COMPUTATIONAL INVESTIGATION OF POTENTIAL URANIUM DIVERSION AT AN ENRICHMENT PLANT DUE TO MATERIAL HOLDUP INSIDE PIPES

A Thesis

by

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ABSTRACT

Several studies have reported the effect of holdup on detectors and the magnitude of holdup in nuclear material processing facilities. However, scenarios how diversion of special nuclear material occurs in the presence of holdup have not studied as of yet with respect to Gaseous Centrifuge Enrichment Plants (GCEP) safeguards perspective. The potential uranium diversion within the uncertainty of the NaI detector in the presence of holdup of uranium in the centrifuge pipe was analyzed. In addition, the possibility of an unnecessary inspection resulting from holdup of uranium was also studied. In order to accomplish the objectives, Monte Carlo N-Particle (MCNP) 6.2 code had been used. A centrifuge pipe modeled in MCNP 6.2 was NPS ½ and a 2” × 2” NaI detector was utilized and the detector was surrounded by 3cm thickness lead as a collimator. Given the condition holdup, UO_2F_2, is 0.7 at%, the simulations had been conducted for various cases ranging from 6 at% to 10 at% ^{235}U enrichments in increments of 1% contained in the pipe. The variations in the holdup thickness on the pipe wall are also studied through the simulations. Separative Work Unit (SWU) was also calculated to be compared the SWU values when nothing has changed with the SWU values when the ^{235}U enrichments of feed and waste has been manipulated. Load-Cell Based weighing System (LCBS) was introduced and considered to accomplish the uranium diversion scenario. Other simulations had been executed to figure the possibility of an unnecessary inspection. While holdup had 5 at% ^{235}U enrichments, the DUF_6, NUF_6, $LEUF_6$ (1 at% to 4 at%) were assumed as the material flowing in the pipe and simulated. Also, $LEUF_6$ (1 at% to 5 at%) was simulated with 0.7

at% ^{235}U enrichment as holdup material. Based on the observation, 1 Significant Quantity (SQ) of uranium could be diverted due to holdup without being detected and the unnecessary inspection may also be called for.

DEDICATION

I dedicate this work to my wife, Rangkyeong. Without her support, this work would have not been possible and completed. Thanking you for your endless encouragement.

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CONTRIBUTORS AND FUNDING SOURCES

Contributors

This work was supervised by a thesis committee consisting of Professor Sunil S. Chirayath [advisor] and Professor John R. Ford [committee member] of the Department of Nuclear Engineering [Home Department] and Professor Andrew L. Ross [committee member] of the Department International Affairs [Outside Department].

All other work conducted for the thesis was completed by the student independently based on the project idea and advice provided by Prof. Sunil S. Chirayath [advisor].

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NOMENCLATURE

UO_2F_2	Uranyl Fluoride
UF_6	Uranium Hexafluoride
HF	Hydrogen Fluoride
at%	Atomic Percent
wt%	Weight Percent
MCNP	Monte Carlo N-Particle
keV	kilo Electron Volt
s^{-1}	Per Second (SI unit for inverse time)
α	False Alarm Probability
β	Nondetection Probability

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1. INTRODUCTION

1.1. Background

When it comes to Special Nuclear Material (SNM) diversion, civilian uranium Gaseous Centrifuge Enrichment Plants (GCEPs) are regarded as a threat and concern to the nuclear nonproliferation community. This is because GCEPs are generally large facilities and they can produce required quantity of Highly Enriched Uranium (HEU) for use in a nuclear weapon within a few months if misused. Also, centrifuges are modular, so it can be easily configured into either a cascade for the production of HEU or Low Enriched Uranium (LEU). Each cascade can be independently and simultaneously operated. Therefore, it is very important to enforce nuclear safeguards in GCEPs with the objectives of timely detection of diversion of SNM and their misuse. In nuclear safeguards, Significant Quantity (SQ) and SNM conversion (to weapons useable form) time are two important parameters required to be analyzed. An SQ refers to the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded.[1] The SQ values for SNM has been set by the International Atomic Energy Agency (IAEA) and relates to the IAEA's inspection goal. The LEU with less than 5% ^{235}U enrichment is the product from GCEPs and it is categorized as indirect (weapons) useable material. The SQ value for LEU is 75 kg of ^{235}U. The conversion time for LEU to weapons useable material ranges from 3 months to

1 year by according to the stipulations by the IAEA. The range is due to the possibility of different ^{235}U enrichment within the LEU category.

"Holdup" is defined as the deposition of SNM occurring in nuclear material processing facilities, for example, inside pipes and ducts.[2] In a GCEP, the holdup occurs inside the centrifuge pipes due to the deposition of UF_6 in a solid form, UO_2F_2. UO_2F_2 is a direct product of the reaction of UF_6 with the moisture in the air as shown in the Eq.1.[3]

$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF \qquad \text{Eq.1}$$

The reaction depicted in Eq. 1 is possible whenever UF_6 comes in contact with moisture that may have leaked into the centrifuge piping. A GCEP consists of thousands of centrifuges connected by long piping and ducts. Hence, there is a potential for leakage of moisture and the formation of UO_2F_2 deposited in the internals of the piping. This type of deposit can result in a detection error while measuring ^{235}U enrichment. Based on gamma radiation detection, the measurement of ^{235}U enrichment can be affected due to self-attenuation by uranium present in the UO_2F_2 holdup in the internals of the piping. Another cause of error can be due to the gamma radiation emitted from the uranium holdup itself, which can result in undesired enhancement of signal in the sodium iodide (NaI) gamma radiation detector. Hence, there can be a mismatch between the measured ^{235}U enrichment compared to the actual enrichment inside the pipe, which needs to be analyzed to quantify the aforementioned discrepancy.

Gamma radiation spectroscopy, one of Non-Destructive Assay (NDA) methods, is usually used to determine the ^{235}U enrichment of UF_6 gas flowing in pipes. It shall be noted here that the gamma radiation spectroscopic detectors are simple, reliable and portable to use. Those features of a gamma radiation detector, for example a NaI detector, make the determination of ^{235}U enrichment in UF_6 rapid and accurate. High enrichment of uranium in a GCEP can take place and can be concealed using the presence of holdup if the amount of holdup is unaccounted. In this study, radiation transport modeling and simulations of UF_6 pipe using MCNP6.2 code is carried out for a variety of ^{235}U enrichment cases ranging from Depleted Uranium (DU) to 10 at% to estimate the possible uranium diversion within the uncertainty of ^{235}U enrichment determination due to the interference from holdup. In order to figure out how much amount of holdup is required to veil a higher value of ^{235}U enrichment than what has been stated by the facility, parametric variation in the holdup thickness inside the pipe wall is also analyzed using the MCNP6.2 radiation transport simulations incorporating a NaI detector in the simulation model. Furthermore, the simulations of UO_2F_2 using MCNP6.2 code is conducted by adjusting the concentration of ^{235}U in holdup deposition ranging from depleted uranium to 5 at% ^{235}U enrichment to evaluate the influence of enrichment of holdup on gamma spectroscopy of UF_6. International Target Values (ITVs) provided by the IAEA for a NaI detector are applied for the uncertainty quantification of uranium enrichment measurement.

1.2. Objective and Scope

The main objective of this research is to study the effect of uranium deposit or holdup (UO_2F_2) in the internals of the pipe on the measurement of ^{235}U enrichment in UF_6 gas in GCEPs while employing the NaI detector. Modeling and simulations of pipe with UF_6 gas flow in a GCEP are carried out for parametric variations of the holdup thickness as well as ^{235}U enrichment in the gas. Simulations were performed by using the Monte Carlo radiation transport code, MCNP6.2.[4] Simulations supported the estimation of the effect of holdup or deposit of UO_2F_2 in the pipe while measuring ^{235}U enrichment of UF_6 gas flowing in the pipe. Since measurement of holdup typically has a high uncertainty in the range of 20%~50% or more,[5] it is not enough to confirm the exact amount of holdup in the pipe. Moreover, the NaI detector widely used in GCEPs has a relatively high measurement uncertainty compared to HPGe detector or other spectroscopies. Holdup in the pipe wall can make the gamma radiation signal weaker or stronger depending on the abundance of ^{235}U in holdup. In other words, determination of the ^{235}U enrichment of UF_6 flowing in the pipe can be interrupted by a relative error (inclusive of random and systematic error) of the NaI detector and effect of holdup on the NaI detector. Preliminary studies indicate uranium diversion is potentially possible by hiding within the measurement error, if uranium holdup on the pipe walls are not appropriately accounted for. The results of this study can be used to quantify the potential errors in ^{235}U enrichment measurements at GCEPs due to uranium holdup on pipes and will be useful to avoid frequent inspections.

1.3. Previous Work

There have been some studies with respect to the accurate measurement of SNM in the nuclear material processing facilities using the NaI detector under the condition that material holdup exists. Generalized-geometry holdup (GGH) measurement method is one such study that has been tested and verified several times by Russo *et al*.[6] The GGH is the formalism coming from USNRC regulatory guides,[7] which assumes SNM is distributed as a point source, a line source, or area source for the measurement of holdup. This concept is well depicted by Sprinkle, Jr. *et al*. [8] In Sprinkle, Jr.'s study, generalized geometry calibration equations are newly proposed by collecting numerous data from experiments to increase the accuracy of the SNM measurement.[8] Another holdup measurement method reported is the Holdup Measurement System 4 (HMS4) which is developed by the Y-12 National Security Complex and the Oak Ridge National Laboratory under the support of US Department of Energy Office.[9] In HMS4, more accurate corrections for finite source and gamma radiation self-attenuation are newly applied to improve the measurement of SNM. Since GGH technique had focused on holdup measurement of plutonium and HEU until 2005, new GGH approach which is directly applicable to LEU facilities is studied by Belian *et al*.[10]

A research for the purpose of GCEP safeguards, which is named the Hexapartite Safeguards Project (HSP) has been established by the EURATOM, IAEA, Australia, Germany, Japan, Netherlands, the United Kingdom, and the United States in an international forum.[11] In order to improve the safeguards for undeclared LEU production, advanced safeguards approaches are studied and verified by Boyer *et al.*[12] Heinonen proposed a Material Unaccounted For (MUF) calculation concerned with the holdup in terms of verifying decommissioning of South Africa's nuclear weapons.[13] However, any of these studies or research has not analyzed and reported the potential uranium diversion scenario within the uncertainty of gamma radiation spectroscopy in the presence of uranium holdup inside UF_6 pipe wall.

1.4. Steps of the Thesis Study

Thesis study was performed as follows;

1. Simulate gamma radiation measurement of UF_6 gas centrifuge enrichment pipe with the incorporation of a NaI detector using MCNP6.2 code to obtain the gamma radiation spectrum from two uranium sources (UF_6 flowing in the pipe and UO_2F_2 holdup inside the pipe wall).

2. Acquire the pulse height distribution of ^{235}U and ^{238}U gamma radiation at characteristic energies respectively at 186 keV and 1001 keV (by using the F8 pulse height tally featured in MCNP6.2 code) through Monte Carlo simulation.

3. Estimate the quantities (or thicknesses) of holdup required to conceal a higher value ^{235}U enrichment of UF_6 within the uncertainty of the measurement (5.8% for LEU[14]) through MCNP6.2 simulations for variations in thickness and ^{235}U enrichment itself.

4. Evaluate the number of centrifuges required to divert ^{235}U equivalent to one SQ (75 kg of ^{235}U) in 3 months based on the results of the simulations.

5. Studies on the variations of the ^{235}U enrichment of feed and waste to make the quantity of feed and waste as same as a normal operation (5% ^{235}U enrichment product from natural uranium feed and no diversion occurs).

6. Evaluate the possibility of an unnecessary inspection depending on the enrichment and the amount of holdup.

7. Analyze the potential uranium diversion and an unnecessary inspection due to holdup unaccounted and recommend safeguards approaches to prevent nuclear proliferation risk due to uranium holdup in GCEP piping.

1.5. Significance

The goal of the thesis is to investigate how much amount of holdup is needed to conceal the diversion of uranium within the uncertainty of ^{235}U enrichment measurement while using NaI gamma detector. A second objective is to evaluate the effect of unaccounted uranium holdup on inspection frequencies in GCEP due to the error holdup causes in ^{235}U enrichment measurement. Calculation of ^{235}U mass difference and UF_6 flow rate is also part of the study to estimate how many centrifuges are needed to divert one SQ of uranium given the material holdup scenario. The results of the study have significance in modifying the safeguards approach in GCEPs.

2. LITERATURE REVIEW

2.1. Uranium Gas Centrifuge Enrichment

2.1.1. Uranium

Uranium, is well known as the material used for a nuclear weapon, naturally consists of three prominent isotopes, ^{234}U, ^{235}U, and ^{238}U. Among them, ^{235}U is used in a nuclear weapon. The abundance of ^{235}U is only 0.711 wt% in natural uranium and ^{238}U is at 99.2 wt%, with a minor ^{234}U fraction of 0.0054 wt%. Therefore, separation techniques of ^{235}U from natural uranium is important for the purpose of military (^{235}U enrichment > 90 wt%) as well as civilian use (^{235}U enrichment < 5 wt%).

The separation of uranium isotopes focuses on enriching ^{235}U than the 0.711 wt% present in natural uranium. The enrichment of uranium product varies from 3% to over 90% depending on the utilization of uranium. Usually 4%~5% ^{235}U enrichment is used in Light Water Reactors (LWRs), which are the widely operated nuclear power plants in the world for civilian production of electricity. This type of uranium is categorized as LEU where the ^{235}U enrichment is less than 20%. The uranium product with more than 20% of ^{235}U is categorized as HEU. The residual uranium, after the separation of ^{235}U isotope from natural uranium, is called tails or depleted uranium.

2.1.2. Centrifuge

There are at least four (electromagnetic, diffusion, centrifuge, laser) methods to enrich ^{235}U which are practiced in the world. Among them, enrichment of UF_6 gas using centrifuges is currently the most popular method due to its energy efficiency. [15]

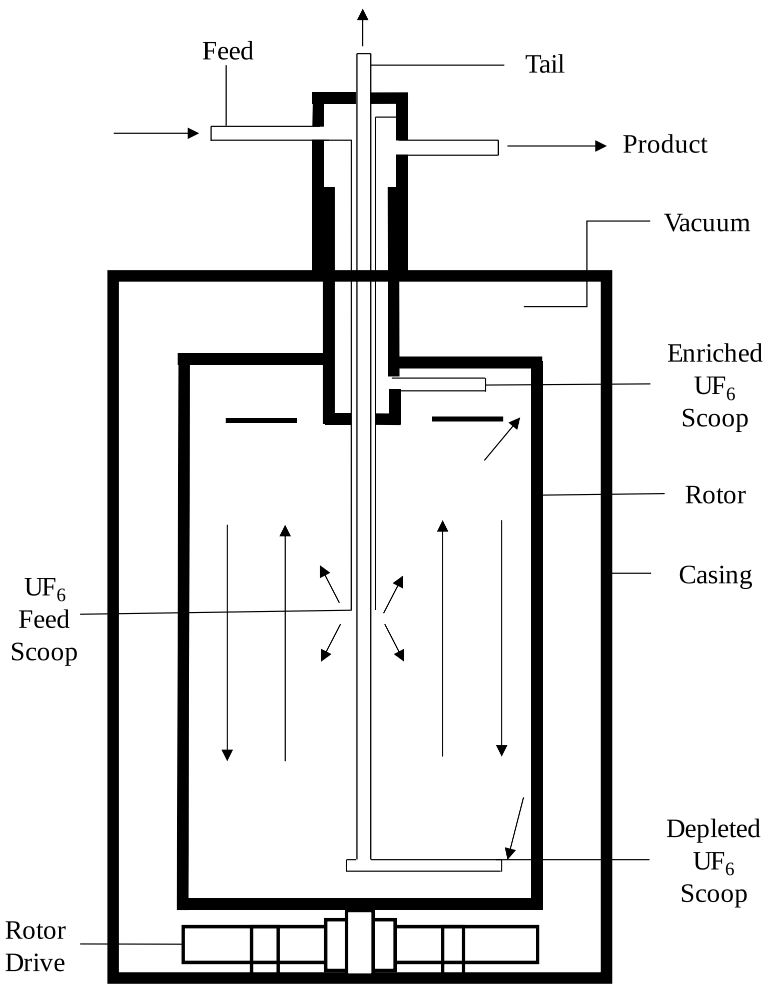

Figure 2.1 A schematic of countercurrent gas centrifuge with internal circulation ;adapted from [16].

Figure 2.1 depicts the schematic of a centrifuge and is modified from "Uranium Enrichment Processes: Gas centrifuge (2012)" published by USNRC. The centrifugal force makes it possible to separate of the isotopes of uranium by taking advantage of the difference in their atomic masses. The gaseous phase of UF_6 is injected into the cylinder (centrifuge) through the feed pipe. The gas circulating inside the centrifuge is forced to separate the heavier isotope (^{238}U) from the lighter isotope (^{235}U) and locate the heavier isotopes at the outer radius than center as shown in Figure 2.2. Figure 2.2 is adapted from "Uranium Enrichment" published by *Villani S*. The difference in atomics masses, however, is not large, so strong centrifugal forces must be required to produce a million times the centrifugal acceleration of gravity, which means the rotor depicted in Figure 2.1 have to spin at a very high speed. The speeds vary depending on the efficiency, circulation rate, and separative capacity of the centrifuge. For example, at room temperature if UF_6 gas is subjected to rotation in a centrifuge at a spin speed of 500 m/s, the separation factor is 1.162, which means the ratio of ^{238}U to ^{235}U at the outer radius is 1.162 times greater than at the center and the pressure at the outside is 46 million times greater than at the center. [17] Hence, by this centrifuge process, the relatively enriched ^{235}U is located at the center of the centrifuge and fed to the next set of centrifuges until the desired enrichment of ^{235}U is achieved in the product. The depleted UF_6, which is at the farther radial distance in the centrifuge, further gets depleted and is discarded when the abundance of ^{235}U goes down to about 0.2 wt%.

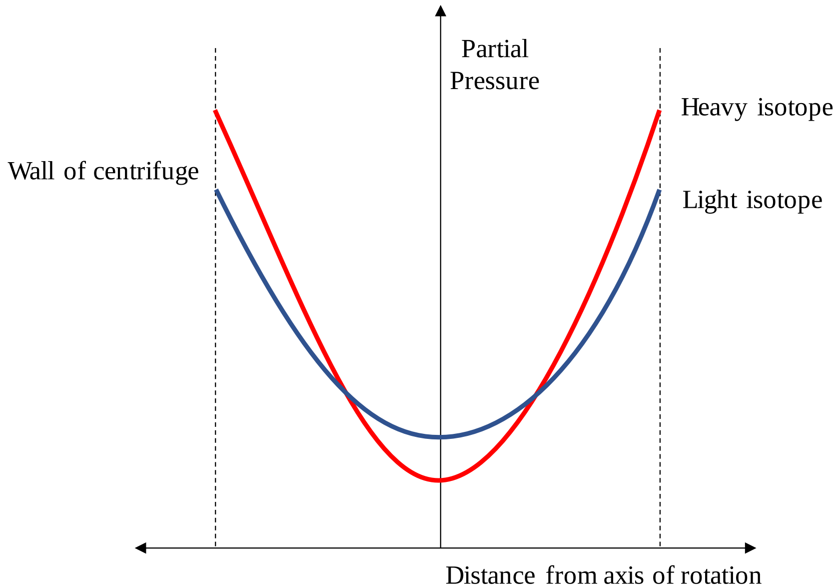

Figure 2.2 A distribution of light and heavy isotopes in a centrifuge with respect to the profile of a centrifuge; adapted from [18].

2.1.3. Cascades

In a GCEP, thousands of centrifuges are connected in a particular way to form a cascade to produce a desired enriched uranium product. One centrifuge is generally regarded as the smallest element of a GCEP, which is also called a separating unit.[17] A set of centrifuges connected in parallel is called a stage. In a stage, the abundance of ^{235}U in its feed is same and exiting is also same, but at higher abundance for the product pipe and at lower abundance for the tail pipe. The number of centrifuges in a stage depends on the amount of mass flow of uranium, which also dictates the final product mass. A serially connected group of stages is usually necessary to produce the desired enrichment of ^{235}U. This serially connected group of stages is called a cascade. In Figure 2.3, a separating unit, or a centrifuge is illustrated by a cylinder and it shows the flow of feed, product, and tails in a stage. Also, the parallel group of centrifuges in Figure 2.3 shows a stage.

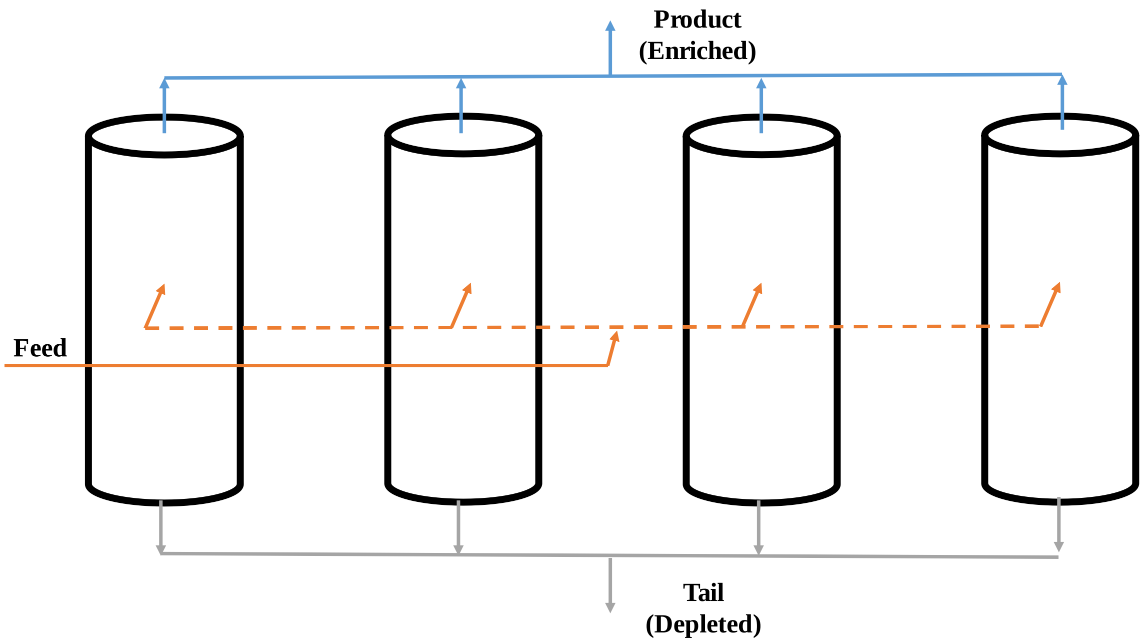

Figure 2.3 An example of a group of separating units (centrifuges) connected in parallel to form a stage.

As shown in Figure 2.4, a cascade consists of three categories of stages, feed stage, enriching stage, and stripping stage. Besides the feed stage, both enriching and stripping stages have multiple stages. Enriching stages, or positive integer stages, are for producing the ^{235}U enrichment higher and higher as the enrichment stage number increases. Stripping stages, or negative integer stages, process the residue from a previous stage. At the largest negative number stage, the tails (waste) is pulled out. The number of stages in a cascade can be different depending on the ^{235}U concentrations in feed, product, and waste.

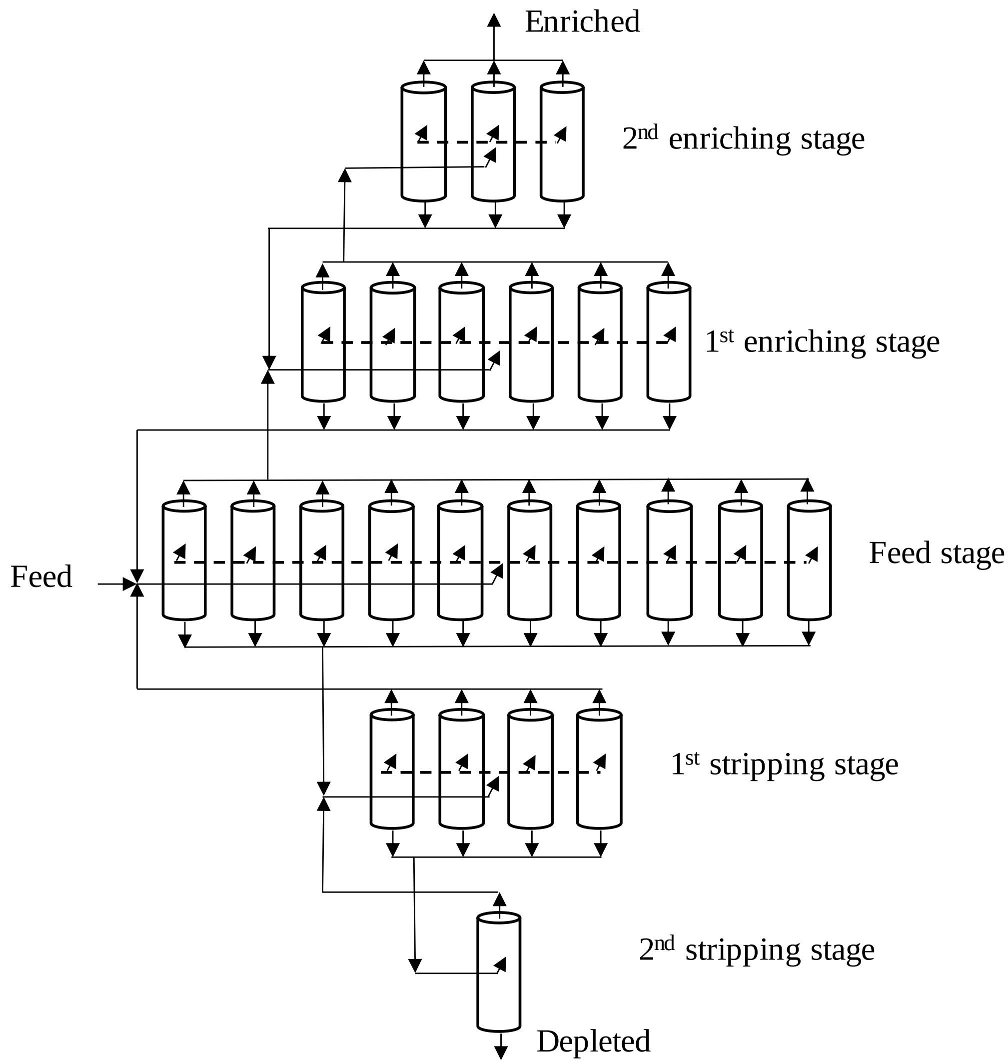

Figure 2.4 An example of a cascade shows it consists of numerous stages, which are categorized in enriching, feed, and stripping, to make the desired product.

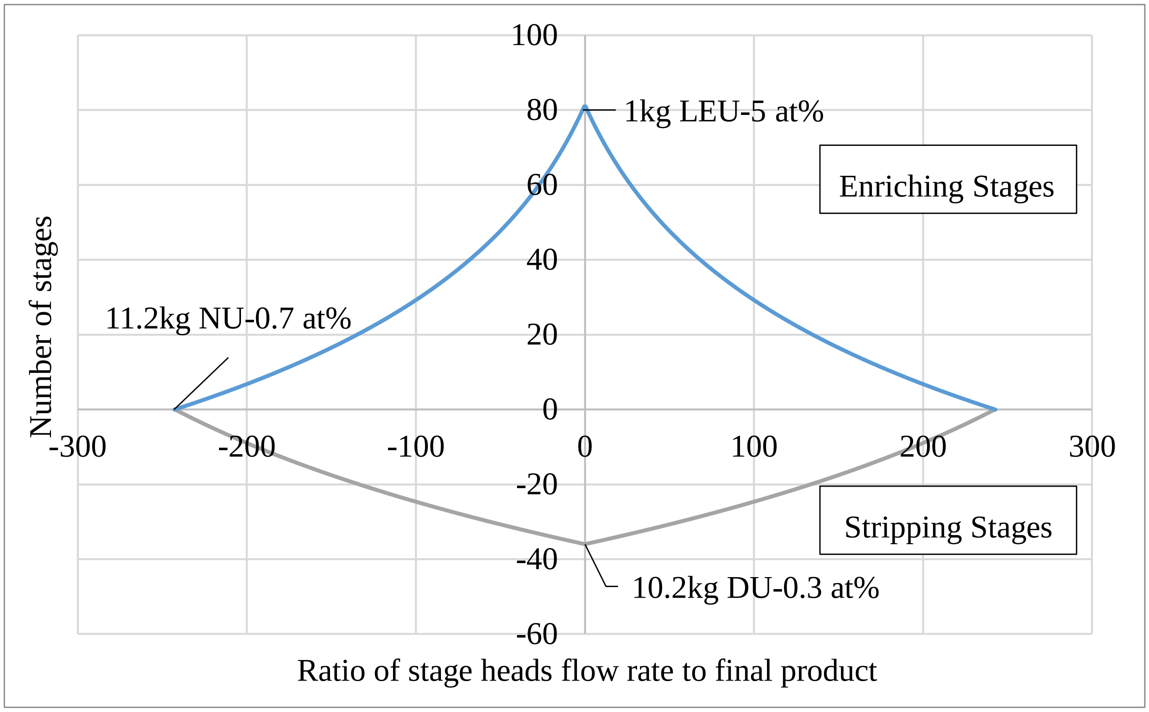

Figure 2.5 An example of an ideal cascade showing how many stages are required to make 5 at% ^{235}U UF_6 with natural uranium when the ^{235}U concentration of waste is 0.3 at%.

Figure 2.5 shows the relation between the number of stages and ratio of heads flow rate (at each stage) to the final product (in this case it is 1kg of product-LEU). In Figure 2.5, the difference between the number of enriching stages and stripping stages are noticeable. The general cascade arrangement in GCEP is similar to that shown in Figure 2.5. The ideal cascade is known as the most efficient cascade model in terms of production of SNM and the cost. As it is seen in Figure 2.5, every stage has a little different flow rate due to its shape, so there are no more than two stages with the same size and number of centrifuges. [19] The constraint makes the construction of the ideal cascade impossible. To solve this problem in practical cascade design, the approximation of the ideal cascade by a small number of square cascade segments was developed and it was named "squared-off cascade". [19] The squared-off model is a method to make an achievable and efficient cascade. An example of squared-off cascade is well depicted in Figure 2.6.

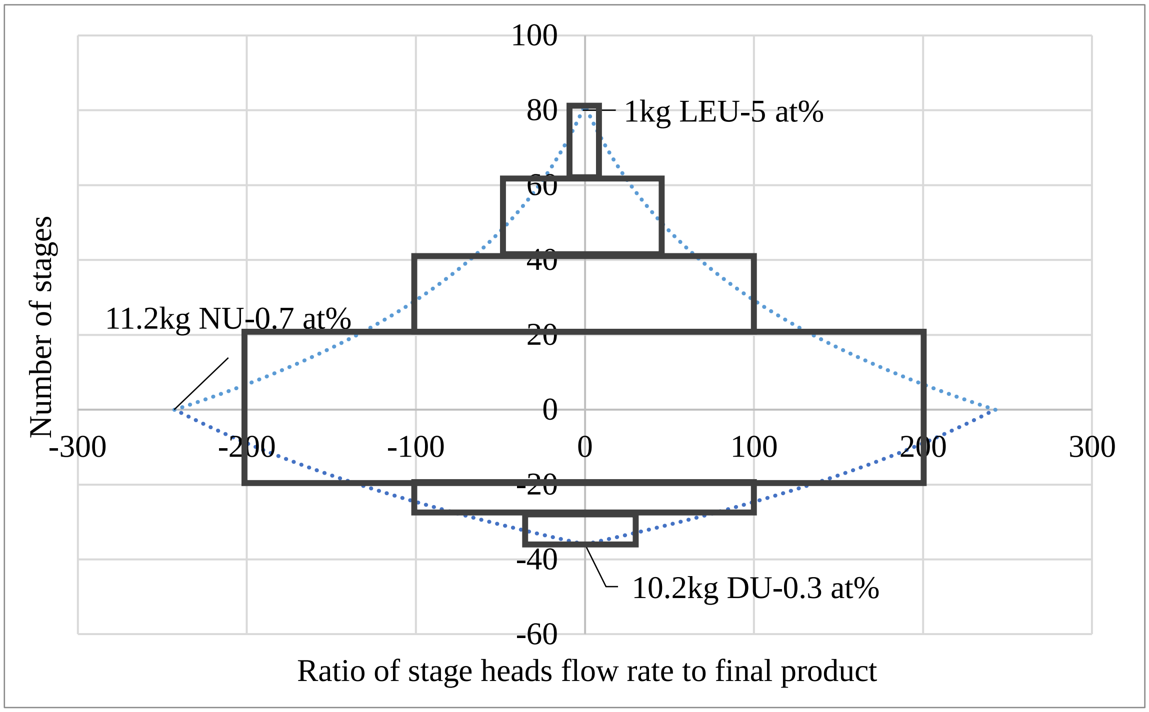

Figure 2.6 An example of a squared-off cascade (black) shows an approximation to an ideal cascade.

Figure 2.6 suggests that the uranium mass flow rate is the same value once stages are in the same square cascade. This means the number of centrifuges of each stage in the square cascade is the same unless various type of centrifuges are used in the cascade and the centrifuges may have different capacity for UF_6. This advantage makes it easy for the construction of cascade and arrangement of centrifuges.

2.1.4. Holdup (UO_2F_2)

Holdup, the accumulation of SNM inside the processing equipment, generally happens in multiple irregular geometrical locations such as bends and is hard to detect and evaluate the accurate amount of holdup. Measuring holdup is important from both safety (with regards to criticality) and safeguards (with regards to nuclear material accounting and control) perspectives because of the high economic value of nuclear material and the risk that the theft nuclear material can be produced for nuclear weapon against a certain society, state, or country.[2] Therefore, studies have been conducted to minimize the buildup of holdup, to measure the magnitude of holdup and determine the location, and to remove it. [2] The amount of uranium holdup have been reported to 0.3 – 1g/m length of the pipe in a nuclear material processing facility [2,20] and a similar magnitude can be expected at GCEPs.

2.2. International Target Values (ITVs)

International Target Values (ITVs) are the standards for the measurement error of nuclear material accounting. The measurement error consists of random error, which varies in an unpredictable way under repeatability conditions, and systematic error, which are caused by some kind of bias in the detector system. From those two errors, an uncertainty of a measurement can be written as

$$u = \sqrt{{\sigma_s}^2 + {\sigma_r}^2} \qquad \text{Eq.2}$$

Where:

u : the uncertainty of measurement

σ_s : systematic error

σ_r : random error

ITVs have been established as the relative errors of the measurements when Destructive Analysis (DA) and Non-Destructive Analysis (NDA) are used. The values are provided by the IAEA. Among them, a few examples of NDA are given in Table 2.1. Load-Cell Based weighing System (LCBS) is commonly used for weighing nuclear material to check if any diversion has happened. The method is usually used for the detection of mass difference between shipper and receiver. Inspector Multi-Channel analyzer with NaI detector (IMCN) is usually used when ^{235}U abundance NDA measurement is conducted. The ITVs of IMCN have different values in accordance with the ^{235}U abundance of UF_6. These values are also listed in Table 2.1 and is used in this study for the SNM diversion calculation.

Table 2.1 A few examples of ITVs when NDA is used in the measurement of nuclear material.

Method	Measurement	Uncertainty component (% relative)		ITV (% relative)
		σ_r	σ_s	
LCBS	Mass	0.05	0.05	0.07
IMCN	DUF_6	20	8	21.54
	NUF_6	10	3	10.44
	$LEUF_6$	5	3	5.83

2.3. Separative Work Units

The Separative Work Unit (SWU) is defined as the separation work effort required to enrich ^{235}U concentration in uranium compared to ^{238}U. The SWU is measured in units of kg and usually manipulated economical units to determine cost ($) per SWU and kWh per SWU. Figure 2.7 below can help understand the correlation between the enrichment of ^{235}U, the amount of product, and SWU. Figure 2.7 is modified from an article about uranium enrichment printed in World Nuclear Association. As HEU or weapon-grade uranium is produced using the same amount of feed quantity, the SWU will be higher and the quantity of product will be lower than if LEU was the original goal of ^{235}U enrichment.

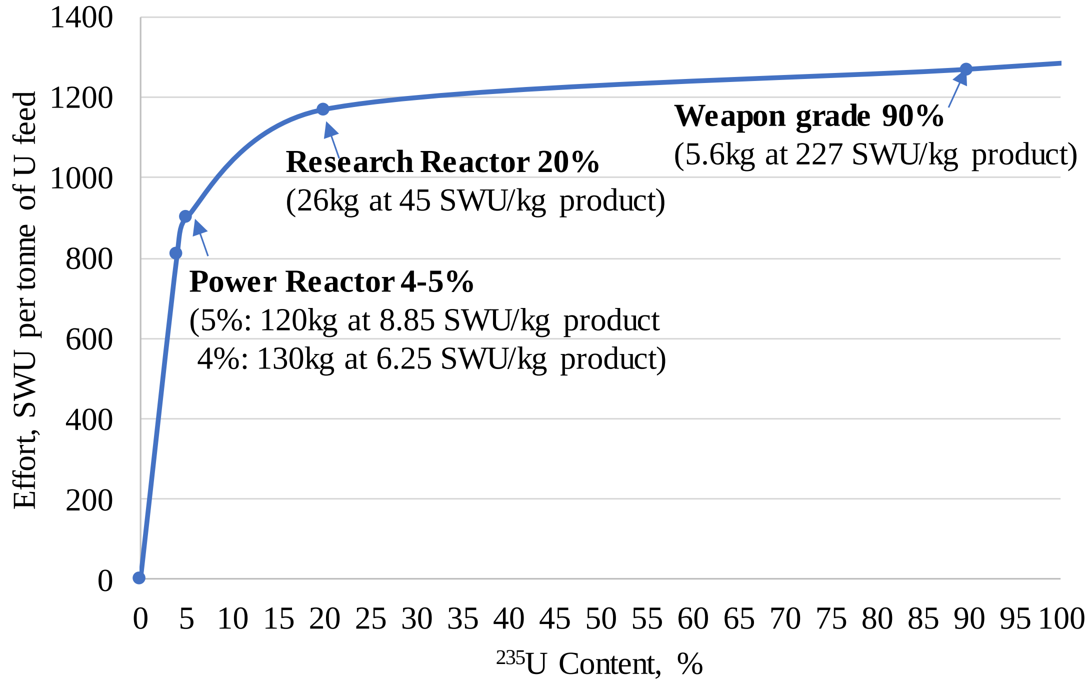

Figure 2.7 Graph showing how much product of different ^{235}U enrichment of uranium will be produced when one ton of natural uranium is used as the feed; adapted from [21].

2.4. False Alarm Probability and Nondetection Probability

The IAEA has established a concept of diversion detection with respect to statistical methods and this is called as “Type-II error”. The type II error means that inspectors draw a conclusion that diversion did not occur when in fact it did occur. [1] The probability, β, of a Type-II error is commonly referred to as the nondetection probability and less than 20% is recommended. [1] High Type-II error may lead to SNM diversion in nuclear material processing facility. In contrast, the detection method of nuclear material can indicate diversion has occurred in fact it did not occur. This type of error is named “Type-I error”, and the probability of committing a type I error is termed as “False alarm probability (α)”. [1] Type-I error committed will indicate that an amount of nuclear material is missing when, in fact, no diversion has occurred. [1] The value of α is recommended by IAEA to be set at 5% or smaller. The false alarm probability is usually applied when gamma spectroscopy is used for the detection of nuclear material diversion and is also combined with the nondetection probability (β). [1] A pictorial representation of false alarm probability and nondetection probability are shown in Figure 2.8.

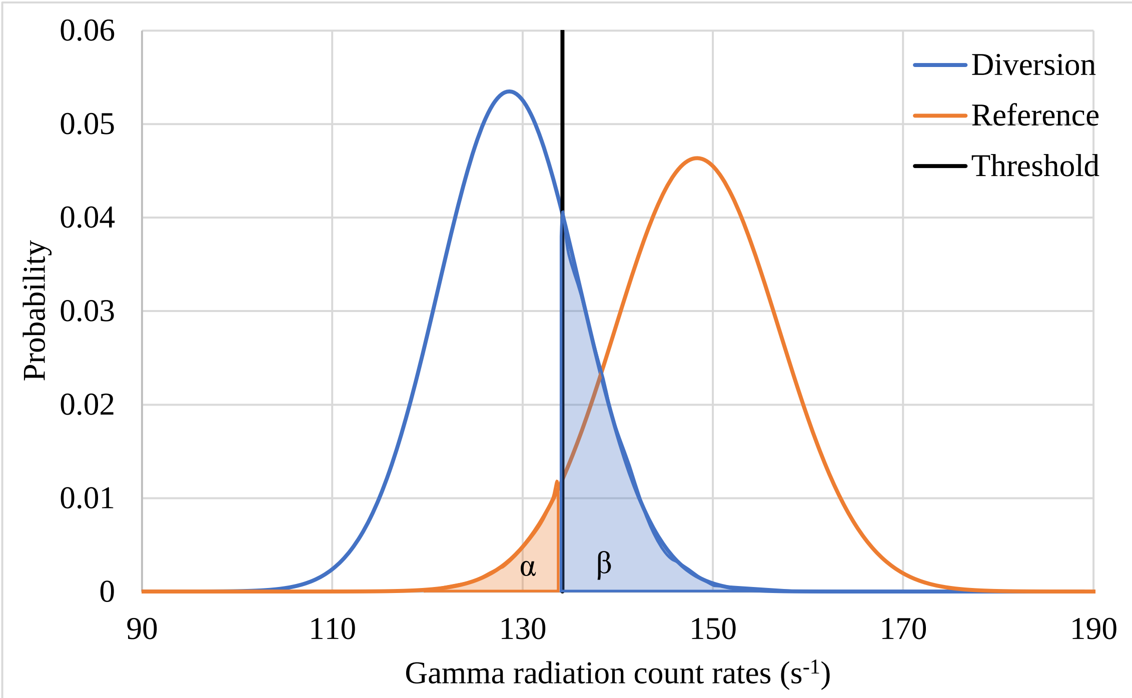

Figure 2.8 Depiction of false alarm probability (α) and nondetection probability (β) given the threshold of detection. α is 5% and β is set to 20% in the figure.

In Figure 2.8, two Gaussian distribution graphs are plotted, the orange one is the reference of the gamma radiation count rate of nuclear material and the blue one is the gamma radiation count rate of nuclear material when a diversion has occurred. If diversion has occurred in a nuclear material facility, the blue one is moving to left-side and the nondetection probability β is decreasing. To avoid the risk from diversion and unnecessary inspection, the threshold of gamma radiation detection has been set by taking into account both false alarm probability (α) and nondetection probability (β). The operator should determine the applicable value of nuclear material diversion detection threshold and the nondetection probability should be less than 20%.

Unlike aforementioned depiction of false alarm probability and nondetection probability, Figure 2.9 shows the case when a GCEP is misused and more highly enriched uranium is produced than what operator have stated, which is studied and discussed in this thesis. If the facility has been misused, the orange distribution is moving to right-side and the probability β is decreasing. When the probability β is less than 20%, the facility will be inspected to check if the facility is being misused. However, holdup in a GCEP pipe make a problem to detect the misused facility. This is due to that holdup can reduce gamma radiation signal significantly as analyzed previously.

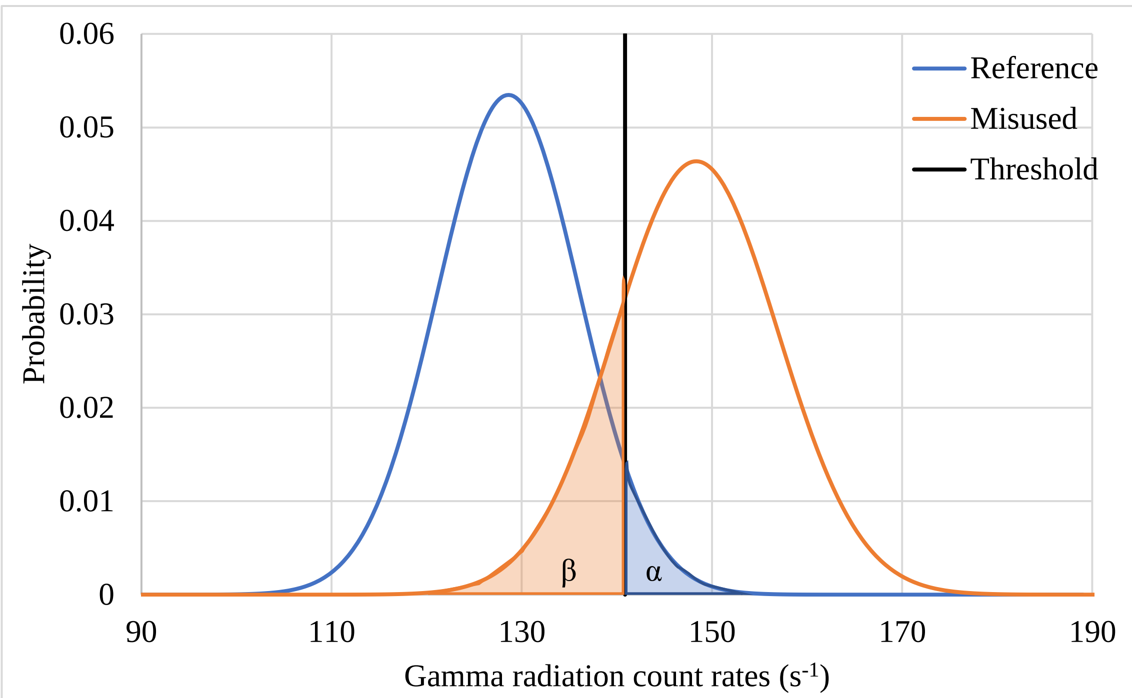

Figure 2.9 Another depiction of false alarm probability (α) and nondetection probability (β) given the threshold of detection. α is 5% and β is set to 20% in the figure.

3. METHODOLOGY

3.1. MCNP6.2 Modeling

Radiation transport simulations were performed using MCNP6.2 code to evaluate the effect of uranium enrichment gamma radiation signal from UF_6 gas in a NaI detector due to solid uranium (UO_2F_2) holdup in the internals of the centrifuge piping. Through the simulations, gamma radiation spectroscopic evaluations employing a NaI detector model mounted on a typical centrifuge pipe were carried out to determine ^{235}U enrichment in UF_6. All figures were rendered by the Vised software. [22]

3.1.1. Modeling a General Configuration in MCNP6.2

In order to evaluate the gamma radiation measurement of ^{235}U enrichment in UF_6 in a GCEP using MCNP6.2, several parameters had to be assumed in the simulations. Figure 3.1 shows a geometry model of the gamma-ray measurement of the simulations; the NaI detector (light green) is placed in a lead collimator (red) which reaches outside the pipe (green). Except for the amount of UO_2F_2 (light blue) and UF_6 (blue), all parameters were unchanged in terms of the configuration. As the thickness of holdup, or the amount of holdup, increases, the space for UF_6 flow decreases because the inner diameter of pipe has not changed through the entire simulations. The details about changes in the amount of UO_2F_2 and UF_6 will be discussed in the section 3.1.2

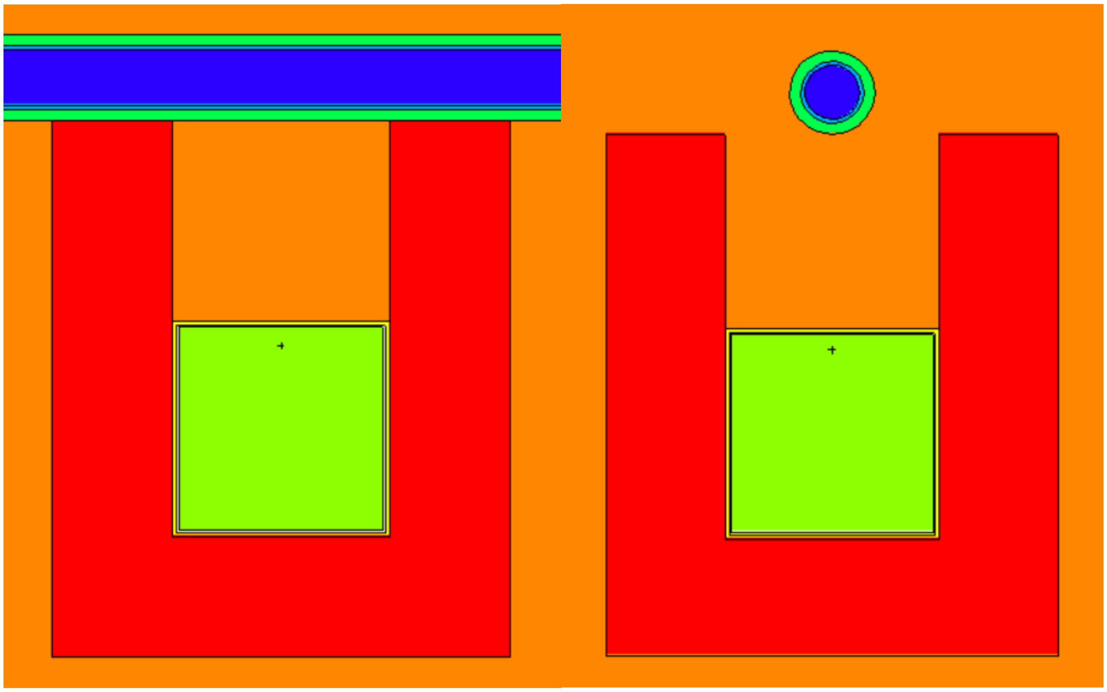

Figure 3.1 Gamma measurement configuration modeled in MCNP6.2 rendered by the Vised software. It shows the front view configuration (Right) and the side view configuration (Left)

In MCNP6.2, the specification of the NaI detector used is 5.08cm diameter and 5.08cm height in a 0.5cm thick aluminum can, and empty space between the NaI crystal and the aluminum. The NaI detector is surrounded by 3cm thick lead collimator. In order to simplify the geometry, the photomultiplier and tube base was not taken into account in the model. The details of NaI model are shown in Figure 3.2 below.

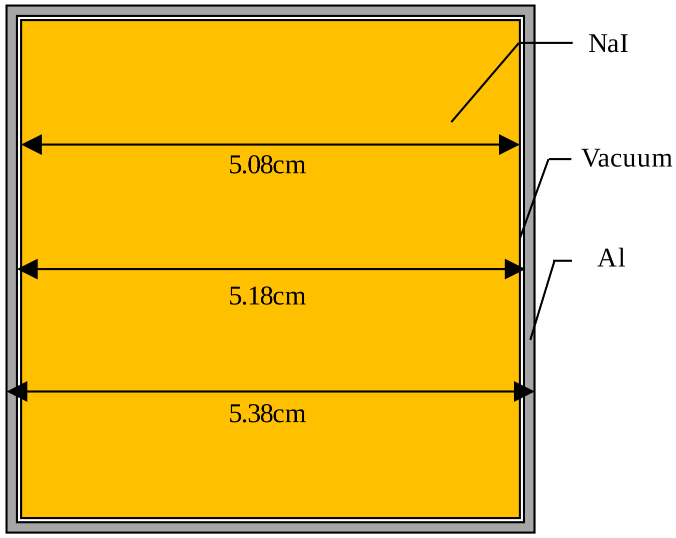

Figure 3.2 The schematic of the NaI detector shows the parameters and materials to be simply assumed throughout the simulations.

Gas centrifuge enrichment facility has lots of pipework consisting of head pipes as well as thin pipes in each centrifuge itself to process UF_6. Since the specification of pipes in each centrifuge has not been fully revealed in public yet some data of head pipes and various size of centrifuges have been reported in literatures. As such, a method used in North America, which called Nominal Pipe Size (NPS), was assumed for the modeling purposes. The American Society of Mechanical Engineers (ASME) has published numerous editions about the standard pipe information and defined the size of pipe in terms of Outer Diameter (OD) and wall thickness. [23] A few examples are listed in Table 3.1.

Table 3.1 An example of NPS established by ASME used for setting the outer diameter and wall thickness of stainless-steel pipe in MCNP6.2

NPS	DN (dimensionless)	OD (mm)	Wall thickness (mm)				
			Sch.5s	Sch.10s /20	Sch.30	Sch.40s /40/STD	Sch.80s /80/XS
¼	8	13.72	1.245	1.651	1.854	2.235	3.023
½	15	21.34	1.651	2.108	2.413	2.769	3.734
¾	20	26.67	1.651	2.108	2.413	2.870	3.912
1	25	33.40	1.651	2.769	2.896	3.378	4.547
2	60	60.33	1.651	2.769	3.175	3.912	5.537
3	80	88.90	2.108	3.048	4.775	5.486	7.620

The specification of the pipe used in the simulations has 21.34mm of an outer diameter with 2.769mm of wall thickness and a length of 50cm.

3.1.2. Gamma Energy Source Definition

In MCNP model, the radiation source type and geometrical shape must be defined through its SDEF (source definition) input. The SDEF input represents source definition, which requires a variety of parameters for accurate representation in a numerical simulation. [4] Among the parameters, the radius and length of the source (since source in this study is in cylindrical shape) and the gamma radiation energy of each isotope are important. The simulations consisted of two cases, one where the gamma radiation source is only defined due to UF_6 gas in the pipe and the other where solid deposit of UO_2F_2 is the only gamma radiation source. Therefore, results from two separate simulations are needed to evaluate the composite NaI gamma radiation spectrum of the GCEP pipe modeled. In one parametric variation of the simulation, for example, 6 at% of ^{235}U enrichment is present in UF_6 gas flowing in the pipe and 0.1cm UO_2F_2 holdup material with 0.7 at% ^{235}U in it, hence the NaI gamma radiation spectra obtained from the simulations of each of the sources (UF_6 and UO_2F_2) are merged to evaluate the composite gamma radiation spectrum from the pipe.

The radius of the gamma source in SDEF is different for each case. From the assumptions made and discussed in section 3.1.1 regarding the specification of the pipe used in MCNP6.2 model, the inner radius of pipe is 0.79cm and its length is 50cm. From the amount of holdup (g/m), density of UO_2F_2 (g/cm^3), the length of the pipe (cm), and the inner radius of the pipe (cm), the thickness of holdup (cm) was estimated. Throughout the simulations, the amount of holdup is varied from 1g/m to the amount that is found to conceal the corresponding parametric variation of ^{235}U enrichment of UF_6.

In gamma radiation spectroscopy, the magnitude and the counts under the photo peaks (created due to the photoelectric effect in the NaI detector) obtained for the respective gamma radiation emitted by each isotope is used to estimate the concentration of each isotope. In this study, only selected gamma radiation energies emitted by ^{235}U and ^{238}U isotopes were simulated, which are respectively 186 keV and 1001 keV. [2] In order to perform gamma radiation spectroscopy using MCNP6.2, the probability (or the weighting factor) of energy of each isotope must be given, which means gamma radiation source strength of ^{235}U and ^{238}U inside the pipe must be taken into consideration.

Subsequently, the pulse height distribution of gamma energies in the NaI detector can be evaluated. The gamma radiation source strength (γ/s) for each energy was calculated by multiplying the mass (g) of an isotope with the gamma activity (γ/g·s) of the corresponding isotope, 4.32×10^{4}γ/g·s for ^{235}U and 7.34×10^{1}γ/g·s for ^{238}U. [2] When 5 at% ^{235}U enriched UF_6 gas is used, the masses of ^{235}U and ^{238}U are 15.51g and 294.67g respectively, thus the gamma radiation source strengths are 6.70×10^{5} γ/s for ^{235}U and 2.16×10^{4} γ/s for ^{238}U. Hence, probabilities of emission of each gamma radiation energy (186 keV and 1001 keV) used as input in MCNP model are 0.969 for ^{235}U and 0.031 for ^{238}U.

As mentioned previously, the simulations were executed with two cases for each of the parametric variations because the pipe has two different gamma radiation sources, UF_6 gas flowing in the pipe and UO_2F_2 deposited in the internals of the pipe. The source particle plots are rendered by the Vised software. [24] Figure 3.3 represents when only UF_6 is used as the gamma radiation source and Figure 3.4 shows when only UO_2F_2 is used as the gamma radiation source. In Figures 3.3 and 3.4, the assumption made is that 100g/m of holdup is present in the internal of the pipe and the thickness of the holdup is 0.033cm.

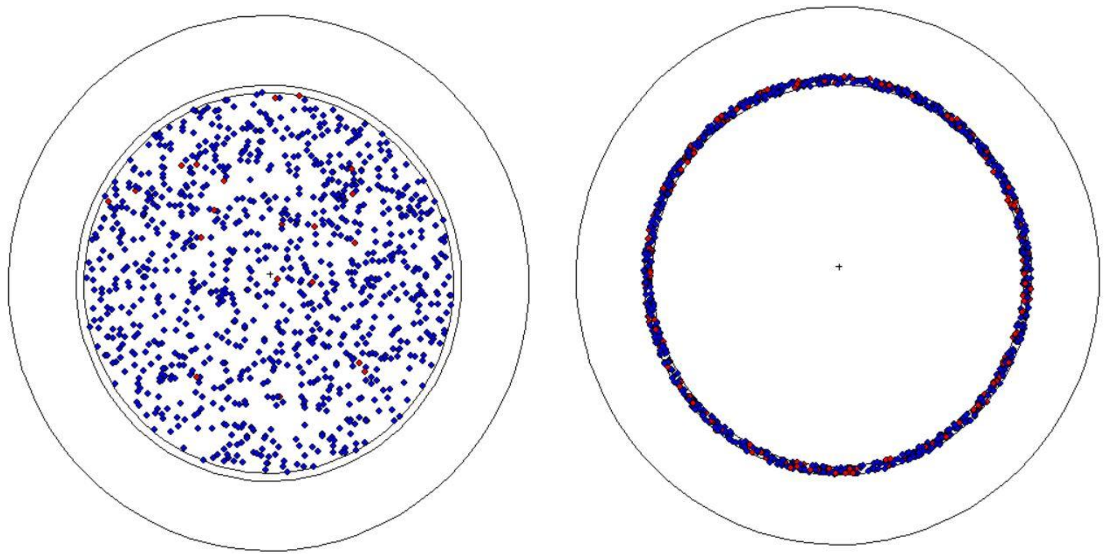

Figure 3.3 1000 source particles were generated to observe the gamma-ray production for 10 at% ^{235}U enriched UF_6 gas (left) and 0.7 at% ^{235}U of UO_2F_2 (right). The blue dots and red dots represent ^{235}U and ^{238}U gamma radiation respectively. The inner circle is the gaseous UF_6 flowing in the pipe. The inner ring is the solid matter of holdup (UO_2F_2) deposited in the pipe wall.

3.2. MCNP6.2 Simulations

3.2.1. Pulse Height Distribution (F8 Tally)

MCNP6.2 has a few detector functions, called as tallies. These tallies are instrumental in deriving the user required output such as particle flux, energy deposition, and pulse height distribution, etc.[4] Each tally has been numbered from 1 to 8 and the pulse height distribution is 8th tally in MCNP6.2, so it is called F8 tally. Tally is also applicable to the various types of particles, neutron, photon, electron, etc. In all the simulations carried out for various parametric variations studied and reported in this thesis, gamma radiation was tallied using F8 tally in order to acquire gamma radiation spectrum from the pipe using NaI detector. The F8 tally works well to obtain the gamma radiation pulse height spectrum.[4] The pulse height tally scores the energy deposited in a detector by each source particle and the secondary particles. That is, the tally collects its history generated in MCNP6.2 once it is deposited in a designated cell, which is the NaI detector in this case.

When F8 tally is used in MCNP6.2 simulation, a set of pulse height bins are created to collect the energy distribution of gamma radiation. Since NaI detector has a low resolution for gamma radiation, it was appropriate to use an energy bin width of 20 keV. An energy bin to score particles that travel through the detector without energy deposition is also created (0 to10^{-4} keV).[4] When the size of bins are different, the accurate pulse height distribution cannot be acquired. This is due to fact that the F8 tally records the scores (energy deposition) within the bins. Therefore, dividing the pulses by each energy bin width is important to obtain the accurate pulse height distribution.

Special treatments for tallies are required in order to represent the Gaussian Energy Broadening (GEB) phenomenon in detectors while using the F8 tally. The energy broadening is represented by the parameter, Full Width at Half Maximum (FWHM). The relationship between FWHM and gamma radiation energy can be written as

$$FWHM = a + b\sqrt{E + cE^2} \qquad \text{Eq.3 [4].}$$

Where,

E: the gamma energy of the radiation (MeV)

a (MeV), b ($MeV^{-1/2}$), and c (MeV^{-1}) are fitting constants.

The three constants can be evaluated through an experiment where an FWHM value each is evaluated for three different gamma radiation energies. These three pairs of

FWHM and gamma energies can used to create three equations with three unknowns thus making it possible to evaluate the constants, a, b, and c. These are called as GEB constants and they vary depending on the type and size of the gamma detector used. For the NaI detector used in this study, these GEB constants were evaluated through an experiment using two gamma radiation sources, ^{60}Co and ^{137}Cs. ^{60}Co emits two gamma radiation energies, 1173 keV and 1332 keV, and ^{137}Cs has emits one gamma radiation energy, 662 keV. Using each photo peak for each of the three energies, three FWHM values were calculated. Thus, the GEB constants a, b, and c were evaluated as -0.00789, 0.06769, and 0.21159, respectively and these values were used in MCNP6.2 input files for the GEB special treatment command.

The results from MCNP6.2 using F8 tally represent pulses within the energy bin and not the gamma-ray counts. In order for the gamma-ray spectrum to be plotted, the gamma-ray counts must be acquired. The results from F8 tally shows pulses occurring including the zero bin. The energy bin width average and corresponding numbers of pulses are plotted as a gamma spectrum. The total source strength of each isotope was multiplied with each average value of pulses in an energy bin to produce the energy dependent gamma-ray counts. For the reasons mentioned above the simulations were conducted as two cases for detecting gamma radiation from UF_6 gas and UO_2F_2 deposit, count spectrum from both simulations of UF_6 gas in the pipe and UO_2F_2 deposit in the internal of the pipe was added to acquire the total gamma radiation counts in each energy bin. The differences in the isotope abundances of uranium in UF_6 and UO_2F_2 were considered while the source strengths were applied to the pulse height distribution obtained from the MCNP simulations. The gamma radiation spectrum thus obtained is shown in Figure 3.5. Figure 3.5 shows the total gamma radiation counts obtained for the centrifuge pipe for a typical case as a function of energy and significant energy peaks at 186 keV and 1001 keV.

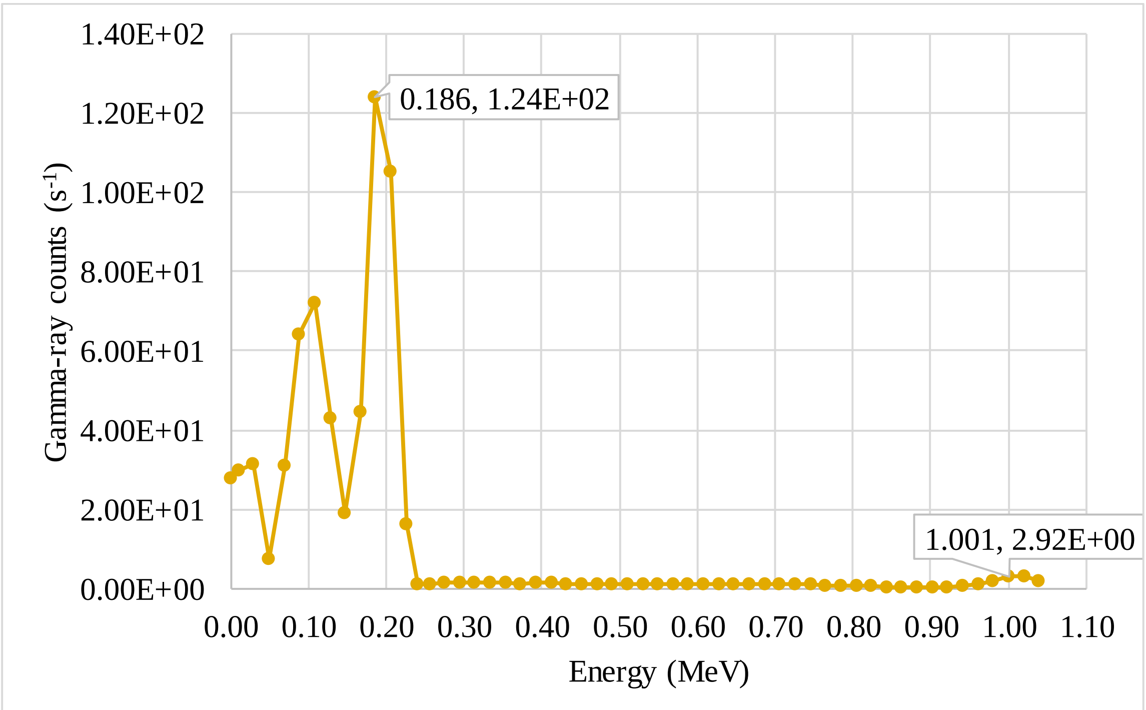

Figure 3.4 The NaI gamma-ray spectrum using F8 tally in MCNP6.2 code when 8 at% ^{235}U enriched UF_6 flows in the pipe deposited with 150g/m of holdup material (0.7 at% ^{235}U UO_2F_2)

3.2.2. Modification in ^{235}U Abundance of Holdup Material

For determining the effect of holdup on the gamma-ray spectroscopy, it is important to modify ^{235}U abundance in UO_2F_2 because the holdup material can make the gamma radiation counts in the NaI detector higher or lower depending on the abundance of ^{235}U in the material. In the simulations, the amount of ^{235}U content was varied from 0.3 at% to 5 at%. Figure 3.5 shows the difference in gamma radiation rendering when the holdup material has 0.7 at% or 5 at% ^{235}U in UO_2F_2. These two ^{235}U abundance of the holdup material had been mainly applied for the simulations. In the case of study of uranium diversion, 0.7 at% ^{235}U of UO_2F_2 was generally considered as the default value of the atomic percentage of uranium in the holdup material to analyze the potential uranium diversion scenario depending on the ^{235}U enrichments of UF_6 flowing inside the pipe. For the study on the frequency of inspection, 5 at% ^{235}U of UO_2F_2 was assumed as the enrichment of holdup material to determine whether an unnecessary inspection could happen when depleted, natural, or low enriched uranium of UF_6 flows in the pipe.

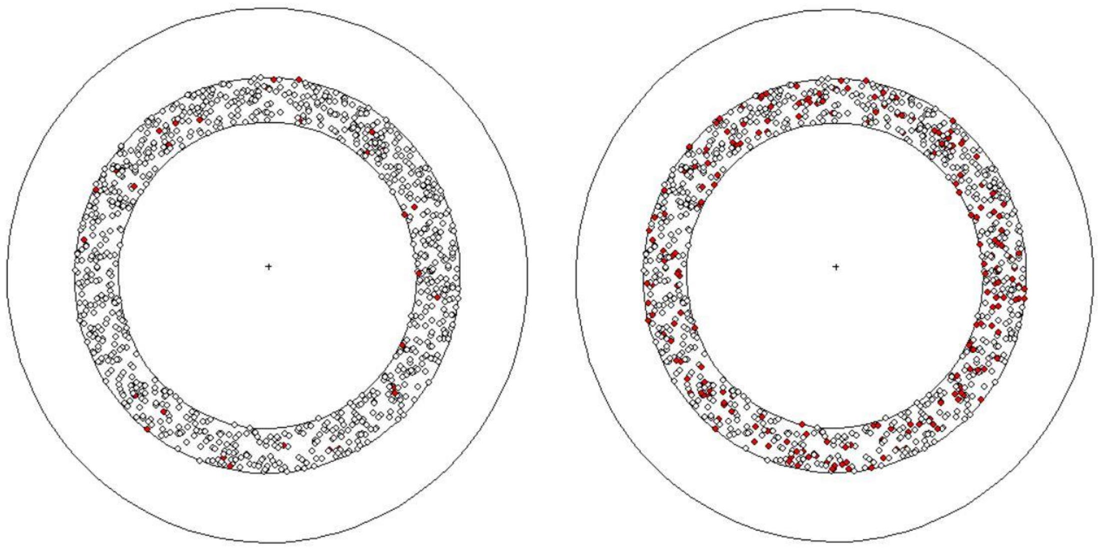

Figure 3.5 1000 Source particles in holdup material rendered by the Vised software. The red dots are ^{235}U and the white dots are ^{238}U. The inner ring represents the source particles that are generated in UO_2F_2 deposited on the pipe wall; 0.7 at% ^{235}U of UO_2F_2 is located on the left-side of the figure and 5 at% enriched ^{235}U of UO_2F_2 is located on the right-side of the figure.

3.3. Study on the Separative Work Units Variation

The SWU is directly connected with the energy usage in a uranium enrichment facility, so SWU needs to be the same value in order for concealing uranium diversion. A relatively high use of electricity in a GCEP producing LWR useable uranium (< 5% ^{235}U enrichment) within a short term would indicate the potential production of HEU in the facility. Therefore, SWU variation calculations were conducted to check if any manipulation of SWU makes it possible for the diversion of uranium within the conversion time stipulated by the IAEA without being detected. In order to obtain a desired amount of enriched UF_6, a certain amount of UF_6 (usually natural uranium) is required to be fed into the centrifuges and UF_6 as depleted uranium to be sent out as tails (waste). By using these three masses (feed, tails, and product) and the corresponding ^{235}U weight percentages, a mass balance equation can be written as

$$x_F M_F = x_P M_P + x_T M_T \quad \text{Eq.4}$$

Where:

x_F: the ^{235}U concentration of the feed (wt%)

M_F: the mass of the feed (kg)

x_P: the ^{235}U concentration of the product (wt%)

M_P: the mass of the product (kg)

x_T: the ^{235}U concentration of the tail (wt%)

M_T: the mass of the tail (kg)

The Eq.4 using conservation of mass ($M_F=M_P+M_T$) can be written as

$$M_F = \frac{x_P - x_T}{x_F - x_T} M_P \quad \text{Eq.5}$$

By using Eq.5, the quantity of feed and waste can be calculated if the quantity of product and the concentration of product, feed, and tail are known using

$$V(x) = (1-2x)ln\left(\frac{1-x}{x}\right) \quad \text{Eq.6}$$

Where:

x: the enrichment concentration (%)

Separative work can be expressed in terms of a function V(x) as shown in the Eq.6. This is also called as elementary value function, or separative potential.

Then, SWU can be calculated using the quantity of feed, product, and tail and Eq.6 using

$$SWU = M_P V(x_P) + M_T V(x_T) - M_F V(x_F) \quad \text{Eq.7}$$

Under the condition that the quantity of product is 1 kg, the SWU calculation using Eq.7 was conducted by varying the enrichment of feed and waste so that the values of SWU will be the same even though the enrichment of UF_6 product is different.

4. RESULTS AND DISCUSSION

4.1. Uranium Diversion

4.1.1. Estimation of Holdup Required

The MCNP results of pulse height distribution of gamma radiation in the NaI detector obtained using the F8 tally were used to construct the gamma radiation spectra due to UF_6 gas and UO_2F_2 deposit in centrifuge pipe. Please note that the spectra simulated is only due to gamma radiation energies of 186keV from ^{235}U and 1001keV from ^{238}U. In order to determine how much uranium deposit (holdup) is required in the internals of the centrifuge pipe in order to conceal a higher value of ^{235}U enrichment of UF_6 than declared, the gamma radiation counts of ^{235}U and ^{238}U from the spectra were calculated. The counts considered come from the area under the ^{235}U photo peak which ranges from 146keV to 226keV and the ^{238}U peak ranging from 961keV to 1041keV, the ranges are due to the gaussian energy broadening for gamma radiation energy in the detector discussed in chapter 3. A sample gamma radiation spectrum and how the photo peak energy deposition counts are accounted for is shown in Figure 4.1.

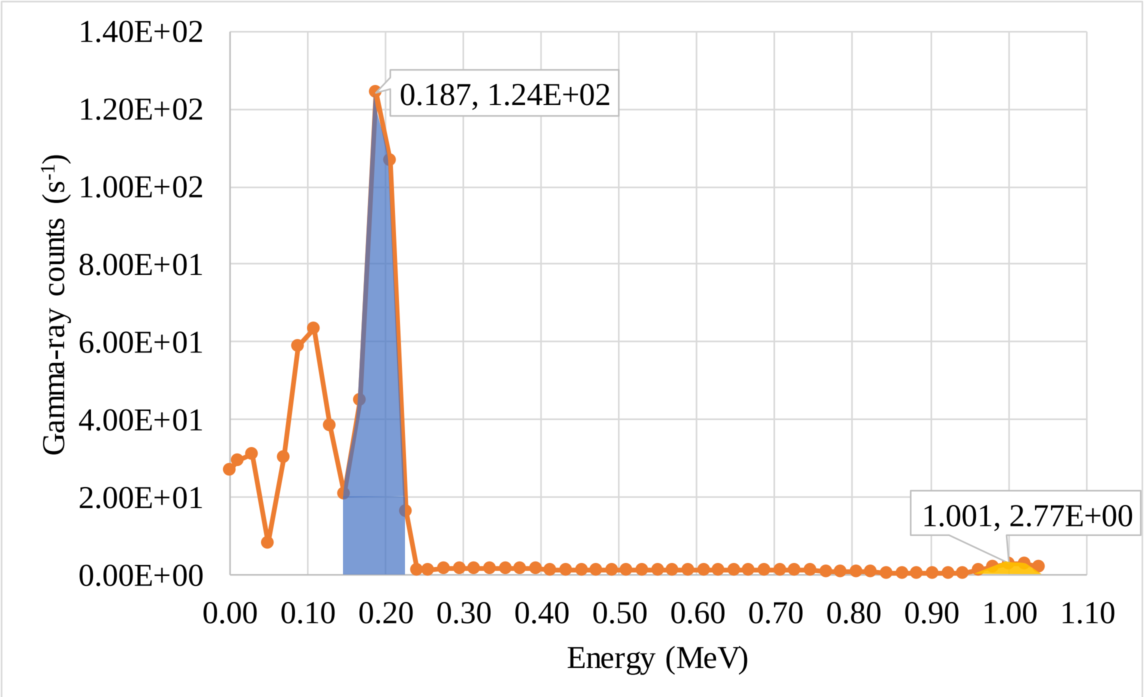

Figure 4.1 The colored areas (blue for ^{235}U and yellow for ^{238}U) are taken into consideration due to Gaussian broadening when calculating the total gamma radiation counts.

From the total gamma radiation count rates calculated from MCNP simulations, Gaussian distribution analyses (as described in section 2.4) were introduced to estimate the amount of potential uranium diversion. In all the simulations, the false alarm probability, α was set to 5% and the associated nondetection probability, β was estimated. Since the case assumed had more than 5 at% ^{235}U enrichment, the gamma radiation count rates recorded are higher than the one of reference, which is 5 at% enrichment of UF_6 and is colored as blue curve in each case. Hence, it gives a misuse signal when the facility is inspected. However, the concealment of a misuse signal can be possible when holdup has low concentration of ^{235}U and is deposited enough to conceal it. Therefore, as the amount of holdup varied, an amount of holdup that will make exceed the nondetection probability, β greater than 20% was determined when LEU greater than 5 at% enrichment is under production. In the case of ^{235}U enrichment measurement of LEU using NaI detector, the uncertainty (ITV) of detection has been established by the IAEA. The ITV is set at a relative error (inclusive of random and systematic errors) of 5.8%. Accordingly, the ITV of 5.8% are taken into consideration as 1-σ in Gaussian distributions.[1] Figures 4.2 and 4.3 respectively shows the Gaussian distributions of 6 at% ^{235}U UF_6 gas and 10 at% ^{235}U UF_6 gas with different amount of holdup for the case study where 0.7 at% ^{235}U enrichment is assumed to present in the holdup.

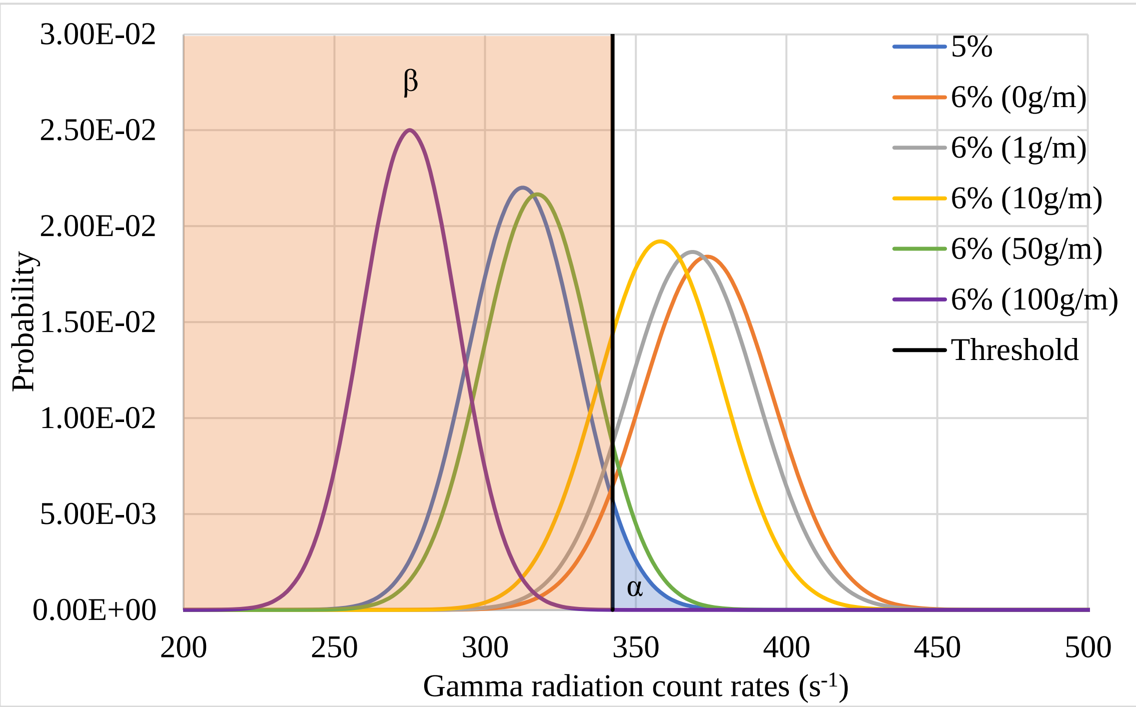

Figure 4.2 Gaussian distributions of gamma radiation count rates when 6 at% UF_6 is flowing in the pipe and a certain amount of 0.7 at% holdup is deposited in the internals of the pipe.(on the legend) α and β are applied.

The more the amount of holdup (0.7 at% ^{235}U) in the internals of the pipe, the less gamma radiation count rates for 6 at% UF_6 enrichment misuse are recorded as depicted in

Figure 4.2. In Figure 4.2, The left-side of threshold is filled by orange color and named β because the areas under the gaussian distributions are the probability β of each distribution. The probabilities are listed in Table 4.1 when different amount of holdup is accounted for.

Table 4.1 Changes in β when 6 at% UF_6 flows in the pipe and the amount of holdup (0.7 at%) is increasing.

The amount of holdup (g/m)	0	1	10	50	100
β (%)	7.37	10.80	22.34	91.24	100.00

Table 4.1 shows the probability β is above 20% when about 10g/m 0.7 at% holdup is deposited and suggests that 6 at% UF_6 flowing in the pipe with over 10g/m 0.7 at% holdup inside will not be inspected even though nondetection probability is greater than 20%. Figure 4.3 and Table 4.2 shows a similar trend. Since Figure 4.3 and Table 4.2 are assumed 10 at% UF_6, the required holdup amount is higher than the case of 6 at% UF_6. When 10 at% UF_6 flows in a GCPE pipe with over 170g/m 0.7 at% holdup, the facility would not be inspected due to higher probability of β than 20%.

Similarly, Gaussian distributions used in uranium diversion analysis are provided for various parametric variations of ^{235}U enrichment and UO_2F_2 holdup in Appendix A.

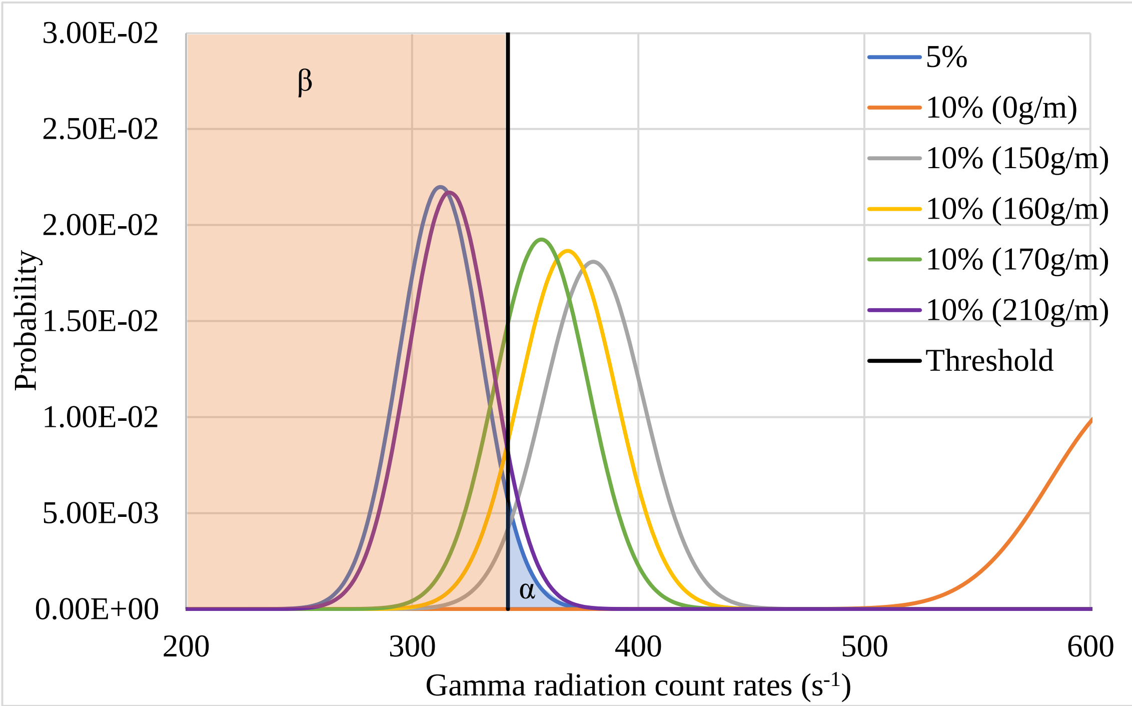

Figure 4.3 Gaussian distributions of gamma radiation count rates when 10 at% UF_6 is flowing in the pipe and a certain amount of 0.7 at% holdup is deposited in the internals of the pipe.(on the legend) α and β are applied.

Table 4.2 Changes in β when 10 at% UF_6 flows in the pipe and the amount of holdup (0.7 at%) is increasing.

The amount of holdup (g/m)	0	150	160	170	210
β (%)	0	4.32	10.83	23.63	91.87

4.1.2. Potential Uranium Diversion Calculation

The amounts of holdup required to conceal higher than the declared ^{235}U enrichment (5 at%) in UF_6 has been evaluated and are listed in Table 4.3. Also, the values listed in Table 4.3 are the corresponding amount of uranium mass in UF_6 in the pipe length section that has been modeled for this study.

Table 4.3 Mass of uranium calculated at each ^{235}U enrichment showing an increase in ^{235}U content (in potentially diverted mass) as well as the corresponding amount of holdup required for concealing the higher ^{235}U enrichment.

^{235}U enrichment (at %)	The amount of holdup required (g/m)	Uranium mass in UF_6 (g) [25][1]	^{235}U mass in UF_6 (g)
5	0	310.17	15.32
6	10	307.68	18.24
7	60	295.25	20.43
8	100	285.31	22.56
9	135	276.60	24.61
10	165	269.14	26.61

The ^{235}U mass was calculated by multiplying the uranium mass with ^{235}U weight percent, not atomic percent and they are respectively 4.94 wt%, 5.93 wt%, 6.92 wt%, 7.91 wt%, 8.90 wt%, and 9.89 wt% for the corresponding 5 at%, 6 at%, 7 at%, 8 at%, 9 at%, and 10 at%.

[1] The density of UF_6 gas is 4.68 g/cm^3

Table 4.4 Uranium daily flow rate calculated from UF_6 flow rate; adapted from [17].

Stage	UF_6 flow rate (g/s)	U flow rate (g/s)	U flow rate (kg/day)
Feed	0.0317	0.0212	1.835
Product	0.0142	0.0095	0.822
Tail	0.0175	0.0117	1.013

Table 4.4 shows the daily uranium flow rate in a centrifuge, which is used for the calibration of the results from the MCNP6.2 simulations. The simulations have been executed under the assumption that UF_6 exists only in the pipe length section modeled, so the amount of UF_6 used in the mass calculation is different with UF_6 mass in a centrifuge in the real world. For this reason, the calibration is necessary to determine the potential diversion of uranium and predict how many centrifuges are required to divert 1-SQ of LEU. In order to determine uranium diversion, uranium mass flow rate was calculated by multiplying the uranium fraction in UF_6, 0.67, with UF_6 flow rate. The unit for the flow rate is changed from g/s to kg/day. The ^{235}U mass difference (Ref. Table 4.3) between mass at 5 at% compared to that for a given ^{235}U enrichment listed in Table 4.3 was calculated. Based on these uranium mass flow rate and ^{235}U mass difference inputs, the number of centrifuges required to divert 1-SQ of LEU was evaluated and is shown in Table 4.5. By diving the uranium product flow rate (kg/day) with each uranium mass in UF_6 listed in Table 4.3, the calibration values were acquired and are given in Table 4.5. Since the simulation and calculation focuses on enriched uranium product, which means product pipe, or it can be the entire enriching stages from the perspective of facility, the uranium flow rate at product pipe was used as the parameter to calculate the calibration values.

Table 4.5 ^{235}U potential diversion at the last product stage.

^{235}U enrichment (at %)	^{235}U mass difference (g)	Calibration value (/day)	^{235}U potential diversion at the product stage (g/day)	^{235}U potential diversion at the product stage (kg/3mo)
5	0	0	0	0
6	2.92	2.67	7.80	0.70
7	5.10	2.78	14.21	1.28
8	7.24	2.88	20.85	1.88
9	9.28	2.97	27.59	2.48
10	11.28	3.05	34.47	3.10

The potential amount of uranium diversion due to holdup was obtained and is listed in Table 4.5. The potential amount of uranium diversion (g/day) was multiplied with the ^{235}U mass differences and the corresponding calibration values. The unit (g/day) was changed to evaluate the possibility of uranium diversion in 3 months because IAEA has set the minimum conversion time for LEU as 3 months and maximum as 1 year. The timeliness goal set by IAEA for detection of LEU diversion is no more than 1 year. However, 3 months of conversion time was postulated to minimize the amount of potential uranium diversion in the calculation. 1-SQ, 75 kg of ^{235}U, was divided by the amount of potential diversion in 3 months (kg/3mo). However, the amount of potential diversion in Table 4.5 represents that mass comparison between the last stage and the stage at 5 at%. In other words, the values did not take into consideration the other stages before the final product stage. Please note that a correction is needed since the enrichment has increased to 9 at% in this example compared to the 5 at% through more product stages. In order to make the correction, the number of enriching stages must be taken into account. For acquiring the number of stages, the ideal cascade is assumed as the model. The number of stages at each ^{235}U enrichment goal is listed in Table 4.6.

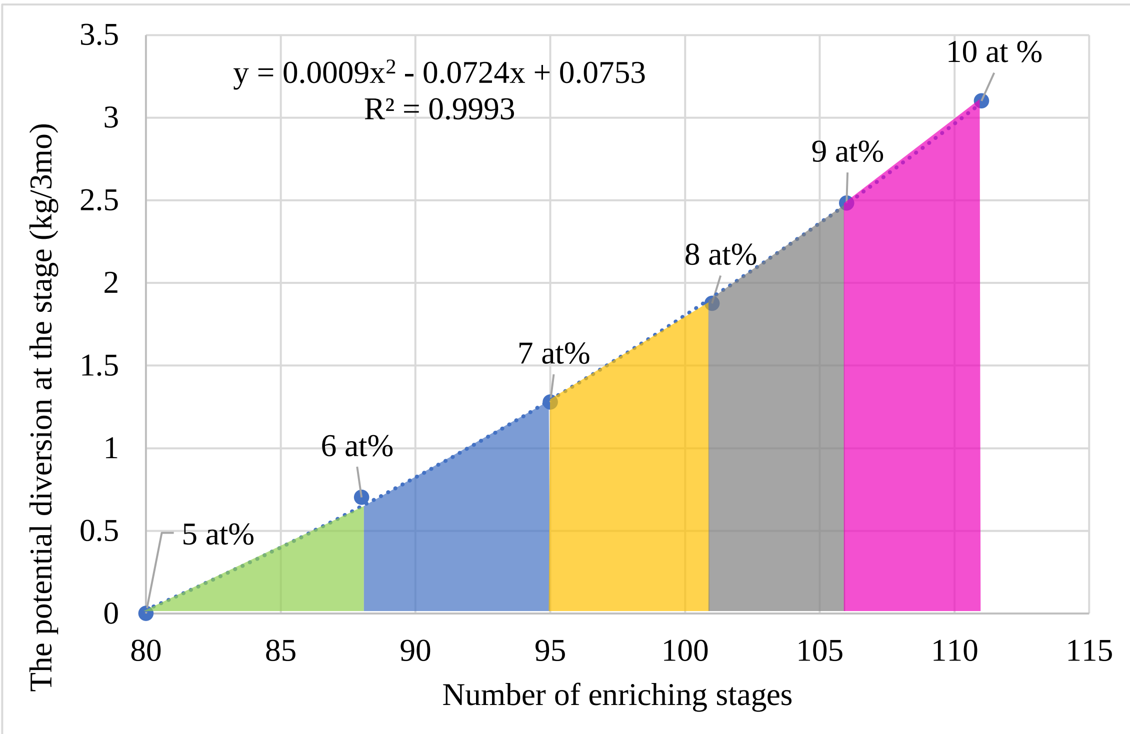

Figure 4.4 The number of enriching stages with respect to the potential diversion at the stage shows polynomial and the area under the graph to calculate the potential diversion in the cascade. Trend lines and the coefficient of determination (R^2) are also added on the graph.

The Figure 4.4 shows curve depicting the diversion of ^{235}U as a function of number of enriching stages. The number of centrifuges each stage is not predictable in the ideal cascade unless the total number of centrifuges is known. Thus, the calculations were conducted under the assumption that the all the stages after 5 at% ^{235}U enrichment occurred has the same number of centrifuges per stage. A new approach of where the squared-off cascade is combined with the ideal cascade is proposed to calculate how many centrifuges are required to divert 1-SQ of uranium between the uranium diversion range in GCEP due to holdup, which is between 5 at% and 10 at%. Figure 4.5 shows the new cascade model proposed to make the centrifuge calculation simple.

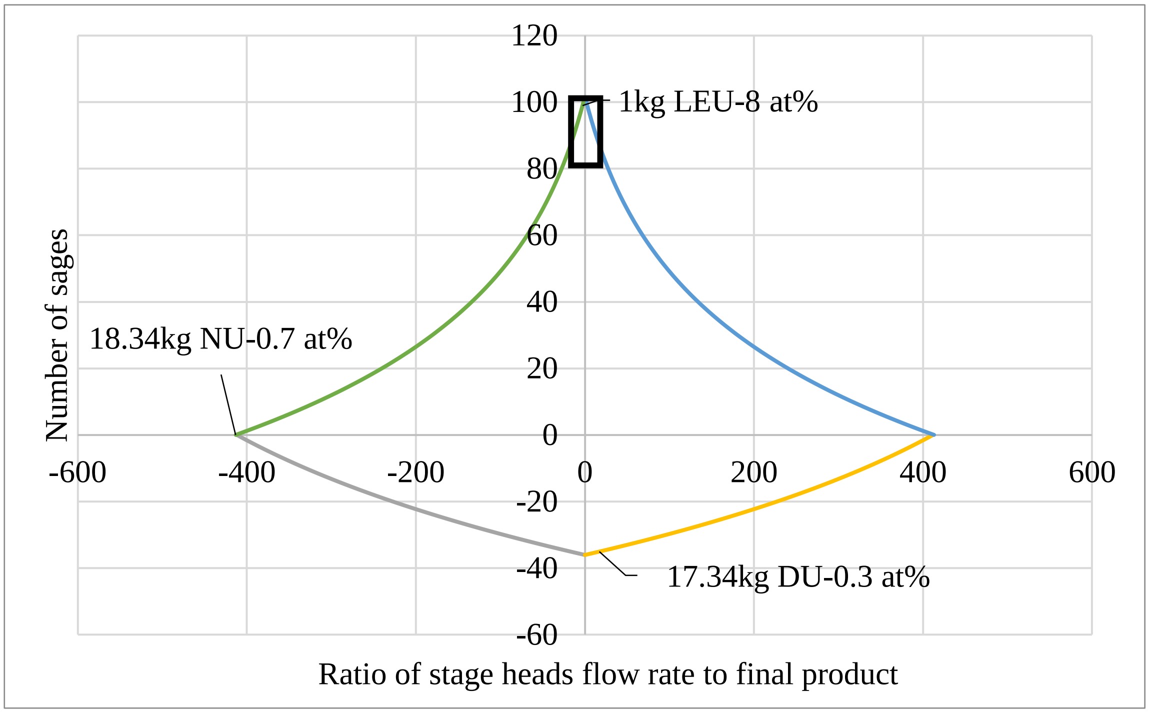

Figure 4.5 The cascade model modified for the corrected potential diversion calculation when 8 at% ^{235}U is the desired product.

Except for the stage range from 81 to 100, every stage follows the ideal cascade. The enriching stages from 81 to 100 follow the squared-off cascade, which is the potential uranium diversion range and is represented as a black rectangle. Therefore, the centrifuge calculation was conducted under the assumption that the uranium diversion is occurring from the square cascade section shown in Figure 4.5 and the potential amount of uranium diversion should be divided by 1-SQ of uranium, then the centrifuges required is acquired at each stage after 5 at% ^{235}U processing stage, which is the enriching stage number 80 in this case. The total centrifuges required after 5 at% ^{235}U processing stage are also calculated by multiplying the number of centrifuges required at each stage and the number of enriching stages in the diversion range and is shown in Table 4.6. For example, the potential diversion in the cascade is obtained by the summation of the green triangle and blue trapezoid in Figure 4.4 and the number of centrifuges at each cascade are deduced by dividing the potential diversion in the cascade with 1-SQ of uranium, 75 kg of ^{235}U. The results are listed in Table 4.6.

Table 4.6 The potential diversion and the number of centrifuges required after the correction.

^{235}U enrichment (at %)	Potential diversion at the stage (kg/3mo)	Enriching stages	^{235}U potential diversion in the cascade (kg/3mo)	Centrifuges required at each stage	Centrifuges required in the cascade after 5 at% ^{235}U
5	0	80	0	N/A	N/A
6	0.70	88	2.81	27	216
7	1.28	95	9.74	8	120
8	1.88	101	19.21	4	84
9	2.48	106	30.11	3	78
10	3.10	111	44.07	2	62

Please note that an increment of 216 centrifuges are required to produce 6 at% enrichment of UF_6 and GCEP consists of thousands of centrifuges. It means 216 centrifuges is not a significant number. The diversion of LEU could happen in 3 months due to holdup based on the study presented here.

4.1.3. Results from Feed and Waste Manipulation

Table 4.7 SWU, UF_6 quantity, the number of stages, and corresponding ^{235}U wt% of product under no change of ^{235}U wt% in feed and waste.

^{235}U wt%			SWU	UF_6 quantity (kgU)			Number of stages	
Product	Feed	Waste		Feed	Product	Waste	Stripping	Enriching
4.940	0.711	0.296	7.13	11.20	1	10.20	35	80
5.929	0.711	0.296	9.09	13.57	1	12.57	35	88
6.918	0.711	0.296	11.08	15.96	1	14.96	35	95
7.907	0.711	0.296	13.09	18.34	1	17.34	35	101
8.896	0.711	0.296	15.12	20.72	1	19.72	35	106
9.886	0.711	0.296	17.16	23.10	1	22.10	35	111

Section 4.1.2 focuses on up to which ^{235}U enrichment of the LEU product diversion can be concealed due to holdup occurring in the pipe. the evaluation described in section 4.1.2 indicates that the concealment of enriching more than the typical 5 at% enrichment of ^{235}U is possible. However, the concealment by only considering the product pipe is not enough in diversion analysis of 1-SQ of uranium in 3 months. The manipulation of ^{235}U enrichment in feed and waste pipes also need to be considered. In Table 4.7, the study on the LEU (^{235}U enrichment > 5 at%) production are listed, which includes SWU, throughput, and the number of stages. The SWU value and UF_6 mass were calculated using equations 4 and 6. The UF_6 mass of the product is set to 1 kgU. As it is seen in Table 4.7, the SWU, UF_6 mass of feed and waste, and the number of enriching stages increase

as the ^{235}U enrichment increases even when the ^{235}U enrichment of feed and waste is kept constant The SWU value is directly connected with electricity use, so LEU (^{235}U enrichment > 5 at%) production will easily be detected if the electricity expense is inspected.

Table 4.8 SWU, UF_6 quantity, the number of stages, and corresponding ^{235}U wt% of product with changes of ^{235}U wt% in feed and waste.

^{235}U wt %			SWU	UF_6 quantity (kgU)			Number of stages	
Product	Feed	Waste		Feed	Product	Waste	Stripping	Enriching
4.940	0.711	0.296	7.13	11.20	1	10.20	35	80
5.929	0.850	0.350	7.24	11.16	1	10.16	35	81
6.918	0.950	0.400	7.61	11.85	1	10.85	35	83
7.907	1.100	0.450	7.62	11.47	1	10.47	36	83
8.896	1.300	0.550	7.05	11.13	1	10.13	35	81
9.886	1.400	0.600	7.32	11.61	1	10.61	34	83

In Table 4.8, the SWU values, UF_6 mass, and the number of enrichment stages are listed for the parametric variations (manipulation) of ^{235}U enrichment in feed and waste. Even though the ^{235}U enrichment of product increases, the SWU, UF_6 mass of feed and waste, and the number of stages do not change as much. If a GCEP is operating under this condition given in Table 4.8, the undeclared uranium diversion could occur.

To determine whether ^{235}U enrichment of feed and waste can concealed due to holdup, more simulations using MCNP6.2 were executed. A new model is created where the feed pipe contains 0.3 ^{235}U at% in holdup and 1.5 ^{235}U at% in UF_6 gas as well as, the waste pipe contains 0.3 ^{235}U at% in holdup and 0.6 ^{235}U at% in UF_6 gas. This new model was used to carry out MCNP simulations Unlike LEU abundance detection, the relative errors of Depleted Uranium (DU) and Natural Uranium (NU) are 22 % and 10%, respectively.

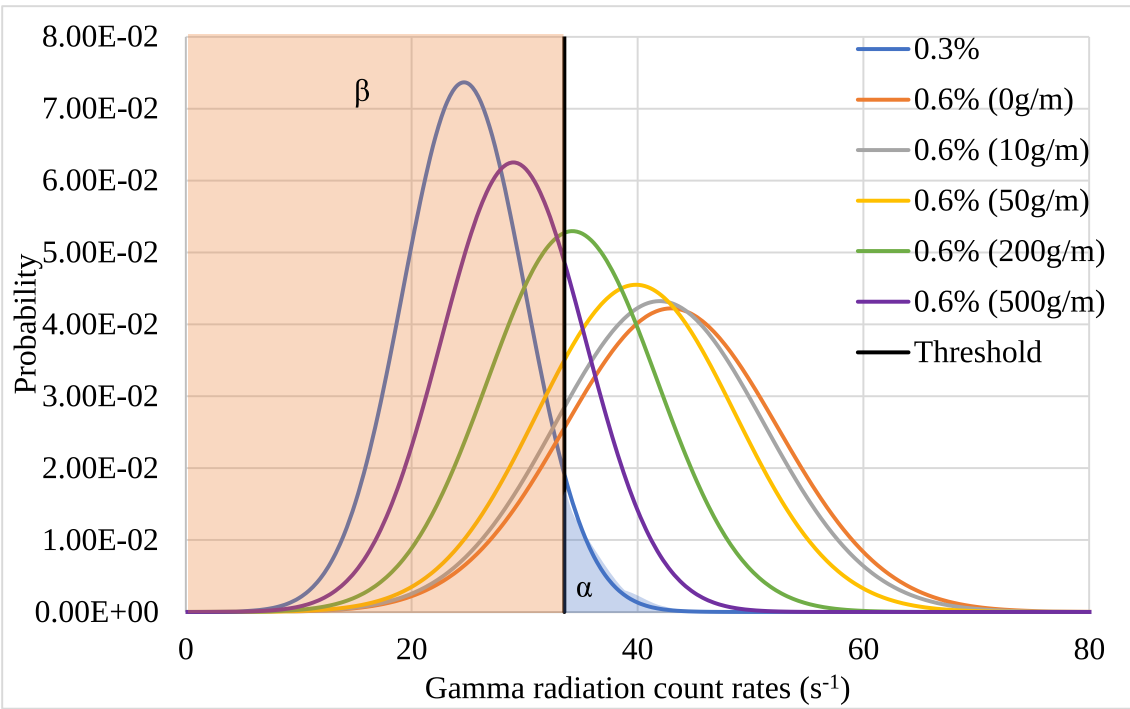

Figure 4.6 Gaussian distributions of ^{235}U gamma radiation count rates when 0.6 at% ^{235}U of UF_6 is flowing in the pipe and 0.3 at% ^{235}U of holdup is deposited. The amount of holdup varies.

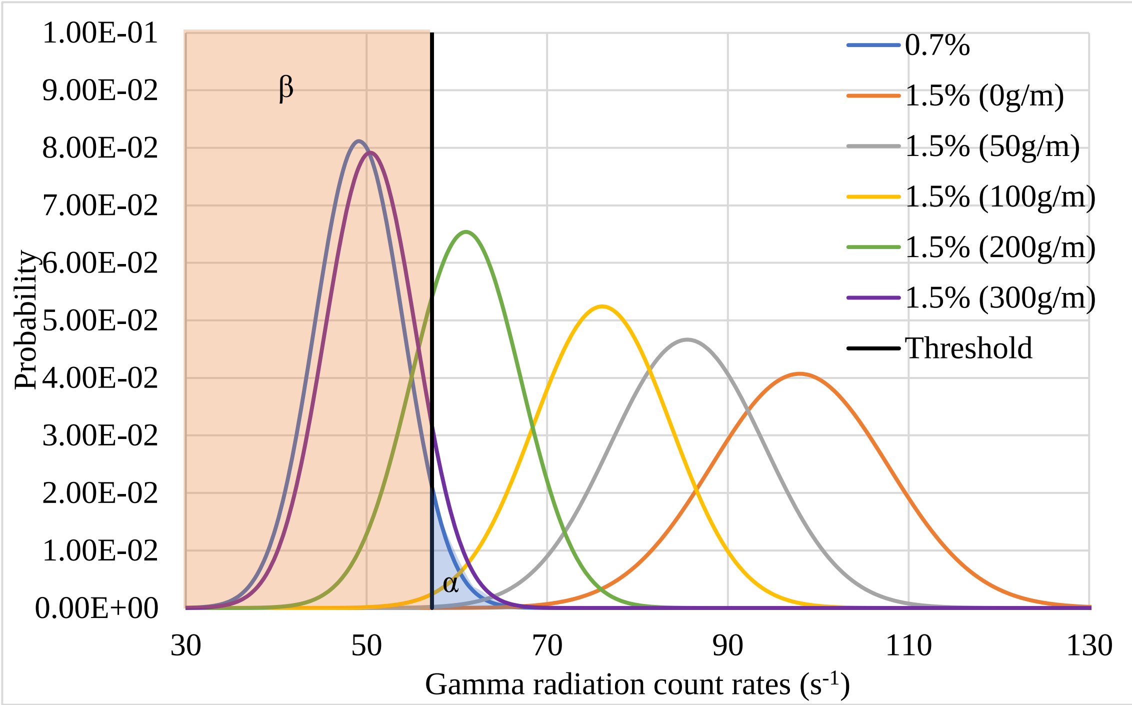

Figure 4.7 Gaussian distributions of ^{235}U gamma radiation count rates when 1.5 at% ^{235}U of UF_6 is flowing in the pipe and 0.3 at% ^{235}U of holdup is deposited. The amount of holdup varies.

Figure 4.6 and Figure 4.7 show Gaussian distributions of ^{235}U gamma radiation count rates. Each figure has commonly 0.3 at% ^{235}U of holdup material, but ^{235}U concentration of UF_6 for each figure is different as ^{235}U enrichments were 0.6 at% and 1.5 at%, respectively. From the analyses, 50g/m of holdup is required for the concealment of 0.6 at% ^{235}U and 200g/m of holdup is required to hide 1.5 at% ^{235}U. This feature indicates that low ^{235}U concentration of UF_6 is also difficult to detect by gamma spectroscopy alone if the amount of holdup exists enough to make nondetection probability more than 20%. As a result, the manipulation of ^{235}U abundance in feed and waste could also lead to nondetection of GCEP misuse.

4.1.4. Load-Cell Based System Calculation

Load-Cell Based weighing System (LCBS) is one of the NDA measurements to measure mass of materials. Currently, portable LCBS is widely used to determine the mass of UF_6. [26] Since the uranium product weighing and enrichment measurement are for materials in its bulk form and not an item, a relative error in the bulk material measurements need to be applied for material accounting. The measurement error values (ITVs) recommended by IAEA in this case of uranium bulk mass measurement is 0.07 % (inclusive 0.05% of system error and 0.05% of random error). Using the relative error of the weighing system and the fact that 1kg of UF_6 reacting with 0.1 kg water yields to a deposition of 0.88kg of UO_2F_2[27] in the internals of the pipe, the amount of holdup was able to be estimated and is shown in Figure 4.10. This indicates that the estimation of amount of UO_2F_2 can be produced in the internals of the pipe when UF_6 reacts with water. The blue dots in Figure 4.8 show the relation between the amount of holdup created and the UF_6 loss. The linear plot comes from the aforementioned discussion that 1kg of UF_6 can produce 0.88 kg of UO_2F_2. The yellow line in Figure 4.10 is the maximum acceptable amount of UF_6 loss in 3 months to draw the conclusion that there is no diversion occurring in GCEP. According to USNRC, typical throughput for a modern gas centrifuge is 1.7kgU/day, which is about 926kg UF_6/year. (multiplying 365 days and dividing by 0.67, uranium ratio in UF_6) The measurement uncertainty is 0.07%, so the amount is about 0.162 kg of UF_6 in three months. This three-month uncertainty, 0.162kg, of UF_6 is equivalent to about 143g of holdup, which means 143g of UO_2F_2 can be produced without the detection of uranium diversion in one centrifuge within 3 months using LCBS. That amount, 143g, of holdup can be rephrased as 286g/m if the length of pipe has been assumed to be 50 cm modelled in this simulation and all holdup generated in the centrifuge is deposited in the internal of the pipe. (the thickness of holdup can be different depending on the length of pipe) The results prove the aforementioned analyses about holdup and it could be possible theoretically. Therefore, 1-SQ diversion of uranium can occur without detection using the NaI detector if an operator of GCEP have an intention to divert by concealing it within the holdup.

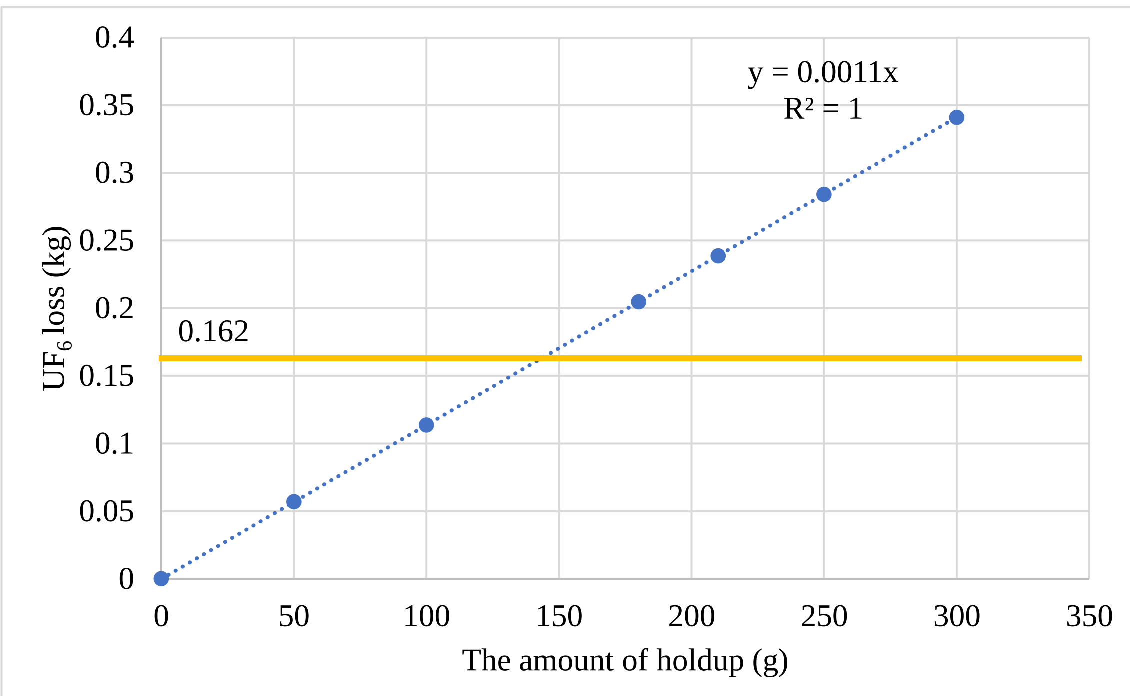

Figure 4.8 The production amount of holdup in accordance with UF_6 loss coming in contact with water (blue dots) and the maximum acceptable UF_6 loss amount in 3 months (yellow line).

4.1.5. Gas Flow Meters Nondetection

Once the material holdup is deposited in the internals of pipe, the cross-section of gas flowing in the pipe will be reduced. Accordingly, the velocity of gas will increase. If the velocity of gas increase is substantially high, then it will be noticeable. Hence, the analysis on gas flow meters was performed to evaluate what amount of holdup can be hidden without getting noticed by the flow velocity changes. In GCEP, gas flow meters are built in the facility, but what type of gas flow meters have been used is not directly available in literature. Therefore, the general measurement error of gas flow meters was assumed as 5% in this study. Usually, the measurement error of gas flow meters is less than 2%. When 1 error bar is considered, the maximum error of gas flow meters is 4% since it can be overlapped. Also, it has about 1% of repeatability error, so maximum 5% error was considered. Since the inner radius of pipe was assumed as 0.79cm, the amount of holdup inside the pipe corresponding to a 5% flow rate error not being noticed could be determine.

Table 4.9 The increment of velocity of UF_6 with the corresponding amount of holdup.

The amount of holdup (g/m)	Inner pipe radius with holdup (cm)	Inner cross-section with holdup (cm^2)	The portion of area of holdup (%)	The ratio of velocity of UF_6 with holdup
10	0.7868	1.9450	0.80	1.008
15	0.7852	1.9371	1.20	1.012
30	0.7805	1.9136	2.40	1.025
50	0.7740	1.8822	4.00	1.042
60	0.7708	1.8665	4.80	1.050
100	0.7577	1.8037	8.01	1.087
286	0.6937	1.5117	22.90	1.297

Table 4.9 shows the ratio of velocity of UF6 gas under the existence of holdup and suggest that maximum 60 g/m of holdup can be deposited in the internals of pipe without being detected given the unnoticeability in flow rate is about 5% buried in the flow rate measurement error

In section 4.1, it is show that 286g/m of holdup can be deposited in a centrifuge with LCBS calculation error alone, but it will block about 23% of inner area of pipe and increase the UF_6 gas velocity by about 30%, so gas flow meter will detect it. During uranium diversion analyses, three parametric measurement methods are considered and gives maximum 60g/m of holdup production boundary. This amount of holdup can conceal up to 7 at% ^{235}U of UF_6 due to holdup.

4.2. Unnecessary Inspection

Safeguards risk coming from holdup is not just only the uranium diversion, but also that may call for unnecessary inspections. The unnecessary inspection here means that nondetection probability is less than 20% and gamma radiation count rates is recorded less or more than the case of reference, but in fact, no diversion has occurred or facility had not been misused. This unnecessary inspection due to holdup result from two cases, one is that a relatively high enriched uranium of holdup makes gamma signal strong and the other is that a relatively low enriched uranium of holdup makes gamma signal weak. Study on scenario about unnecessary inspection had been conducted. Plots used in these analyses are attached in Appendix B.

4.2.1. Weak Gamma Signal from Holdup

The case study that holdup is deposited as 0.7 at% ^{235}U was performed because the total flow rate of natural uranium in GCEP is extremely high compared to the other stages. Thus, the number of centrifuges built in GCEP is the most, so it is easy to produce holdup. The significance of this study is that the goal ^{235}U enrichment of LWR, 3 wt% to 5 wt%, can cause false alarm easily with a relatively low amount of 0.7 at% ^{235}U holdup. Table 4.10 shows the required holdup amount to indicates that diversion has occurred in the facility in fact no material is missing with corresponding ^{235}U enrichment of UF_6.

In Table 4.10, the estimated ranges of holdup amount are listed. This shows about 50g/m of 0.7 at% ^{235}U holdup can cause false alarm when 3, 4, and 5 at% of UF_6 are flowing in the pipe. Indeed, the exact amount of holdup, which make nondetection probability 20%, is not studied in this thesis, but it is possible to estimate what amount of holdup can cause false alarm with Table 4.10 and Figure 4.9. In Figure 4.9, the more amount of holdup exists in the pipe, the less gamma radiation count rates are recorded. This cause to low nondetection probability and unnecessary inspection of facility.

Even considering the boundary of holdup, 3 at% to 5 at% enrichment of UF_6 can call for an inspection which shall be avoided if the holdup is appropriately quantified.

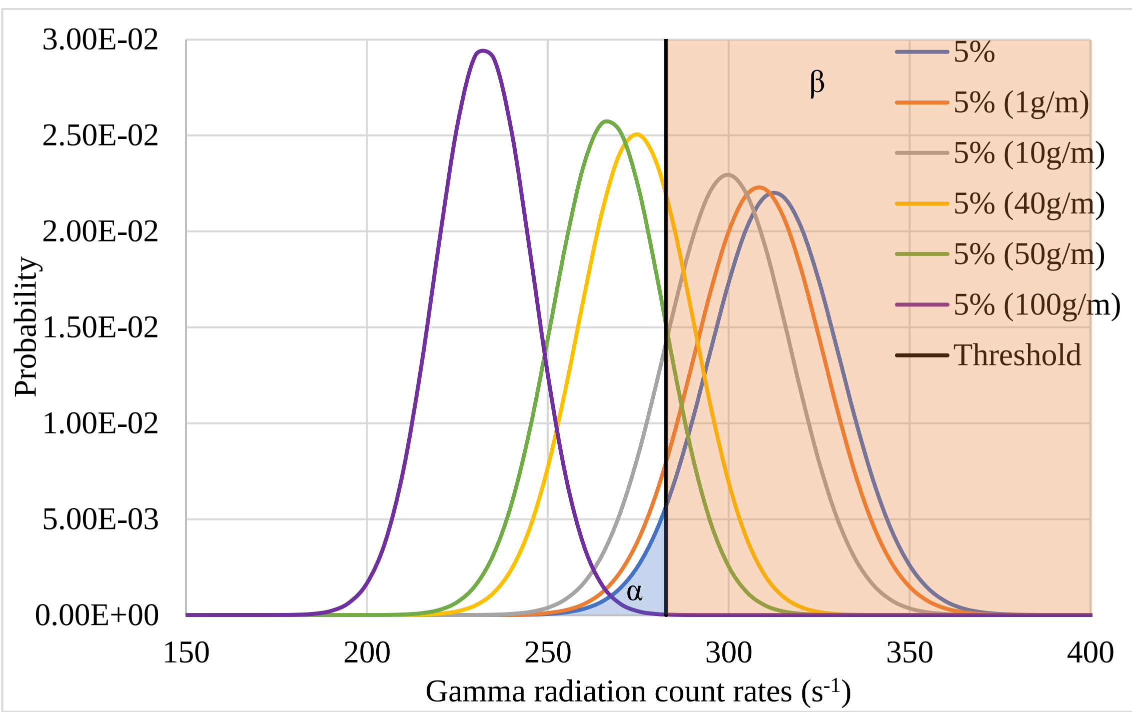

Figure 4.9 Gaussian distributions of gamma radiation count rates when 5 at% UF_6 is flowing in the pipe and a certain amount of 0.7 at% holdup is deposited in the internals of the pipe.(on the legend) α and β are applied.

Table 4.10 The estimated ranges of nondetection probability and the amount of holdup to cause false alarm.

^{235}U enrichment of UF_6 (%)	Nondetection probability (%)	The amount of holdup to cause false alarm (g/m)
1	12.83 ~ 27.51	200 ~210
2	21.21	75
3	26.37 ~ 14.44	50 ~ 60
4	18.65	50
5	14.98 ~ 30.16	40 ~ 50

4.2.2. Strong Gamma Signal from Holdup

The study on what happens if holdup uranium emits strong gamma signal was carried out. For example, when the ^{235}U abundance of UF_6 is 0.3 at%, 0.7 at%, 1 at%, 2 at%, 3 at%, or 4 at%, the holdup consists of 5 at% ^{235}U. This condition may cause false alarm and make unnecessary inspection because strong gamma signal can give misuse signal to gamma radiation detection system. The amounts of holdup that can cause a false alarm was evaluated and is shown in Table 4.11. Since DUF_6 and NUF_6 have a relatively higher uncertainty compared to $LEUF_6$, the amount of holdup causing false alarm is similar to 1 at% ^{235}U despite of lower ^{235}U abundance, which means the higher uncertainty of measurement makes the detection of DUF_6 and NUF_6 inaccurate. The false alarm resulting from holdup would be impossible if 5 at% ^{235}U of UF_6 flows in the pipe with 5 at% ^{235}U of holdup. This does not give significant enhancement or reduction of gamma radiation count rates. All Gaussian distributions of gamma radiation count rates are added in Appendix B.

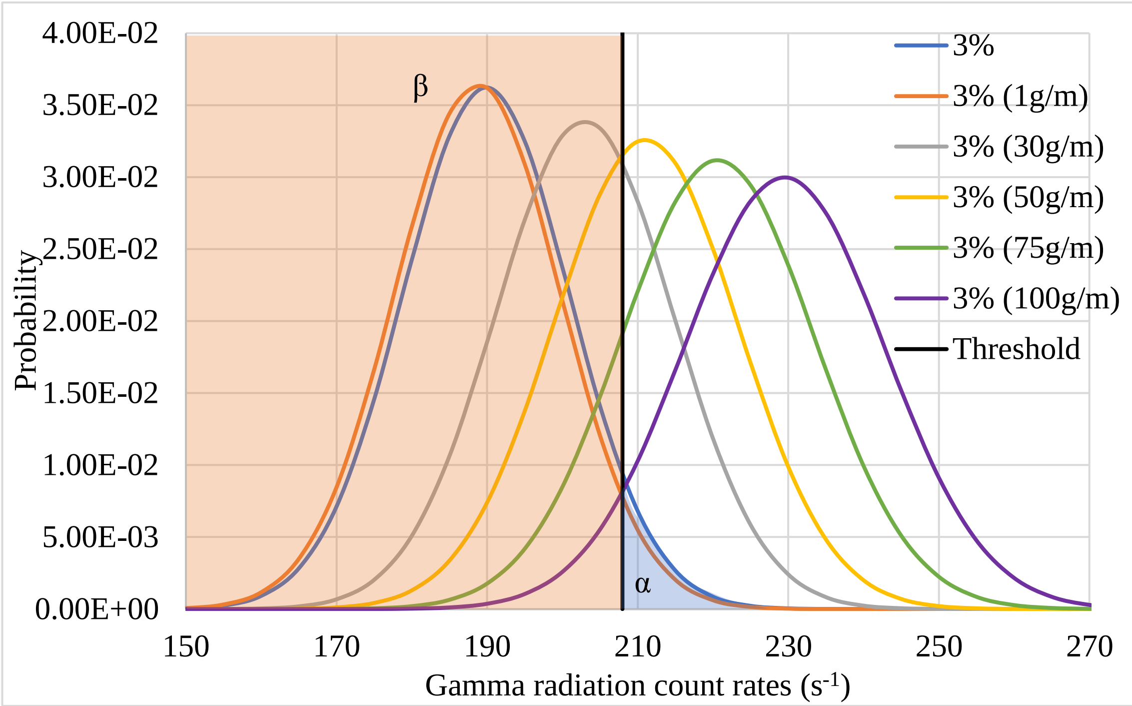

Figure 4.10 Gaussian distributions of gamma radiation count rates when 3 at% UF_6 is flowing in the pipe and a certain amount of 5 at% holdup is deposited in the internals of the pipe.(on the legend) α and β are applied.

Table 4.11 The estimated ranges of nondetection probability and the amount of holdup causing false alarm.

^{235}U enrichment of UF_6 (%)	Nondetection probability (%)	The amount of holdup causing false alarm (g/m)
0.3	21.06	15
0.7	15.64 ~39.37	15 ~10
1	6.63 ~28.45	15 ~10
2	19.25	30
3	16.11 ~40	75 ~50
4	16.86 ~25.68	250 ~200

Holdup could produce a false alarm based on the discussions made above. Especially, LEU less than 2% will cause the false alarm with small amount of holdup and the ranges are in the boundary of maximum amount of holdup. This circumstance would happen when a cascade in GCEP undergoes a modification, where the centrifuges processing 5 at% UF_6 are moved and relocated to enrich 1 or 2 at% UF_6. The appropriate detection and inspection of holdup is needed to avoid unnecessary inspection due to the false alarm when the cascade modification occurs.

4.3. Thickness of Pipe Variation

A possible uranium diversion scenario which was not studied in this research is the variation of pipe thickness. Since GCEP has a long piping and various type of pipes, the variation of pipe thickness should be considered as a cause for reduction or increase in gamma radiation signal. If the thickness of pipe or the material of pipe is different, the gamma radiation should be interfered. For example, if the thickness of pipe increases, the gamma radiation should be reduced due to attenuation. In figure 4.11, one example of pipe thickness variation is depicted. In this study, NPS ½ Sch.40s pipe was assumed as the model of piping, so it was deemed as the reference and colored blue (top on the legend). The details about thickness of pipe is listed in Table 3.1. (Section 3.1.1)

From the variation of pipe thickness, false alarm problem and uranium diversion might happen. If LEU more than 5 at% produces with thicker pipe, the production of LEU more than 5 at% can be hidden since the attenuation from the thickness of pipe. Also, If the pipe has less thickness, then it should give misuse signal and lead to an unnecessary inspection. In addition, the combination with other parameter could happen to make 1-SQ diversion of LEU without being detected.

Although the effect of the thickness of pipe was not studied in this research, the variation of thickness needs to be discussed when it comes to the potential uranium diversion including the measurement methods for assessment of pipe thickness

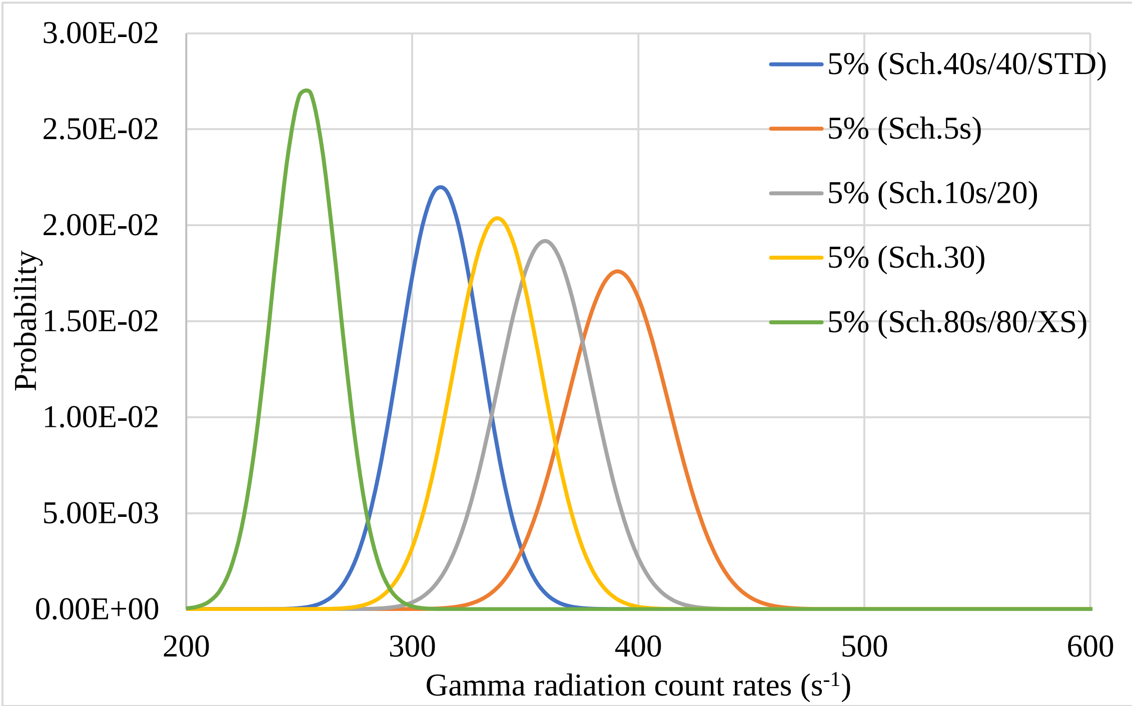

Figure 4.11 Gaussian distributions of gamma radiation count rates of 5 at% ^{235}U. The thickness of pipe varies.

5. CONCLUSIONS

The potential uranium diversion within the uncertainty of the NaI detector in the presence of the holdup of uranium in the centrifuge pipe was analyzed. The amount of holdup required to conceal more highly enriched uranium was studied. In addition, the possibility of an unnecessary inspection resulting from the holdup of uranium was also studied. Monte Carlo radiation transport code, MCNP6.2, was used throughout the simulations. The simulations using F8 detector pulse height tallies were conducted to acquire gamma-ray spectrum of the pipe that UF_6 flows and UO_2F_2 is deposited on the pipe wall. With the results from the MCNP6.2 simulations, the analyses on the uranium diversion and the possibility of an unnecessary inspection has been conducted. ITVs had been used as the reference of the relative error when the statistical analyses had been conducted in this study.

The analysis proved that 1-SQ of uranium could be diverted within 3 months without being detected if the certain amount of holdup exists. It was confirmed clearly that the higher the ^{235}U enrichment of UF_6, the more the amount of holdup is required to hide the highly enriched UF_6. In order to ensure that the uranium diversion could be occur in GCEP due to holdup unaccounted for, the number of centrifuges needed in a cascade to divert 1-SQ uranium within 3 months was analyzed. The analysis suggests the number of centrifuges required after UF_6 is enriched to 5 at% is reasonable value and the 1-SQ of uranium diversion could happen in GCEP with small number of centrifuges. The number of centrifuges range from 62 to 216 depending on the ^{235}U enrichment of UF_6. Meanwhile, the new cascade model was proposed to calculate the number of centrifuges in the cascade.

One of the parameters for safeguards inspection, SWU, was also studied to check if the concealment of the production of more highly enriched uranium is possible. By parametrically varying the ^{235}U abundance of feed and waste, it is proved that the operator could make that SWU does not change significantly as well as the quantity of feed, product, and waste and the number of stages. Further, the UF_6 loss created by holdup was calculated to figure how much holdup is needed for nondetection of diversion using LCBS and the amount of UF_6 and holdup were found to be 0.162kg and 143g per centrifuge, respectively.

Gas flow meters was introduced to measure the boundary of the amount of holdup generated without being detected as another parameter because the measurement of gas flow, or velocity, has a relatively low error and the value was assumed as 5%. It suggested that holdup can be produced up to 60g/m without being detected with gas flow meters. Correspondingly, 7 at% ^{235}U of UF_6 is the boundary of uranium diversion scenario occurring due to 60g/m holdup.

The possibility of an unnecessary inspection was analyzed and it was proved that the unnecessary inspection may be called for. The analyses considered of two cases; one is strong gamma radiation signal from holdup the other is weak gamma radiation signal from holdup. 5 at% ^{235}U of holdup had been assumed for the strong gamma radiation and 0.7 at% ^{235}U of holdup had been assumed for the weak gamma radiation. Based on the observation, LEU goal enrichment, 3 at% to 5 at%, is the range causing false alarm under the existence of 0.7 at% ^{235}U holdup. (weak gamma radiation signa) Also, below 2 at% is feasible to cause false alarm and make an unnecessary inspection when the material holdup consists of 5 at% ^{235}U. (strong gamma signal) Therefore, holdup should be accounted to prevent the unnecessary inspection and save the cost.

In conclusion, holdup unaccounted can interfere the accurate gamma-ray measurement in GCEP and could yield to 1-SQ of uranium diversion as well as a false alarm. If an operator of GCEP have the intention of diverting uranium and follow the scenario studied, the 1-SQ of uranium diversion is possible without being detected. Moreover, holdup can cause false alarm problem and call for unnecessary inspections. However, the system for the determination of presence of holdup has still a huge uncertainty even though the safeguards risk from holdup was brought up earlier and the detection system have been developed for the past several decades. A new technique reducing the measurement error of holdup is required to decrease the risk of uranium diversion from holdup and to save the IAEA inspection cost.

As future work, more parametric analysis needs to be considered such as thickness of pipe variation or material of the pipe. Also, a technique, which makes the uncertainty of holdup measurement lower should studied.

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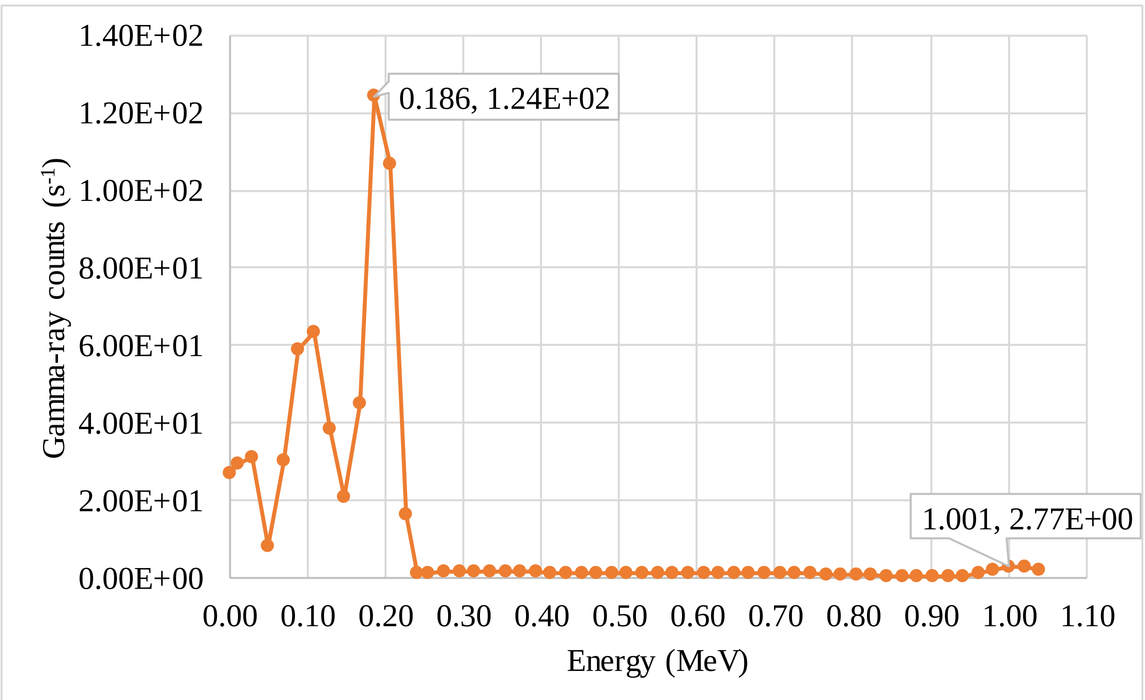
APPENDIX A

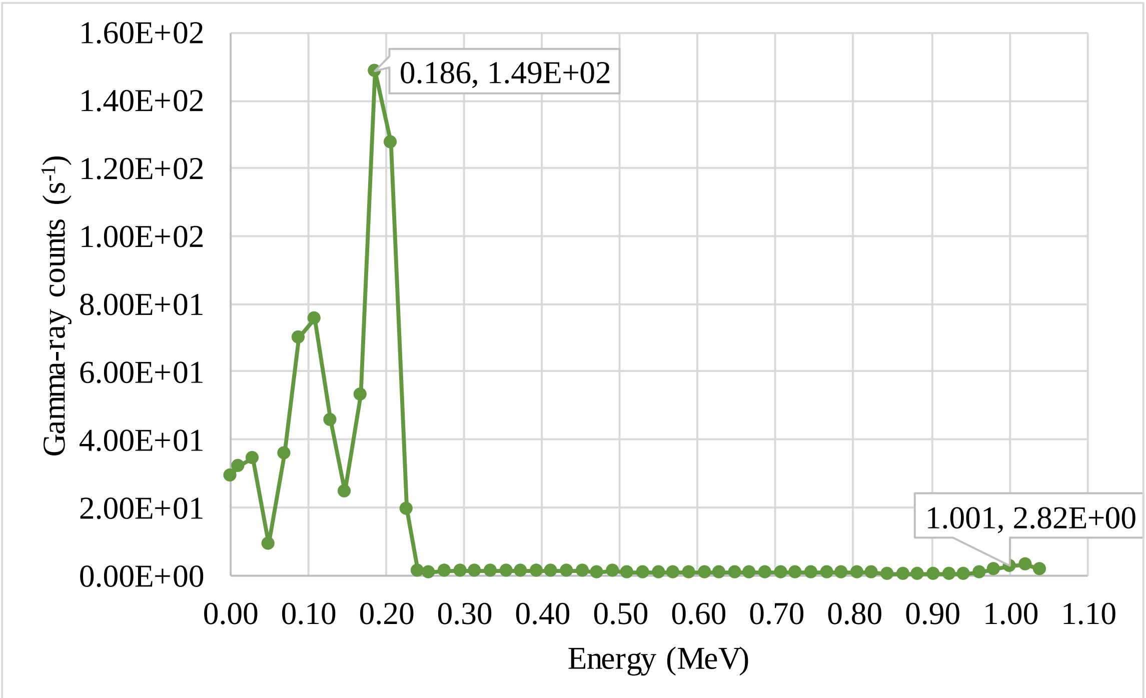
In Appendix A, three type of plots are included. Each type is categorized as A, B, C and description is concluded. The ^{235}U enrichment ranges from 5 at% to 10 at%. The plots of each enrichment have different color. These plots are to prove the simulations data used in this thesis, but not is included and explained in the text.

A. Gamma-ray spectrum when no holdup occurring in the pipe.

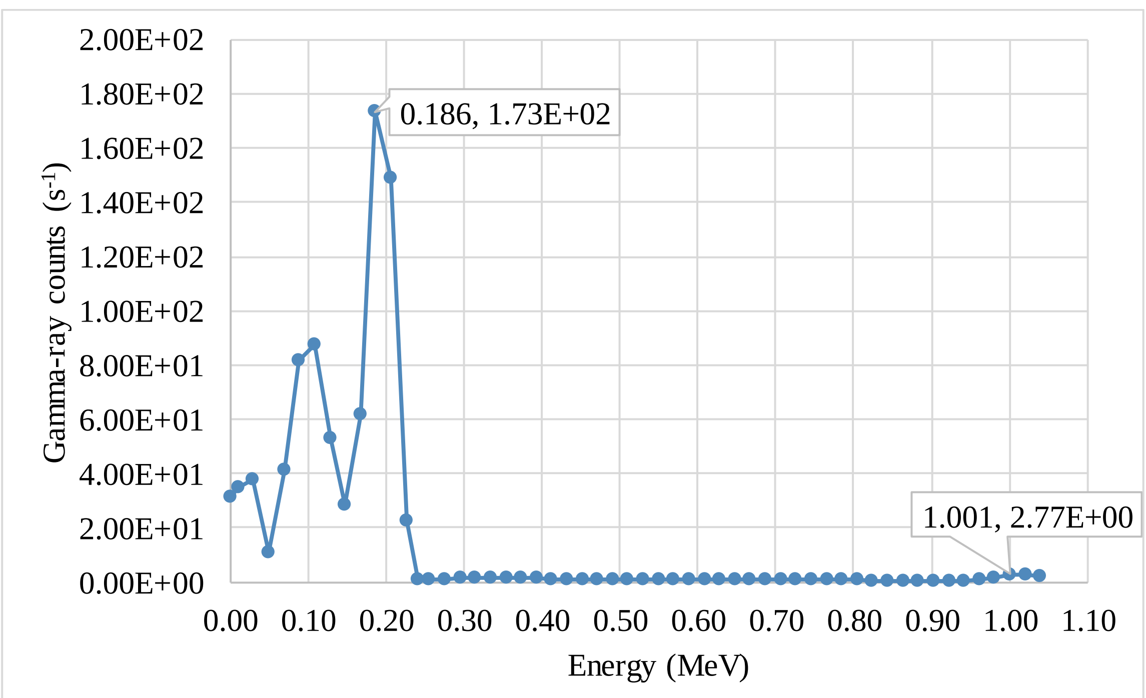
(a) 5 at% ^{235}U



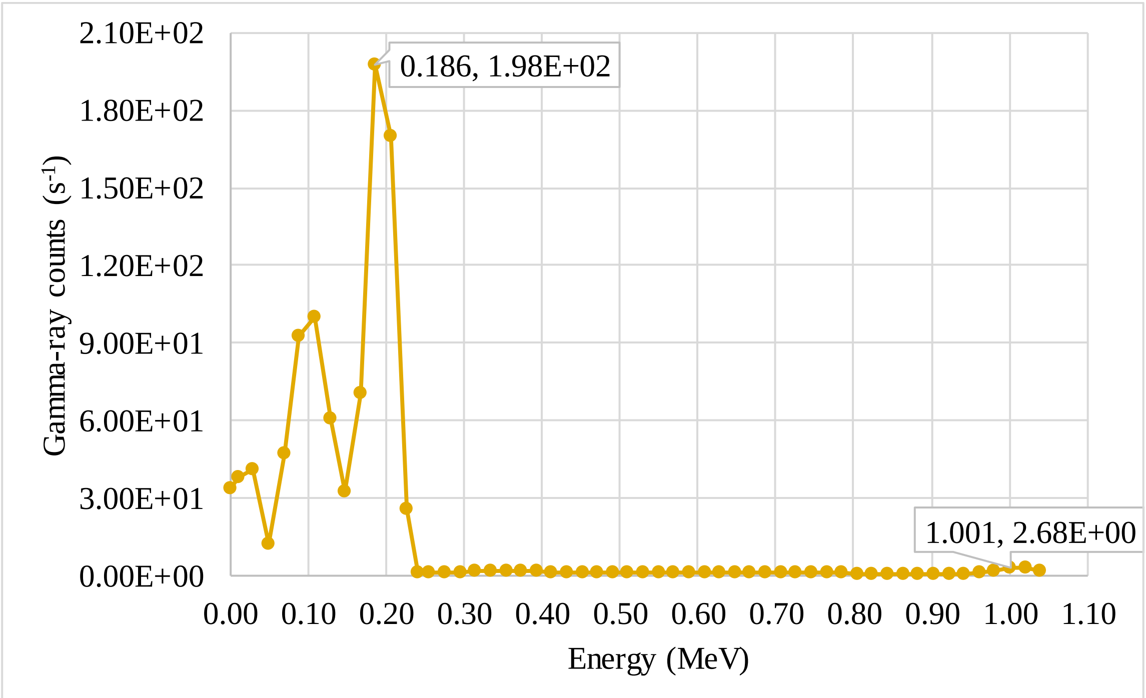
(b) 6 at% ^{235}U



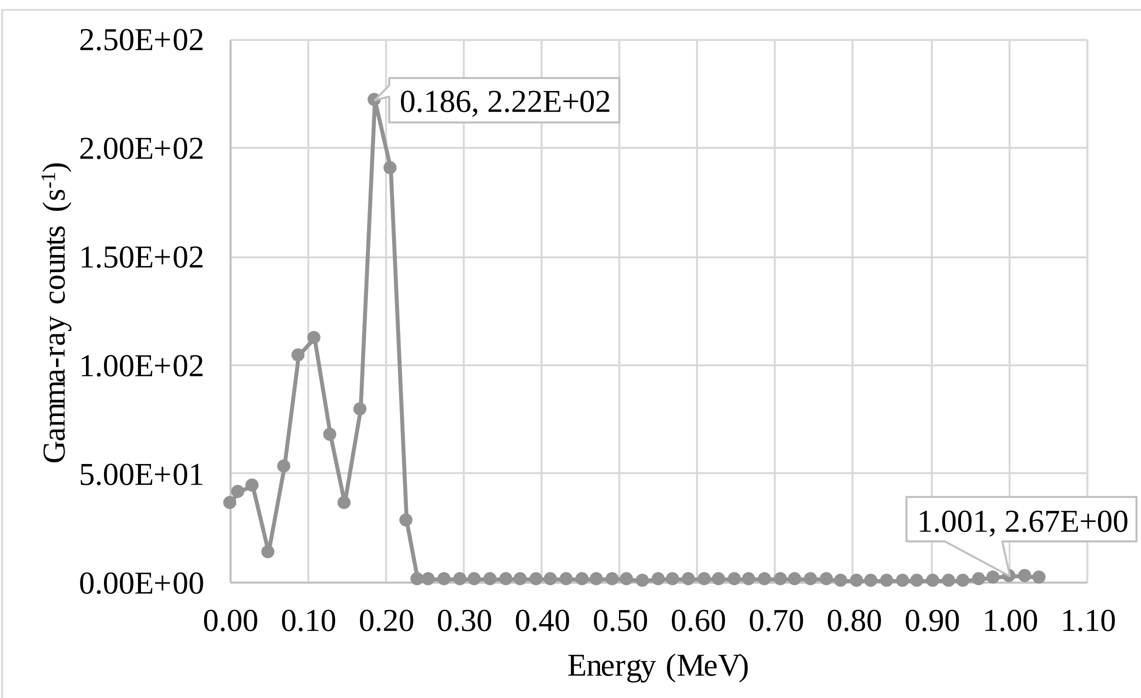
(c) 7 at% ^{235}U



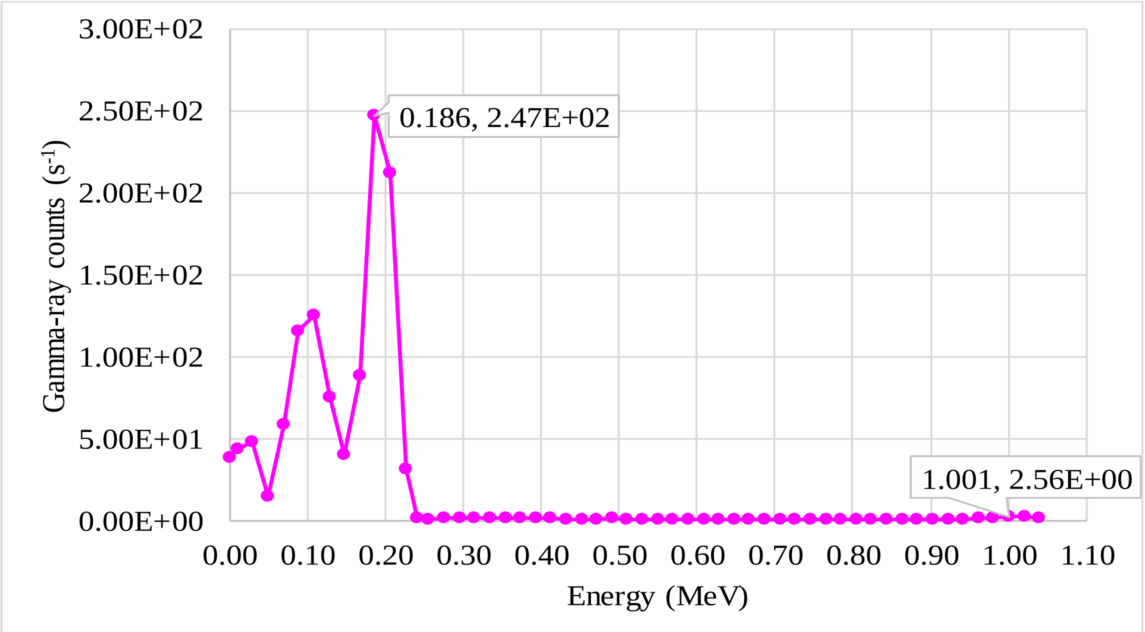
(d) 8 at% ^{235}U



(e) 9 at% ^{235}U

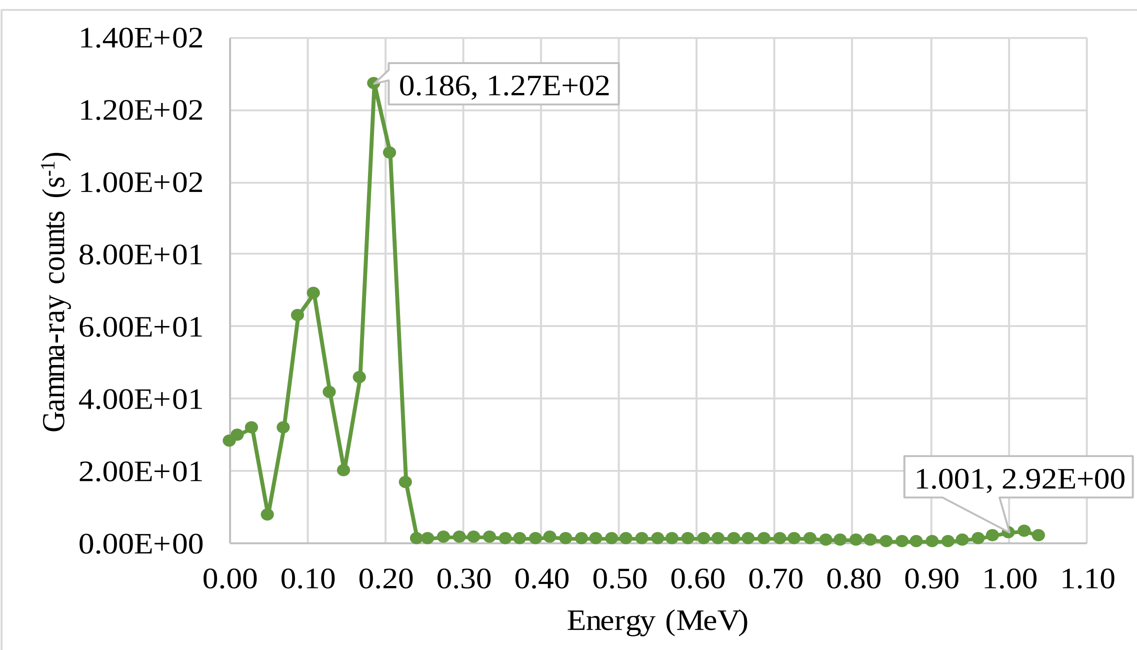


(f) 10 at% ^{235}U

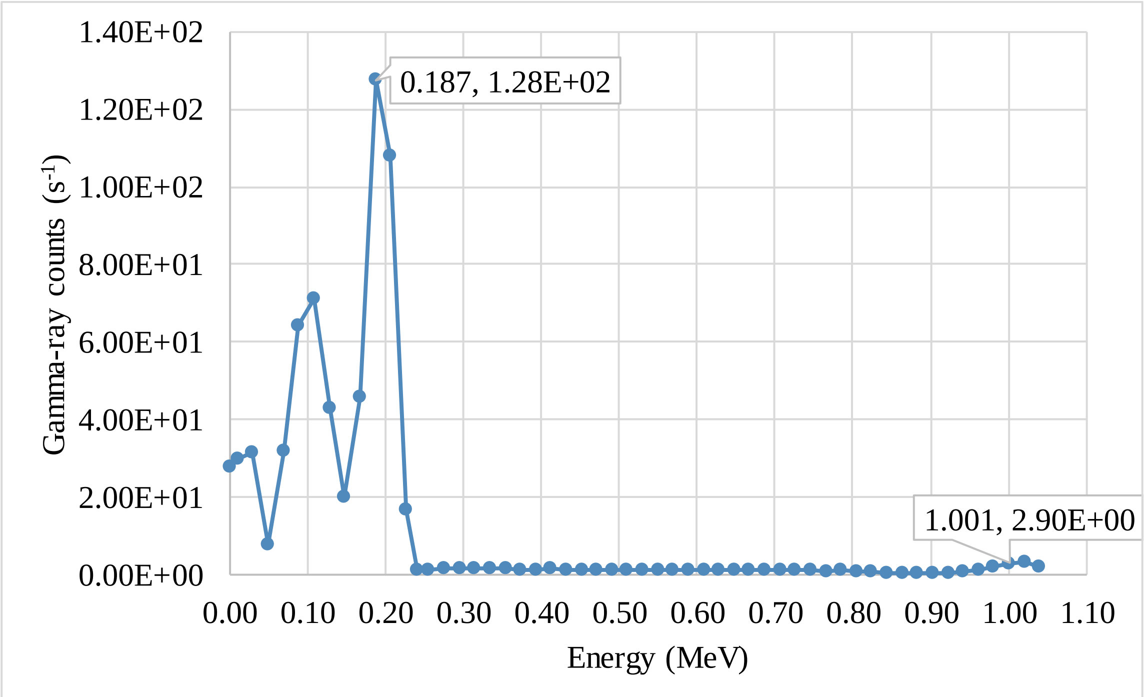


B. Gamma-ray spectrum when a certain amount of holdup required to conceal more highly enriched UF_6 is deposited in the pipe.

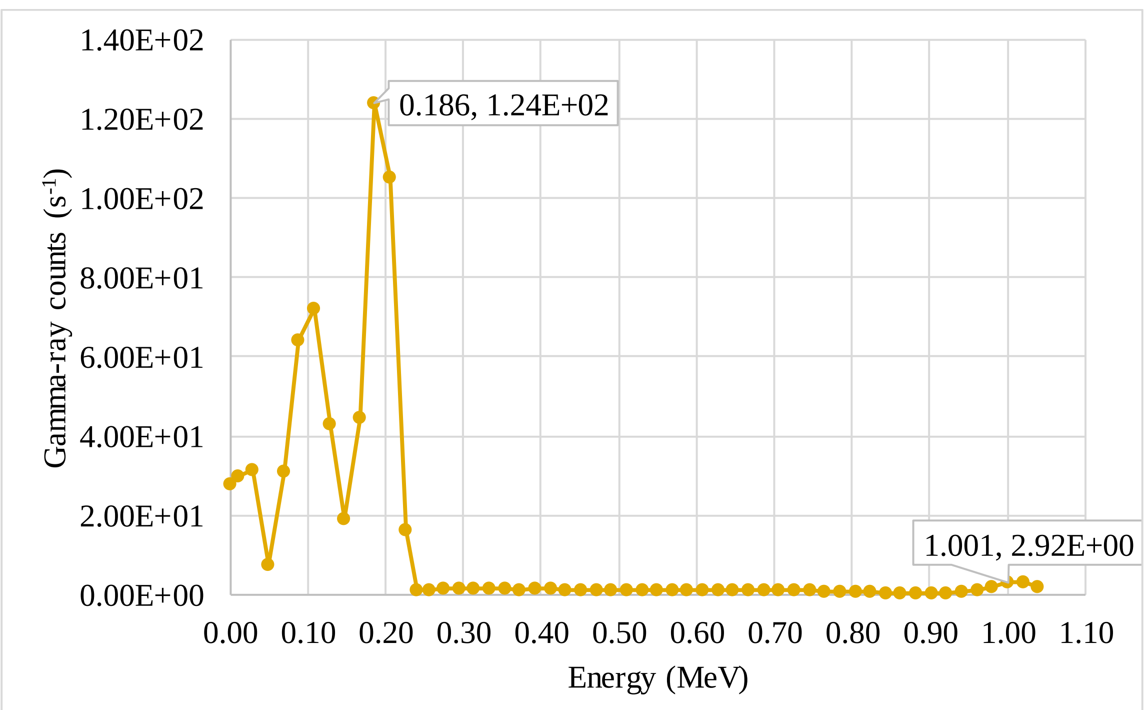
(a) 6 at% ^{235}U when 50 g/m holdup is deposited in the pipe.



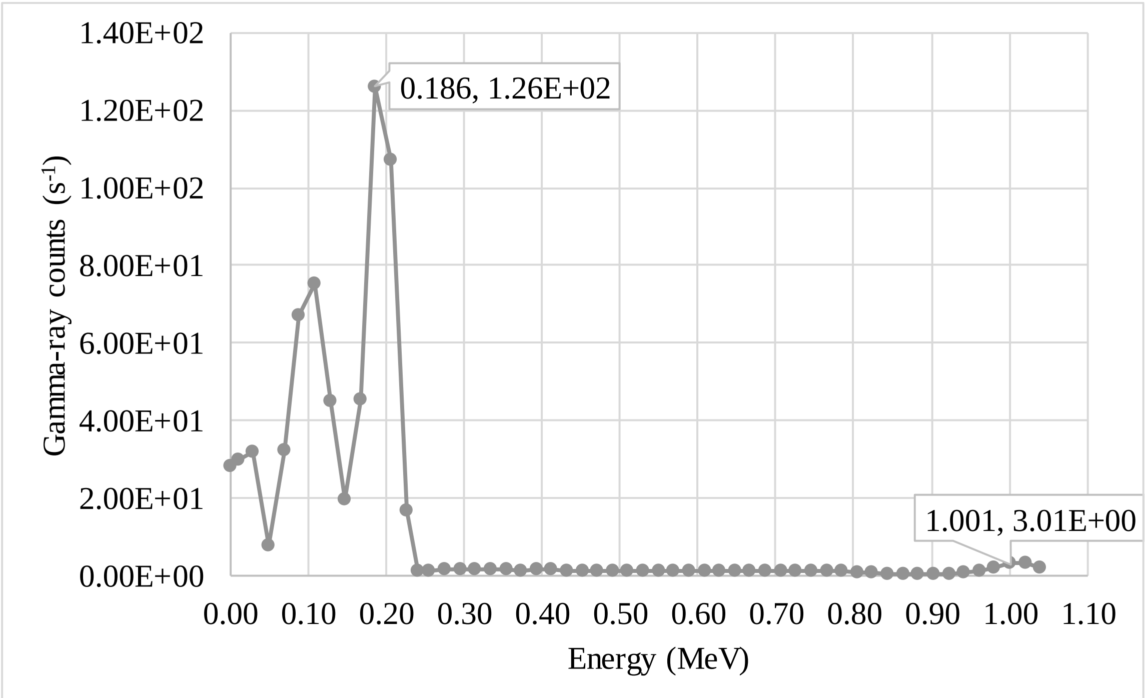
(b) 7 at% ^{235}U when 100 g/m holdup is deposited in the pipe.



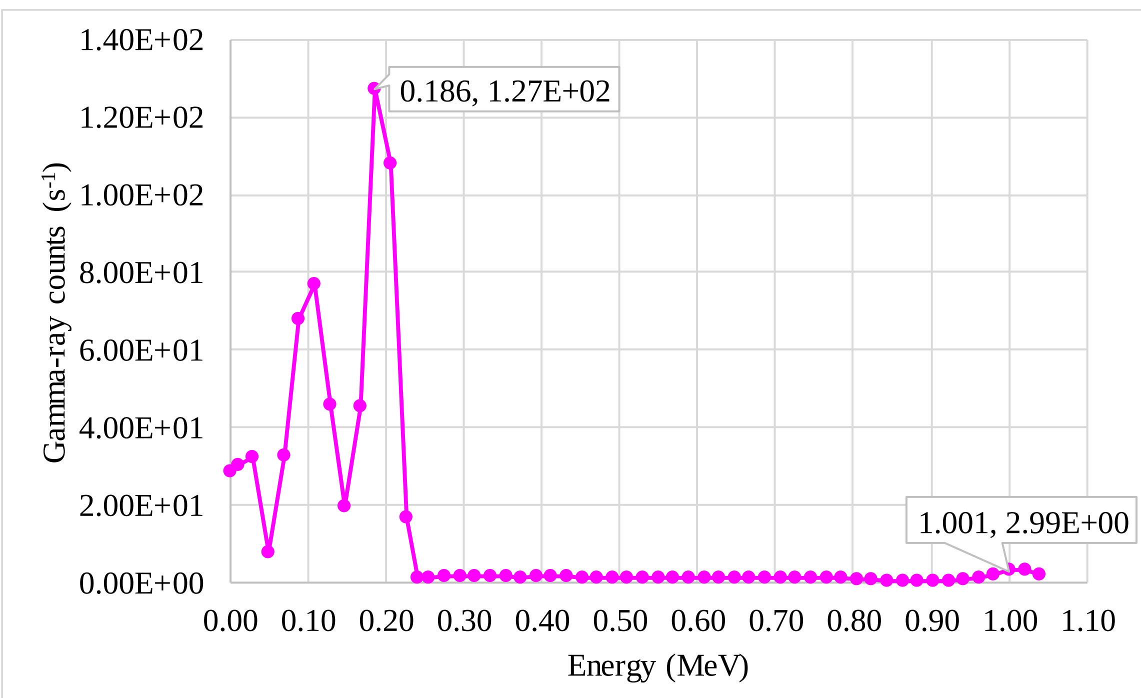
(c) 8 at% ^{235}U when 150 g/m holdup is deposited in the pipe.



(d) 9 at% ^{235}U when 180 g/m holdup is deposited in the pipe.

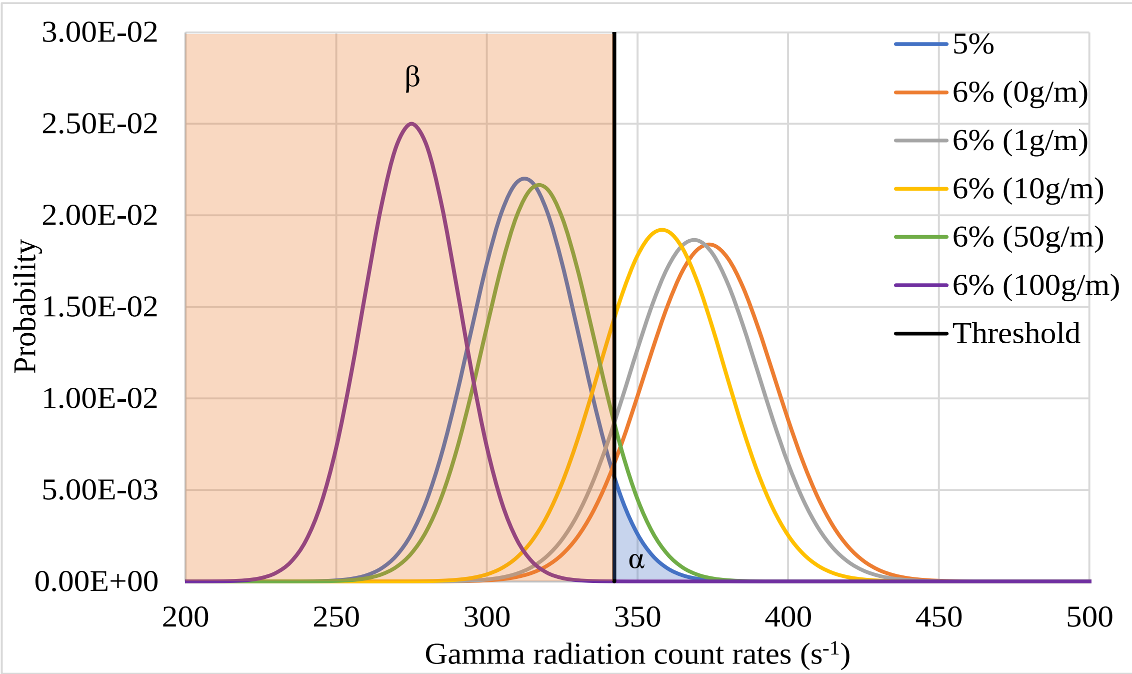


(e) 10 at% ^{235}U when 210 g/m holdup is deposited in the pipe.

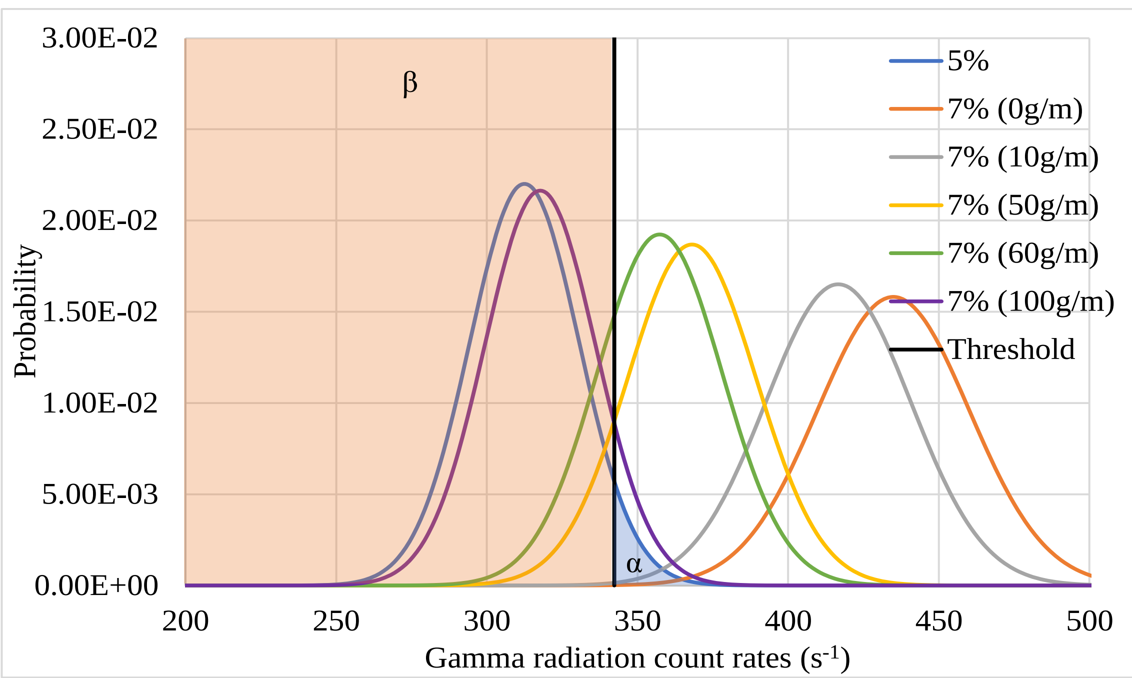


C. LEU (> 5 at% ^{235}U) with 0.7 at% ^{235}U Holdup

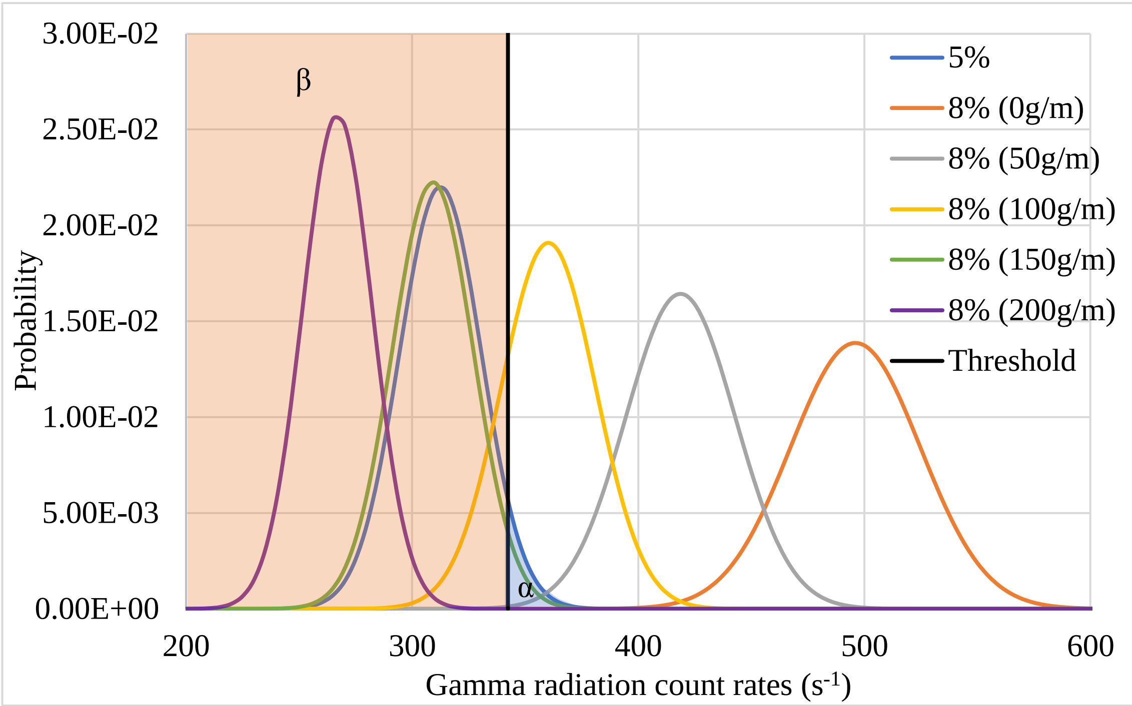
(a) 6 at% ^{235}U UF_6



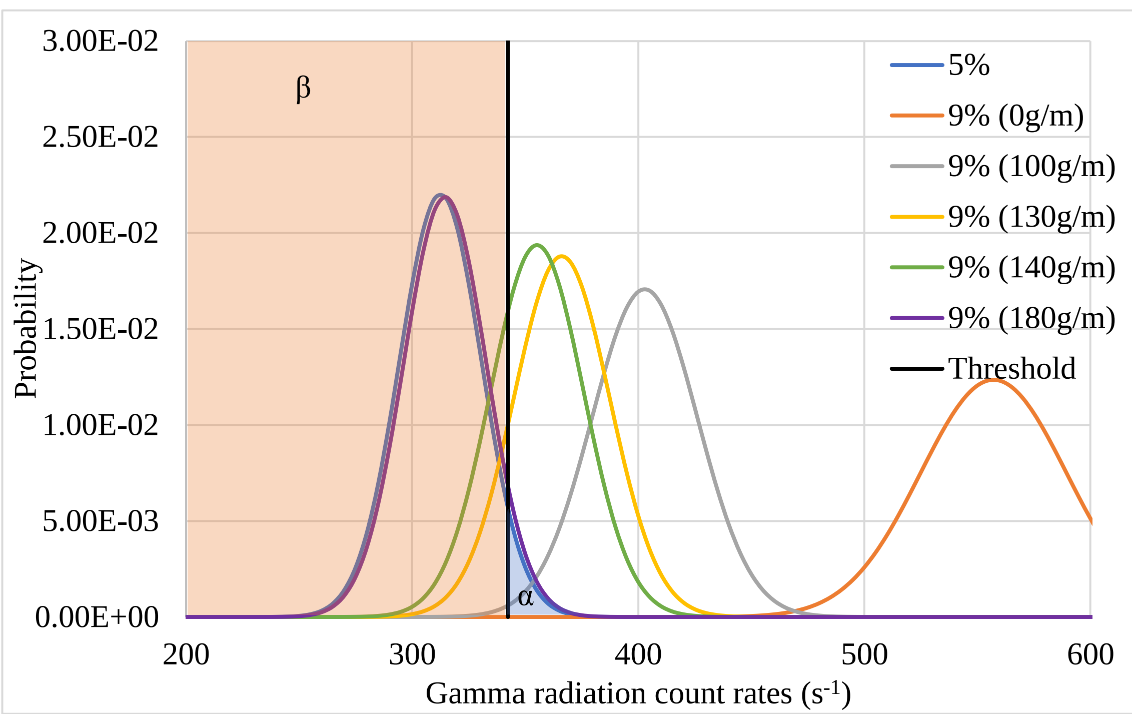
(b) 7 at% ^{235}U UF_6



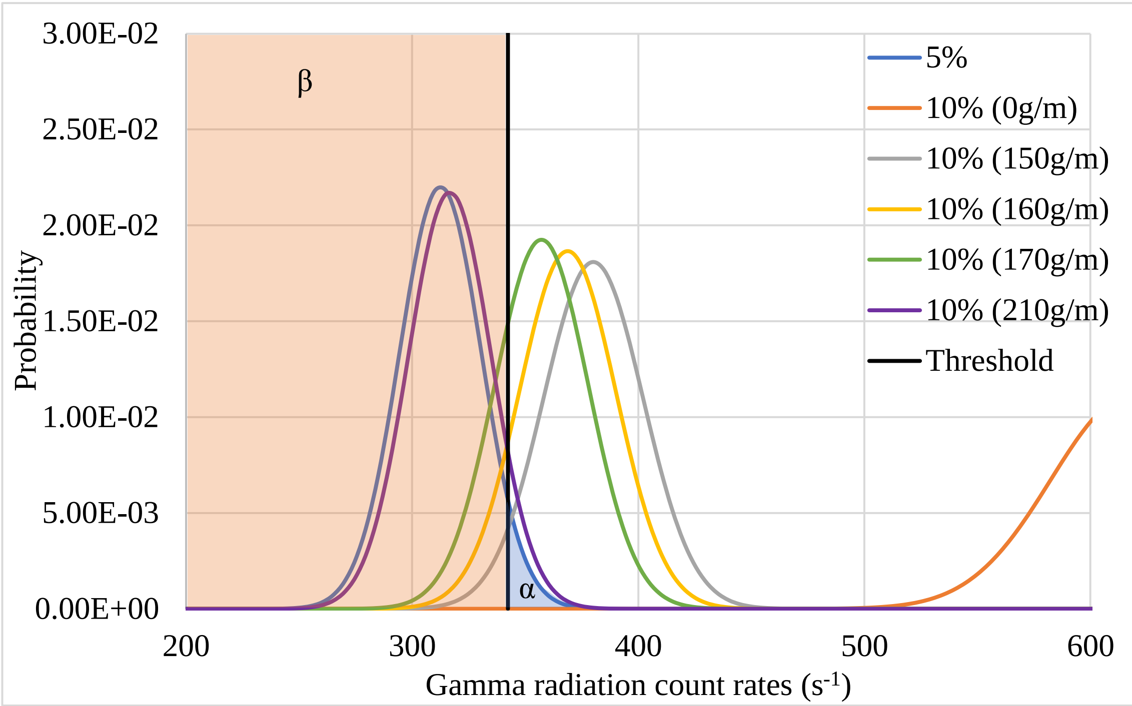
(c) 8 at% ^{235}U UF_6



(d) 9 at% ^{235}U UF_6



(e) 10 at% ^{235}U UF_6

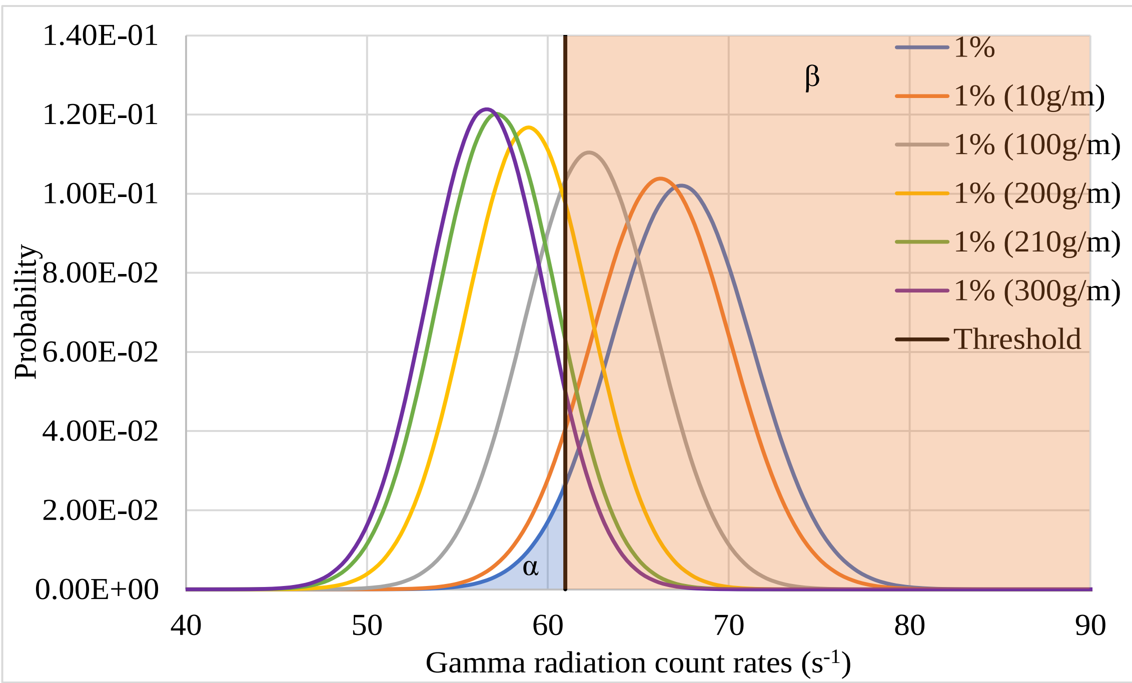


APPENDIX B

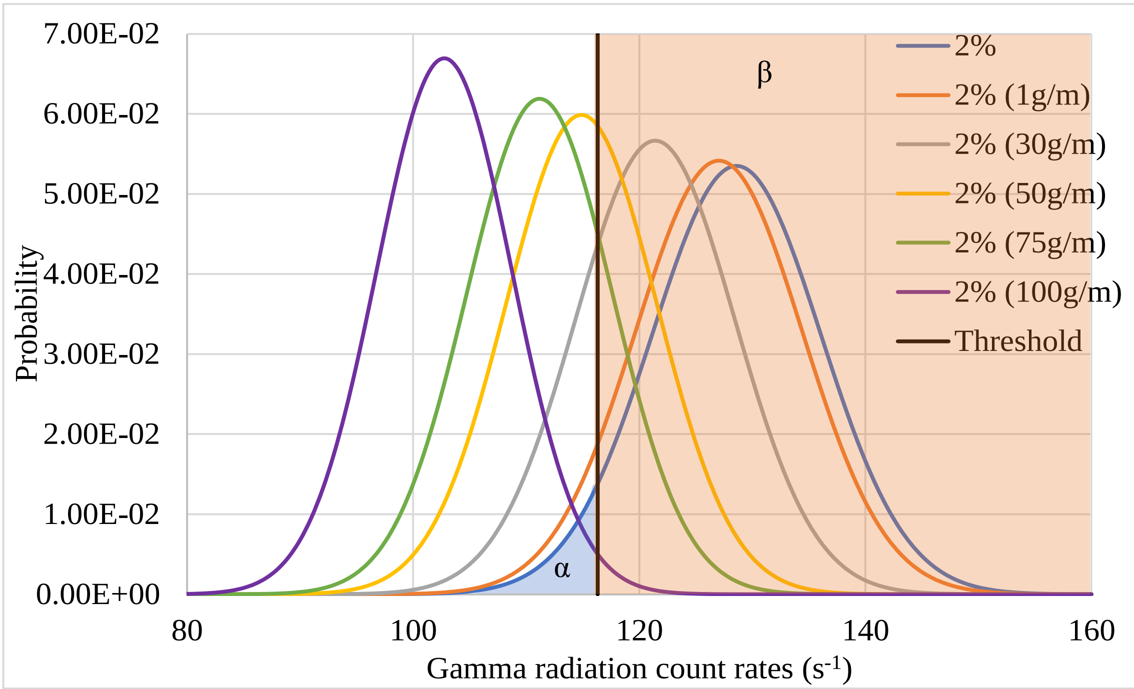
All plots used to depict false alarm probability and nondetection probability is listed in this Appendix.

D. Low Enriched UF_6 (< 5 at% ^{235}U) with 0.7 at% ^{235}U Holdup

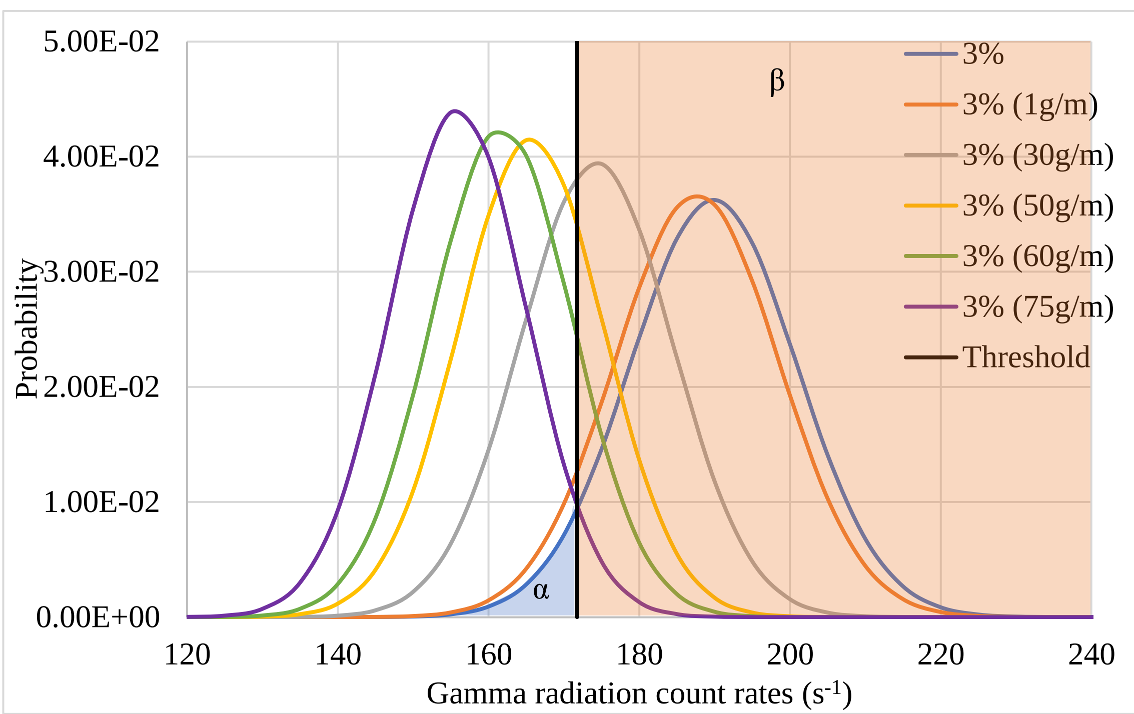
(a) 1 at% ^{235}U UF_6



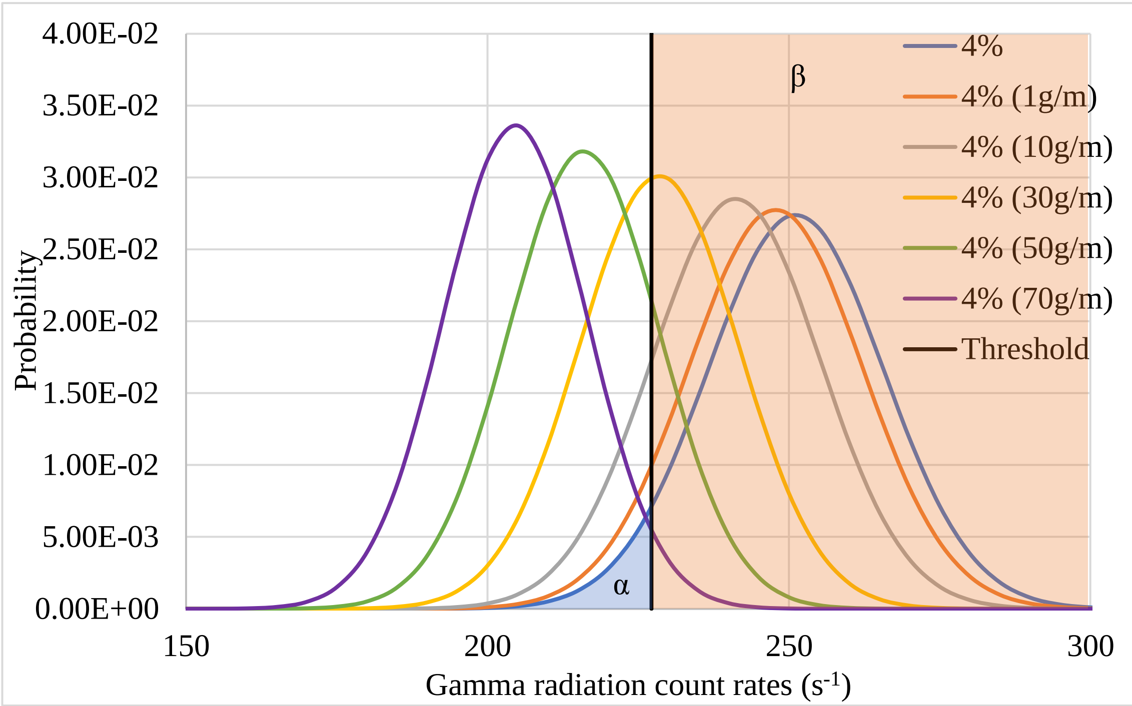
(b) 2 at% $^{235}U\ UF_6$



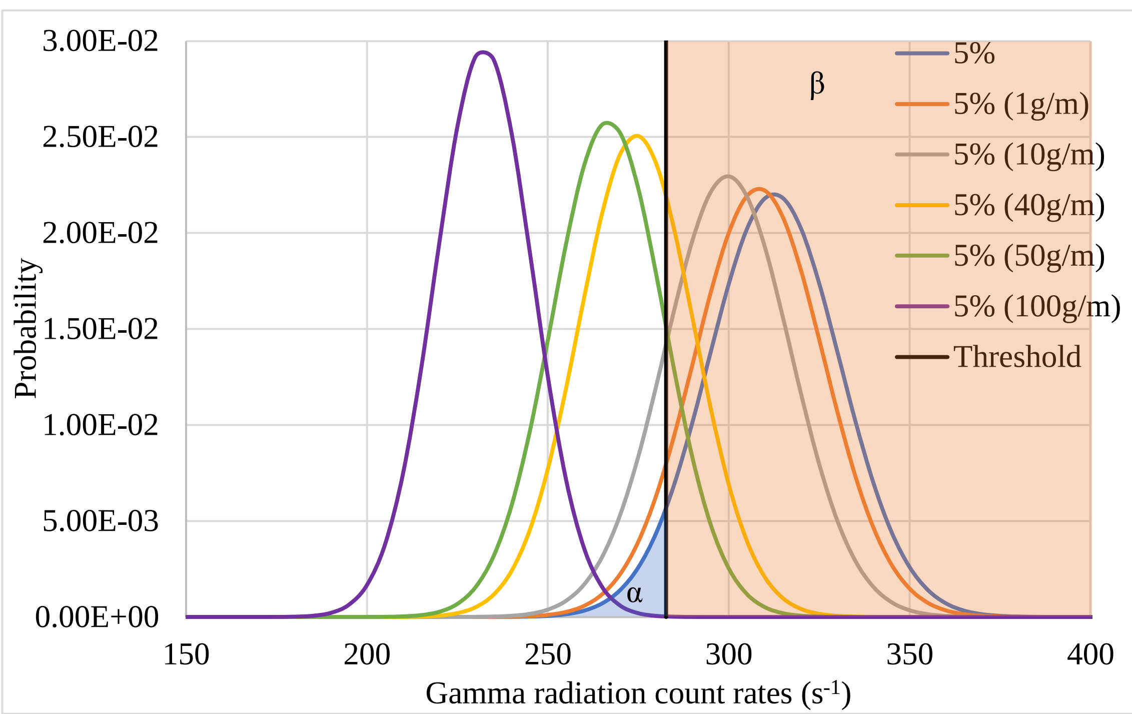
(c) 3 at% $^{235}U\ UF_6$



(d) 4 at% ^{235}U UF_6

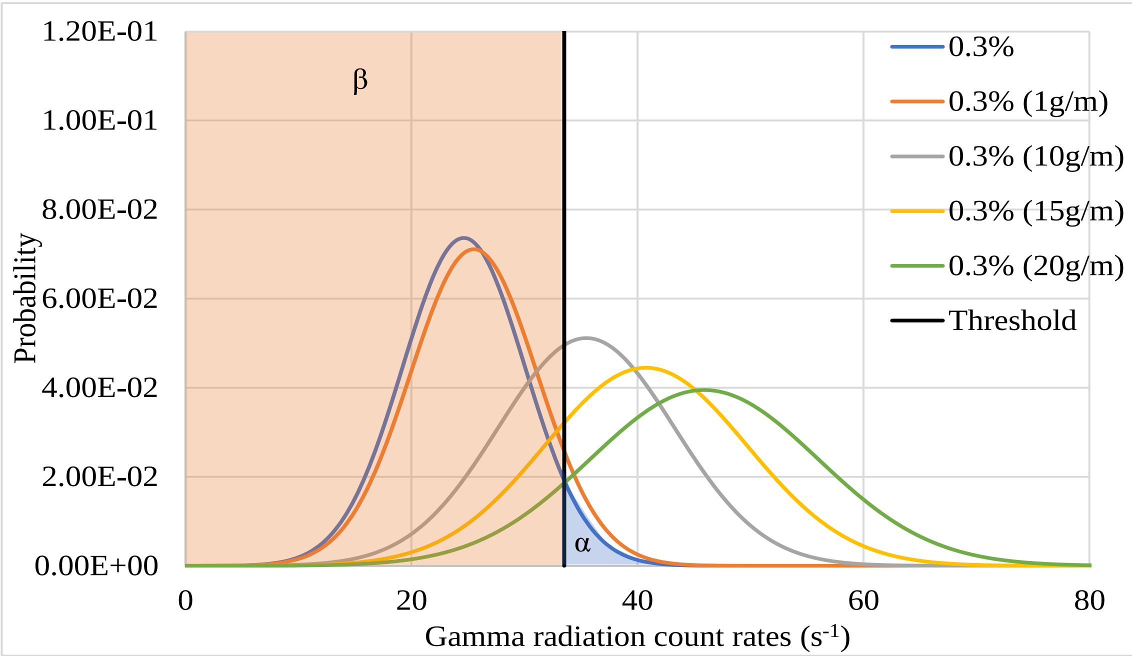


(e) 5 at% ^{235}U UF_6

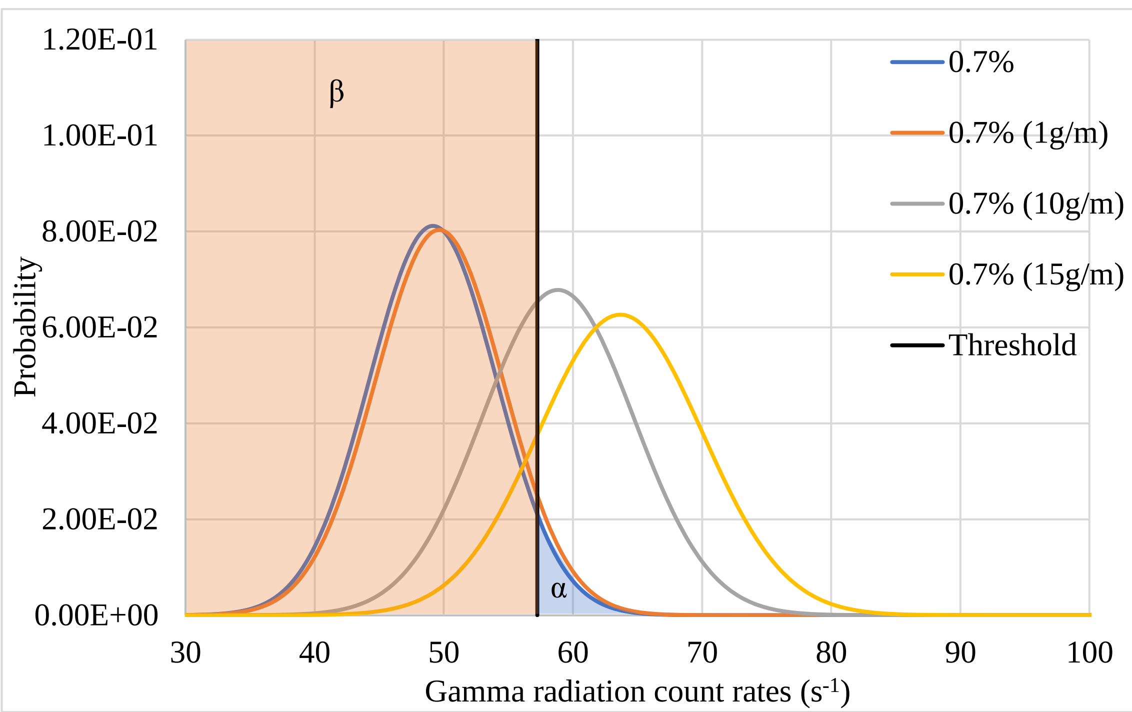


E. Depleted, Natural, and Low Enriched UF_6 (< 5 at% ^{235}U) with 5 at% holdup

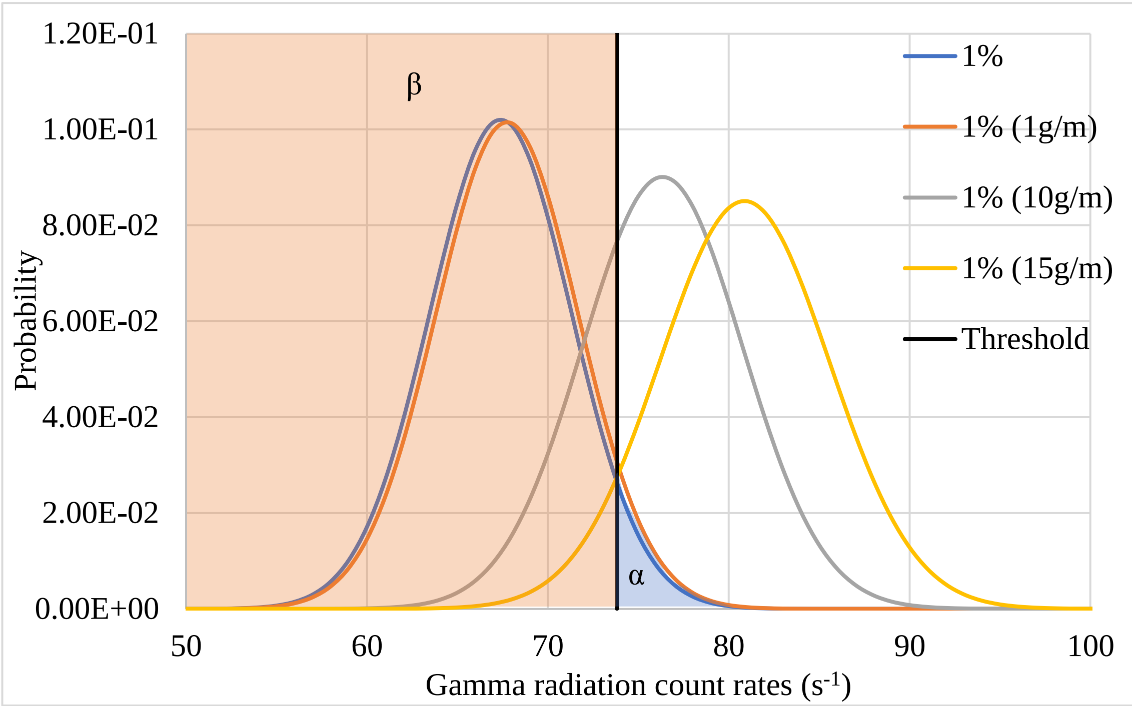
(a) 0.3 at% ^{235}U UF_6



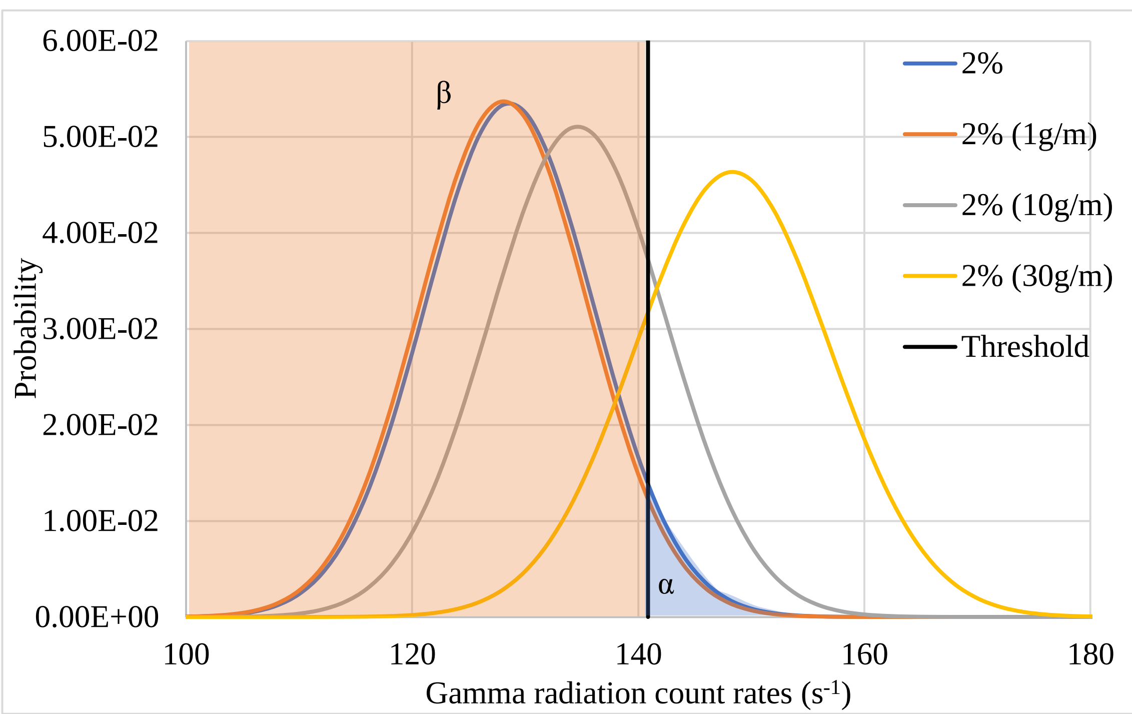
(b) 0.7 at% ^{235}U UF_6



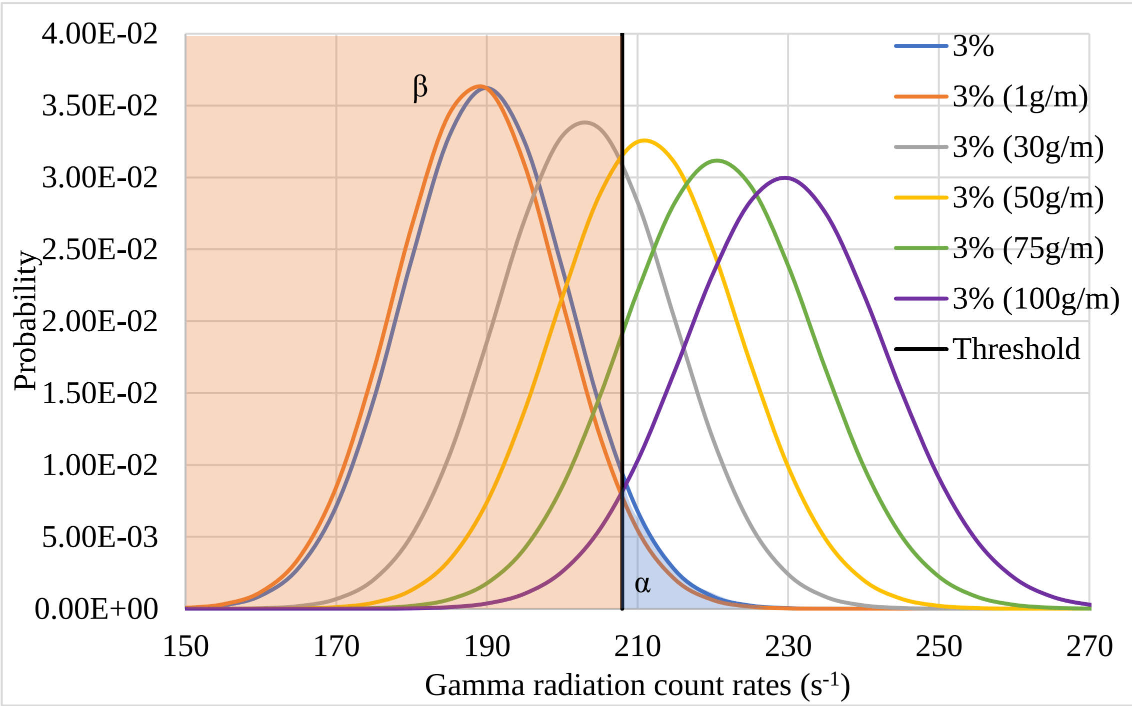
(c) 1 at% $^{235}U\ UF_6$



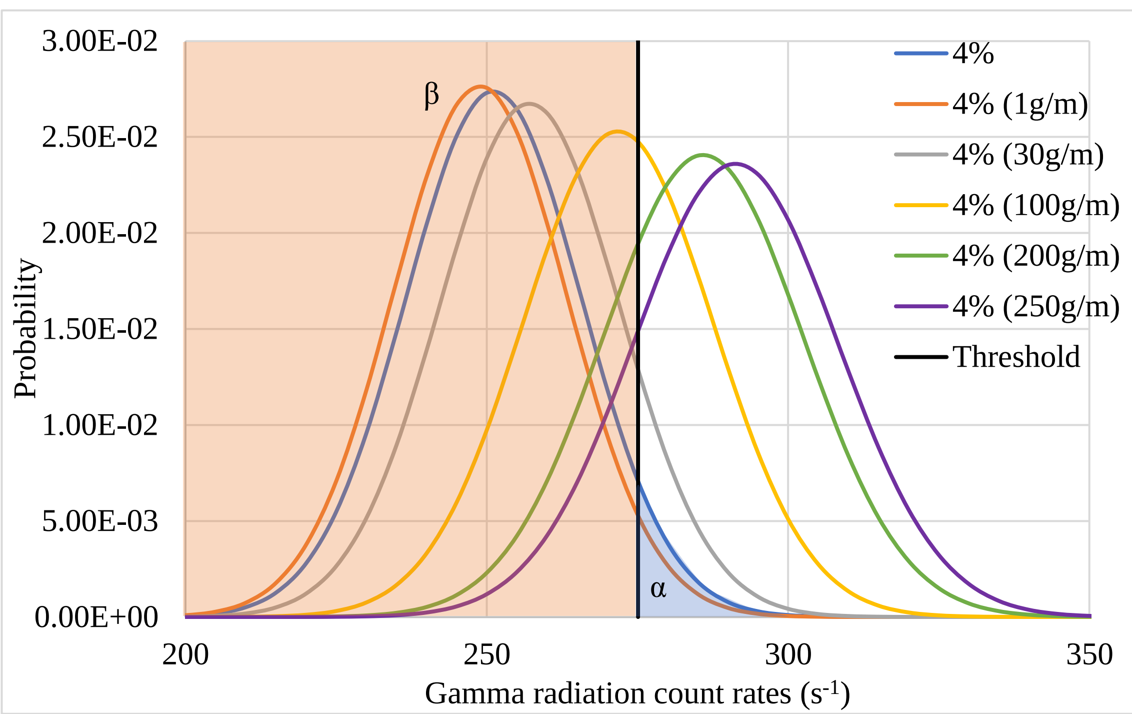
(d) 2 at% $^{235}U\ UF_6$



(e) 3 at% ^{235}U UF_6



(f) 4 at% ^{235}U UF_6



(g) 5 at% $^{235}U\ UF_6$