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# The Uranium Cylinder Assay System for Enrichment Plant Safeguards

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## Abstract

Safeguarding sensitive fuel cycle technology such as uranium enrichment is a critical component in preventing the spread of nuclear weapons. A useful tool for the nuclear materials accountancy of such a plant would be an instrument that measured the uranium content of $UF_6$ cylinders. The Uranium Cylinder Assay System (UCAS) was designed for Japan Nuclear Fuel Limited (JNFL) for use in the Rokkasho Enrichment Plant in Japan for this purpose. It uses total neutron counting to determine uranium mass in $UF_6$ cylinders given a known enrichment. This paper describes the design of UCAS, which includes features to allow for unattended operation. It can be used on 30B and 48Y cylinders to measure depleted, natural, and enriched uranium. It can also be used to assess the amount of uranium in decommissioned equipment and waste containers. Experimental measurements have been carried out in the laboratory and these are in good agreement with the Monte Carlo modeling results.

## Introduction

Safeguarding sensitive fuel cycle technology such as uranium enrichment is a critical component in preventing the spread of nuclear weapons. Independently-verifiable material accountancy is a fundamental measure in detecting diversion of nuclear material. To achieve material balance over an entire enrichment plant, it is essential to know the mass and enrichment of $UF_6$ inside the facility as well as transferred in and out of the facility. The bulk of $UF_6$ moving in and out of the plant is contained in 30B and 48Y cylinders. In addition, there may be uranium by-products in waste containers and decommissioned equipment (i.e., cold traps, centrifuges, etc.).

Traditionally, the uranium mass contained in $UF_6$ cylinders has been determined using load cells or electronic scales. They are often used by both the operator and the inspector, making authentication difficult. These systems also require reference weights for calibration and a valid tare weight for each cylinder. A study of load cell performance conducted by URENCO UK Ltd. showed them to have reliability issues and raised concerns over the cost of realizing an authenticated system.[1]

We have developed the Uranium Cylinder Assay System (UCAS), which uses passive neutron detection to determine uranium mass in $UF_6$ cylinders as well as waste containers and decommissioned equipment. UCAS was designed for Japan Nuclear Fuel Limited (JNFL) for use in Rokkasho Enrichment Plant. Two units were fabricated: (1) a fixed-geometry system for assaying 30B and 48Y cylinders on the facility's transfer trolley and (2) a mobile unit for assaying waste containers and decommissioned equipment. A mass measurement system based on passive neutron detection is an alternative that can be used in lieu of traditional load cells and electronic scales or as a redundant system to provide additional safeguards assurance.

UCAS uses total neutron counting to determine uranium mass assuming the enrichment is known. The primary source of neutrons in enriched $UF_6$ comes from the alpha bombardment of fluorine, where $^{234}U$ is the dominant alpha emitter. Because the enrichment of $^{234}U$ follows that of $^{235}U$, total neutrons give an indirect measure of the enrichment.[2] The high penetrability of neutrons through $UF_6$ means that total neutrons also track the uranium mass. Thus, with a known enrichment, the uranium mass can be determined from the total neutron count rate. Conversely, total neutron counting can be used to determine enrichment given the uranium mass, but the $^{234}U/^{235}U$ ratio must also be known.[3,4]

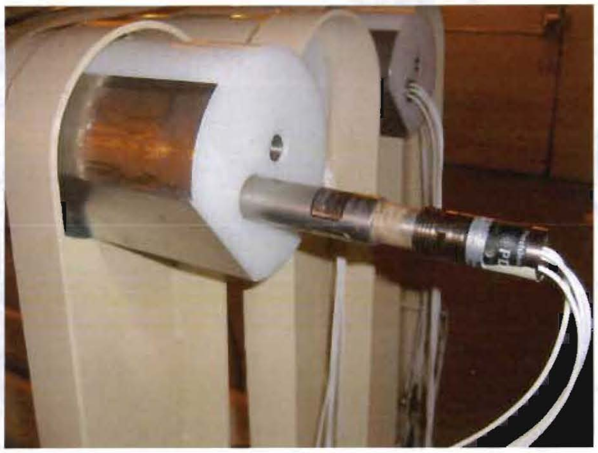
The following sections describe the design and characterization of UCAS. All of the physics calculations were performed using Monte Carlo N-Particle Extended (MCNPX), and the characterization measurements were performed at Los Alamos National Laboratory (LANL).

## Mechanical & Electrical Components

Each UCAS unit consists of four identical detector pods containing two $^3He$ tubes per pod (one upper and one lower tube). The tubes are embedded in a cylindrical block of polyethylene, which is partially wrapped in cadmium. The detector pods are enclosed in aluminum cases. A photo of one of the mobile pods is shown in Fig. 1. The fixed-geometry pods are similar except that they are not bolted to a wheeled cart. Instead, a sliding mechanism will move the pods into the measurement position on the trolley. Also, the cable connections for the fixed-geometry pods are inside the cover so that they can be used in unattended mode. The weight of each pod is 73 kg not including the cart.

Fig. 1. Photo of a mobile UCAS pod.

With two $^3He$ tubes per pod, there are a total of eight tubes per system. Each tube has 4 atm of $^3He$, an active length of 121.9 cm (48 in.), and a diameter of 2.54 cm (1 in.). The detector/moderator configuration is shown in Fig. 2. Fig. 2(a) shows how the detector is assembled. The cylindrical polyethylene moderator fits inside the aluminum case. The partial cadmium wrapping on the outside of the polyethylene can also be seen in the photo. The $^3He$ tube slides into the polyethylene moderator.



(a)

(b)

Fig. 2. Detector/moderator configuration shown in (a) exploded view and (b) assembled view.

Fig. 2(b) shows the assembled detector/moderator configuration. The cylindrical moderator has a notch taken out of the round surface in the direction of the sample. The position of the tube inside the moderator is optimized for maximum count rate. Fig. 3 shows the count rate as a function of the depth of the tube within the polyethylene. The count rate is maximized when there is 1.9 cm (0.75 in.) of polyethylene between the outer radius of the $^3$He tube and the surface of the moderator. The extra polyethylene on the back side of the tube also provides shielding.

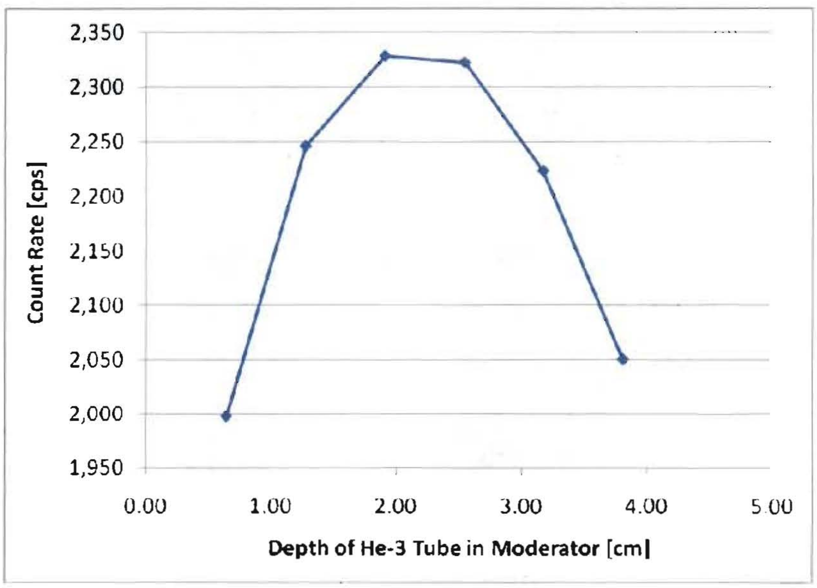



Fig. 3. Count rate as a function of $^3$He tube depth in the polyethylene moderator.

The detectors are accessed through the blue side cover (see Fig. 1). The fixed-geometry system may be used in unattended mode, so the cable connections are inside the security cover, which can be fitted with a tamper-indicating device. The hole where the cables run out of the cover was designed to be used with the standard tamper-indicating conduit used by the IAEA. Because the mobile system will only be used in attended mode, the cable connections are on the outside of the frame for easy access. The cover also has two viewing windows that allow the operator to see the LEDs on the Precision Data Technology (PDT) preamplifiers to ensure that they are registering counts.

UCAS was designed for singles mode only, but standard shift register coincidence settings are used: 4.5 μsec pre-delay, 64 μsec gate width, and a high voltage of 1680 V. The signals from each of the eight detectors are wired separately back to an OR box and then to a JSR-15 shift register. The coincidence data can be useful for state-of-health and authentication purposes. Each system comes with a data analysis computer that uses IAEA Neutron Coincidence Counting (INCC) software (version 5.1.2 or later).

## Detector Characterization

The physics calculations used to optimize the design and characterize sensitivities for UCAS were performed using MCNPX. The neutron energy spectra and source strengths for various $UF_6$ enrichments were calculated using another code called SOURCES. The uranium isotopics and $UF_6$ source strengths calculated with SOURCES are given in Tables 1 and 2.

Table 1. Uranium Isotopics.

| Enrichment | U-234 [atom %] | U-235 [atom %] | U-238 [atom %] |
|---|---|---|---|
| Depleted uranium | 0.001 | 0.20 | 99.8 |
| Natural uranium | 0.0055 | 0.72 | 99.2745 |
| 2.00% enriched | 0.01901 | 2.00 | 97.98099 |
| 3.50% enriched | 0.0288 | 3.50 | 96.471 |
| 5.00% enriched | 0.049827 | 5.00 | 94.950173 |

Table 2. $UF_6$ Source Strengths Calculated with SOURCES.

| Enrichment | (α,n) Source Strength [n/sec–cm$^3$] | Spontaneous Fission Source Strength [n/sec–cm$^3$] | Total Source Strength [n/sec–cm$^3$] |
|---|---|---|---|
| Depleted uranium | 0.0574 | 0.0430 | 0.1004 |
| Natural uranium | 0.1447 | 0.0428 | 0.1875 |
| 2.00% enriched | 0.4061 | 0.0423 | 0.4484 |
| 3.50% enriched | 0.5974 | 0.0416 | 0.6390 |
| 5.00% enriched | 1.0030 | 0.0410 | 1.0440 |

<u>30B and 48Y Cylinders</u>

The fixed-geometry unit will be used to assay 30B and 48Y cylinders containing feed, product, and tails. The calculated efficiency of the unit for a maximum-filled 30B cylinder containing 3.5% enriched $UF_6$

is 1.2%. Figs. 4 and 5 show the count rates in 30B and 48Y cylinders as a function of uranium mass. In both plots, the count rate increases with increasing uranium mass.

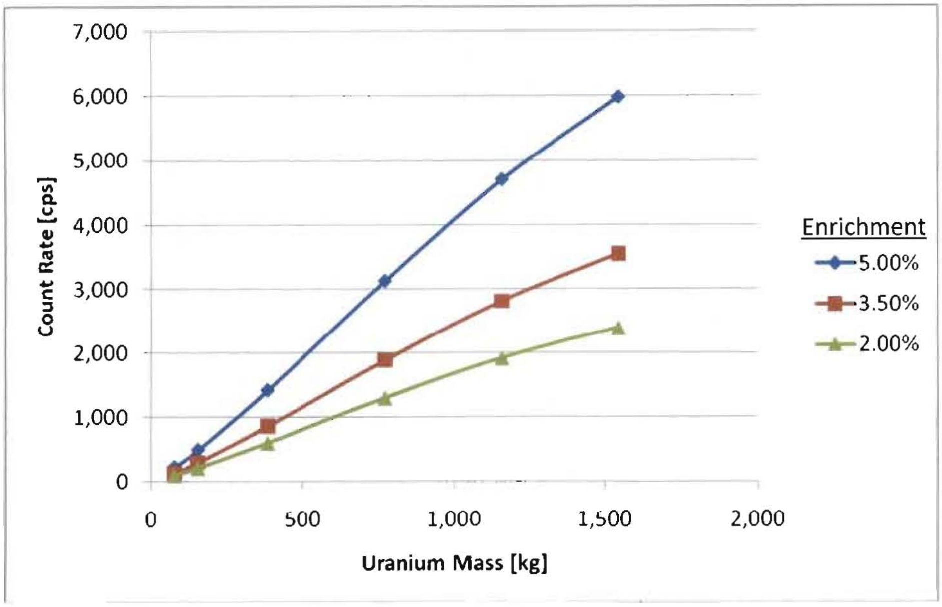



Fig. 4. UCAS response for a 30B cylinder containing low-enriched $UF_6$.

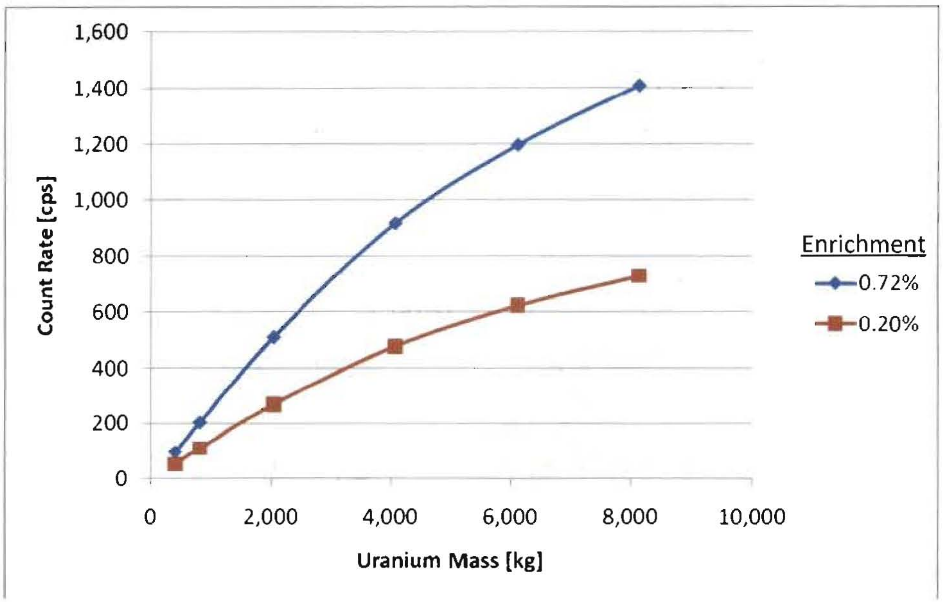



Fig. 5. UCAS response for a 48Y cylinder containing natural and depleted $UF_6$.

The biggest source of uncertainty for the fixed-geometry unit is the distribution of $UF_6$ within the cylinder. Counting statistics should be less than 1%, background is low in the transfer hall where the cylinders will be measured, and uncertainties in the calibration and $^{234}U/^{235}U$ ratio should also be relatively low compared to the source distribution term.

The distribution of $UF_6$ within a cylinder depends on how it was filled, the last operation made on it (e.g., sampling in liquid phase after homogenization), and the storage conditions.[5] When a cylinder is filled in liquid phase, most of the material remains in the lower part of the cylinder with a thin deposit on the upper part of the wall. When it is filled by desublimation, the solid $UF_6$ adheres evenly to the cylinder wall, creating an annular ring. Over time, some of the $UF_6$ on the upper part of the cylinder will slough off and fall to the bottom. We conducted a modeling study to assess the magnitude of the effect that source distribution has on the count rate. Based on several perturbations of $UF_6$ distribution within a 30B cylinder, we determined that the source distribution introduces an uncertainty of approximately 5% to the measurement.

In addition, we calculated the minimum detectable mass (MDM) of $UF_6$ in 30B and 48Y cylinders. These values are given in Table 3 in terms of uranium and equivalent $UF_6$ mass. The MDM is calculated as 3σ above background. The background rate in the transfer hall was estimated to be 0.32 cps based on previous measurements by another instrument. The background rate was scaled based on the volume of $^3$He in each instrument and shielding effects. The cylinder count time was assumed to be 15 minutes.

Table 3. Minimum Detectable Mass for 30B and 48Y Cylinders.

| Container | Enrichment [% U-235] | MDM [g U] | MDM [g $UF_6$] |
|---|---|---|---|
| 30B cylinder | 5.00 | 18 | 27 |
| 30B cylinder | 3.50 | 30 | 44 |
| 30B cylinder | 2.00 | 42 | 62 |
| 48Y cylinder | 0.72 | 251 | 371 |
| 48Y cylinder | 0.20 | 476 | 704 |

Waste Containers and Cold Traps

The mobile UCAS unit was designed to assay waste containers (i.e., crates and 208-liter drums) and equipment removed from the enrichment plant (i.e., cold traps). These items introduce additional measurement challenges. For waste measurements, there are uncertainty terms associated with the composition and density of the waste matrix and the uranium enrichment. For cold trap measurements, there is uncertainty in the chemical form of the uranium.

When $UF_6$ comes into contact with the moisture in the air, it forms $UO_2F_2$. For waste measurements, we assume that all of the source material is $UO_2F_2$. The waste containers at Rokkasho Enrichment Plant hold three categories of waste: reduced flammable, reduced resistance to flammable, and compacted incombustible. Because the waste matrix is not well characterized, each type of waste was modeled as a different mixture of iron and polyethylene at a fraction of full density. We also assume the $UO_2F_2$ is evenly distributed throughout the matrix. The uranium enrichment introduces another uncertainty term in waste measurements. Waste containers can contain varying enrichments, so the value is estimated from operational data.

Cold traps removed from the plant contain very little source material, but it is important to quantify that amount. The lack of source material means that counting statistics may become a significant factor in the total measurement uncertainty. The chemical form of the source material also plays into the uncertainty terms. Because the equipment has been removed from operation, it may contain a mixture of $UF_6$ and $UO_2F_2$. The INCC software allows the operator to choose the most likely combination (all $UF_6$, half $UF_6$ and half $UO_2F_2$, or all $UO_2F_2$) based on the history of the cold trap.

The additional unknowns in the waste and cold trap measurements combined with smaller uranium amounts mean that there is more uncertainty in these measurements compared to the cylinder measurements; however, MCNPX simulations give us a good understanding of how sensitive UCAS is to the uncertain parameters. Many of the uncertainties will also be better understood after the on-site calibration.

Characterization Measurements

UCAS was characterized by creating response profiles along the axial, radial, and vertical axes of the pods. This was done experimentally using a $^{252}Cf$ source, which was moved along each axis in 10 cm increments. The measurements were then compared to MCNPX simulations of the same test. The results are shown in Figs. 6, 7, and 8. Each of the plots shows measurements from a single pod. For the radial profile case (Fig. 7), the zero-point in the MCNPX simulation was 6 cm from the geometric centerline, which is why the measurement locations are shifted. As seen in the plots, there is excellent agreement between the Monte Carlo simulations and the experimental data in all three cases. These validation measurements give credibility to the MCNPX-based characterization of UCAS.

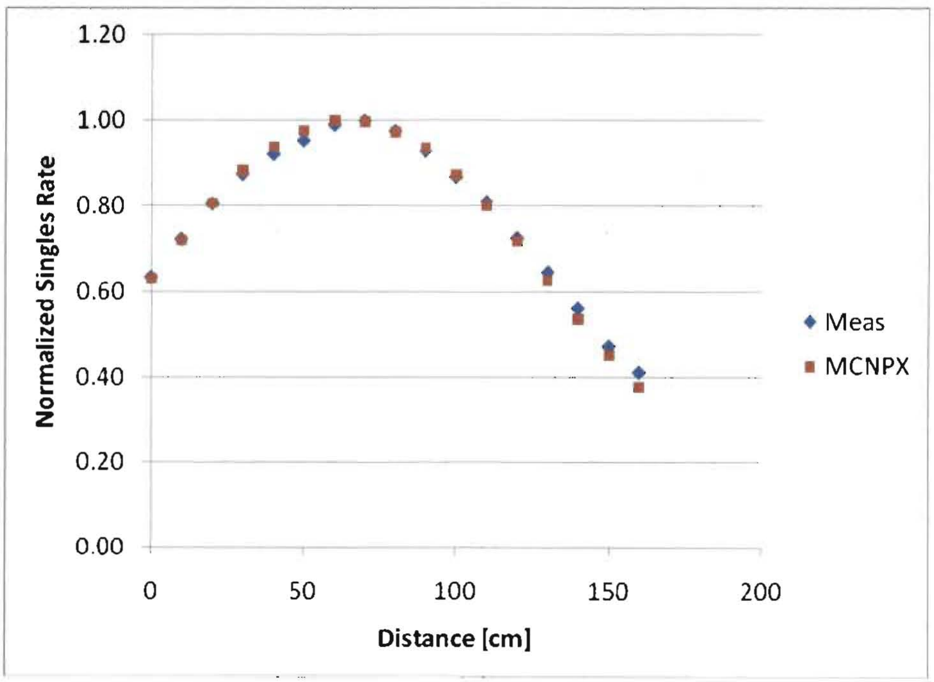



Fig. 6. UCAS axial response profile—measured vs. MCNPX simulation.

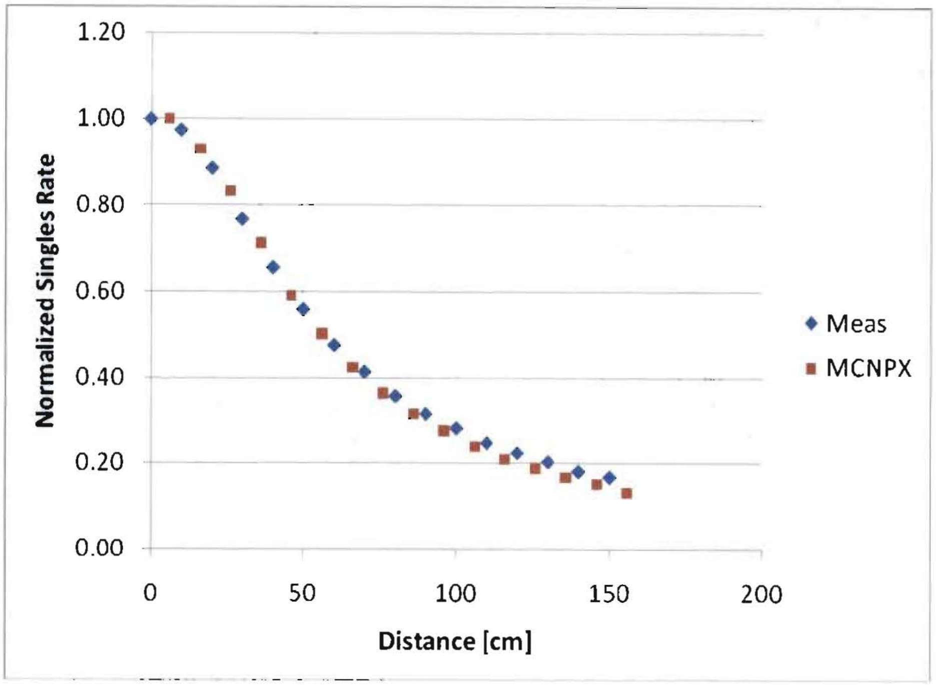



Fig. 7. UCAS radial response profile—measured vs. MCNPX simulation.

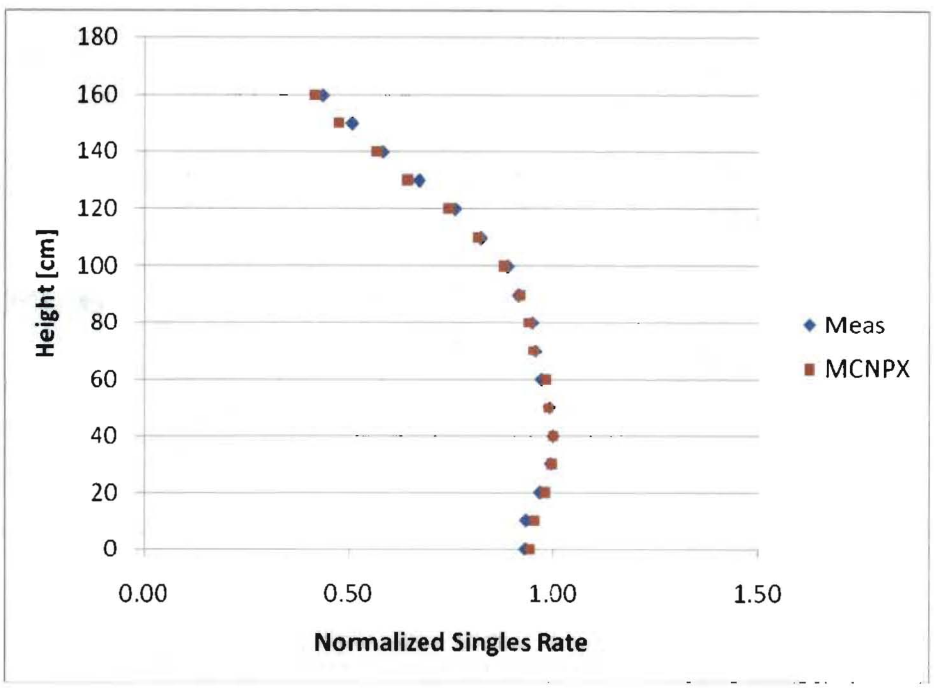



Fig. 8. UCAS vertical response profile—measured vs. MCNPX simulation.

## Conclusions

In this paper, we have introduced a new instrument to determine uranium mass in $UF_6$ containers assuming a known enrichment. UCAS was designed specifically for use at Rokkasho Enrichment Plant to assay 30B and 48Y cylinders, waste containers, and equipment removed from the plant. Two units were fabricated: (1) a fixed-geometry system for product, feed, and tails cylinder measurements on the facility's transfer trolley and (2) a mobile unit for measuring other items. UCAS uses total neutron counting to determine uranium mass. The detection principle is based on the correlation between $^{234}U$ and $^{235}U$ in enriched uranium.

The detector design was optimized using MCNPX simulations. Each system consists of four pods, with two $^{3}He$ tubes per pod. The tubes are embedded in a polyethylene moderator that is partially covered with cadmium. Each unit uses a JSR-15 shift register and INCC software.

Modeling studies showed that the largest source of uncertainty in the cylinder measurements is the distribution of $UF_6$ within the cylinder. For waste measurements, the dominant uncertainty is the waste matrix, and for cold traps, it is the chemical form of the uranium. MCNPX simulations provide us with a good understanding of how these uncertainties affect the count rate. Characterization measurements made with $^{252}Cf$ matched MCNPX simulations very well and give credibility to the Monte Carlo modeling studies.

Future work for UCAS includes installation and calibration at Rokkasho Enrichment Plant in May 2010. Because of the relatively high penetrability of neutrons through $UF_6$ cylinders and potential for unattended mode operation, neutron methods may also offer an alternative (or complement) to gamma-ray methods for enrichment determination. Menlove, Swinhoe, and Miller are currently studying a technique that uses total and correlated neutrons as well as the cadmium ratio to simultaneously determine uranium enrichment and mass in $UF_6$ containers.[6] These types of advances in nondestructive assay techniques represent important steps in independently-verifiable nuclear material accountancy for enrichment plant safeguards.

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