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Detection of Illicit HEU Production in Gaseous Centrifuge Enrichment Plants using Neutron Counting Techniques on Product Cylinders

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Abstract

Innovative and novel safeguards approaches are needed for nuclear energy to meet global energy needs without the threat of nuclear weapons proliferation. Part of these efforts will include creating verification techniques that can monitor uranium enrichment facilities for illicit production of highly-enriched uranium (HEU). Passive nondestructive assay (NDA) techniques will be critical in preventing illicit HEU production because NDA offers the possibility of continuous and unattended monitoring capabilities with limited impact on facility operations. Gaseous centrifuge enrichment plants (GCEP) are commonly used to produce low-enriched uranium (LEU) for reactor fuel. In a GCEP, gaseous UF_6 spins at high velocities in centrifuges to separate the molecules containing ^{238}U from those containing the lighter ^{235}U. Unfortunately, the process for creating LEU is inherently the same as HEU, creating a proliferation concern. Insuring that GCEPs are producing declared enrichments poses many difficult challenges. In a GCEP, large cascade halls operating thousands of centrifuges work together to enrich the uranium which makes effective monitoring of the cascade hall economically prohibitive and invasive to plant operations. However, the enriched uranium exiting the cascade hall fills product cylinders where the UF_6 gas sublimes and condenses for easier storage and transportation. These product cylinders hold large quantities of enriched uranium, offering a strong signal for NDA measurement. Neutrons have a large penetrability through materials making their use advantageous compared to gamma techniques where the signal is easily attenuated. One proposed technique for detecting HEU production in a GCEP is using neutron coincidence counting at the product cylinder take off stations. This paper discusses findings from Monte Carlo N-Particle eXtended (MCNPX) code simulations that examine the feasibility of such a detector.

Introduction

For nuclear energy to meet global energy needs without the threat of nuclear weapons proliferation, innovative and novel safeguards approaches are needed. Part of these efforts will include passive verification techniques that can monitor uranium enrichment facilities to watch for illicit production of High Enriched Uranium (HEU).

In a Gaseous Centrifuge Enrichment Plant (GCEP), many centrifuges are used to separate ^{235}U from ^{238}U in UF_6 gas. The centrifugal force is the mechanism that separates the UF_6 containing the lighter ^{235}U isotope from those which have ^{238}U. In each centrifuge, a very small degree of separation occurs, requiring a GCEP to have a cascade of several thousand centrifuges working in concert to achieve a high degree of separation. In a typical commercial GCEP natural uranium will be enriched to 3-5% ^{235}U.

Monitoring the enrichment inside the cascade hall of centrifuges is not practical for many reasons. First, it is uneconomical to monitor every centrifuge, and if a small sample is chosen to represent the whole, an intelligent adversary can easily defeat the measurement. Second, the centrifuges and design of the cascade hall are proprietary, making plant operators unwilling to grant anything more than minimal access. Thus, for any design to be practical for implementation it must have a minimal impact

to cost and operations of the GCEP, and at the same time be robust enough to function as a significant impediment to illicit production of HEU. Non Destructive Assay (NDA) techniques on select equipment in a GCEP can provide a means to monitor for HEU production without significant interference in plant operations. Two possible locations for using neutron based NDA techniques in a GCEP are at the vacuum system cold traps and the product cylinder takeoff stations. Monitoring these locations has the advantage that they are located outside of the cascade hall, require a small number of detection systems to be installed, and would be difficult to circumvent the measurement devices unnoticed. This work examines the feasibility of using neutron coincidence counting on the product cylinder bays inside a GCEP to detect illicit HEU production[1]. Evaluation of the vacuum systems cold trap for monitoring the production of HEU has already been documented [2]. Other work using coincidence counting around product cylinders exist. One study examines using coincidence counting with bare and cadmium covered detectors to determine uranium enrichment and fill fraction [3].

Neutron Monitoring at the Product Cylinder Bay

UF_6 enriched in ^{235}U is collected in an array of large cylinders. These product cylinders are kept in chilled bays so that the UF_6 gas, which sublimes slightly below room temperatures, can be condensed into a solid for greater storage capacity. A 48Y container is often used for storage and holds several thousand kilograms of UF_6. This large collection of enriched product creates an ideal place for passive measurement systems to detect illicit production of HEU. Monte Carlo Neutral Particle eXtended [4] (MCNPX) models were created to simulate an array of ^3He tubes on each side of the product cylinder bays. Neutron coincidence counting, along with the mass of UF_6 in the product cylinder is used to determine the concentration of ^{235}U being produced at a GCEP.

After exiting the centrifuge cascade hall, the enriched uranium is pumped to the product cylinders, for collection and holding until further needed. The product cylinders are housed in an array of insulated bays. One major advantage of monitoring at the product cylinder bays is that relatively few places of measurements need to be taken that represent the product enrichment throughout the plant. Another advantage, particularly over monitoring at the cold traps, is that thousands of kilograms of UF_6 collects in the 48Y containers, creating a strong signal for measurement, greatly reducing count time while improving accuracy. If the operator wanted to bypass the product cylinders and divert material, a portable take off station would need to be used which will be extremely obvious.

Coincidence Counting

When an isotope fissions, either spontaneously or induced, there is a probability that multiple neutrons will be released simultaneously. This is in contrast to other sources of neutrons, such as (α, n) reactions, where exactly one neutron is emitted. Coincidence counting uses the time correlation between neutron detections to distinguish fission events (Doubles) from single neutron events (Singles), such as (α,n) reactions. This gives information about the quantity of fissile material in the item being measured. Details of coincidence counting are available in the PANDA manual [5].

In a small UF_6 item, the major source of Doubles counts is from the spontaneous fission of ^{238}U isotopes. The main source of Singles counts is from (α,n) reactions between the fluorine and the alpha decay of the ^{234}U isotope.

Table I gives the spontaneous fission and (α,n) yields per kilogram per second for different uranium isotopes in UF_6. While the spontaneous fission yield for ^{238}U per kg is not grossly larger than the spontaneous fission yield for ^{234}U, the greater abundance of ^{238}U atoms makes it the dominant source

of spontaneous fission for all but very highly enriched items. As a result, the ratio of Doubles counts to Singles counts (D/S ratio) may be used as an indicator of the ratio of ^{238}U to ^{234}U. The enrichment of ^{234}U follows with the enrichment of ^{235}U, thus the D/S ratio may be used as an enrichment indicator.

Table I. Spontaneous fission and (α,n) yields for key uranium isotopes in UF_6.

Isotope	Spontaneous Fission Yield [n/s-kg]	(α,n) Yield [n/s-kg]
^{234}U	5.0	580,000
^{235}U	0.4	80
^{238}U	13.6	28

In larger mass UF_6 items, the effect of multiplication dominates the number of Doubles counts. This effect is due to neutrons inducing fission in ^{235}U. Items above several kilograms, particularly in compact geometries, the ^{235}U will contribute to a significant number of Doubles counts. The effect of multiplication causes the Doubles to Singles ratio to become a complex function of the enrichment and multiplication. Therefore, with larger mass items, the D/S ratio may still be used as an indicator of enrichment.

MCNPX Modeling

To study the sensitivity of a neutron monitor to detect the production of HEU in a GCEP many Monte Carlo simulations were performed. These simulations modeled the detector response for many different variables, such as the enrichment, geometry, and isotopic concentrations. The basic MCNPX model created is seen in Figure 1. A half-inch thick cylindrical steel container roughly 299.7-cm long by 123.2-cm diameter represents the 48Y cylinder. The 48Y cylinder (product cylinder) is inside a low temperature take off station. The design details of the take off station are not known exactly and the model is approximated by a steel bay with 15.2-cm of insulation inside. The steel bay is roughly 190.5 x 91.5 x 91.5-cm in size and with a 1.9-cm thick wall. A concrete floor that is 61.0-cm thick and extends from each edge of the bay by 61.0-cm is also included in the model. On the left and right bay walls is a 230 x 7.7-cm polyethylene slab that extends the height of the bay. Inside each slab is an array of equally spaced ^{3}He tubes that are 2.54-cm diameter and 91.5-cm long. Two detector designs were modeled, one with 44 and the other with 12 ^{3}He detectors distributed in the polyethylene slabs. The majority of the modeling was done with the 44 ^{3}He tube design.

A density of 5.0 g/cm^3 was used for the UF_6 accumulated inside the product cylinder, resulting in a UF_6 mass range from 870 to 12,200 kg. SOURCES 4C Code [6] was used to determine the (α,n) and spontaneous fission rates of each UF_6 enrichment. This information allowed the MCNPX results to be converted to the number of Doubles and Singles that would be detected for a given count time.

The 30B cylinder, which holds less mass than a 48Y, is a probable candidate for the storage of enriched uranium in the product cylinder bays. The conclusions in this study are for the larger 48Y container. We believe that responses measured on product bays containing 30B cylinders will still exhibit the same behaviors and trends discussed in this work.

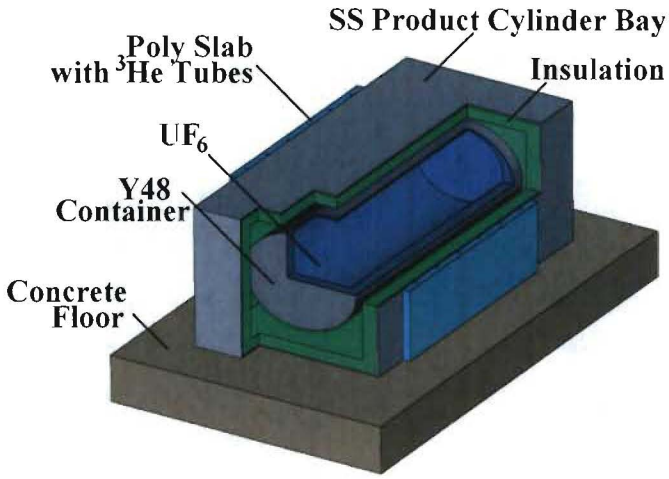

Figure 1. MCNPX cutout model of a 48Y cylinder inside of a product cylinder bay with ^{3}He detector arrays on two sides of the bay. In this model, each slab contains 6 ^{3}He tubes.

Geometry of accumulated UF_6

Considering the dimensions of the 48Y cylinder and assuming that the UF_6 collected inside has a density of 5 g/cm^3, the 48Y cylinder can hold over 10,000 kg of UF_6. The large mass of uranium makes the geometry of the UF_6 deposited in the cylinder particularly important. Compact geometries will result in higher multiplication throughout the item. Unfortunately, the details of how, geometrically, the UF_6 collects inside the cylinder is unknown. Three different cases of the UF_6 geometry are examined and are presented in Figure 2. The first geometry shows one extreme case where the UF_6 enters the product cylinder, sublimes, and falls to the bottom, filling the container in the same manner that a liquid would. This scenario is referred to as the 'liquid' model. The next case considers that the UF_6 doesn't sublime until it contacts the chilled walls of the product cylinder. In this scenario, the UF_6 sticks to the walls and creates a cylindrical shell of uniform thickness against the inner walls of the 48Y container. This scenario is referred to as the 'ring' model. We believe the most likely geometry is between these two extreme cases. As the sublimed UF_6 collects on the wall, the mass increases, and at some point the weight of the material hanging on the walls overcomes the forces holding it to the wall and it falls to the bottom of the can. This concept is represented in the third case, which is referred to as the 'crescent' model.

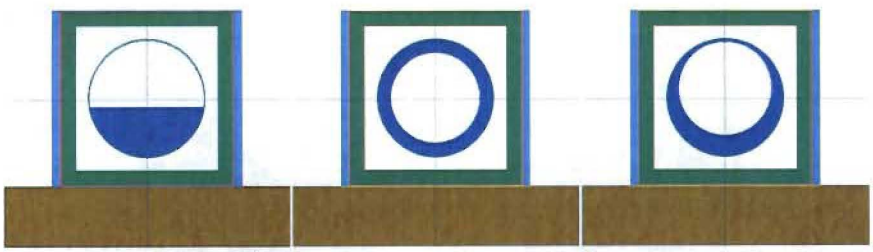

Figure 2. Shown are the three different geometries of UF_6 accumulation inside of the product cylinder that were modeled. Left: UF_6 accumulation in the 'liquid' model. Center: UF_6 accumulation in the 'ring' model. Right: UF_6 accumulation in the 'crescent' model.

Figure 3 shows the D/S ratio calculated for the three different UF_6 accumulation geometries at three enrichment values of 4%, 10%, and 20. The results are very similar for the 'ring' and 'crescent' geometries at each enrichment. However, the 'liquid' model deviates noticeably from the other two models as enrichment is increased.

For a given mass, the true D/S ratio can range anywhere between the two extremes, 'liquid' and 'ring'. With the product cylinder only 20% full, the range created by unknown geometry can be resolved between HEU and 4% enriched. Furthermore, there is no reason to believe that the UF_6 would fill a cylinder differently based on enrichment. Therefore, the upper bound extreme for one enrichment would not need to be resolved from the lower bound extreme of a different enrichment. The variations in deposition geometry will not yield a large variation in the D/S ratio and this will result in very clear separation of the D/S ratio for the different enrichments. Modeling from this point forward only considers the 'crescent' geometry.

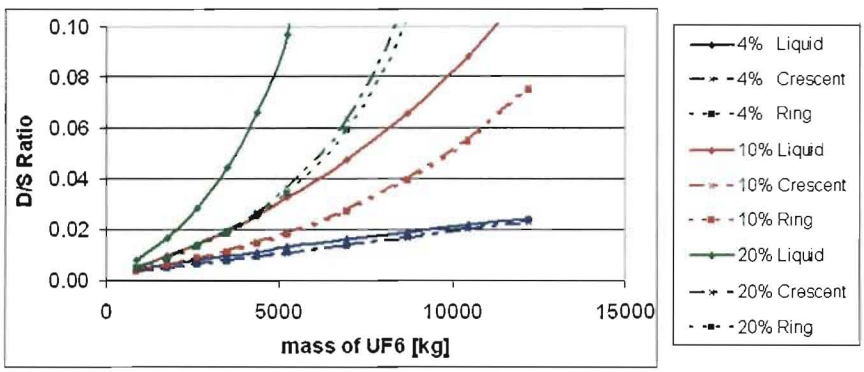

Figure 3. Doubles to Singles ratio as a function of UF_6 mass inside the product cylinder for three enrichment values in three different geometries. Detector array uses 44 ^{3}He tubes.

Uranium Isotopics

A possible source of uncertainty in this analysis is the relative concentration of ^{234}U and ^{235}U. Uranium feed can come from many different sources, including uranium mined in different parts of the world, depleted uranium for re-enrichment, and reprocessed uranium. For these different sources the relative abundance of ^{234}U to ^{235}U will vary. Therefore, the ratio of $^{234}U/^{235}U$ varies in the enriched uranium product, as shown in Table II for 4%, 10% and 20% enriched uranium. Small changes in the quantity of ^{234}U can impose significant changes to the number of Singles, (α,n), neutrons created in an item, also given in Table II. Therefore, a systematic error is introduced into the D/S calculation from variation of the $^{234}U/^{235}U$ ratio. This is particularly important for smaller items.

As the mass increases, induced fission of ^{235}U becomes the dominate contributor to the D/S ratio and the systematic error to the D/S ratio from the variability in the ^{234}U becomes negligible. This occurs because the ^{234}U (α,n) source contribution to the Singles also contributes to the Doubles in a proportional manner by inducing fissions in ^{235}U. In scenarios where induced fission of ^{235}U is a large contribution to the total neutron source strength, taking the ratio of D/S can significantly reduce the uncertainty created from not knowing the ^{234}U concentration. Fortunately, neutron monitoring of the product bay provides items that are usually very large in mass.

Table II. The amount of ^{234}U in an item as a percentage of ^{235}U for three different enrichments of uranium. The *Expected* column is the value used for the D/S ratio. The *Upper Limit* and *Lower Limit* represent the maximum range for different feed values.

Enrichment ^{235}U [%]	^{234}U / ^{235}U [%]			(α,n) [n/s·cm3]		
	Expected	Upper Limit	Lower Limit	Expected	Upper Limit	Lower Limit
4	0.64	1.22	0.55	1.01	1.84	0.88
10	0.65	1.43	0.59	2.45	5.24	2.23
20	0.70	1.67	0.63	5.15	12.10	4.65

Information on different ratios of $^{234}U/^{235}U$ along with SOURCES 4C code was used to calculate the upper and lower bound (α,n) and spontaneous fission rates. Calculating the D/S ratio using the source strength of the upper and lower bounds gives the systematic uncertainty due to not knowing the ^{234}U concentration of the feed stream, and ultimately, the product stream. This uncertainty can be calibrated for and is only important in GCEPs that regularly change the feed source.

Error Analysis

A count time of 1 hour is not unreasonable and is used for this work. The product cylinder remains in position for much longer than one hour, and a longer count time would improve the statistical error. However, this must be balanced against changes in the source strength that may happen during a counting period. For example, if a measurement is being taking while the product cylinder is being filled and the quantity of UF_6 inside the product cylinder triples during that time, then the D/S ratio will not be a representative measurement of the nuclear material in the product cylinder when the measurement is finished. Ultimately the count time will need to be a compromise between the rate of mass deposited and the statistical error.

The statistical uncertainty of the MCNPX calculation was small enough to be considered negligible and was not included. The MCNPX statistical uncertainty should not be confused with any errors inherent in the MCNPX models created, which are unaccounted for in this analysis. The uncertainty presented in the results are calculated by adding 3σ of the counting statistics uncertainty in quadrature with the ^{234}U uncertainty limits. If one can ensure that the same feed type is always used, the uncertainty from $^{234}U/^{235}U$ ratio will be reduced significantly. If the error bars of the D/S ratios do not overlap, a change in the enrichment produced, is detectable by the proposed neutron monitoring system, within the scope of the errors considered by this work.

Detection of HEU using the D/S ratio and Mass

For this technique to work as described above, the mass of UF_6 in the product cylinder would need to be known. Typically the facility monitors the mass with a load cell that is integrated into the low-temperature take off stations. Combining these pieces of information would allow for easy determination of the product enrichment. Continuously monitoring load cells has been studied [7] and could easily be applied to this application.

Detecting the production of undeclared enrichments with as little UF_6 inside the product cylinder as possible creates the best safeguards measure. The D/S ratio as a function of UF_6 mass inside the product cylinder for enrichments of 4%, 10% and 20% ^{235}U with the 44-tube model is shown in Figure 4.

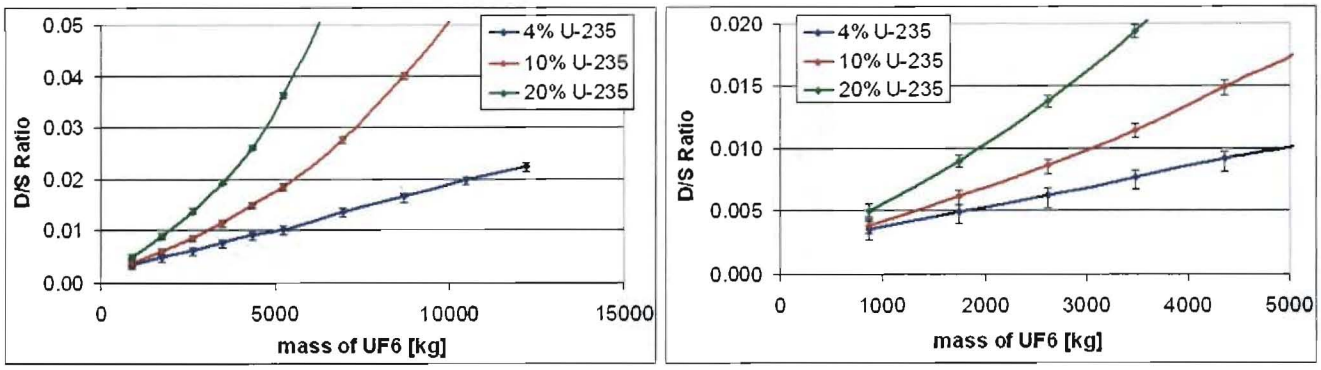

Figure 4. Doubles to Singles ratio as a function of UF_6 mass for the crescent model. D/S ratio and mass scale zoomed-in on the right for convenience. Detector array uses 44 ^{3}He tubes and a 1 hour count time.

The large mass of UF_6 that 48Y containers hold shows a strong relationship between ^{235}U present and the D/S ratio. For the 20% and 10% enriched uranium, the D/S ratio increases exponentially with mass. The 4% ^{235}U case does not exhibit this pattern because it does not have enough ^{235}U by volume to let multiplication dominate the D/S ratio. Only 5% (871 kg) of the product cylinder needs to be filled for the production of 4% enriched uranium to be distinguished from 20% enriched with a count time of one hour. The 44 ^{3}He tube model offers excellent detection capabilities for illicit production of HEU.

Ideally, the mass of UF_6 would be known; however, there are a few options available if the mass is unknown. Figure 4 shows the D/S ratio trend is very different between 4% enriched and. Therefore, trend analysis of the D/S ratio as the product cylinder fills may provide a means to detect HEU production without knowing the mass. Second, two pieces of information are produced by the neutron monitor: Singles and Doubles. By considering only one value, the D/S ratio, not all of the information available is being utilized. Considering either the Doubles or Singles in conjunction with the D/S ratio provides another way to detect HEU production without knowing the mass [1].

Array of Five Product Cylinder Bays

The product cylinder bay area is a collection of individual bays modeled at 91.5 cm apart. Understanding the amount of cross-talk that will occur from neighboring product cylinder bays is important in quantifying the detection sensitivity. The model consists of an array of 5 product cylinder bays and is depicted in Figure 5. The bay of interest, the central one, varies enrichment while the remaining 4 bays contain 4% enriched uranium to represent an expected background from neighboring bays and are 70% full (12197 kg) to increase their source strength. This configuration was chosen since it is expected to contribute the largest possible cross-talk interference.

Figure 6 shows the calculated D/S ratio in a single product cylinder bay and an array of 5 product cylinder bays. The addition of large quantities of 4% enriched UF_6 in neighboring product cylinder bays have a negligible effect on the ability of the system to detect HEU production. The contribution of detected neutrons from the additional product cylinder bays is an order of magnitude less than the contribution from the product cylinder of interest. This impact is further trivialized when the ratio of Doubles to Singles is taken.

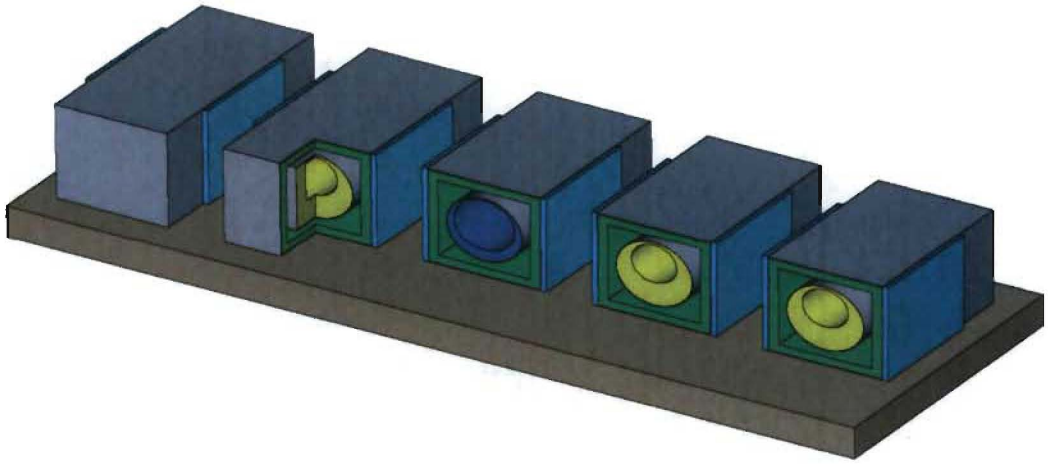

Figure 5. MCNPX model cutout of five product cylinder bays in an array. The central product cylinder bay varies its uranium enrichment and mass of UF_6 (blue); while the peripheral bays contain 4% enriched uranium and stay 70% full (yellow).

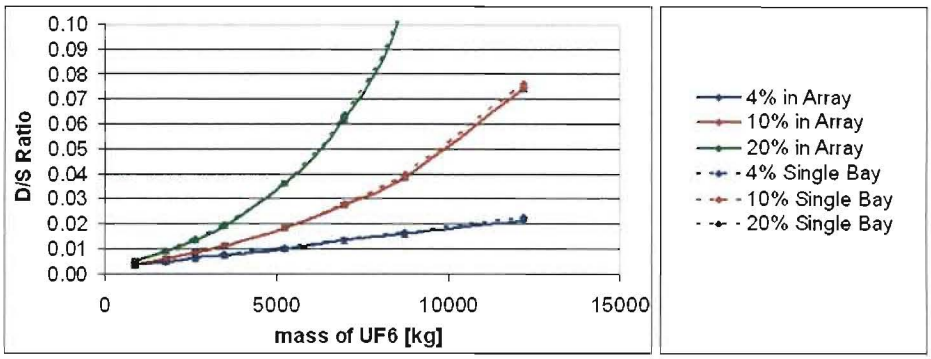

Figure 6. Calculation comparing the D/S ratio between a single product cylinder and an array of product cylinders. Calculated using the 44 tube model with a 1 hour count time.

Increasing the enrichment inside the central product cylinder bay reduces the effect of the signal from neighboring bays full of 4% UF_6. At higher enrichments and greater mass, the D/S ratio is dominated by the induced fissions, not the initial source of neutrons (spontaneous fission and (α,n) reactions). Neutrons that originate from neighboring bays, are detected and skew the D/S ratio results, however, they also contribute to the induced multiplication inside the item being measured, which has the effect of bringing the D/S ratio back to what it would be without the surrounding UF_6.

In practice, each product cylinder bay in the array would have its own set of ^3He tubes, providing additional information. From this information, a system can be set up to account for the noise of neighboring bays, and adjust the D/S ratio accordingly, further reducing what little impact is observed in 4% enriched case.

12 Tube Model

The current cost of ^3He makes elaborate detector designs of high efficiency impractical for wide distribution. Therefore, the effect of using significantly less ^3He tubes is examined. A MCNPX model

was created that uses only 12 ^{3}He tubes instead of 44. The 12-tube detector system is about a fifth as efficient at detecting neutrons from the product bay, which is particularly damaging to the accuracy of coincidence counting. Figure 7 shows the results of the 12 tube model.

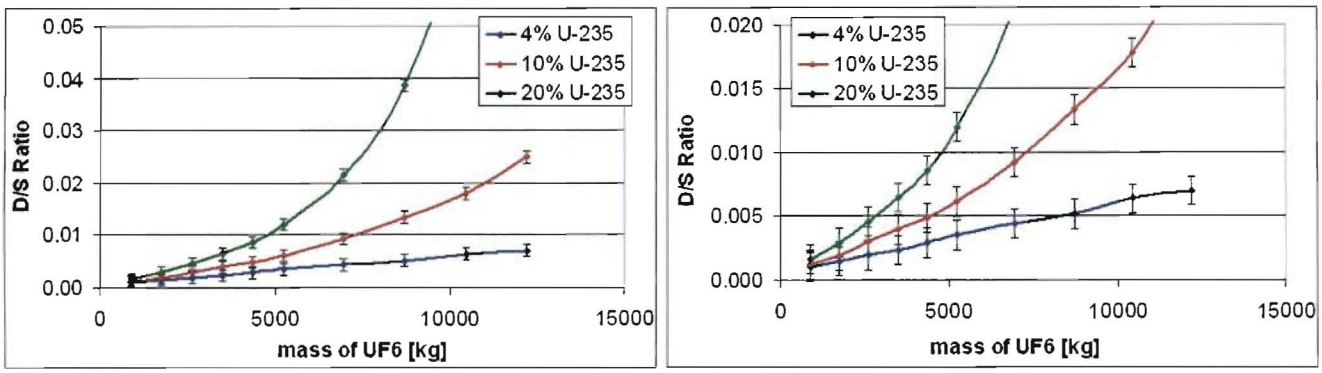

Figure 7. Doubles to Singles ratio as a function of UF_6 mass for the crescent model. D/S ratio and mass scale zoomed-in on the right for convenience. Detector array uses 12 ^{3}He tubes and a 1 hour count time.

While the D/S ratio for different enrichments in the 12-tube model is not as cleanly resolved as the 44-tube model, production of 20% enriched uranium can still be distinguished from 4% enrichment when the product cylinder is only 10% full (1742 kg). At least 25% of the product cylinder needs to be filled (4356 kg) to determine if the product is enriched to 10% ^{235}U instead of 4%. Depending on the plant operating procedures, it may be possible to reduce the counting statistics by increasing the measurement time. To further improve detection limits, the systematic error may be decreased with knowledge of the ^{234}U/^{235}U ratio. Operator declaration of ^{234}U or using a similar monitoring method around the feed cylinders to bound the possible range of ^{234}U concentration are possible solutions.

Conclusion

Neutron coincidence counting on the product cylinder bay offers an opportunity to monitor relatively few items that represent the overall activity of the GCEP. Using 44 ^{3}He tubes, on two sides of the product cylinder bay, measuring the Doubles to Singles ratio can determine the enrichment of uranium being produced at the GCEP. Assuming that the mass of product UF_6 in the cylinder is also measured, production of 20% enriched uranium is distinguishable from 4% enriched uranium when the product cylinder is only 5% full.

Calculations suggest that uncertainty in the geometry of how UF_6 collects in the product cylinders are not a detriment to neutron coincidence counting providing a means for unattended monitoring for HEU production. Similarly, this study found that material in neighboring product cylinders will not significantly impede detection capabilities.

The systematic error of not knowing the ^{234}U concentration creates bias in the (α,n) source term, which raises the lower level detection limit. However, obtaining knowledge about the ^{234}U concentration from operator declaration or by using a neutron monitoring system at the feed cylinders stations offers the potential to mitigate this systematic error.

When using only 12 ^{3}He tubes for neutron monitoring, the decreased detector efficiency does not pose a significant obstacle. Comparing the D/S ratio and mass allows resolution between the production of 20% enriched uranium and 4% when the product cylinder is at least 15% full. If less fidelity to detect HEU production is tolerable, even less ^{3}He tubes are needed.

Without knowledge of mass, detection of HEU production is still possible. As UF_6 mass inside the product cylinder increases, the D/S ratio for different enrichments have different trends. Therefore, trend analysis of the D/S ratio as the product cylinder fills may provide a means to detect HEU production without knowing the mass. Also, using the D/S ratio with the Singles or Doubles can be substituted for knowledge of mass inside the product cylinder.

This study found monitoring the product cylinder bays with neutron coincidence counting has high potential as a significant safeguard against illicit production of HEU in Gaseous Centrifuge Enrichment Plants. However, effective and complete safeguards at a gas centrifuge facility will most likely need several systems integrated together to achieve effective safeguards. This approach is often referred to as defense in depth. Any one safeguards system can be defeated but several systems integrated together will be much more difficult to circumvent. The neutron monitor used in conjunction with a load cell monitoring, tags and seals, and camera surveillance is one approach for effective safeguards at an enrichment facility.

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