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Author(s): Howard O. Menlove
Martyn T. Swinhoe
Karen A. Miller

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A More Accurate and Penetrating Method to Measure the Enrichment and Mass of UF_6 Storage Containers Using Passive Neutron Self-Interrogation

Howard O. Menlove, Martyn T. Swinhoe, Karen A. Miller

Los Alamos National Laboratory, Nuclear Nonproliferation Division, Los Alamos, NM
E-mail: hmenlove@lanl.gov, swinhoe@lanl.gov, kamiller@lanl.gov

ABSTRACT

This paper describes an unattended mode neutron measurement that can provide the enrichment of the uranium in UF_6 cylinders. The new passive neutron measurement provides better penetration into the uranium mass than prior gamma-ray enrichment measurement methods. The Passive Neutron Enrichment Monitor (PNEM) provides a new measurement technique that uses passive neutron totals and coincidence counting together with neutron self-interrogation to measure the enrichment in the cylinders. The measurement uses the neutron rates from two detector pods. One of the pods has a bare polyethylene surface next to the cylinder and the other polyethylene surface is covered with Cd to prevent thermal neutrons from returning to the cylinder. The primary neutron source from the enriched UF_6 is the alpha-particle decay from the ^{234}U that interacts with the fluorine to produce random neutrons. The singles neutron counting rate is dominated by the ^{234}U neutrons with a minor contribution from the induced fissions in the ^{235}U. However, the doubles counting rate comes primarily from the induced fissions (i.e., multiplication) in the ^{235}U in enriched uranium. The PNEM concept makes use of the passive neutrons that are initially produced from the ^{234}U reactions that track the ^{235}U enrichment during the enrichment process. The induced fission reactions from the thermal-neutron albedo are all from the ^{235}U and provide a measurement of the ^{235}U. The Cd ratio has the desirable feature that all of the thermal-neutron-induced fissions in ^{235}U are independent of the original neutron source. Thus, the ratio is independent of the uranium age, purity, and prior reactor history.

INTRODUCTION AND BACKGROUND

For safeguarding uranium enrichment plants, it is essential to verify the mass and enrichment of the UF_6 located in the plant and the mass that is transferred in and out of the facility. A large fraction of the UF_6 mass is contained in the 30B and 48Y cylinders that hold the feed uranium (natural uranium), the product output (2-5% typical enrichment), and the tails (depleted uranium). These cylinders are large in size and have heavy steel walls. The gamma-ray enrichment measurement is difficult because of the heavy steel wall and, because the 186 keV gammas can only penetrate a small thickness of UF6. Thin deposits of UF_6 on the inside of the walls from prior use of the cylinders can cause erroneous results. For the gamma-ray based enrichment measurements, a separate thickness measurement is required to correct for the attenuation of the 186 keV gamma rays by the steel. The IAEA is considering a measurement station at the input/output of the enrichment plant that would measure all cylinders that enter or leave the plant [1]. This paper describes an attended or unattended mode neutron measurement that can provide the enrichment and mass of the uranium in the UF_6 cylinders. The passive self-interrogation neutron measurement provides better penetration into the uranium than prior enrichment measurement methods. This same basic measurement technique was used to measure the ^{235}U mass in highly enrichment 5A cylinders in 1986 by Menlove and Stewart [2].

PNEM CONCEPT

This paper provides a description of the PNEM measurement technique that uses passive neutron totals and coincidence counting together with neutron self-interrogation to .measure the enrichment and the UF_6 mass in 30B and 48Y cylinders. The measurement uses the passive neutron singles and induced-fission doubles rates from two neutron detector pods. One of the pods has a bare polyethylene surface next to the cylinder and creates thermal neutrons that return to the cylinder, and the other polyethylene surface is covered with Cd or boron to prevent thermal neutrons from returning to the cylinder. The two-detector configuration is illustrated in Fig. 1. The primary neutron source from the enriched UF_6 is the alpha-particle decay from the ^{234}U that interacts with the fluorine to produce (α,n) neutrons. Another weaker source of neutrons is ^{238}U spontaneous fission. These spontaneous fission neutrons are an order of magnitude less abundant than the (α,n) neutrons for the enriched uranium, but the spontaneous fission neutron fraction is similar to the ^{234}U neutrons for the natural uranium. For depleted uranium, the spontaneous fission source becomes equal to the ^{234}U component

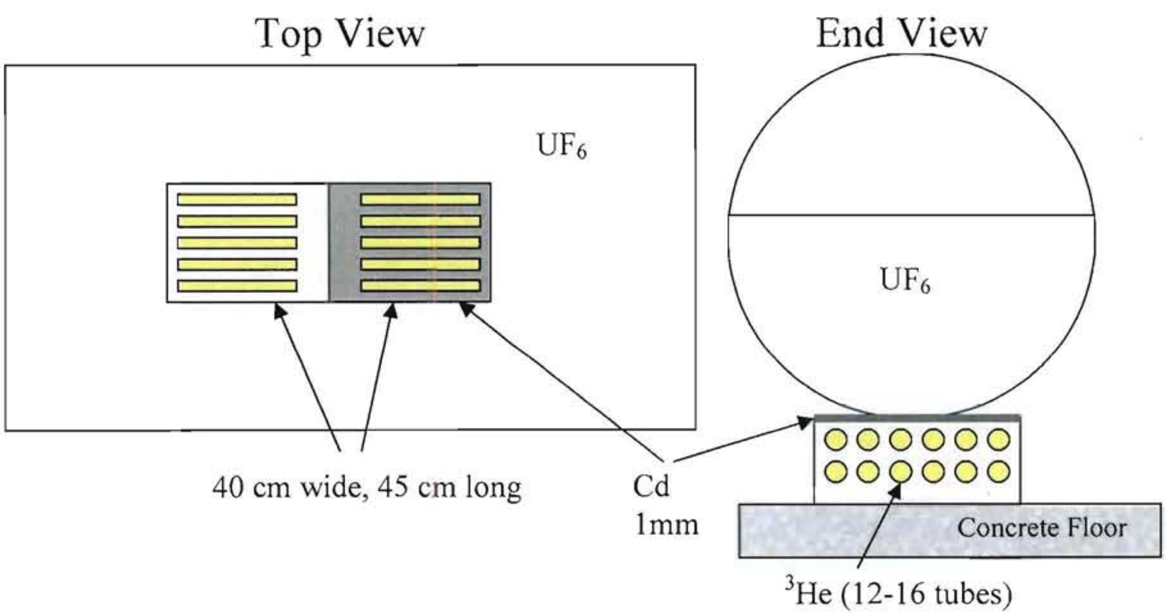

Fig. 1. Top and end views of the PNEM concept for an installed system showing the bare and Cd-lined detectors positioned near the bottom surface of a 30B UF_6 cylinder.

For enriched uranium, the singles neutron count rate is dominated by the ^{234}U (α,n) neutrons with a minor contribution from the induced fissions in the ^{235}U. However, the doubles counting rate comes primarily from the induced fissions (i.e., multiplication) in the ^{235}U in the enriched uranium. The PNEM concept makes use of the passive neutrons from the ^{234}U reactions that track the ^{235}U enrichment during the enrichment process. For non-recycle UF_6, a passive neutron measurement on the outside of the cylinder can provide the mass of ^{234}U in the cylinder but not the enrichment.

The Cd Ratio Measurement

The induced fission reactions from the thermal-neutron reflection are all from the ^{235}U and provide a measurement of the ^{235}U. The 30B cylinders contain a large mass of UF_6 that is infinitely deep to the thermal-neutron interrogation, so all enrichments of uranium absorb all of the reflected thermal neutrons, but the penetration distance increases as the enrichment decreases. As the penetration depth increases for the lower enrichments, the parasitic absorption of neutron in the ^{238}U increases, and the induced fissions in the ^{235}U per source neutron decrease. Thus, the Cd ratio increases as a function of the enrichment and the ratio provides an enrichment measurement.

The Cd ratio, which is measured by the ratio of the counts in the two detector slabs, has the desirable feature that all of the thermal-neutron-induced fissions in ^{235}U are independent of the original neutron source. *Thus, the Cd ratio is independent of the uranium age, purity, and prior reactor history.* For attended mode measurements, the Cd ratio can be obtained using only one detector slab by manually removing the Cd liner that is positioned between the UF_6 and the detector slab. For installed unattended mode applications, two detector slabs can be used to eliminate the need for any mechanical motion. The PNEM system then becomes very similar to a host of unattended neutron coincidence counters that are in routine use by the IAEA [3].

Doubles/Singles Ratio

In addition to the Cd ratio, the data can be analyzed for both totals rates (S) and coincidence doubles rates (D). For the UF_6 cylinders, there is fast-neutron multiplication that peaks in the center of the UF_6 mass and thermal-neutron-induced fission that peaks near the polyethylene boundary for the detector that has no Cd cover. Both the D/S and Cd ratios are independent the neutron source rate, which cancels in the ratios. The D counts in the Cd-covered detector are primarily from fast-neutron fission in the UF_6; whereas, the D counts in the bare detector are from both fast-neutron fission and thermal-energy fission because of the reflected thermal neutrons.

Table 1 lists the measured parameters that can be used to solve for the variables that include the ^{235}U enrichment, the ^{234}U mass, the uranium mass, and the multiplication (M). The UF_6 cylinders have a known size and shape, so the relationship between the density, enrichment, and multiplication can be estimated using Monte Carlo N-Particle Extended (MCNPX) simulations [4].

Table 1. Measured Parameters and Variables of Safeguards Interest.

Measured Parameter	Cd	Variable of Key Interest
Singles	no	^{234}U mass +235U concentration
Singles	yes	^{234}U mass
Singles (Cd ratio)	no/yes	^{235}U enrichment
Doubles	no	^{235}U enrichment, M
Doubles	yes	^{235}U enrichment, M
Doubles (Cd ratio)	no/yes	^{235}U enrichment, M
D/S (no Cd)	no	^{235}U enrichment, M, ^{235}U mass
D/S (Cd)	yes	^{235}U enrichment, M, ^{235}U mass

EQUIPMENT DESCRIPTION

For enrichment plant applications, the detectors could be operating in the attended or unattended mode using a shift register equivalent for the data collection. MCNPX simulations have been performed to estimate count rates and statistical precision. The simulated detector contained two rows of ^{3}He tubes to provide high efficiency near the surface of the UF_6 cylinders as illustrated in Fig. 1. The ^{3}He tubes were 2.54 cm diameter with a length of 40 cm and embedded in high-density polyethylene. The Cd liner that was positioned on the face of the one detector slab was 1 mm thick. Various detector tube configurations were evaluated to optimize the signal rates and the thermal-neutron-reflection fraction. High efficiency is required for the doubles counting statistics, and the front row of ^{3}He tubes need to be approximately 2 cm back from the detector face so as not to interfere with the thermal-neutron return into the UF_6 cylinder.

RESULTS AND EVALUATION

MCNPX simulations were used to calculate the S and D rates as a function of enrichment and fill mass fraction. The fill mass fraction is the percentage of the maximum fill level of the cylinder. The results for the S rates are illustrated in Fig. 2 where we can see that the rates depend on both the enrichment and the fill mass. Fig. 3 shows the D/S ratio after making a correction for the ^{238}U spontaneous fission neutrons. Figs. 2 and 3 show that the measured response is a function of both the enrichment and the fill mass fraction. For enrichments above 2%, this correction is very small. The D/S ratio tracks the enrichment with only a small (< 10%) dependence on mass for UF_6 fill levels that are more than half full.

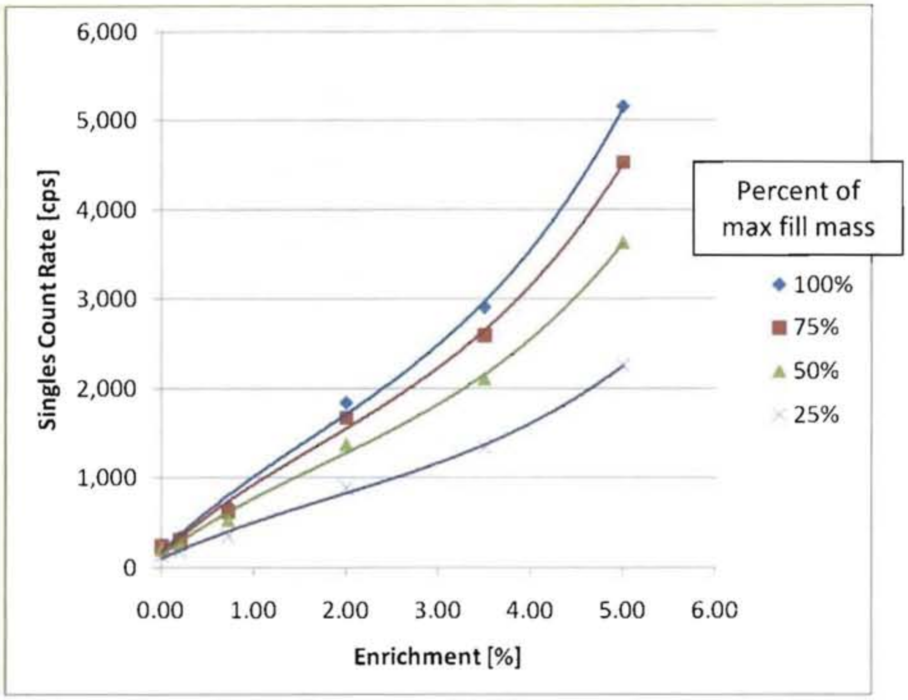

Fig. 2. Singles rate as a function of ^{235}U enrichment and percentage of maximum cylinder fill mass for a 30B UF_6 cylinder.

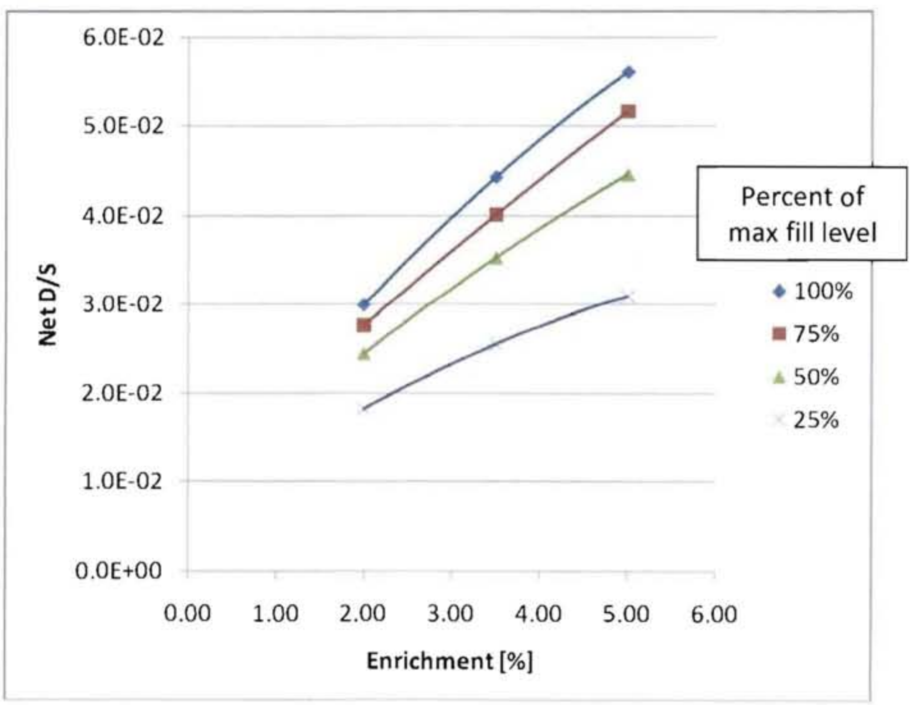

Fig. 3. Doubles/singles ratio as a function of ^{235}U enrichment for a 30B UF_6 cylinder containing low-enriched UF_6.

For the enrichment case using natural uranium, the ratio of $^{234}U/^{235}U$ is expected to have a known relationship to the enrichment for a particular plant. Both the enrichment and the mass fraction can be determined using Figs. 1 and 2 with an iterative method because the S rate is much more dependent on the fill mass than the D/S ratio. For enrichment levels above 2%, we can use the D/S ratio to determine the enrichment with the full mass calibration assumption. Then with the enrichment from Fig. 1, we can use the S response in Fig. 2 to determine the corresponding fill mass. We next return to the D/S calibration to determine the more accurate value of the enrichment. This iterative process is repeated to the convergence of the enrichment and mass for enrichment levels above ~ 2%. For natural and depleted uranium in 48Y cylinders the D/S ratio departs from the curves in Fig. 1, and we can measure the cylinders using the Cd ratio.

The measurement statistical uncertainties for the PNEM method are reasonably good. The count rates for the simulated detector were high enough to provide good counting statistical uncertainties in less than five minutes. The singles rates were approximately 2,000 cps for an enrichment of 3.5%; thus, a few seconds of counting provides better than 1% statistics for the singles rate. On the other hand, the doubles rates are much less and about 600 seconds of counting is required to obtain 1.5% statistical precision. The statistical precision to match the current gamma-ray based techniques can be obtained in less than one minute [5].

For the 48Y cylinders, which hold natural and depleted uranium, the ^{234}U and ^{235}U mass fractions become very small and the spontaneous fission from the ^{238}U becomes the dominant source for the D rate. The D/S rate is then dependent on the ^{238}U spontaneous fraction of the neutron emission rate. However, the Cd ratio is still sensitive to the ^{235}U enrichment. Fig. 4 shows the MCNPX simulation of the singles rate Cd ratio results that can be used for the natural and depleted uranium cases. The singles Cd ratio tracks the enrichment with good sensitivity for the depleted and natural uranium cases.

The Cd ratio for the D rate is focused on the UF_6 enrichment near the interface of the bare detector (i.e., no Cd) and UF_6 cylinder. Fig. 5 shows that the enrichment measurement is relatively independent of the fill mass because the induced fissions are from the local volume that is near the interface. The ratio tends to saturate for enrichment levels above 2%, so the Cd ratio for D would be used for 48Y cylinders that contain the natural and depleted uranium.

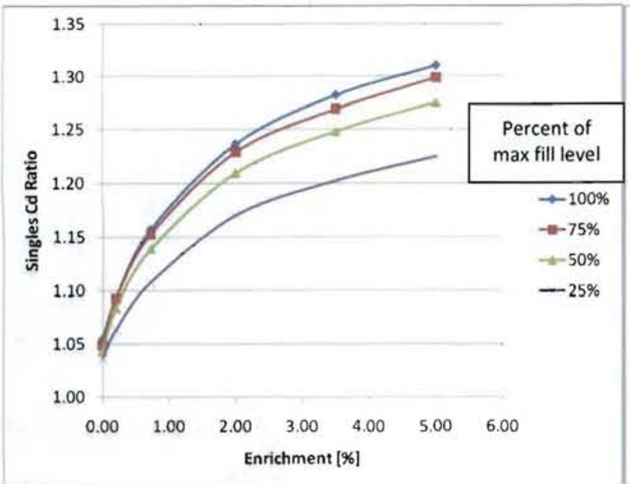

Fig. 4. Singles Cd ratio as a function of ^{235}U enrichment and percentage of maximum cylinder fill level.

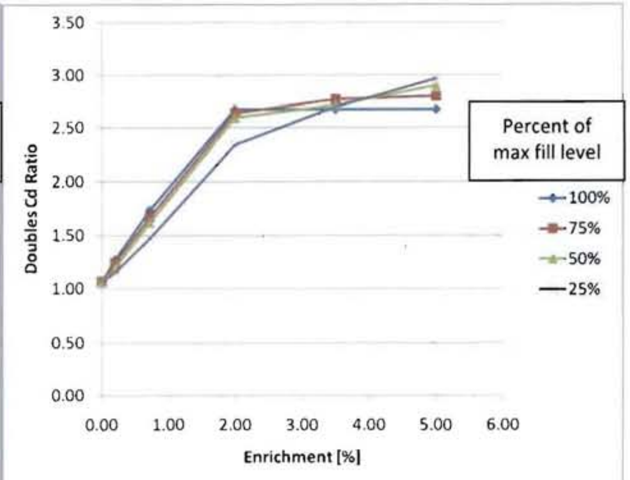

Fig. 5. Doubles Cd ratio as a function of ^{235}U enrichment and fill level showing the enrichment measurement is almost independent from the fill mass for the 48Y type cylinders.

SUMMARY

The PNEM method has been evaluated for the verification of the enrichment and mass of 30B and 48Y cylinders containing UF_6. The method requires a high efficiency neutron detector located adjacent to the UF_6 cylinder. The detector can be located under the cylinder or along the lower sides of the cylinder. We have evaluated the measured response for a ^{3}He-based detector configuration, but other high efficiency neutron detectors could also be used.

The PNEM method provides the following benefits:

1. The measured ^{235}U enrichment is independent of the uranium prior history in reactor use, as well as the UF_6 purity and age.
2. The measured ^{235}U enrichment is independent of the UF_6 cylinder wall deposits (heels), and the ^{235}U mass is independent of the operator's cylinder tare weight and weighing system. .
3. The method can be applied in the unattended mode with continuous operation similar to other IAEA unattended measurement systems.

The measured ^{235}U mass determination still requires knowledge of the ^{234}U/^{235}U ratio as a function of enrichment, but the enrichment measurement is independent of this ratio. The prior work with 5A cylinders shows that the method can be applied to higher enrichment uranium in smaller diameter cylinders [2].

A prototype PNEM detector is in the design and fabrication stage to demonstrate the technique. In addition, high efficiency neutron slab detectors at the IAEA will be evaluated for the PNEM measurement technique.

ACKNOWLEDGEMENTS

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