

A New Technique for Uranium Cylinder Assay Using Passive Neutron Self-Interrogation

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Abstract. To achieve material balance over an entire uranium enrichment plant, it is essential to verify the mass and enrichment of uranium hexafluoride (UF₆) transferred in and out of the facility. A large portion of the UF₆ is contained in 30B and 48Y cylinders. Traditional gamma-ray methods for UF₆ cylinder assay measure the net counts in the 186-keV peak and rely on a second measurement of the cylinder wall thickness for the attenuation correction. Limiting factors of gamma-ray techniques include difficulties with non-homogenized cylinders, measurement of natural and depleted uranium, and operation in unattended mode. Neutrons provide more penetration into UF₆ cylinders than gamma rays and can easily be used in unattended mode. This paper describes a new Passive Neutron Enrichment Meter (PNEM) approach that uses total and coincidence neutrons to simultaneously determine the mass and enrichment of UF₆ within 30B and 48Y cylinders. The system consists of Helium-3 tubes embedded in polyethylene with a removable cadmium cover. It can be used to assay feed, product, and tails cylinders. The primary source of neutrons in enriched UF₆ comes from the alpha bombardment of fluorine, where U-234 is the principal alpha emitter. In general, the enrichment of U-234 follows that of U-235. The (alpha,n) neutrons dominate the singles count rate with a minor contribution from induced fission in U-235. The doubles count rate in enriched uranium comes primarily from induced fission (i.e., multiplication) in U-235. The PNEM concept makes use of the doubles counts, which provide a measure of U-235 enrichment, and singles counts, which can be used to infer uranium mass when the U-235 enrichment is known. The cadmium ratio is a useful relationship for natural and depleted uranium, where the doubles are dominated by spontaneous fission of U-238. The cadmium ratio has the desirable feature that all of the thermal-neutron-induced fissions in U-235 are independent of the original neutron's source. Thus, the cadmium ratio is independent of the uranium age, purity, and prior reactor history.

1. Introduction

Safeguarding sensitive fuel cycle technology such as uranium enrichment is a critical component in preventing the spread of nuclear weapons. To achieve material balance over an entire uranium enrichment plant, it is essential to verify the mass and enrichment of UF₆ transferred in and out of the facility. A large portion of the UF₆ is contained in 30B and 48Y cylinders. 30B cylinders contain the product UF₆, with typical enrichments of 2-5% ²³⁵U. 48Y cylinders hold the feed (natural uranium) and tails (depleted uranium).

Traditionally, enrichment is verified using a gamma-ray technique that measures the 186-keV peak from ²³⁵U. The 186-keV gamma can only penetrate a small distance in the UF₆, making it difficult to get a representative sample of a heterogeneous cylinder. For example, there is often a heel of UF₆ plated on the inner surface of the cylinder from previous use, which can cause erroneous results. 30B and 48Y cylinders are physically large and have a thick steel wall. The technique requires knowledge or a measurement of the cylinder wall thickness to correct for attenuation in the steel. A small error in the wall thickness (e.g., from rust or crud on the cylinder) can result in a large error in the measured enrichment.

The mass of uranium inside the cylinder is typically determined using a load cell or electronic scale. These systems are often used by both the operator and the inspector, making authentication difficult. They also require reference weights for calibration and a valid tare weight for each cylinder. Furthermore, there is no indication of whether the material inside the cylinder is nuclear material. More recently, a system called the Uranium Cylinder Assay System (UCAS) was installed at Rokkasho Enrichment Plant in Japan that uses total neutron counting to determine uranium mass in 30B and 48Y cylinders [1]. It has the added benefit of

verifying that the material is, in fact, nuclear material. However, UCAS was designed to be an operator system, not an inspector system.

This paper describes a new Passive Neutron Enrichment Meter (PNEM) approach that uses total and coincidence neutrons to simultaneously verify the full-volume mass and enrichment of UF₆ within feed, product, and tails cylinders. The system consists of two briefcase-sized detector pods with twelve ³He tubes each. The tubes are embedded in polyethylene with a removable cadmium cover. The remaining sections of this paper address the PNEM design optimization, physics calculations, and expected sources of uncertainty.

2. Design optimization

PNEM was designed to be a portable instrument. Each pod has dimensions of 61×34×10 cm³, weighs approximately 20 kg, and has a handle on one side for carrying. The ³He tubes are at a pressure of 4 atm. Fig. 1 shows the proposed measurement position with respect to a 30B cylinder (cylinder cradle not shown). The measurements can be done in the storage area of the enrichment plant by placing the detector pods on the floor on either side of the cylinder. The measurement position is similar for 48Y cylinders.

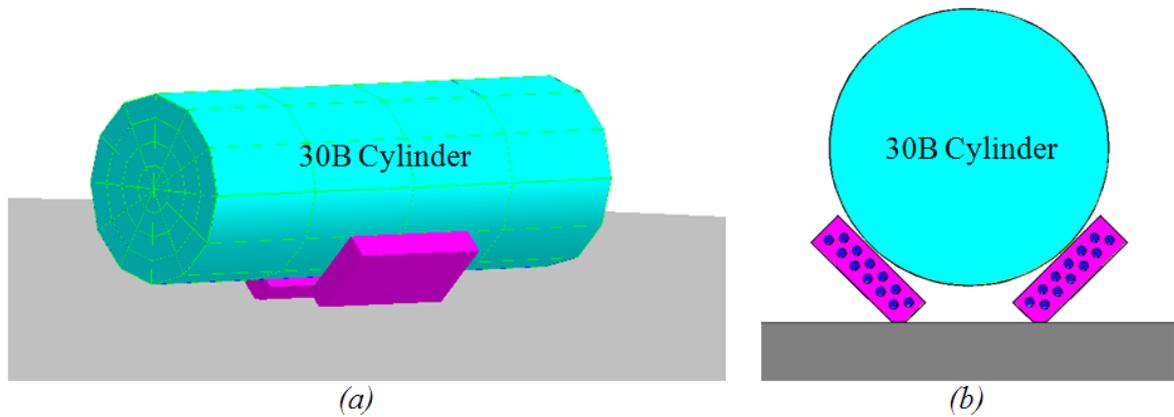


Fig. 1. PNEM measurement position with respect to a 30B cylinder (cylinder cradle not shown): (a) 3-D view and (b) front end view showing ³He-tube positioning.

The amount of polyethylene between the top of the pod and the first row of tubes was set at 1.9 cm, which was the optimal tube depth for the UCAS unit. In the PNEM measurements, the accidental (A) rate will be one or two orders of magnitude larger than the reals (R) rate. Under this condition, the error in A dominates the statistical uncertainty. Menlove, Beddingfield, and Schear showed that if A is much larger than R, then the relative uncertainty in the reals is given by [2]:

$$\frac{\sigma_R}{R} \approx \frac{\sqrt{A}}{R}.$$

Thus, we can write

$$\frac{\sigma_R}{R} \approx \frac{\sqrt{GT^2}}{R},$$

where G is the coincidence gate width and T is the totals rate. Because the totals rate is proportional to the efficiency (ϵ) of the detector, the reals rate is proportional to the efficiency squared (ϵ^2), and the gate width is proportional to the die-away time in the detector (τ), we can say that the relative uncertainty is proportional to the square root of the die-away time divided by the efficiency:

$$\frac{\sigma_R}{R} \propto \frac{\sqrt{\tau}}{\epsilon}.$$

The position of the second row of tubes was optimized by maximizing a figure of merit (FOM) defined as

$$FOM = \frac{\varepsilon}{\sqrt{\tau}}$$

In other words, the FOM identifies the tube positioning that minimizes the statistical uncertainty in the detector. Monte Carlo N-Particle Extended (MCNPX) was used to simulate PNEM measurements of a fully-filled 30B cylinder containing various enrichments of UF₆. Five different tube positions were modeled, and the results are shown in Fig. 2.

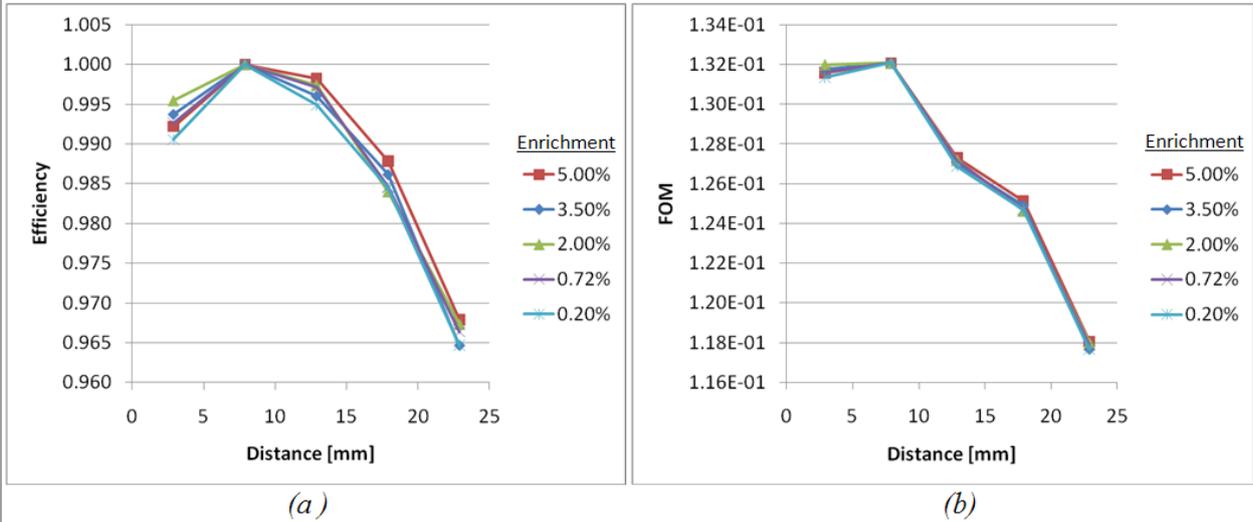


Fig. 2. Results showing the optimization of the ³He tube positioning in PNEM: (a) normalized efficiency and (b) figure of merit, both as a function of the distance between the first and second rows of tubes.

Fig. 2(a) shows the normalized efficiency as a function of the vertical distance between the first and second row of tubes (between the outer radii). It shows a clear peak at a distance of 8 mm for all five enrichments. Fig. 2(b) shows the FOM as a function of the distance between the rows of tubes. The die-away time was calculated using the following relationship from the PANDA Manual [3]:

$$\tau = -\frac{-G_{32}}{\ln\left(\frac{R_{64}}{R_{32}} - 1\right)},$$

where G_{32} refers to a 32 μ sec gate width and R_{32} and R_{64} are the real values for simulations using 32 and 64 μ sec gate widths, respectively. The FOM curves also peak at a distance of 8 mm, indicating the optimal tube spacing to minimize the statistical error.

3. Physics calculations

The primary source of neutrons in enriched UF₆ comes from the alpha bombardment of fluorine, where ²³⁴U is the principal alpha emitter. In general, the enrichment of ²³⁴U follows that of ²³⁵U. The (α ,n) neutrons dominate the singles count rate with a minor contribution from induced fission in ²³⁵U. The doubles count rate in enriched uranium comes primarily from induced fission (i.e., multiplication) in ²³⁵U. The PNEM concept makes use of the doubles counts, which provide a measure of ²³⁵U enrichment, and singles counts, which can be used to infer uranium mass. The cadmium ratio is a useful relationship for natural and depleted uranium, where the doubles are dominated by spontaneous fission of ²³⁸U. The cadmium ratio has the desirable feature that all of the thermal-neutron-induced fissions in ²³⁵U are independent of the original neutron's source. Thus, the cadmium ratio is independent of the uranium age, purity, and prior reactor history. The physics calculations for 30B and 48Y cylinders described below were performed using MCNPX.

3.1. 30B cylinders

For the 30B cylinder test cases, we modeled cylinders containing enrichments ranging from 2-5% ²³⁵U. Three different fill levels (100%, 90%, and 80%) were modeled for each enrichment. The fill levels are

correlated to the mass of uranium in each cylinder. Fig. 3 shows the results. Given a known or operator-declared uranium mass, the enrichment can be determined using the doubles-to-singles ratio shown in Fig. 3(a). The mass can then be verified using the singles rate, shown in Fig. 3(b).

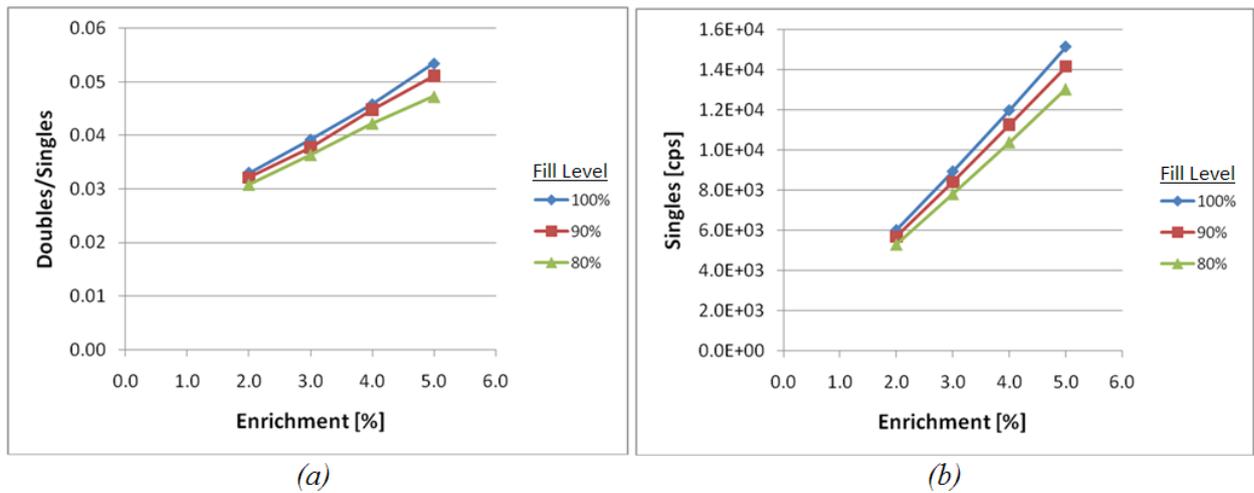


Fig. 3. MCNPX 30B cylinder modeling results showing (a) the doubles-to-singles ratio as a function of enrichment and (b) the singles count rate as a function of enrichment, both for various fill levels.

3.2. 48Y cylinders

For the 48Y cylinder test cases, we modeled cylinders containing natural (0.711% ^{235}U) and depleted UF_6 (0.30% and 0.20% ^{235}U). Again, three different fill levels were modeled for each enrichment, which are correlated to the mass of uranium in each cylinder. Fig. 4 shows the results. The enrichment can be determined using the doubles-to-singles ratio shown in Fig. 4(a). The doubles rate is primarily from ^{238}U spontaneous fission that is almost constant, so the negative slope of the double-to-singles ratio is a result of the increase in the ^{234}U singles rate. One feature to notice is that the doubles-to-singles ratio is fairly independent of the uranium mass. The mass can also be verified using the singles rate, shown in Fig. 4(b).

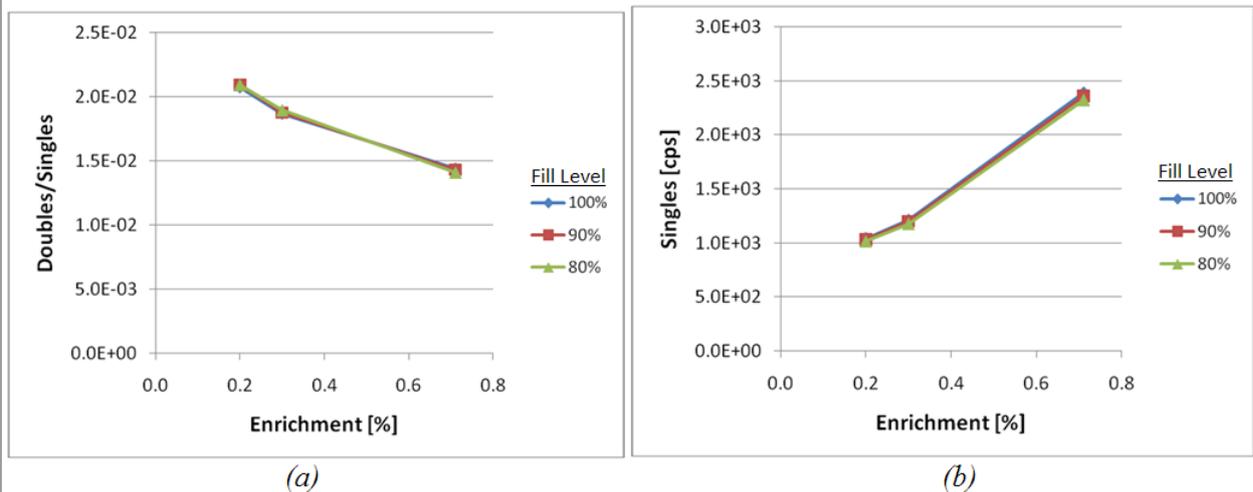


Fig. 4. MCNPX 48Y cylinder modeling results showing (a) the doubles-to-singles ratio as a function of enrichment and (b) the singles count rate as a function of enrichment, both for various fill levels.

In addition to the doubles-to-singles ratio, the cadmium ratio also shows potential as a signature for the uranium enrichment in 48Y cylinders. This is shown in Fig. 5 for the fully-filled 48Y cylinder. The cadmium ratio is independent of the ^{234}U content of the UF_6 . The advantages and disadvantages of both signatures need to be studied further to determine which one should be used.

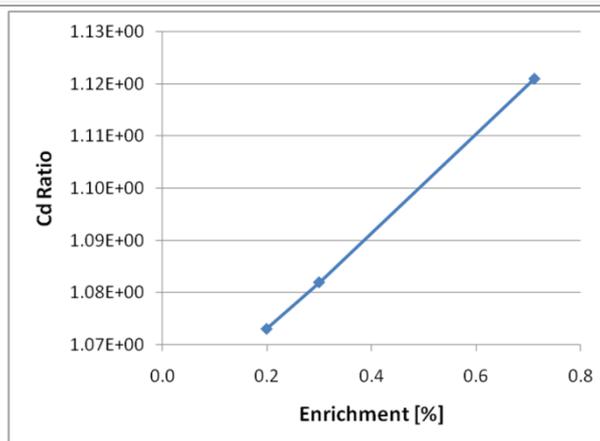


Fig. 5. MCNPX 48Y cylinder modeling results showing the cadmium ratio as a function of enrichment.

4. Uncertainties

4.1. Random uncertainty

Statistical uncertainties are very complex in coincidence counting because the input pulse train contains overlapping random and correlated events. The estimated statistical uncertainties for 30B and 48Y cylinders containing various enrichments of UF_6 are shown in Fig. 6. The results are for cylinders that are at the maximum fill level. In the 30B case, a 1% uncertainty can be achieved in a fifteen-minute count time. In the 48Y case, the depleted cylinders achieve 2% uncertainty in fifteen minutes, but the natural cylinder requires nearly thirty minutes to reach 2% uncertainty.

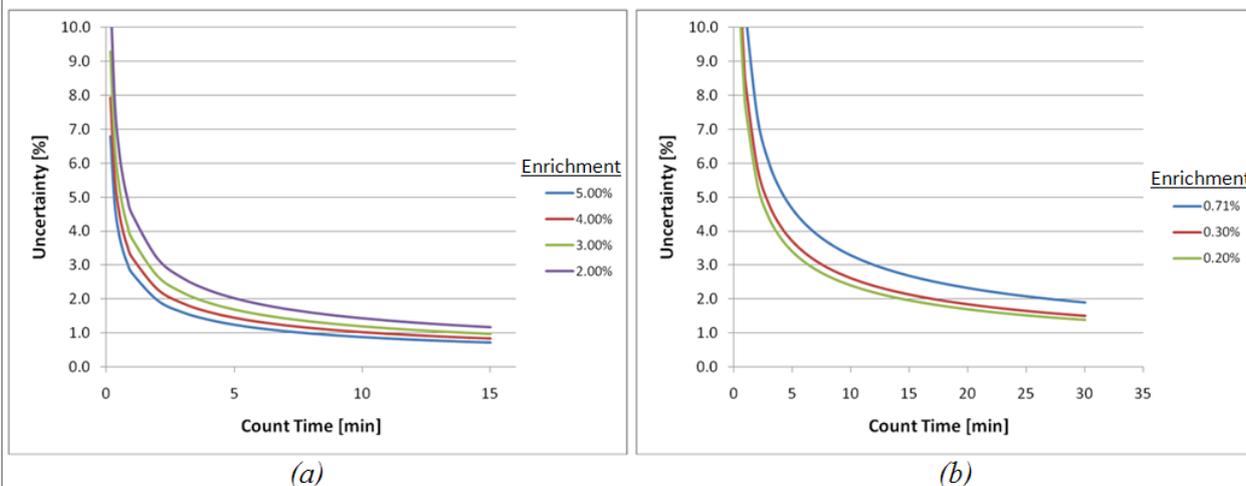


Fig. 6. Statistical uncertainty as a function of count time for (a) 30B cylinders and (b) 48Y cylinders.

4.2. Systematic uncertainty

The biggest sources of systematic uncertainty for PNEM stem from variations in the ^{234}U content of the uranium and the distribution of UF_6 within the cylinder. The ^{234}U content is only a factor for the mass verification because it affects the singles count rate. In the doubles-to-singles and cadmium ratios, the effects of the ^{234}U content essentially cancel out. The ^{234}U fraction can vary depending on how the plant is operated. For example, the ^{234}U content depends on the type of feed material that is enriched. Ore-based (i.e., not reprocessed) natural UF_6 has a very consistent ^{234}U content of about 52-55 ppm. If reprocessed or depleted UF_6 is used as feed material, the ^{234}U content will vary for a given enrichment. The magnitude of these variations and whether they can be accounted for in calibration are topics that require further study. For enrichment plants that have relatively consistent operating parameters, it is possible to estimate the ^{234}U content based on the enrichment level with reasonable accuracy.

The distribution of UF_6 within the cylinder is also a factor in the systematic uncertainty. The geometry

effects are more pronounced in 30B cylinders, where multiplication plays a bigger role in the doubles count rate, than in 48Y cylinders. The UF_6 profile inside the cylinder depends on how the cylinder was filled and the storage conditions. Berndt, Franke, and Mortreau used the filling profiles shown in Fig. 7 in their modeling study of geometry effects on a theoretical total neutron counter for UF_6 cylinders [4]. The x-factor describes the percentage of UF_6 covering the inner cylinder walls with a layer of constant thickness.

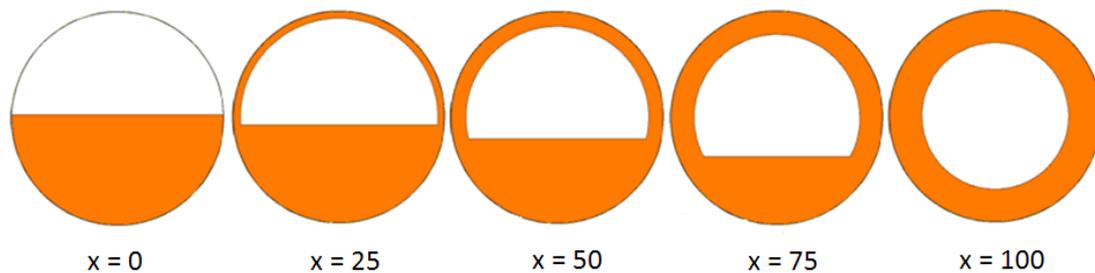


Fig. 7. UF_6 filling profiles [4].

Natural UF_6 cylinders from conversion plants are filled in liquid phase, meaning the UF_6 collects at the bottom of the cylinder. This is illustrated by the $x=0$ case. Product and tails cylinders are generally filled by desublimation, where solid UF_6 adheres primarily to the cylinder wall, creating an annular ring. This is illustrated by the $x=100$ case. Over time, the UF_6 on the upper part of the wall will slough off and fall to the bottom ($x=25, 50, \text{ and } 75$).

The filling profiles shown in Fig. 7 represent the extreme bounding cases. In practice, the true range of filling profiles for most cylinders is a smaller subset of Fig. 7. The size of that subset is something that needs further study. Preliminary MCNPX modeling results show that a good signature for the x-factor can be obtained by placing a third detector pod on top of the cylinder and looking at the ratio between the top and bottom pods. To get a better understanding of the true range of filling profiles, measurements should be taken on a large population of cylinders. This type of measurement campaign would help quantify the systematic uncertainty associated with the distribution of UF_6 inside 30B and 48Y cylinders.

5. Summary and conclusions

In this paper, we have presented a new Passive Neutron Enrichment Meter (PNEM) approach to UF_6 cylinder assay. It uses total and coincidence neutrons to simultaneously verify the full-volume mass and enrichment of UF_6 within 30B and 48Y cylinders. The cadmium ratio can be used for 48Y cylinders and for cylinders containing recycled uranium. The PNEM system consists of two portable, ^3He -based detector pods. The detectors were optimized using a figure of merit that minimizes the statistical uncertainty in the counts. Physics calculations were performed using MCNPX to study the signatures that will be used for verifying mass and enrichment of UF_6 in both 30B and 48Y cylinders. Statistical uncertainties of 1-2% can be achieved with count times of 15 minutes for 30B cylinders and 30 minutes for 48Y cylinders. Finally, the biggest source of systematic uncertainty stems from the distribution of UF_6 within the cylinders.

Future work on this project includes field measurements to prove that the observables can be used to verify uranium mass and enrichment. We also plan to continue studying the magnitude of the systematic uncertainties as well as the sensitivity of the PNEM approach to those uncertainties.

6. Acknowledgments

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