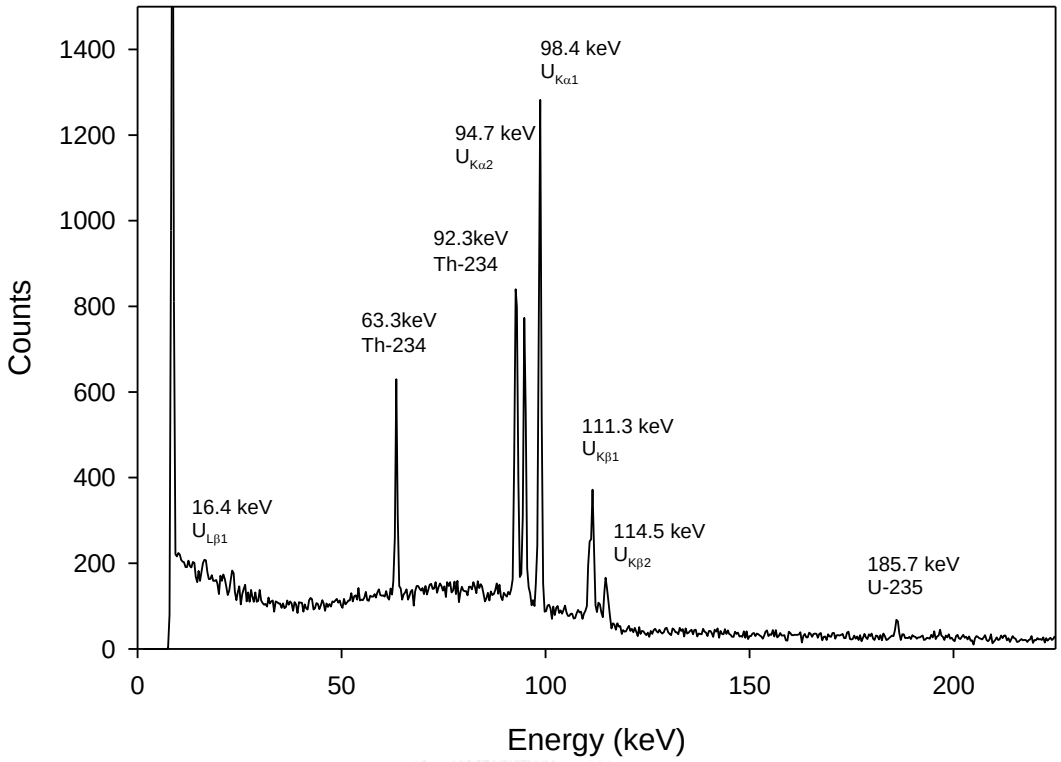

Figure 4-2 Gamma-ray spectrum of Depleted uranium block by LEPS-HPGe

The gamma-ray spectrum for the NUMS was also obtained by the CdTe detector. It can be seen in Figure 4-3 that there are several differences from the spectrum obtained by the HPGe detector. The counts are about 10 times (10x) lower and while the significant or desired peaks are still observable, the lower energy x-rays are not distinguishable. In addition, the background is not as smooth as in the HPGe spectra.

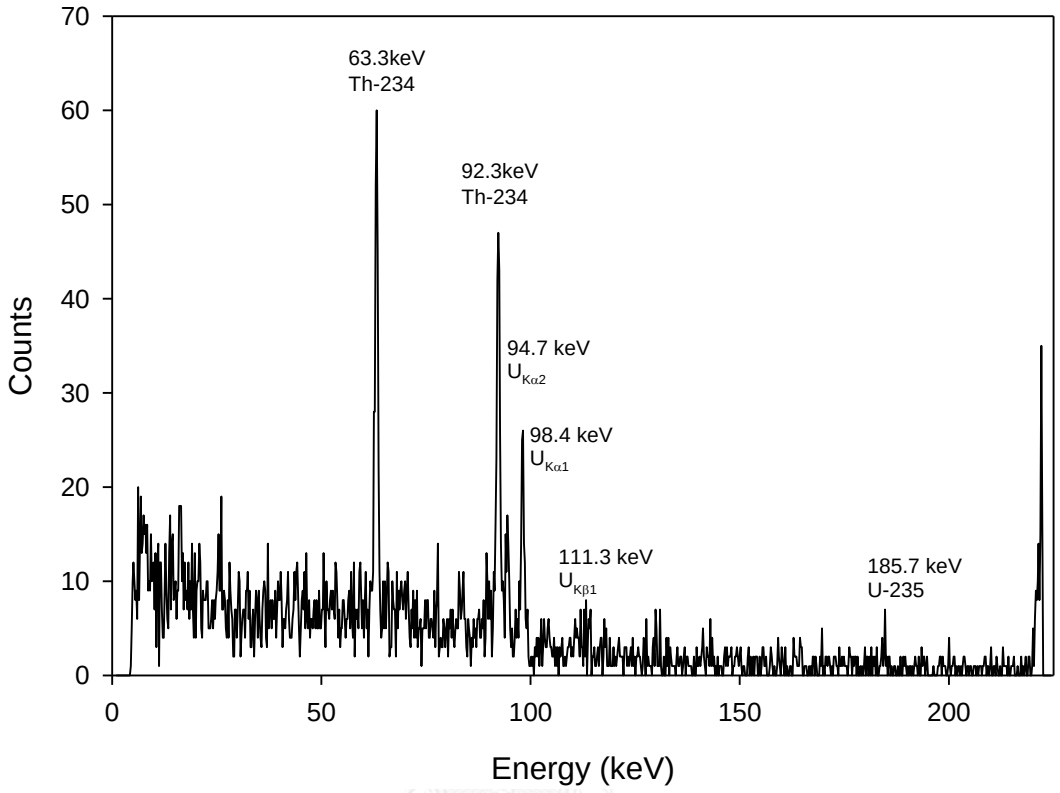

Figure 4-3 Gamma-ray spectrum of Natural Uranium metal standard by CdTe detector

For the gamma-ray spectrum of DU by the CdTe detector it is observed that the counts are also low as compared to that obtained by the HPGe. The 185.7 keV peak is barely distinguishable as seen in Figure 4-4. We can state that both CdTe and HPGe detectors can obtain the gamma spectra for each NUMS and DU but with major differences in counts and intensity.

All these observations are tabulated in Table 4-1 which summarizes the area of each significant peak from the gamma-ray spectra of the NUMS and DUB as measured by both CdTe and HPGe detectors. The ratio of the area under each peak (A*) to the area under the most intense peak (A**) is shown in the last column of the Table 4-1. These data verifies the observation that the most intense peak for the NUMS is the 63.3 keV while for the DUB it is the 98.4 keV peak. The tabulated data also shows that both the counts and area measured by the CdTe detector are 10x lower than the HPGe as previously mentioned. There is a decrease in the 185.7 keV peak as measured in the DUB compared to NUMS by the HPGe while it cannot be observed by the CdTe.

Table 4-1 Summary of the significant peaks from the gamma-ray spectrum of NUMS and DUB measured by both CdTe and HPGe detectors

Sample	Detector	Energy	Counts	A*	(A*/A**)
NUMS	HPGe	63.3	676	1131	100
		92.3	584	1121	99
		94.7	187	119	10
		98.4	257	522	46
		185.7	73	193	17
	CdTe	63.3	60	208	100
		92.3	47	157	75
		94.7	17	30	14
		98.4	26	70	34
		185.7	7	9	4
DU block	HPGe	63.3	630	887	36
		92.3	840	1048	42
		94.7	773	319	13
		98.4	1282	2493	100
		185.7	68	94	4
	CdTe	63.3	50	164	41
		92.3	83	246	61
		94.7	74	121	30
		98.4	95	403	100
		185.7	1	-	

* - Area under each peak ** – Area under the most intense peak

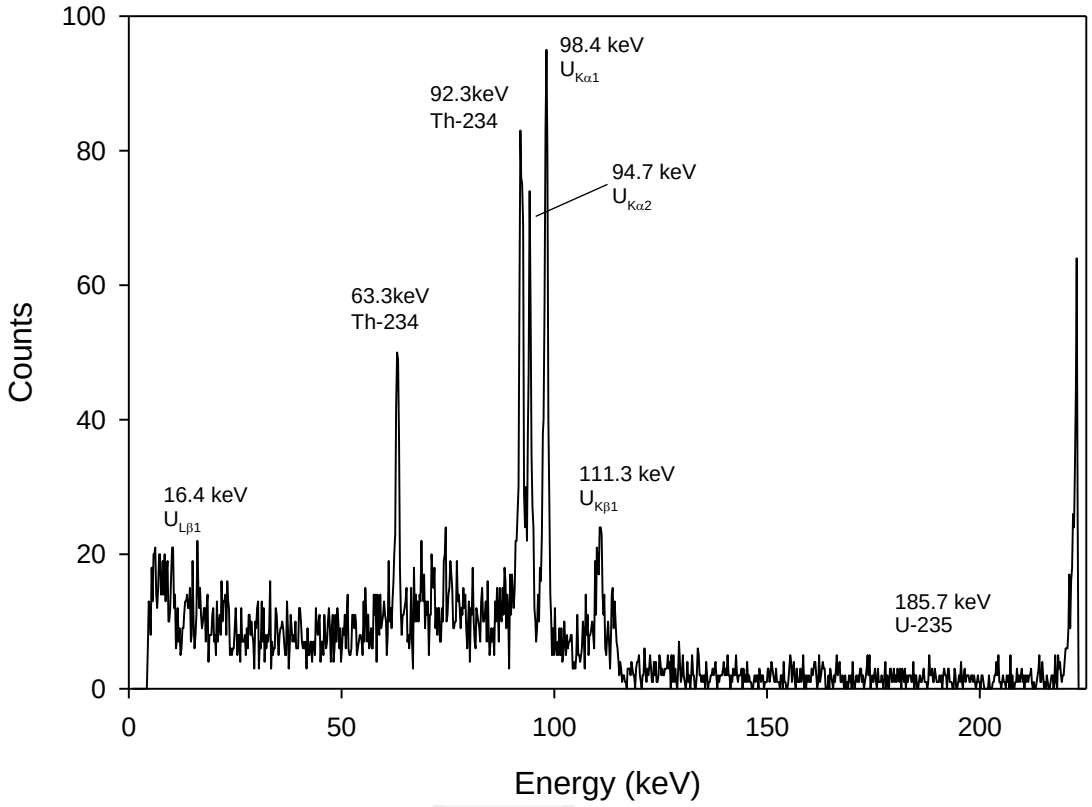

Figure 4-4 Gamma-ray spectrum of Depleted uranium by CdTe detector

Figure 4-5 is an expanded view of normalized values for DU block by both HPGe and CdTe detectors. The HPGe has sharper peaks, lower Full Width at Half Max (FWHM) values as shown in Table 4-2 and therefore better resolution.

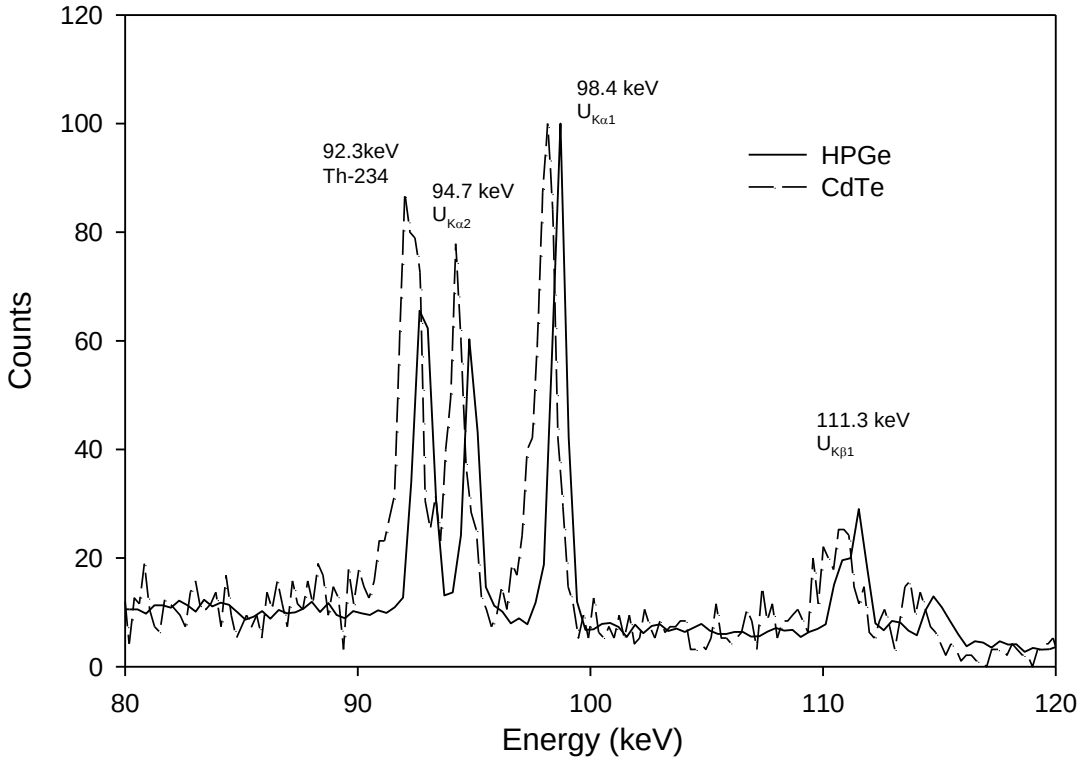

Figure 4-5 Comparison of normalized values for DU block spectra by both the HPGE and CdTe detectors

Table 4-1 Full Width Half Max (FWHM) values for HPGe and CdTe

	FWHM (keV)	
Sample	HPGe	CdTe
NUMS	0.602	0.720
DU block	0.676	0.861

4.2 Variations in parameters

The detectors were tested by varying several parameters and conditions such as changing the distance between the sample and detector and the data acquisition time. The effects of having the presence of other sources and radiation shielding materials in the set-up was also investigated.

4.2.1 Distance between sample and detector

For the HPGe detector, the distance between the sample and detector can be limited by the weight of a sample and by the geometry of the mounting set-up. Lightweight samples may be placed on top of the endcap of the HPGe. For heavier samples, a special support and mounting was placed above the detector and dewar for the samples to be mounted on as shown in Figure 4-6.

Figure 4-6 Mounting platform and support on top of LEPS-HPGe for heavy samples

In considering the Inverse Square Law, it requires the distance of the sample to the detector to be very small to obtain higher counts. For NUMS it can be placed right on top of the endcap. For the DU block and the Industrial radiography projectors, which may weigh from 10 kilograms to 60 kilograms, a mounting set-up was developed to support them. The distance between the detector and a sample placed on the mounting is around 37 mm. The gamma-ray spectra obtained for the DU block (Figure 4-2) was measured at this distance. If the sample is to be measured from the second level,the distance will be around 455 mm. The gamma-ray spectrum for the DU block at this distance is shown in Figure 4-7.

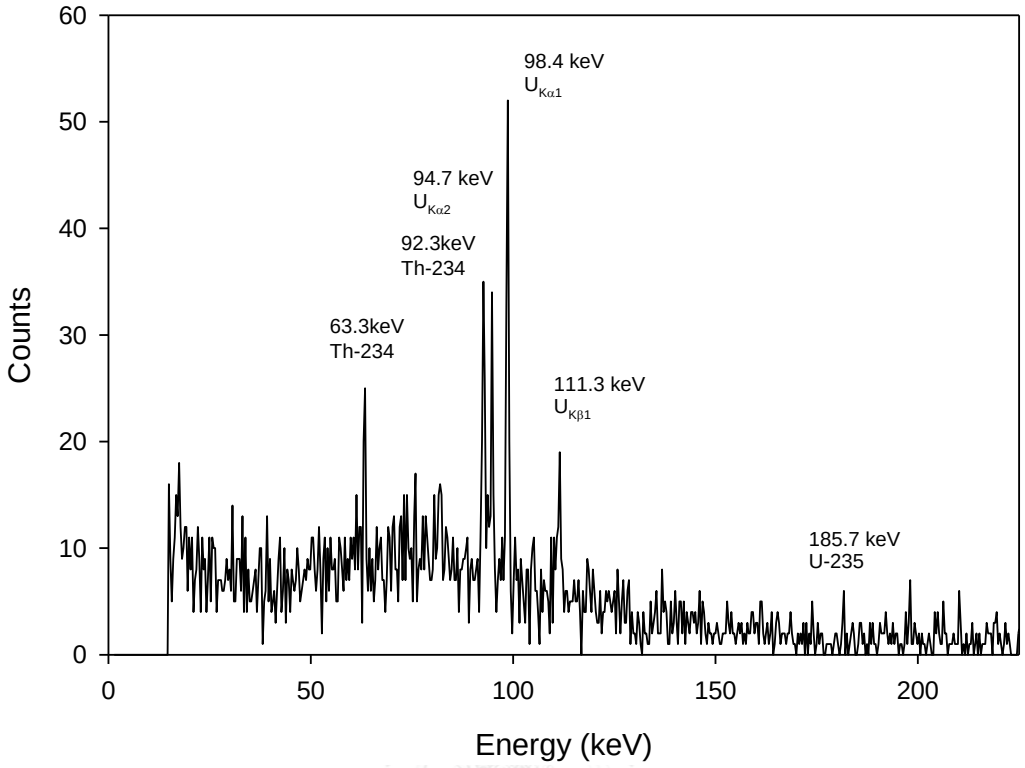

Figure 4-7 Gamma-ray spectrum of Depleted uranium block at 455 mm above HPGe

Compared to Figure 4-2 there is a drastic drop in the counts and because of the low counts, the background is not smooth. However the major peaks are still obtained and distinguishable.

For the CdTe detector, with the use of clamps or just a flat surface as shown in Figure 4-8, the detector and sample can be placed right next to each other, so that they can be in contact or separated easily.

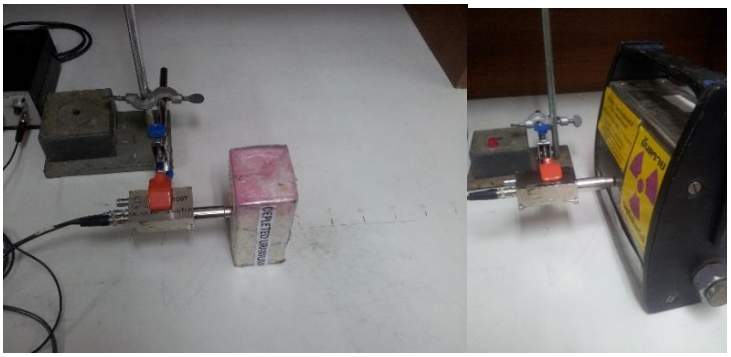

Figure 4-8 Measurement set up of CdTe detector and sample

As with the Inverse Square Law, there is only a certain range between the CdTe and sample before the peaks disappear. Figures 4-9 and 4-10 show the gamma ray spectra of the NUMS and DU block respectively, at various distances from the CdTe detector. In both graphs the relative counts decrease as the distance between sample and detector is increased. Table 4-3 shows the tabulated peak areas for the significant peaks from the gamma-ray spectra of NUMS and DUB at various distances measured by CdTe detector. The decrease in the peak area is very evident for natural uranium in particular at 5cm away. For depleted uranium the decrease is not so significant such that the peaks are still very much distinguishable at 5cm.

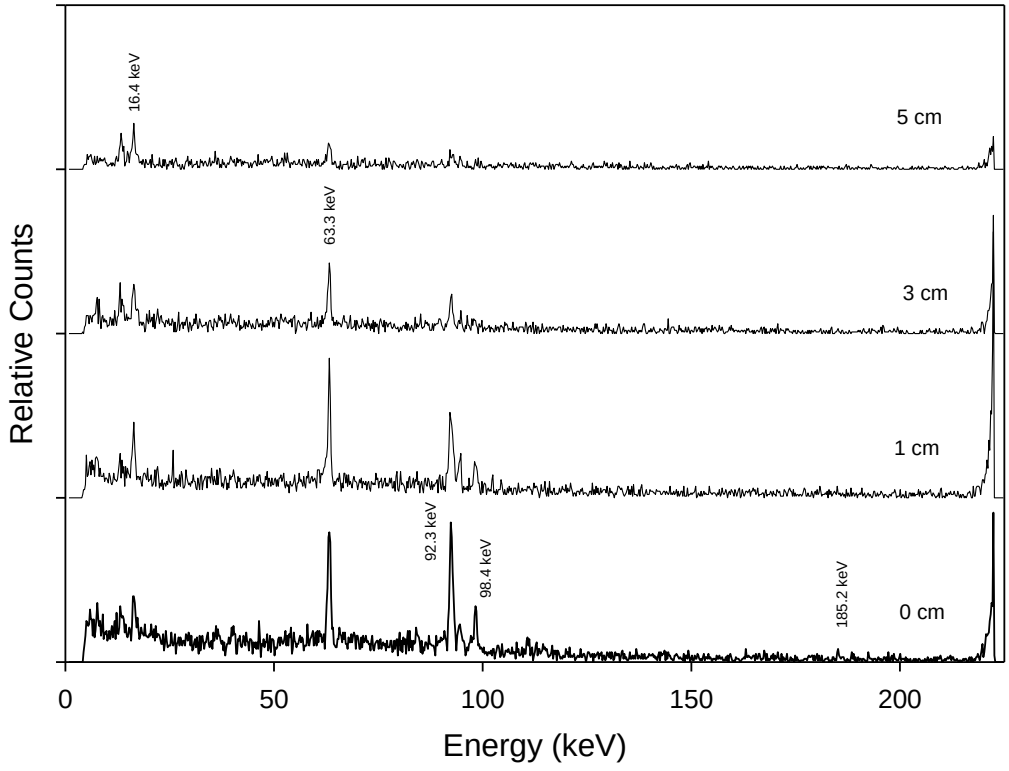

Figure 4-9 Gamma-ray spectra of Natural Uranium Metal Standard at various distance from the CdTe detector

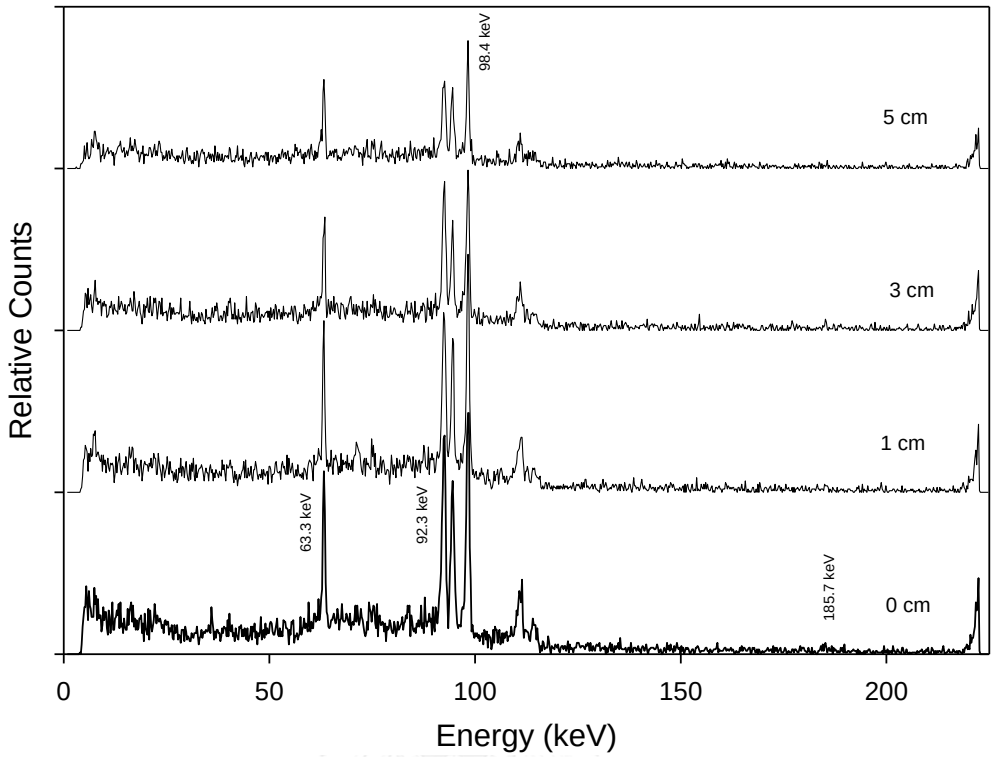

Figure 4-10 Gamma-ray spectrum of Depleted uranium block at various distances from CdTe detector

Table 4-3 Results of peak areas for the significant peaks from the gamma-ray spectrum of NUMS and DUB at various distances measured by CdTe

Energies (keV)			63.3	92.3	94.7	98.4	185.7
Sample	Detector	Distance (cm)	Peak Areas				
NUMS	CdTe	0	288	239	44	126	10
		1	237	200	29	64	0
		3	134	81	23	14	0
		5	77	37	14	23	0

Table 4-3 (continue)

Energies (keV)			63.3	92.3	94.7	98.4	185.7
Sample	Detector	Distance (cm)	Peak Areas				
DUB	CdTe	0	295	303	262	582	2
		1	237	279	170	517	10
		3	162	223	100	365	12
		5	169	126	90	251	9
		9	12	46	24	78	0
		10	73	55	30	67	0
		11.5	27	37	19	31	0
		14	22	25	29	28	0
		15	23	43	16	53	0
		20	3	21	5	15	0

The DU block was moved farther away from the CdTe detector and the results can be seen in Figure 4-11. The peaks are smaller but still distinguishable at 20 cm away. It is interesting to note the change in the peak ratios for the DU block at 10 cm away as shown in Table 4-3. The Th-234 gamma-rays are now higher than the Uranium x-rays. From Figure 4-12 we can note that this change only began at the 10 cm distance.

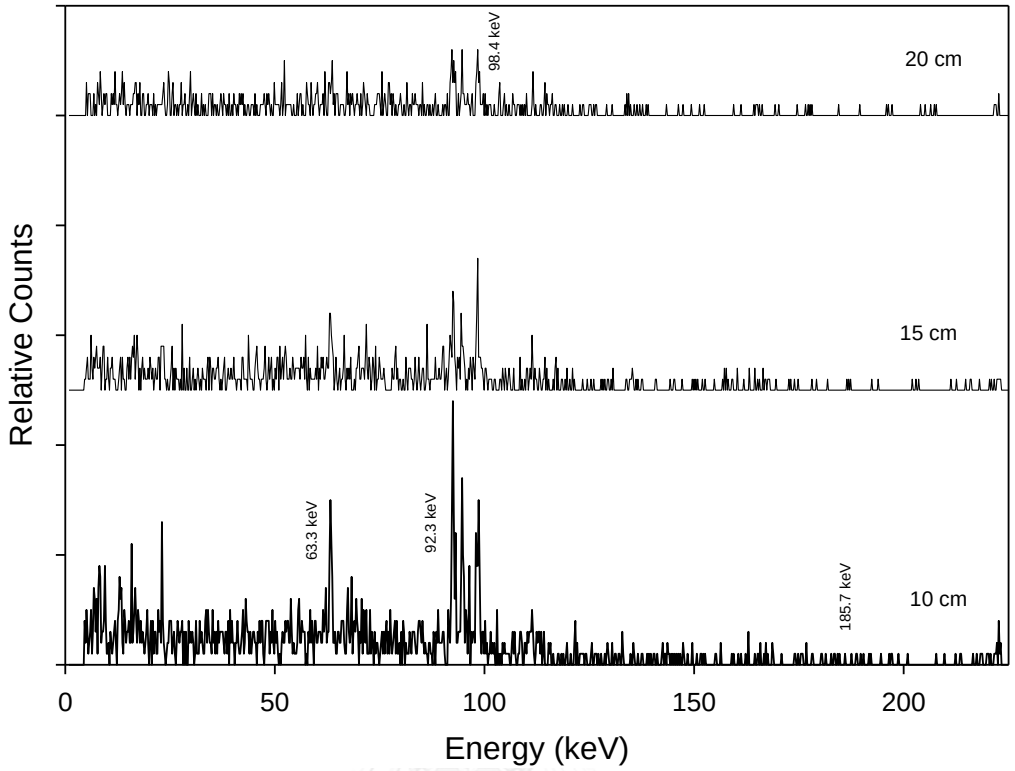

Figure 4-11 Gamma-ray spectra of depleted uranium block at further distance from the CdTe detector

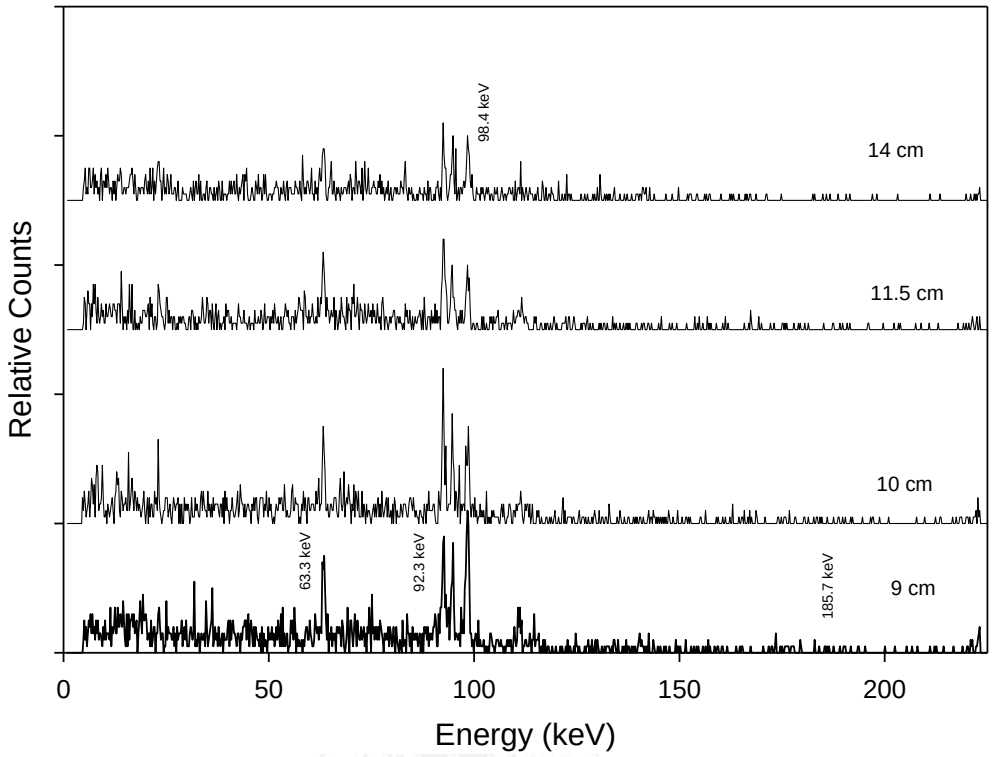

Figure 4-12 Gamma-ray spectra of depleted uranium block between 9 cm to 14 cm away from the CdTe detector

The proper explanation for this is that the weaker x-rays cannot reach the detector anymore starting at this distance of about 10 cm.

4.2.2 Data Acquisition Time

The parameter of acquisition time was varied to detemine the optimum time for these detectors to obtain a respectable spectrum. Selecting the proper acquistion time depends on what is more important for the user. (1) Will you choose the quickest time over a better resolution or shape of the spectra obtained? (2) Does longer acquistion time really give higher counts or better form of the peaks and spectrum? (3) The actual use or application in the field, for we do not want to be an inconvenience to the licensee and the inspector and cause any delay in their work. Both detectors performed gamma spectroscopy on DU block at acquistion times ranging from 10 seconds to 600 seconds. From Figure 4-13 we can see that at only 10 seconds, the HPGe was able to obtain a spectrum of the DU block. The counts are satisfactory but the peaks are still developing.

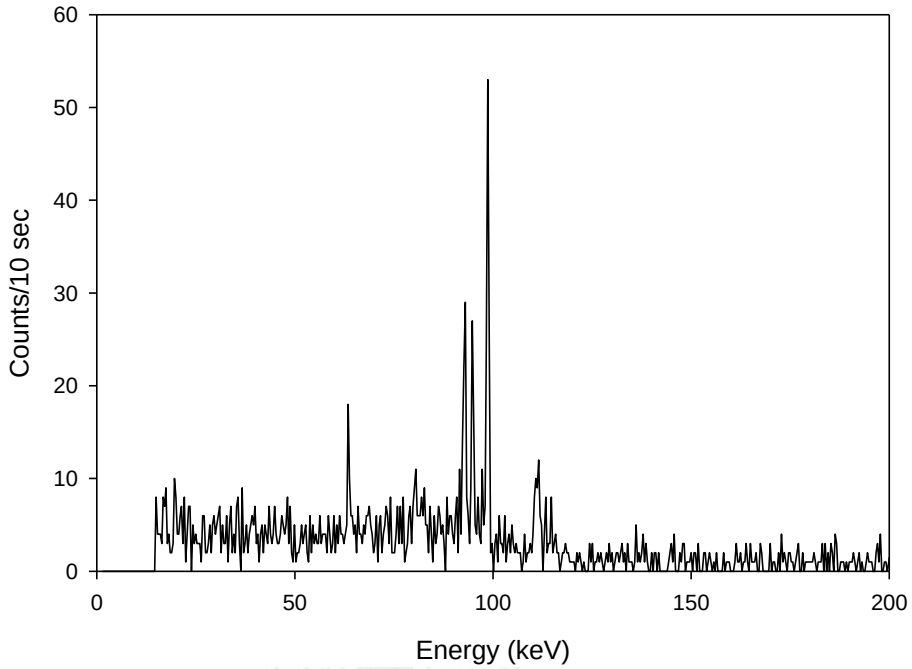

Figure 4-13 Gamma spectrum of DU Block 37 mm above the LEPS-HPGe for 10 seconds

It can be observed in Figure 4-14 that at only 30 seconds, the HPGe was able to obtain a spectrum where the counts and form are more than comparable to the CdTe at 600 seconds. For the CdTe detector, it can also give a spectrum after 10 seconds as shown in Figure 4-15 however it is not as good as that of HPGe detector. The counts are less than 10 so the statistics are not reliable even if the most intensed U x-ray peak is visible.

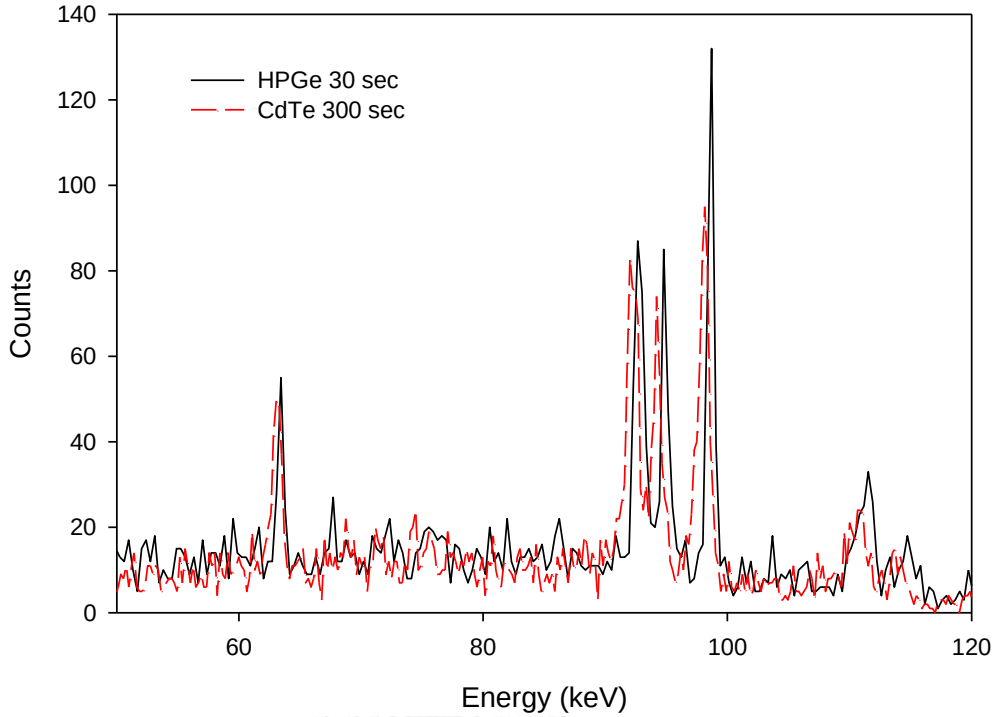

Figure 4-14 Gamma spectra of DU block by HPGe at 10sec and by CdTe at 300sec

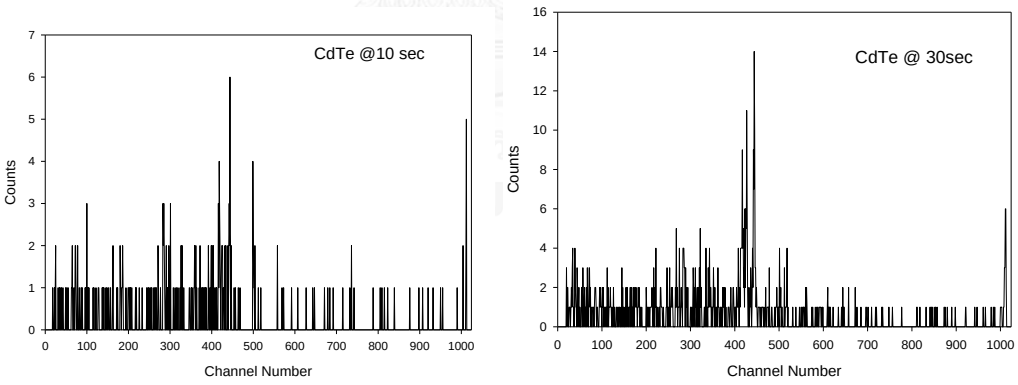

Figure 4-15 Gamma spectrum of DU block by CdTe at 10 and 30 seconds

Figures 4-16 and 4-17 and Tables 4-4 and 4-5 can be used to determine at which counting time is more suitable for each detector to obtain a well resolved spectrum. For the HPGe, there is the luxury of choosing at fast as 60 to 120 seconds due to their acceptable peaks. The time of 300 seconds though would be the most suitable choice, where it is not so long a time and the results give more than manageable counts. As for the CdTe, the 300 seconds acquisition time is enough but other factors may force the

user to use longer time for example if the distance between sample and detector is significantly large.

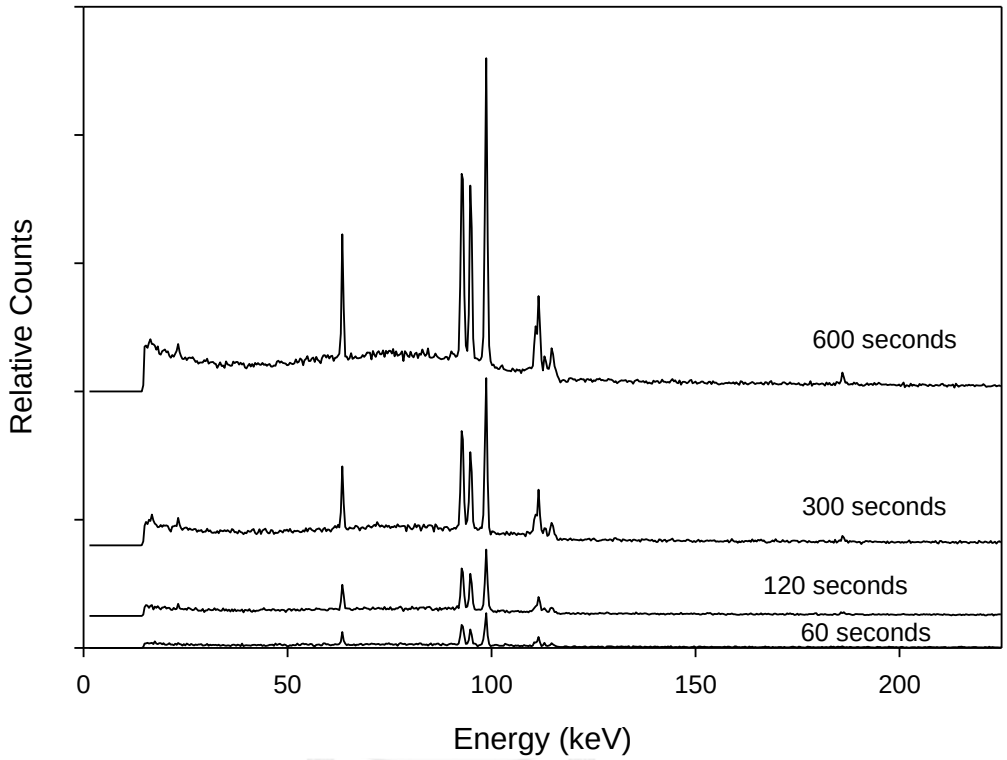

Figure 4-16 Gamma spectra of Depleted Uranium block by HPGe at various acquisition times

Table 4-4 Peak Areas for distinct energies in Gamma-ray spectrum of DUB at different acquisition times by HPGe

Energy (keV)	63.3		92.3		94.7		98.4		185.7	
Acquisition Time (sec)	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area
10	18	23	29	38	27	27	53	91	2	1
30	55	69	87	115	85	80	132	225	7	6
60	124	168	179	252	144	132	271	528	12	9
120	242	350	370	554	328	395	517	965	28	38
180	388	539	534	701	419	476	750	1402	43	67
300	615	790	891	1465	727	805	1303	2365	74	89
600	1225	1568	1697	2902	1605	1846	2598	4756	146	249

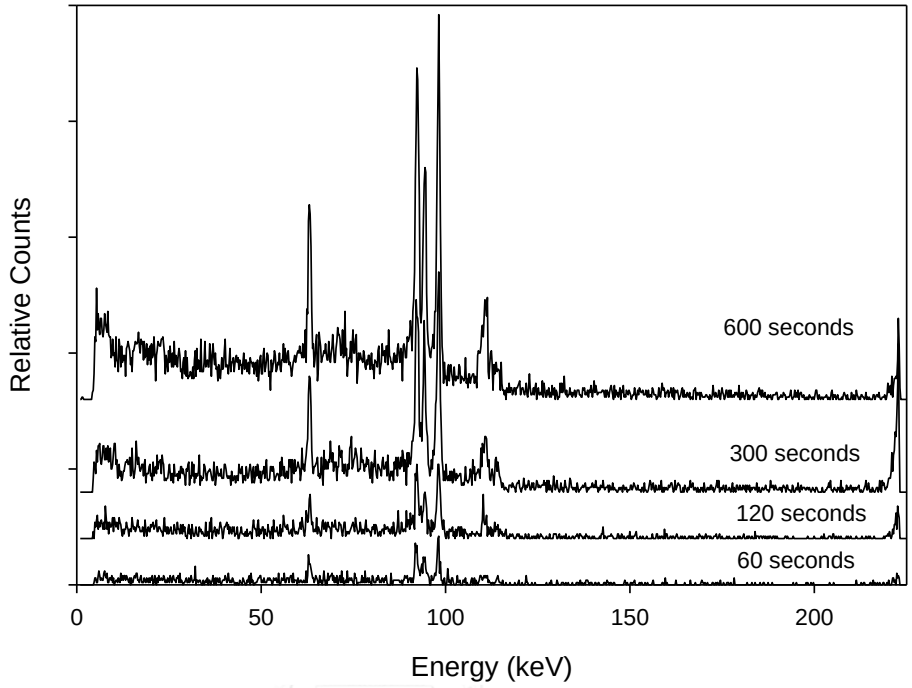

Figure 4-17 Gamma spectra of Depleted Uranium block by CdTe at various acquisition times

Table 4-5 Peak Areas for distinct energies in Gamma-ray spectrum of DUB at different acquisition times by CdTe

Energy (keV)	63.3		92.3		94.7		98.4		185.7	
Acquisition Time (sec)	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area	Counts	Peak Area
10	3	9	4	10	1	0	6	14	0	0
30	4	12	9	20	11	22	14	42	1	1
60	13	40	17	50	12	34	21	63	1	2
120	19	68	32	84	20	46	32	153	1	1
180	33	120	58	151	37	71	52	221	4	3
300	50	170	83	275	74	177	95	427	1	0
600	84	281	143	513	100	252	166	671	1	0

4.2.3 Presence of other radionuclide sources

Depleted uranium is used as shielding material in containers with highly radioactive sources such as Iridium-192, Selenium-75 and Cobalt-60. Of these three sources the laboratory only has Cobalt-60 and its activity is only in the milli and micro Curies range compared to the hundreds of Curies inside industrial radiography projectors. Tests were still carried out using the cobalt-60 mentioned in Table 3-1 and the results can be seen in Figures 4-18, 4-19 and 4-20. There are no Cobalt-60 peaks in this region that will interfere with the U x-rays and Th-234 gammas. The effect is just an increase in the background and the peaks of NUMS and DU riding on the background of the Co-60.

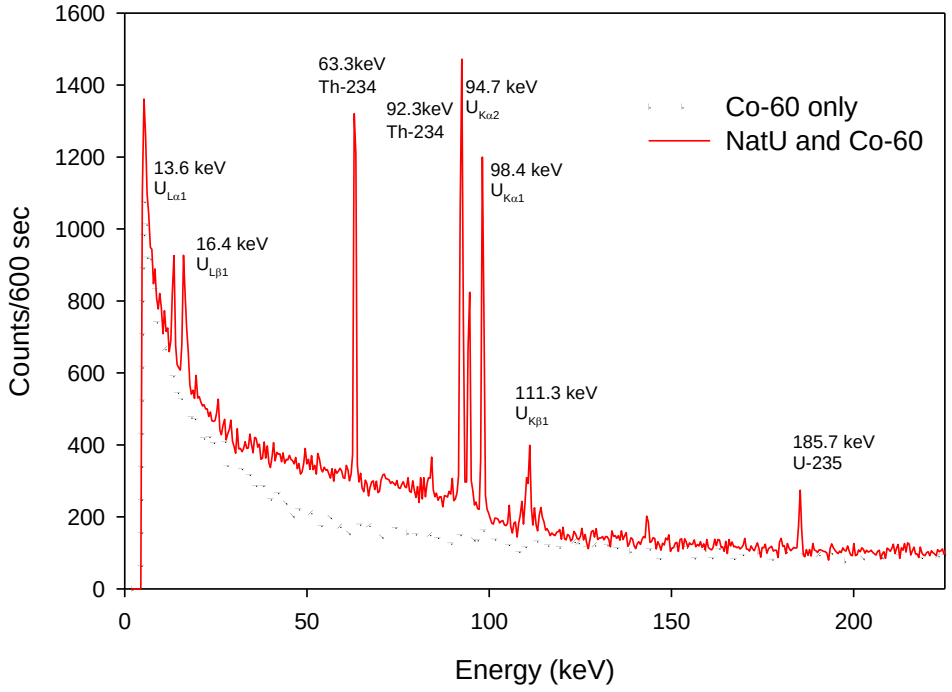

Figure 4-18 Gamma spectrum of Natural Uranium Metal Standard in the presence of a cobalt-60 source measured by HPGe

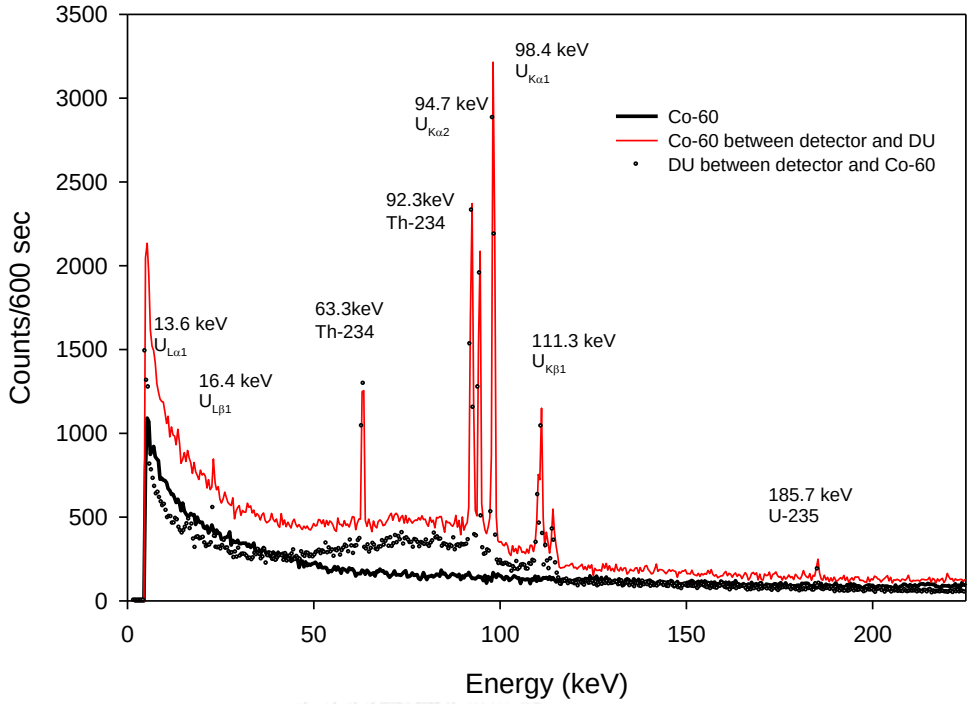

Figure 4-19 Gamma spectrum of DU block and cobalt-60 source by HPGe

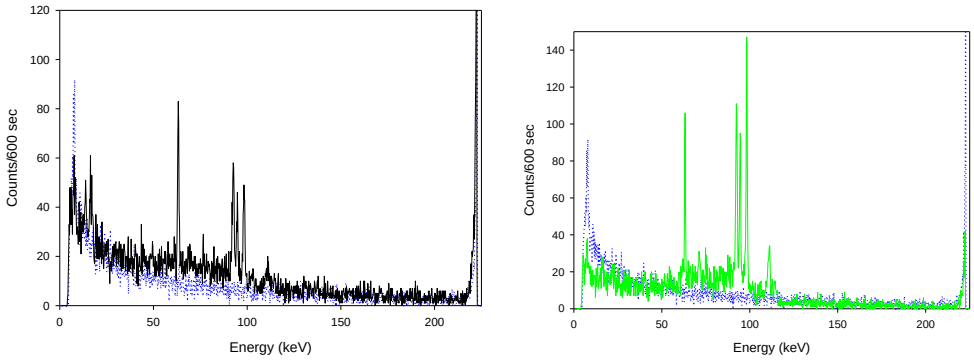

Figure 4-20 Gamma spectra of NUMS and DU block in presence of Co-60 measured by CdTe

4.2.4 Presence of Radiation Shielding materials

Tests were done with known shielding materials such as Aluminum, Iron and Lead. As expected the DU gamma-rays passed through the Al and Fe sheets but many of its low energy x-rays were attenuated thus the huge decrease in counts as shown in Table 4-6. For the lead shielding, there were different results for the HPGe and CdTe.

After 5 mm lead shielding, the uranium x-rays and Th-234 gamma-rays were not detected by the CdTe but for the HPGe, even with a 20 mm lead shielding these same peaks can still be detected by the HPGe as shown in Figures 4-21 and 4-22. This is possible because of the volume of the HPGe detector is way bigger and in particular the whole lead shielding thickness is bigger than the CdTe itself. This brings into light the Medical Cobalt-60 teletherapy which has very thick Lead shielding around the DU as was shown in the Figure 3-5.

Table 4-6 Peaks Areas of significant peaks from the gamma-ray spectrum of DUB with different shielding materials measured by both HPGe and CdTe detectors.

Energies (keV)			63.3	92.3	94.7	98.4	185.7
Detector	Sample	Shielding Material and thickness	Peak Areas				
CdTe	DUB		313	418	292	728	1
		Al 5mm	113	189	135	309	0
		Fe 5mm	40	104	98	211	0
		Pb 5mm	0	0	0	0	0
Hagen	DUB		790	1013	311	2382	108
		Al 5mm	459	679	322	1402	38
		Fe 5mm	65	265	278	670	15
		Pb 1mm	64	127	126	260	8
		Pb 5mm	55	104	66	169	15
		Pb 20mm	6	56	36	83	0

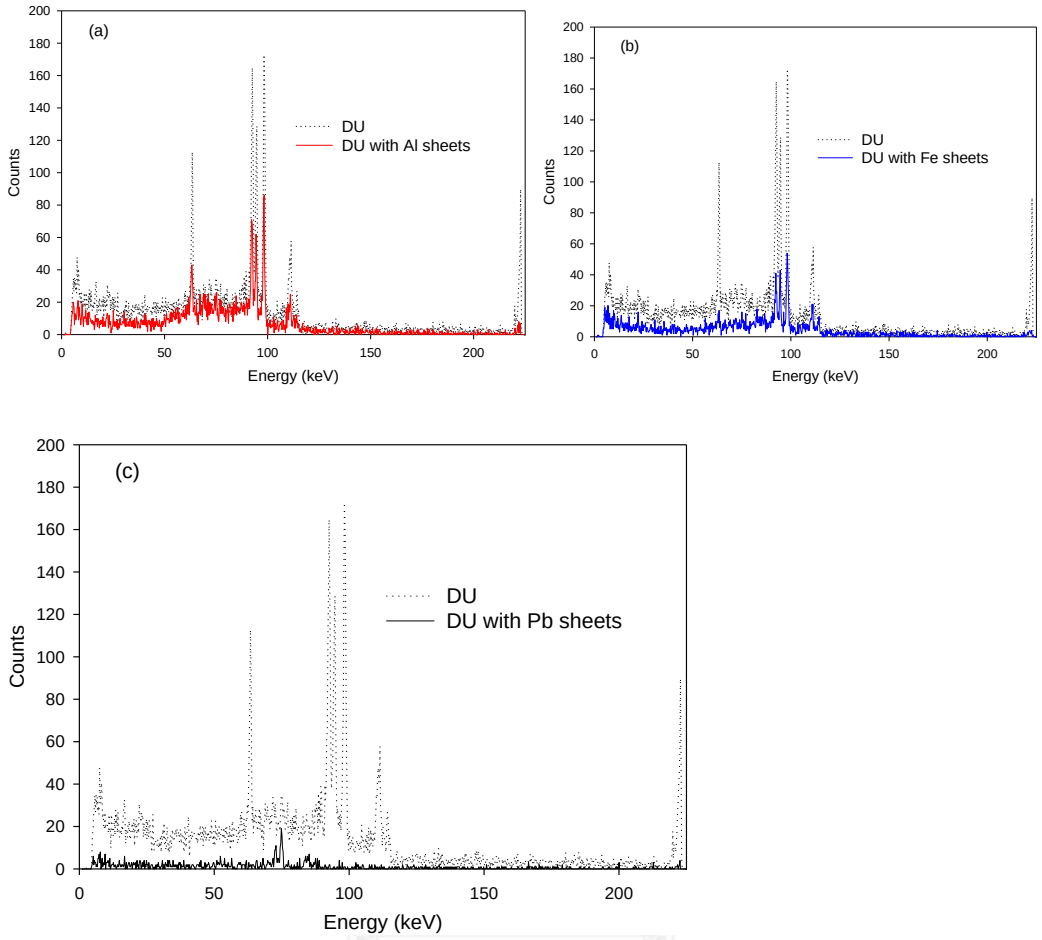

Figure 4-21 Gamma spectra of DU with (a) Al sheets, (b) Fe sheets and (c) Pb sheets by CdTe

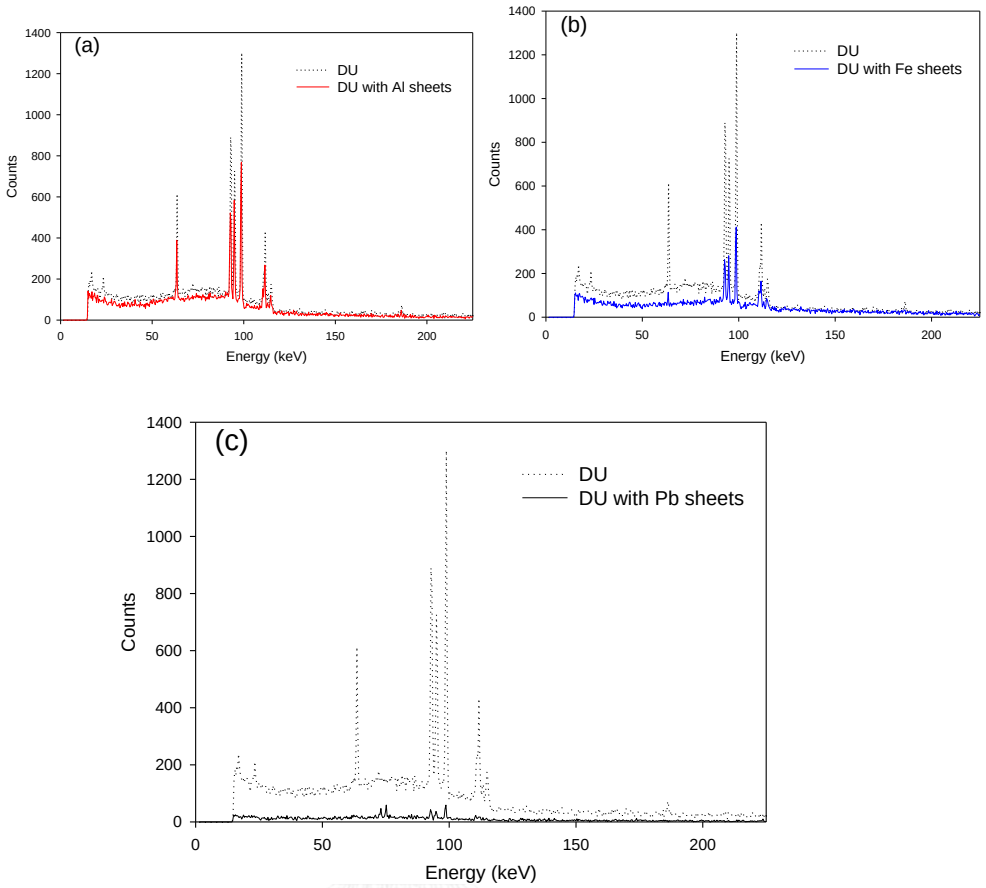

Figure 4-22 Gamma spectra of DU with (a) Al sheets, (b) Fe sheets and (c) Pb sheets by HPGe

4.3 Application of technique in the field

The Laboratory has a Sentinel Model 880 Delta radiography projector (Serial Number D1229) for which we were able to test using both the CdTe and HPGe detectors. Figure 4-23 shows the gamma-ray spectra of a Sentinel Model 880 Delta radiography projector with no source inside by both the CdTe and HPGe detectors. The HPGe spectrum has better resolution and higher counts but both spectra exhibit the same gamma-ray spectrum pattern as that of the DU block (dotted line). The spectra supports that the shielding for this radiography projector is of DU.

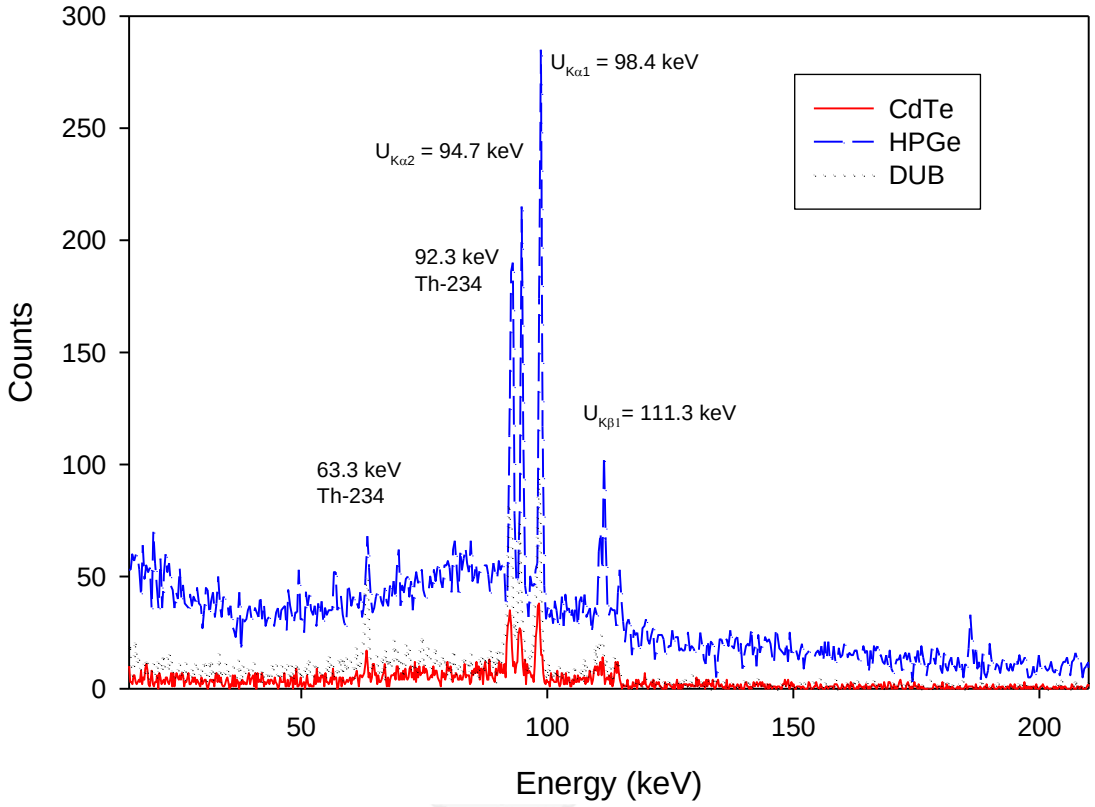

Figure 4-23 Gamma spectra of Sentinel Model 880 Delta radiography projector with no source inside by CdTe and HPGe detector

The Laboratory borrowed a Sentinel Model 660B (Serial Number B3132) radiography projector from the Thai Nondestructive Testing Public Company Limited. A similar spectra can be observed in Figure 4-24 for this Sentinel Model 660B radiography projector which contains an Iridium-192 source whose activity is about 2 Curies (Ci). The background counts can be observed to be higher because of the presence of the Iridium-192 source but all the peaks for a depleted uranium are present thus verifying that the shielding used for this projector is DU.

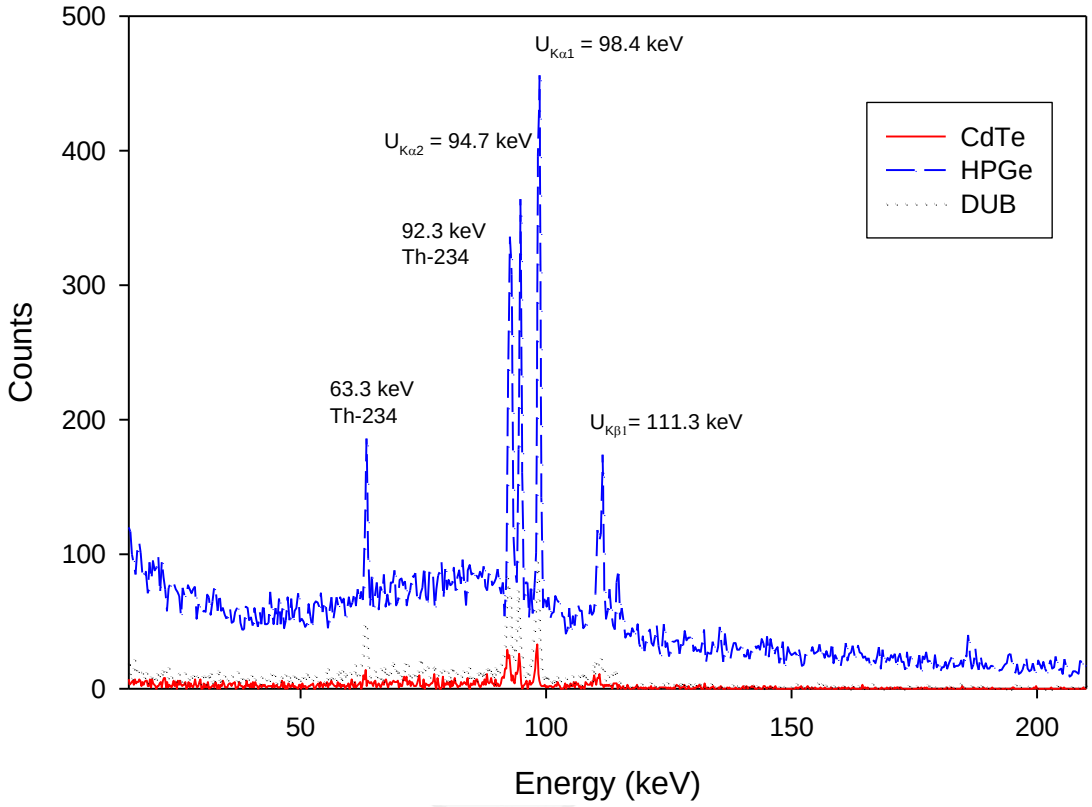

Figure 4-246 Gamma-ray spectra of Sentinel Model 660B radiography projector with 2 Curies of Iridium-192 inside, obtained by CdTe and HPGe detectors

Field tests were done on a Sentinel Model 880 Delta radiography projector at the NDT Division of the Nuclear Technology Service Center of the Thailand Institute of Nuclear Technology (TINT). This particular radiography projector contained Iridium-192 source whose activity was about 40 Curies. This was measured on site with only the CdTe detector system. The HPGe system with its 30 kilo dewar and mounting support was deemed difficult to bring to the field site. The gamma-ray spectrum shown in Figure 4-25 exhibits the (U, Th) x-rays riding on the high background due to the 40 Ci activity of the Ir-192 source. The 63.3 keV Th photon peak is hard to distinguish but given the presence of the other peaks and comparing it with the DUB gamma-ray spectrum, we can deduct that DU is the shielding used for this radiography projector.

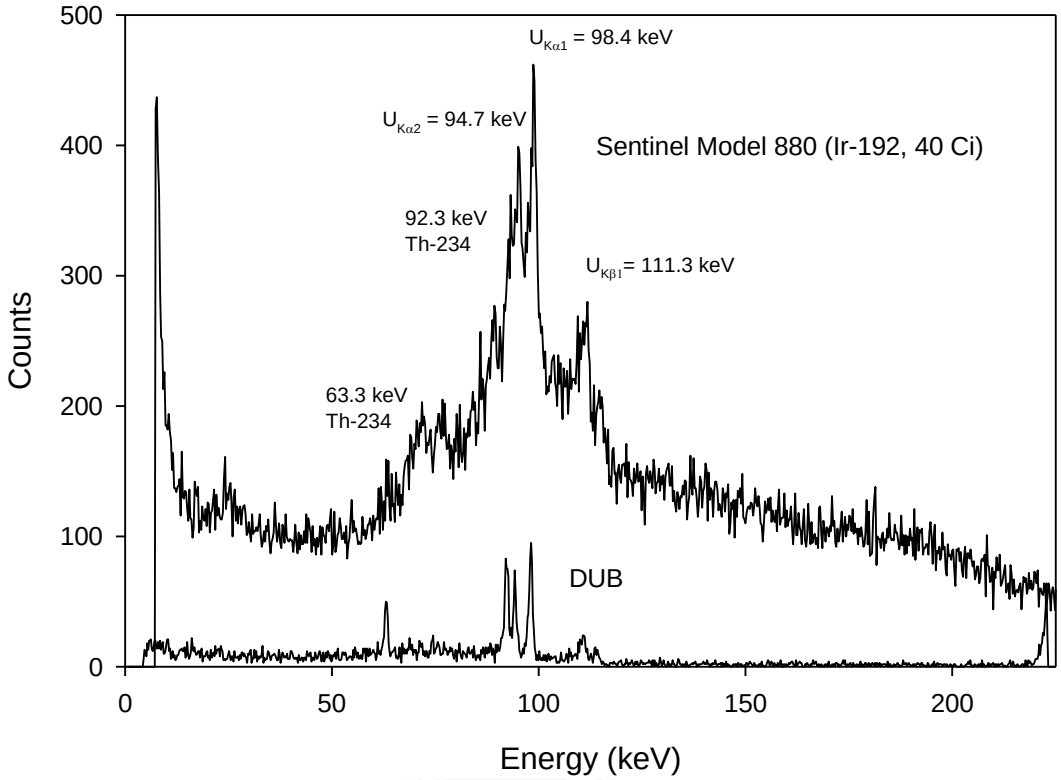

Figure 4-25 Gamma spectrum of a Sentinel Model 880 Delta radiography projector with a 40 Ci Iridium-192 source inside, by CdTe detector

A field test was done at the Thai Nondestructive Testing Public Company Limited where we measured one Iridium-192 projector on their site. This was also a Sentinel Model 880 Delta containing an Ir-192 source whose activity was about 85.88 Ci from an original activity of 98.86 Ci measured September 21, 2015. The gamma-ray spectrum shown in Figure 4-26 exhibits the (U, Th) x-rays riding on the high background due to the 85.88 Ci activity of the Ir-192 source. The 63.3 keV Th photon peak is hard to distinguish but given the presence of the other peaks and comparing it with the DUB gamma-ray spectrum, we can deduct that DU is the shielding used for this radiography projector.

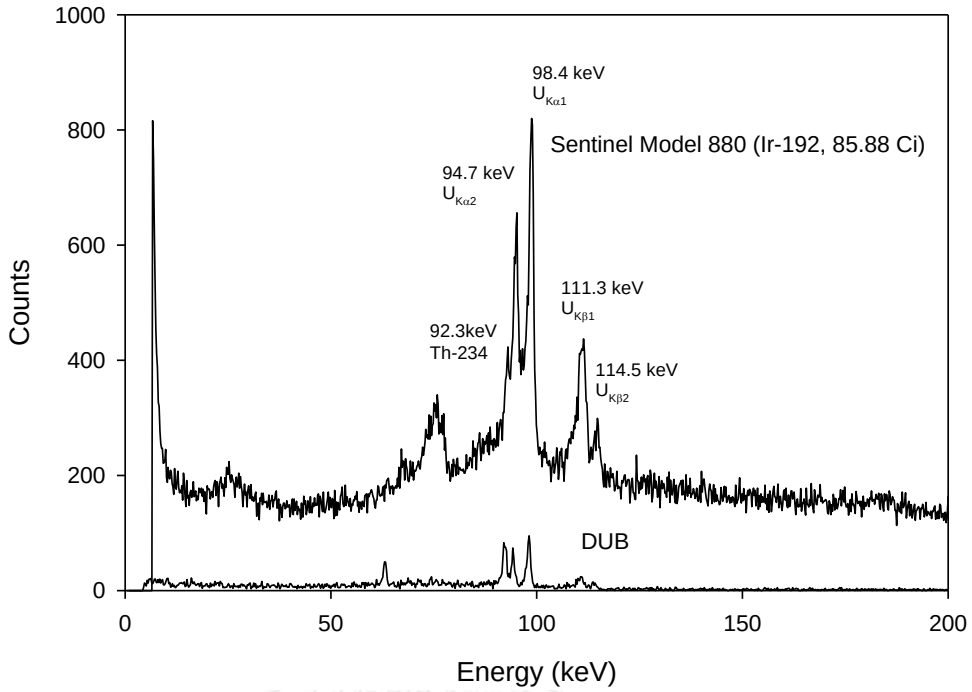

Figure 4-26 Gamma spectrum of a Sentinel Model 880 Delta radiography projector with an 85.88 Ci Iridium-192 source inside, by CdTe detector

A Sentinel Model 680B Cobalt-60 radiography projector was available for measurements during the field test at TINT. The gamma spectrum for this radiography projector taken by the CdTe is shown in Figure 4-27.

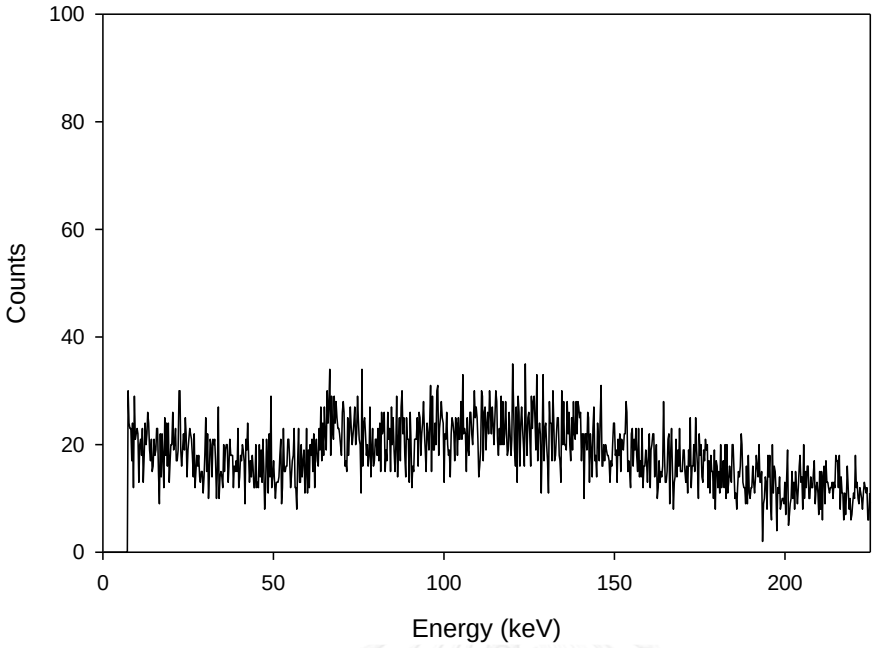

Figure 4-27 Gamma spectrum of Sentinel Model 680B Cobalt-60 radiography projector at the NDT Division of the Nuclear Service Center of TINT

It can be observed that none of the U x-rays nor Th-234 gammas is detected by the CdTe. The spectrum is simply background counts emitting possibly from Cobalt-60 source. The CdTe was not able to detect the DU because of the thick material and casing surrounding the DU and Cobalt-60 source.

We were scheduled to measure the medical Cobalt-60 tele-therapy machine at Ramathibodi Hospital but upon our arrival we were told that the shielding used was tungsten. We deemed it unnecessary to inspect the machine any longer. We tried to look for other hospitals that may have Cobalt-60 tele-therapy machines with DU shielding but could not find one. We could assume based on the diagrams of Figure 3-6 and the results from the Sentinel Model 680B Cobalt-60 radiography projector that we will not be able to detect or obtain a gamma-ray spectrum of DU because of the thick material and Lead shielding surrounding the DU shielding.

4.4 Age determination of depleted uranium

Partial Decay schemes of the naturally occurring uranium isotopes are shown in Figure 4-28. Symbols above the arrows indicate the nature of the radioactive decay. Times above the boxes indicate half-lives for decay. In all cases, the decay chains are stopped at the first long-lived (thousands of years) isotope. The significance of this is that, for a sample of processed uranium (such as DU), only the decays shown would contribute to the detectable activity. Further build-up of other daughter nuclei only occurs in very old (tens or hundreds of thousands of years) uranium samples, such as in uranium ore. This is shown graphically for the U-238 decay chain in Figure 4-29; the short-lived daughters come into equilibrium with the parent after about four half-lives. For U-235, this means that Th-231 comes into equilibrium with the parent within about four days.

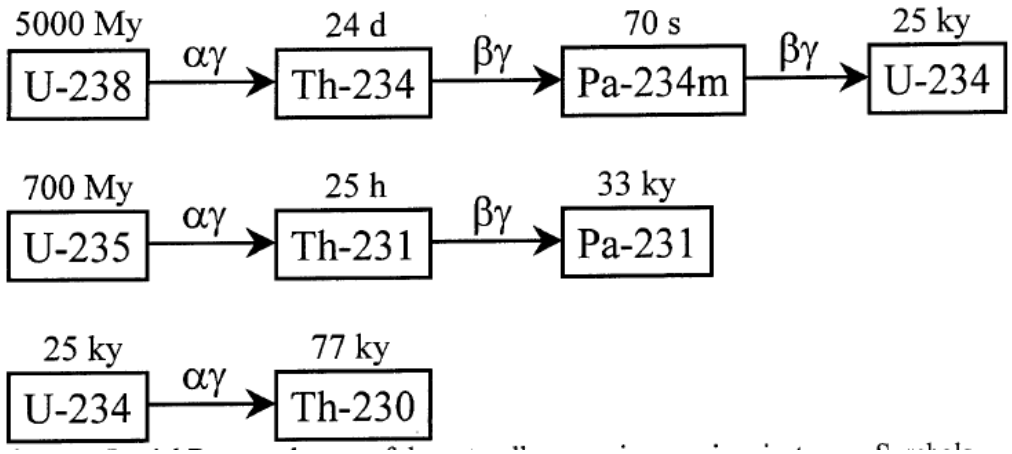

Figure 4-28 Partial Decay schemes of the naturally occurring uranium isotopes[23]

We can observe in Figure 4-29 how the activities of the short-lived daughters are essentially equal to that of the parent after 100 days (approximately 4 half-lives), but that the activity of the long-lived daughter remains very small over the same period. In such a short period we can state that secular equilibrium has been attained therefore our technique will not be able to determine the age of a depleted uranium since after a period of 100 days we will not be able to detect any more changes.

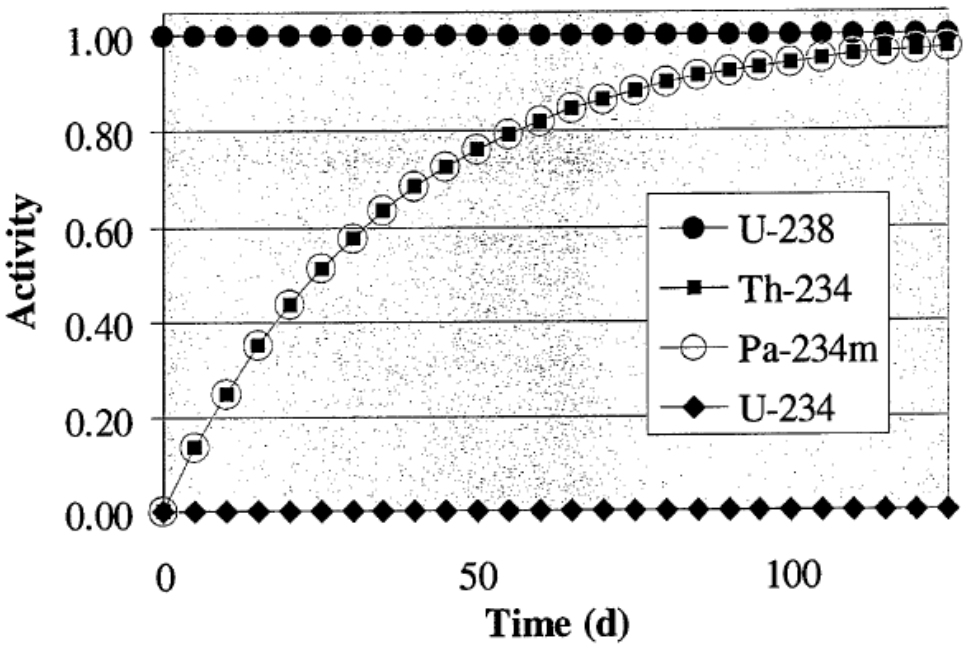

Figure 4-29 Activities of U-238, its two short-lived daughter products, and the long-lived daughter that follows them, as a function of time.[23]

Chapter V

SUMMARY AND CONCLUSIONS

Depleted Uranium metal is commonly used as shielding in medical radiation therapy and industrial radiography equipment because of its high density. DU though is classified as a nuclear material by the International Atomic Energy Agency and must be accounted for and subject to IAEA verification measures. Nuclear Regulating Authorities however lack the proper equipment to detect if DU is really present or not. A simple technique of detecting the presence of DU in the radiation shielding materials of containers with highly radioactive sources was developed by using low energy Cadmium Telluride (CdTe) and High Purity Germanium (HPGe) detectors. Optimum parameters obtained in the laboratory were applied in industrial projectors.

5.1 Conclusions and discussion

This study presented the successful development of a technique to detect the presence of depleted uranium in the shielding materials of containers with highly radioactive sources. The technique was developed based on the performance tests of the low energy gamma-ray CdTe and HPGe detectors. The effect of some parameters on the sensitivity, validation and field testing of the technique were studied. Results suggest about future improvements and successful use in field application. It could be concluded as per following:

5.1.1 Both the Amptek XR-100T CdTe and the Ortec GLP -06165/05 LEPS HPGe could easily obtain the gamma-ray spectrum of natural uranium metal and depleted uranium. The HPGe performed better in the laboratory experiments in the aspect of resolution (lower FWHM, sharper peaks) and higher peak counts.

5.1.2 The gamma-ray spectrum obtained by both detectors established the basis or signature information to determine and differentiate the presence of depleted uranium

in the material. The basis for the presence of DU is that the uranium x-ray peaks are more intense than the U-238 daughter (Th-234) gamma-ray and very low count of the U-235 peak, 185.7 keV.

5.1.3 The vertical orientation of the HPGe and weight of sample dictate the closeness between sample and detector. For lightweight samples, they can be allowed to be placed right on top of the HPGe's endcap. For heavier samples a special mounting was developed. For the CdTe because of its small size and weight, and the utilization of clamps and holders, it can be placed right next to the sample. Results of tests on the variation of distance between sample and detector though proved that the HPGe could still obtain the DU gamma spectra at a longer distance compared to the short range of the CdTe.

5.1.4 The HPGe can obtain a respectable gamma-ray spectrum of DU in just 10 seconds. It was decided that an acquisition time of about 300 seconds was suitable for both detectors.

5.1.5 The effect of the presence of radioactive sources especially high activity sources was for the uranium x-ray and other peaks to ride on the background. The laboratory results were confirmed after field test applications were done on radiography projectors containing high activity Iridium-192 sources.

5.1.6 The HPGe is better than the CdTe in still obtaining the DU spectrum with the presence of shielding materials in particular for Lead in the laboratory setting. Field test on an actual Cobalt-60 tele-therapy did not materialize therefore cannot evaluate if the detectors will be able to detect the DU surrounded by the thick lead shielding as described in the schematic diagram of the tele-therapy machine.

5.1.7 The CdTe detector is more suitable for field test applications due to its size and portability. Results of the field test applications demonstrate the far great advantage of the CdTe over the HPGe.

5.1.8 The developed technique offers a new method for Regulating Authorities to avail of, in determining if the shielding of the industrial radiography projectors they are supposed to account for is really actually depleted uranium.

5.1.9 The developed technique offers the opportunity to improve the accounting and control of nuclear materials within a State.

5.2 Suggestions and Future studies

Suggestions and future studies are as follows:

5.2.1 Further field testing at various sites will help evaluate and establish the applicability of the technique. Different sites, different work practices, to develop the adaptability of the technique.

5.2.2 Actual field test on a medical cobalt-60 tele-therapy machine with DU shielding inside.

5.2.3 Research study on the possible portability of the GLP-06165/05 HPGe. Smaller dewar sufficient for one or two day testing.

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APPENDIX

VITA

Mr. Neil Raymund Guillermo was born on August 05, 1969 in Manila, Philippines. He graduated from the University of the Philippines, Diliman with a Bachelor of Science degree in Physics. He joined the Philippine Nuclear Research Institute (PNRI) in 1992 as a Science Research Specialist. He became the Supervising Science Research Specialist of the Applied Physics Research Section of the Atomic Research Division of PNRI in 2010. After more the 20 years of research in the field of Nuclear Science and Technology, He pursued a Master of Science Degree in Nuclear Technology specializing in Nuclear Security and Safeguards at the Chulalongkorn University in Bangkok, Thailand in 2013.