VALIDATION OF GAMMA-RAY DETECTION TECHNIQUES FOR SAFEGUARDS MONITORING AT NATURAL URANIUM CONVERSION FACILITIES

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Abstract

Recent IAEA circulars and policy papers have sought to implement safeguards when any purified aqueous uranium solution or uranium oxides suitable for isotopic enrichment or fuel fabrication exists. Under the revised policy, IAEA Policy Paper 18, the starting point for nuclear material under safeguards was reinterpreted, suggesting that purified uranium compounds should be subject to safeguards procedures no later than the first point in the conversion process. In response to this technical need, a combination of simulation models and experimental measurements were employed to develop and validate concepts of nondestructive assay monitoring systems in a natural uranium conversion plant (NUCP). In particular, uranyl nitrate ($\text{UO}_2(\text{NO}_3)_2$) solution exiting solvent extraction was identified as a key measurement point (KMP), where gamma-ray spectroscopy was selected as the process monitoring tool. The Uranyl Nitrate Calibration Loop Equipment (UNCLE) facility at Oak Ridge National Laboratory was employed to simulate the full-scale operating conditions of a purified uranium-bearing aqueous stream exiting the solvent extraction process in an NUCP. Nondestructive assay techniques using gamma-ray spectroscopy were evaluated to determine their viability as a technical means for drawing safeguards conclusions at NUCPs, and if the IAEA detection requirements of 1 significant quantity (SQ) can be met in a timely way. This work investigated gamma-ray signatures of uranyl nitrate circulating in the UNCLE facility and evaluated various gamma-ray detector sensitivities to uranyl nitrate. These detector validation activities include assessing detector responses to the uranyl nitrate gamma-ray signatures for spectrometers based on sodium iodide, lanthanum bromide, and high-purity germanium detectors. The results of measurements under static and dynamic operating conditions at concentrations ranging from 10-90g U/L of natural uranyl nitrate are presented. A range of gamma-ray lines is examined, including attenuation for transmission measurement of density and concentration. It was determined that transmission-corrected gamma-ray spectra provide a reliable way to monitor the ^{235}U concentration of uranyl nitrate solution in transfer pipes in NUCPs. Furthermore, existing predictive and analysis methods are adequate to design and realize practical designs. The ^{137}Cs transmission source employed in this work is viable but not optimal for ^{235}U densitometry determination. Validated simulations assessed the viability of ^{133}Ba and ^{57}Co as alternative densitometry sources. All three gamma-ray detectors are viable for monitoring natural uranium feed; although high-purity germanium is easiest to interpret, it is, however, the least attractive as an installation instrument. Overall, for monitoring throughput in a facility such as UNCLE, emulating the uranium concentration and pump speeds of the Springfields conversion facility in the United Kingdom, an uncertainty of less than 0.17% is required in order to detect the diversion of 1 SQ of uranyl nitrate through changes in uranium concentration over an accountancy period of one year with a detection probability of 50%. Although calibrated gamma-ray detection systems are capable of determining the concentration of uranium content in NUCPs, it is only in combination with verifiable operator declarations and supporting data, such as flow rate and enrichment, that safeguards conclusions can be drawn.

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1. INTRODUCTION

Conversion is the process by which natural uranium ore concentrate is purified and converted through a series of chemical processes into uranium hexafluoride gas (UF_6). Conversion has historically been excluded from Comprehensive Safeguards agreements for the ^{235}U -based nuclear fuel cycle. The aim of nuclear safeguards is to deter diversion of nuclear material from peaceful uses by maximizing the chance of early detection. Given the changing global political environment and the increased availability of dual-use nuclear technology, the International Atomic Energy Agency (IAEA) has evolved its policies to emphasize safeguarding this potential feedstock material in response to dynamic and evolving potential diversion pathways. Under INFCIRC 153 Para. 34(C) [2], safeguards commences when materials are suitable for isotopic enrichment or fuel fabrication, which traditionally occurs at the end of conversion with the production of UF_6 cylinders. Under the revised policy, IAEA Policy Paper 18 (PP18), “Safeguards Measures Applicable in Conversion Plants Processing Natural Uranium,” the starting point for nuclear material under safeguards was reinterpreted and a new definition of source material in this category was introduced [3]. Under IAEA PP18, the IAEA suggests that these purified uranium compounds should be subject to safeguards procedures no later than at the first point in the conversion process [4] [5] [6] [7] [8] [9]. Uranium conversion facilities are classified as small, medium, or large with corresponding throughputs of 10, 100, or 1000 metric tonnes of uranium (MTU) per annum. According to the IAEA, a significant quantity (SQ) is “the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded. Significant quantities take into account unavoidable losses due to conversion and manufacturing processes and should not be confused with critical masses” [10]. Due to the high uranium throughput of conversion facilities, meticulous monitoring will be required along the entire conversion process. In the scope of natural uranium conversion plants (NUCPs), 1 SQ translates to 10 metric tons of natural uranium over a period of 1 year with a detection probability of 50% [5].

The IAEA’s criteria for diversion detection of 1 SQ at an NUCP corresponds to a diversion of 10%, 1%, and 0.1% production from small, medium, and large production plants, respectively [5] [6] [7] [8] [9]. Although it may be easier to detect a 10% diversion with a smaller production facility, the nature of the diversion threat becomes increasingly more complex with increasing facility size, and the burden for accurate detection becomes increasingly challenging. Although process monitoring (PM) at a single key measurement point (KMP) will provide essential safeguards data for a small NUCP, the complex structure of large NUCP facilities will require more extensive in-line monitoring points in order to differentiate diversion activities from inventory differences and material unaccounted for (MUF), attributable to measurement bias.

To meet the demand for instrumentation testing at conversion facilities, Oak Ridge National Laboratory (ORNL) developed the Uranyl Nitrate Calibration Loop Equipment (UNCLE) facility to simulate the full-scale operating conditions of a purified uranium-bearing aqueous stream exiting the solvent extraction process in a natural uranium conversion plant [11]. A variety of intermediary uranium compounds are connected with the conversion process. As outlined by Faulkner *et al.* [7] and reinforced in PP18, first point at which uranium becomes

purified follows solvent extraction. UNCLE was designed to reproduce the conditions of the purified aqueous uranyl nitrate stream exiting the solvent extraction process in an NUCP [12]. The scope of this project is focused on this specific phase of the conversion process containing purified aqueous uranyl nitrate.

Pickrell *et al.* and Ladd-Lively *et al.* have benchmarked a Coriolis flow rate meter in addition to testing the 20-array ^3He proportional counter clamp neutron detector developed at Los Alamos National Laboratory (LANL) (now installed at the ORNL UNCLE facility), originally tested at the Springfields NUCP in the United Kingdom [13] [12]. Equipment calibration of the ^3He detector was later completed by Ladd-Lively at the UNCLE facility [11] [14] [15] [16] [17]. Operations monitoring at the UNCLE facility have been preliminarily analyzed by Lee, employing the second-generation Transverse Uranium Neutron Detector (TUrND) developed by LANL, based on the first-generation design by Miller and Pickrell in 2004 [13] [18] [19]. Neutron monitoring efforts have not met performance specifications, nor have they been analyzed for drawing conclusive safeguards conclusions with respect to the 1 SQ requirement by the IAEA [20] [21] [22] [23] [24] [25]. The challenge for passive neutron systems is the weak source strength, in addition to the poor signal-to-noise to ambient and cosmic ray neutrons. This work evaluates whether a technical design basis using gamma-ray spectroscopy can be used for safeguarding nuclear material at the first identified KMP in an NUCP and if the IAEA detection requirements of the detection of 1 SQ can be met.

In order to determine the ^{235}U mass flow over a detection time period, the mass flow of solution, the average concentration of uranium in the solution, and the ^{235}U weight fraction must be known and measured. The flow rate of solution is taken from a calibrated Coriolis flow rate meter. Feed material is known to be naturally enriched and so the uranium concentration can be estimated from a measurement of ^{235}U , which after correction for self-attenuation in the solution is directly proportional to the intensity of the direct 185.7 gamma ray emission. The self-attenuation correction accounts for the possible changing solution concentration over time and is based on a transmission (gamma ray densitometer) technique. In the following section, these concepts will be developed quantitatively in the context of the experimental mock-up facility and set-ups used. This work serves to validate the modeling and simulation tools which might be used to develop an optimize instrument design taking into account constraints which are not yet fully known (being subject to negotiation between all parties). In practice calibration will play a central role in ensuring the target accuracy is met and the test facilities described are representative of what will be needed.

2. THEORY

Characterizing mass flow rate in an NUCP provides auxiliary verification data to aid in determining material throughput. Flow rate data alone provides limited information regarding uranium concentration, especially in cases where material substitution, instrumentation tampering, and falsification of records are potential means of misuse/undeclared activities. As part of a comprehensive nondestructive assay (NDA) safeguards system that accurately provides concentration data, flow rate measurements help correlate mass throughput at an NUCP, as described in Equation 1 [20].

$$M(t) = C(t) \times F(t), \quad [1]$$

where

$M(t)$ = mass flow rate of uranyl nitrate as a function of time, t ,

$C(t)$ = uranium concentration (g U/L), and

$F(t)$ = flow rate (L/hr).

2.1. Algorithm Development for Peak Area Interpretation

To implement Eq. 1 by radiometric techniques requires the nature of the uranyl nitrate solution to be verified. Use of a densitometry source provides a transmission measurement that determines the density of the uranyl nitrate in an NUCP transfer pipe. The transmission, T , is calculated by taking a transmission ratio of the uranyl nitrate-filled pipe (I) to an empty pipe (I_o). From any the candidate sources tested (^{133}Ba , ^{57}Co , ^{137}Cs), the T ratio (I/I_o). is related to the density (ρ) of the solution flowing in the pipe (effective diameter d_{eff}) via the following relation [26]:

$$\rho = -\ln\left(\frac{1}{T}\right) \cdot (\mu_p \cdot d_{eff})^{-1}. \quad [2]$$

where

ρ = uranyl nitrate solution density (g/cm^3),

T = transmission ratio (I/I_o),

μ_p = mass attenuation coefficient (cm^2/g), and

d_{eff} = inner diameter (cm).

In Eq. 2, d_{eff} is treated as a model parameter and accounts for the finite field of view; the material composition (weight fractions of the constituent elements) of uranyl nitrate as a function of density is known and hence the linear attenuation coefficient can be calculated assuming additive Bragg behavior. In the first pass, a nominal operating density (composition) is assumed (between water and the saturation concentration), a refined estimate is made and the calculation repeated. Mathematically, the problem is well behaved and with precise transmission data only a few passes are needed to achieve self-consistency (convergence). Using this relation, the density using dilution measurements provided a calibration for uranyl nitrate solution measurements.

Although densitometry measurements may provide the solution density and potentially uranium concentration through calibration, this still does not tell us about the throughput of ^{235}U , specifically. Consequently, a combination of the transmission data and the 185.7 keV peaks from ^{235}U are required to determine not only the uranium concentration in the NUCP transfer pipe but also the ^{235}U content. The concentration (C) of ^{235}U in the circulating uranyl nitrate is determined by the net peak area count rate of the ^{235}U 185.7 keV signature emission given in Equation 3 [26]:

$$C = \frac{r \cdot CF(AT)}{V \cdot \epsilon \cdot \rho}, \quad [3]$$

where

C = ^{235}U activity concentration of uranyl nitrate solution (Bq/g),

r = net count rate of 185.7 keV emission peak (cps),
 $CF(AT)$ = self-attenuation correction factor,
 V = volume of solution in detector field of view (cm^3),
 ϵ_{peak} = peak efficiency of 185.7 keV (cps/Bq), and
 ρ = uranyl nitrate solution density (g/cm^3).

2.2. Properties of Uranyl Nitrate

A variety of intermediary uranium compounds are connected with the conversion process. UNCLE was designed to reproduce the conditions of the purified aqueous uranyl nitrate stream exiting the solvent extraction process in an NUCP. Dilution measurements were taken for each of the selected gamma-ray detectors with a uranyl nitrate-filled pipe segment. This pipe segment was created from the same Schedule 40 304L pipe used in UNCLE, which was chosen to mimic the Springfields NUCP in the UK. The source uranyl nitrate at 90g U/L was obtained from UNCLE for the dilution experiments. The uranyl nitrate solution circulating through the UNCLE facility contains 90 g of natural uranium dissolved per liter of water, with a measured solution density of $1.122 \text{ g}/\text{cm}^3$ at 20°C . The elemental solution composition was stoichiometrically calculated for natural uranium-bearing uranyl nitrate with a density and concentration of UNCLE. At 90 g U/L, the pipe segment contains 2.62 L of solution with a mass of 2939.64 g, of which ~241 g is natural uranium. The properties of the dilution solutions studied in this work are summarized in Table 1.

Table 1. Material Composition of Uranyl Nitrate Solution in Pipe Segment (2.62 L).

Solution Concentration (g U/L)	90	85	75	50	10
Molarity (M)	0.378	0.357	0.315	0.210	0.042
Density (g/cm^3)	1.122	1.115	1.099	1.064	1.008
Total Uranyl Nitrate Solution Mass in Pipe (g)	2939.64	2921.30	2879.38	2787.68	2640.96
Wt.% U	8.02%	7.62%	6.82%	4.70%	0.99%
Wt.% N	0.71%	0.67%	0.60%	0.41%	0.09%
Wt.% H	9.75%	9.82%	9.95%	10.32%	10.94%
Wt.% O	81.52%	81.88%	82.62%	84.57%	87.98%
Wt.% $\text{UO}_2(\text{NO}_3)^{1.5*}$	12.23%	11.63%	10.41%	7.17%	1.51%
Wt.% H_2O	87.77%	88.37%	89.59%	92.83%	98.49%

* Preparation using acid-deficient UN was conducted because the lower acid concentration led to a quicker precipitation reaction, resulting in a stock solution with a NO_3/U mole ratio of 1.53.

In addition, the uranyl nitrate in UNCLE is ~40 years old and in secular equilibrium. The uranyl nitrate from commercial conversion facilities cannot be assumed to be in equilibrium. As a result, the scope of this project assumes that secular equilibrium cannot be ensured, hence focusing on the direct ^{235}U -based assay signatures, though the other lines are available to measure experimentally in this special situation to add complementary information. The difference between recently purified and aged uranyl nitrate for ^{235}U activity is negligible ($< 4 \times 10^{-6} \%$), given the long half-life of ^{235}U ; thus, we can assume the activity at 44 years (as in UNCLE) for ^{235}U approximates the response from recently purified ^{235}U . All gamma-ray lines $>1\%$ intensity (from which 185.7 keV emission was predominant with 57.2% intensity) were provided in the source definition of the Monte Carlo model (discussed in *Section 3.3*). The activities of ^{235}U at dilution concentrations are summarized in Table 2.

Table 2. ^{235}U Activity for Fresh Uranyl Nitrate at Dilution Concentrations.

Dilution Concentration (g U/L)	Calculated Activity Fresh ^{235}U (Bq)
90	1.59×10^5
85	1.51×10^5
75	1.33×10^5
50	8.84×10^4
10	1.76×10^4

The thickness of the pipe employed in the dilution experiments and in UNCLE are from the same 304L pipe inventory, with measured dimensions summarized in Table 3. The thickness of the 304L pipe in UNCLE was measured to be (0.52 ± 0.05) cm [25]. The pipe specifications for UNCLE were modeled on the conversion facility at Springfields, UK, providing a representative design of NUCP facilities. The linear attenuation coefficient at this energy through 304L pipe was calculated using *XCOM: Photon Cross Sections Database* to be 1.14 cm^{-1} , which results in a 55.4% transmission through 0.52 cm of stainless steel at 185.7 keV [27].

Table 3. UNCLE Pipe Model Dimensions and Composition .

Component	Density (g/cm^3)	Dimensions <i>diameter</i> $\times$ <i>length</i> (cm)
Uranyl Nitrate Solution, 90g U/L	1.122	$7.8\text{Ø} \times 60.3$
Stainless Steel Pipe, 304L	7.9	$8.9\text{Ø} \times 63.7$

2.2.1. Properties of Transmission Sources for Densitometry Monitoring

External transmission sources penetrating the uranyl nitrate-filled pipe provide densitometry information regarding the uranium concentration and uranyl nitrate density of the monitored uranyl nitrate. A transmission-based in-line densitometer using the 662 keV gamma ray from

¹³⁷Cs was initially selected based on source availability for measurements. However, ⁵⁷Co and ¹³³Ba are also investigated through Monte Carlo simulations.

The 122 keV in ⁵⁷Co line is a suitable candidate for assaying ²³⁵U, as it does not interfere with 185.7 keV peak, does not contribute to the Compton continuum under lower energy ²³⁵U photons, and is much more sensitive than ¹³⁷Cs in characterizing attenuation properties of various concentrations of uranyl nitrate because of its higher μ value. Furthermore, the backscatter peak of ¹³⁷Cs also falls near the 185 keV energy; if both ²³⁵U and ¹³⁷Cs, interference from the backscatter peak could provide interference in the measurement of the 185.7 keV emission of the ²³⁵U. However, 122 keV also sits below the major uranium emissions and may be too sensitive to continuum subtraction, unless high-resolution instrumentation is employed (HPGe). In addition, the low energy line is easily attenuated and may require a very strong source to provide sufficient detection statistics. Similarly, the 356 keV photon as a transmission source from ¹³³Ba would have been a sufficient (though a less preferable substitute to ⁵⁷Co), but the available source was too weak for laboratory measurements. ¹³³Ba has three high-intensity photons in the 300–400 keV range, which would interfere in analyzing low-resolution densitometry measurements. The selection of ¹³⁷Cs is theoretically sufficient for densitometry purposes, as it possesses a strong monoenergetic photon emission at 661.7 keV, which will alleviate any transmission peak overlap discrimination with lower resolution measurements.

2.3. Self-Attenuation Correction Principles

The NDA of nuclear material often encounters large sample containers with high self-absorption properties. If the detector efficiency is accurately known as a function of source position and energy and if the geometry and source emission rates are also accurately known, calibration standards are not necessarily required to determine the attenuation correction factor due to self-absorption.

Nondestructive assay of UN flowing in NUCP piping presents a geometry configuration that is susceptible to high self-attenuation. In determining the correction factor for self-attenuation, $CF(AT)$, we are addressing what fraction of the signature photons emitted in the direction of the detector actually reaches the detector. Use of transmission calibration provides a reference for assay using transmission techniques to determine the self-attenuation for a sample, in this case, for UN-filled piping. This section will explore a simple 1-D demonstration of the effects of the relative $CF(AT)$ based on calibration relative to the reference 90 g U/L uranyl nitrate for the 185.7 keV gamma ray from ²³⁵U with a relative transmission of the 122 keV from ⁵⁷Co. A 3-D model of $CF(AT)$ of the uranyl nitrate pipe segment is presented as part of a detailed uncertainty and sensitivity analysis in an accompanying paper using the theory developed by Parker [28], experimental data, Monte Carlo simulations, and Canberra ISOCS® models [29].[‡]

[‡] The slab is given as a simple approximation in the scope of this paper to introduce self-attenuation, but is discussed in more detail in the accompanying paper (and described in [1] *ibid*. As shown in the original work from which this work is being

In defining the relative transmission factor (T_{rel}) from the densitometry source at 122 keV (^{57}Co) for a uranyl nitrate solution (*var*) with respect to the 90 g U/L solution (*ref*):

$$T_{rel.} = \frac{(e^{-\rho \cdot (\frac{\mu}{\rho}) \cdot d})_{var}}{(e^{-\rho \cdot (\frac{\mu}{\rho}) \cdot d})_{ref}} \quad [4]$$

where

ρ = uranyl nitrate solution density (g/cm^3),

μ = linear attenuation coefficient (cm^{-1}), and

d = inner diameter (cm).

var = variable uranyl nitrate solution (85, 75, 50, 10 g U/L)

ref = 90 g U/L uranyl nitrate solution

As given in Table 3, the diameter of uranyl nitrate in the pipe is 7.8 cm and the densities of 10-90 g U/L of uranyl nitrate are given in Table 1. Two key assumptions must be satisfied in the scope of this method in order to ensure that the photon escape-fraction can be computed on a macroscopic scale [28]: First, the gamma-ray source material (i.e., uranyl nitrate solution) is reasonably homogeneous in composition; second, the gamma-ray emitting constituents are small enough such that self-attenuation within the individual particles (i.e., uranium) is negligible.

For a simple approximation to determine the $CF(AT)$, the uranyl nitrate solution can be approximated as a slab:

$$CF(AT) = \frac{(\kappa \cdot \rho \cdot \frac{\mu}{\rho} \cdot d)}{1 - e^{-(\kappa \cdot \rho \cdot \frac{\mu}{\rho} \cdot d)}} \quad [5]$$

where

ρ, μ, d = Defined in Eq. 4

κ = geometrical calibration parameter

As defined by Parker, the slab approximation for is $\kappa \sim 0.8$ [28]. $CF(AT)$ for a cylindrical geometry (< 1) is less than that of a slab or box-shaped sample; also, for far-field approximation for reasonably transparent cylinders $\kappa \sim \pi/4$ [28] [30]. A relative $CF(AT)$ defined with respect to the reference 90 g U/L solution can be written as:

published, the slab is given as a simple approximation and is in good agreement with the cylinder. The scope of this paper is to introduce the concept of self-attenuation as a property of the measurement system.

$$CF(AT)_{rel.} = \frac{CF(AT)_{var}}{CF(AT)_{ref}} \quad [6]$$

where

var = variable uranyl nitrate solution (85, 75, 50, 10 g U/L)

ref = 90 g U/L uranyl nitrate solution

Using mass attenuation coefficient data employed from XCOM [27], the $CF(AT)_{rel.}$ for ^{235}U 185.7 keV gamma ray is plot as a function of the relative transmission factor $T_{rel.}$ for the ^{57}Co 122 keV gamma ray (Figure 1). The objective of using the ^{57}Co 122 keV transmission to was correlate with any self-absorption of the ^{235}U 185.7 keV gamma.[§] This is to provide a simple comparison of the $CF(AT)$ for each of the uranyl nitrate solutions relative to the 90g U/L. Consequently, this illustrates that the 122 keV densitometry source is very sensitive to the uranyl nitrate solution concentration, where the $CF(AT)$ for 185.7 keV gamma can be employed without varying very much over the expected range of operation.

[§] Although experimental data was taken for ^{137}Cs -137 662keV (due to limited source availability in the laboratory), this energy was relatively insensitive to varying concentrations of uranyl nitrate, as discussed in the Results.

Figure 1. Relative CF(AT) relation to relative transmission of 122 keV densitometry source at various densities of uranyl nitrate solution (10-90 g U/L concentration).

3. METHODS

3.1. Dilution Measurements Experimental Setup

Robustness and practical implementation are paramount for safeguards monitoring. A suite of detectors, primarily commercial-off-the-shelf (COTS), was selected for testing detection sensitivity to a range of uranium concentrations of uranyl nitrate. In addition, the variety in detector selection permits comparison of resolution and efficiency parameters in monitoring uranyl nitrate. This selection of detectors was discussed by Dewji *et al.* [24] [25] and included both scintillators – sodium iodide (NaI(Tl)) and lanthanum bromide (LaBr₃) – as well as semiconductors – high-purity/broad-energy germanium (HPGe/BEGe). Detector specifications are outlined in Table 4.

Table 4. Properties of Selected Gamma-Ray Detectors for Validation Experiments [31-35].

Instrument Name	Instrument Weight (kg)	Detector Material	Crystal Dimensions <i>diameter</i> × <i>length</i> (cm)
Canberra 2×2-inch NAIS + Osprey Digital Tube Base	1.28	NaI(Tl)	5.08Ø × 5.08
Canberra Inspector 1000-INPROL-1	2.4	LaBr ₃	3.81Ø × 3.81
Canberra Falcon BEGe	15.5	Ge	2.985 Ø × 2.0

Dilution measurements were taken with a uranyl nitrate-filled 304L pipe segment in a controlled, low-background laboratory facility. This is in order to determine each detector's assay signatures and sensitivity to a range of concentrations of uranium in uranyl nitrate. Both passive ^{235}U and transmission-induced ^{137}Cs densitometry measurements were conducted at UNCLE using the three detectors employed in the dilution experiments. Passive gamma-ray spectra were acquired for 3600 s live time measuring the ^{235}U high-intensity photons (notably 185.7 keV) for uranium concentrations ranging from 10 to 90 g U/L of uranyl nitrate to determine the sensitivity of each of the candidate detector's responses. In addition to passive gamma-ray measurements using HPGe, LaBr₃, and NaI(Tl) detectors, a transmission-based in-line densitometer using the 661.7 keV gamma ray from ^{137}Cs was selected based upon availability. The source activity was 30.7 kBq. The experimental setups with each of the three detectors listed in Table 4. (Falcon BEGe with the tungsten endcap [33] and Osprey 2×2 NaI(Tl) and Inspector 1000 LaBr₃ with the Canberra 7419E Shield/Collimator [36]) are depicted in Figure 2. The detectors were maintained at a fixed geometry in relation to the pipe during the course of all dilution measurements.

Figure 2. Setup for Static Dilution Measurements in Laboratory: (A) Falcon BEGe; (B) Inspector 1000 LaBr₃; and (C) Osprey 2×2 NaI(Tl).

3.2. UNCLE Measurements Experimental Setup

Where the laboratory dilution measurements provided a static, controlled, low-background environment, the series of measurements at UNCLE provided more realistic background interference from adjacent pipes and tanks, in addition to introducing the potential for voiding and temperature variations from fluid flow. Both ^{235}U passive and ^{137}Cs transmission densitometry measurements were conducted at UNCLE using the three detectors employed in the dilution experiments. Detector responses to changes in flow rate (1070 rpm and 500 rpm), shielding configurations (front shield and shadow shield), background determination, and transmission densitometry were assessed in the UNCLE field trials. Ladd-Lively *et al.* previously conducted measurements using the Coriolis flowrate meter installed at UNCLE [17], which benchmarked the mass flow rate of the uranium flowing through UNCLE. The pump speed of 1071 rpm approximates the average flow rate that was used during the field test at the Springfields NUCP [17]. Based on these data, the tests conducted in this work at 1070 rpm and 500 rpm correspond to mass flow rates of 7963.4 kg/h and 3528.9 kg/h, respectively. Photographs of the experimental setup are given in Figure 3 for each of the three detectors. The detectors were situated adjacent to one of the 100 gallon (378.5 L) stainless steel tanks in

UNCLE. Measurement spectra for the 185.7 keV ^{235}U photon were accumulated for 3600 s live time for all configurations. Detectors were maintained at a fixed geometry in relation to the pipe during the course of all flow rate measurements.

(A) (B) (C)
Figure 3. Detector Setup for Dynamic Measurements in UNCLE Operations Facility: (A) Falcon BEGe; (B) Osprey 2Ø x2" NaI(Tl); (C) Inspector 1000 LaBr₃.

3.3. Monte Carlo Validation of Dilution Experiment Setup

Experimental measurements undertaken in the scope of this project provide an overall assessment of the sensitivity of gamma-ray detection technology to variations in concentration, as well as to determine the monitoring capabilities in an operational conversion facility. However, a myriad of variables – both from solution properties and detector efficiency - were not able to be experimentally tested. Thus, simulations provide detector responses to test some variables such as variations in transmission densitometry. Computational models were constructed to simulate the detector responses for passive dilution and transmission measurements. All photon transport simulations were conducted using the MCNPX code [37].

3.3.1. Dilution Experiment Simulation

The modeled uranyl nitrate comprises a solution of 90 g of uranium dissolved per 1000 mL of water, 0.76 wt.% enrichment, with a measured solution density of 1.122 g/cm³. The elemental solution composition was calculated in the uranyl nitrate solution to be 8.0 wt.% U, 0.70% N, 81.5% O, and 9.8% H. As the detector providing the highest resolution signatures, the Falcon BEGe detector was modeled in detail to determine validated detector responses. The dimensions and physical characteristics of the Falcon were modeled to manufacturer's specifications [33] [34] [38]. A VisEd [37] rendering of the detector model is given in Figure 4. As shown, the

HPGe crystal is 13.7 mm from the endcap surface of the collimator, and the detector endcap is maintained at 0.635 cm (1/4-inch) distance from the pipe surface.**

The simulation was modeled after the experimental setup for the dilution measurements, employing lead brick shielding around the perimeter of the collimator, as well as behind the pipe acting as a backshield. The pipe characteristics are given in Table 1 and Table 3, with the geometry depicted in Figure 4. A pulse-height (F8) tally was performed over the active region of the detector crystal to determine the pulse-height spectrum for passive and transmission measurements.

Figure 4. VisEd Model of Falcon BEGe Detector Setup with Uranyl Nitrate-Filled 304L Pipe.

Since actual measurements were accumulated as 8192-channel pulse-height spectra, the MCNPX pulse height spectra were similarly binned and tallied into 8192 channels spanning a total energy range of 1.59 MeV. The Gaussian Energy Broadening (GEB) function [37] in MCNPX, given in Eq. 4, was used to simulate the detector resolution, based on the full-width-at-half-maximum (FWHM) experimental data.

$$FWHM = a + b\sqrt{E + cE^2} \quad [4]$$

where

** Resolution of NaI and LaBr₃ were not sufficient to isolate the 185.7 keV peak for the scope of this paper, in MCNP. However, the accompanying paper discusses the detection efficiency modeled for the NaI and LaBr₃ detectors using Canberra® ISOCS, which discusses detector efficiencies vis-à-vis NaI, LaBr₃, and HPGe (and fully described in [1] Dewji, Safeguards assessment of gamma-ray detection for process monitoring at natural uranium conversion facilities..

$E = \text{Energy (MeV)}$

$\text{FWHM} = \text{full-width-at-half-maximum value of photopeak (MeV)}$.

The fit parameters employed in Eq. 4 for the BEGe detector were:

$$a = (59.5 \pm 6.15) \times 10^{-5}$$

$$b = 0.000763 \pm 0.000147$$

$$c = 0.85487 \pm 0.4942.$$

Since ^{133}Ba and ^{57}Co were unavailable for experimental measurements, use of validated simulations enables assessing the sensitivity of these sources to various concentrations of uranium in uranyl nitrate.

3.3.2. Densitometry Transmission Simulations

3.3.2.1. ^{137}Cs Transmission Dilution Simulations

The uranyl nitrate-filled pipe, collimation, and shielding remained the same as modeled in the passive measurements. In the ^{137}Cs transmission simulations using the same model, a plastic-encased checked source is affixed to the midpoint of the pipe in direct line with the detector's central axis. A VisEd rendering of the transmission source is given in Figure 5.

Figure 5. External Transmission Source Simulation Models with Falcon BEGe.

(A) VisEd Representation of Dilution Experiment; (B) VisEd Simulation of Source Particle-Collisions of Dilution Experiment.

For completeness, the simulations were compared to the experiments on an absolute basis, although the calibration may be applied in a relative sense. The ^{137}Cs source employed in the

experiments was decay corrected from the date of certification, with an activity of 3.06×10^4 Bq (0.83 μ Ci). In the source definition of the external transmission source in the MCNPX model, the 661.7 keV ^{137}Cs source is emitted with an intensity of 85.1% [39]. A pulse height tally was also conducted in this simulation. However, since two sources are employed – ^{235}U in uranyl nitrate and ^{137}Cs transmission source – a *FUn* tally modifier was required in the MCNPX tally card in order to separately tally contributions from each source. Since each tally is normalized to emissions per source particle, each separate tally was calculated using appropriate emission intensities and activities prior to summation to determine an overall detector response.

3.3.2.2. ^{133}Ba and ^{57}Co Transmission Simulations

It can be anticipated that the ^{133}Ba and ^{57}Co used as alternative transmission densitometry sources will prove more sensitive to uranium content based on mass attenuation coefficient interactions of photons at the 356 and 122 keV energies, respectively. Since these sources were unavailable for experimental testing, use of validated simulations enables assessing the sensitivity of these sources to various concentrations of uranium in uranyl nitrate. The simulations emulated the setup previously depicted in Figure 5. The source energy intensities for ^{133}Ba and ^{57}Co are summarized in Table 5.

Table 5. Predominant Source Photon Emissions from ^{133}Ba and ^{57}Co [39].

^{133}Ba		^{57}Co	
Energy (keV)	Intensity (%)	Energy (keV)	Intensity (%)
276.4	7.2	122.1	85.6
302.9	18.3	136.5	10.7
356.0	62.1		
383.9	8.94		

4. RESULTS

4.1. Dilution Measurement Results

Dilution measurements were conducted in a controlled, low-background environment in order to assess the gamma-ray detector responses to various concentrations of uranium, identify the properties of signatures at each concentration, and provide data for simulation modeling efforts. Using the Falcon BEGe, Osprey 2 $\times$ 2 NaI(Tl), and Inspector 1000 LaBr₃ detectors, gamma-ray signatures were analyzed for dilution concentrations of uranyl nitrate at 90, 85, 75, 50, and 10 g U/L. The pipe segments employed in these measurements were to the specifications of UNCLE. Both passive ^{235}U and ^{137}Cs transmission densitometry measurements were conducted for three candidate COTS detectors.

4.1.1. Passive Dilution Measurements

As shown in the photopeak area analysis depicted in Figure 6, the Osprey has the highest peak area for the 185.7 keV ^{235}U peak, whereas the Inspector 1000 has the lowest efficiency in the

507 experimental setups employed. The dilution concentration is linearly correlated with the net peak
508 area of the 185.7 keV ^{235}U emission. From 90 g U/L down to 10 g U/L, the peak area of 8053
509 cpm drops overall by 95% at a rate of 85 cpm per g U/L uranyl nitrate for the Osprey detector.
510 For the Inspector dilution concentrations from 90 to 50g U/L, the peak area of 1133 cpm drops
511 overall by 57% at a rate of 16 cpm per g U/L (Note: At 10 g U/L, the peak for the Inspector was
512 not discernible by Genie as an identifiable peak within the provided confidence limits, and thus
513 no peak area data point is provided at this concentration). Finally, the Falcon net peak area of
514 3963 cpm drops from 90 to 10 g U/L overall by 88% at a rate of 44 cpm per g U/L.
515 Consequently, the Osprey is the most sensitive to concentration variations in ^{235}U , whereas the
516 Inspector is the least sensitive.^{††} The uranyl nitrate solution provided sufficient counts in 3600 s
517 acquisition time to keep statistical error within < 5% for all detectors and concentrations.
518
519

^{††} A measurement aberration was, however, observed for the Osprey from 85g U/L to 75g U/L. A full sensitivity analysis was conducted for the full-energy peak efficiency of this detector using Canberra's ISOCS®. This data is reported in the companion paper and fully described in [1].

Figure 6. Net Peak Area for ^{235}U 185.7 keV Emission as a Function of Uranium Concentration. *Note: Error bars are plot, but are small compared to the scale of the data.*

4.1.2. ^{137}Cs Transmission Dilution Measurement Results

For each of the dilution concentrations at 90, 85, 75, 50, and 10 g U/L, transmission measurements were taken using ^{137}Cs for densitometry measurements. The dilution concentrations are poorly linearly correlated to the peak area fit of the 661.7 keV ^{137}Cs emission from a diagnostic perspective, as shown in Figure 7. Essentially, the trends are flat; the change in U concentrations is not discernable. From 90 to 10 g U/L dilution concentrations, the peak area of 969.5 cpm changes by approximately 11.5% for the Osprey detector. For the Inspector dilution concentrations from 90 to 10 g U/L, the peak area of 248 cpm changes by approximately 11.5%. Finally, the Falcon peak area decreases from 629 cpm at 90 g U/L by ~13% at 10 g U/L. Transmission measurement peak areas are expected to increase with decreasing solution concentration and density; however, in practice, due to mass attenuation properties, self-attenuation, source-detector geometry, and setup limitations in the contamination area, the 661.7 keV photons ^{137}Cs transmission measurements were insensitive to uranium in uranyl nitrate. Additionally, from 10 to 90g U/L solution concentration, the statistical error associated with peak area counts for the Osprey is approximately 3.4%. With the Inspector 1000, the statistical error over the range of dilution concentrations is ~6.5%, whereas the statistical error associated with the Falcon is 4%.

Figure 7. Peak Area for ^{137}Cs Transmission Measurements as Function of Uranium Concentration for All Detectors.

In going to lower gamma-ray transmission energies (^{133}Ba and ^{57}Co), the change in uranium concentration is discernable because the linear attenuation coefficient is sensitive to the uranium photoelectric effect cross section (see 4.3.2 for discussion).

4.2. UNCLE Measurement Results

Both passive ^{235}U and ^{137}Cs transmission densitometry measurements were conducted in field trials at UNCLE using the three detectors employed in the dilution experiments. The dilution measurements provided an indication of instrumentation sensitivity and potential monitoring signatures, which were tested in an operational environment. Under realistic operating conditions, including facility background (pipes/tanks), environmental variables (temperature, pressure), and fluid flow dynamics, the robustness of the Falcon, Inspector, and Osprey were tested.

4.2.1. Change in Flow Rate Measurements

As shown in Figure 8, there is a minimally discernable difference in the detected count rates as a function of flow rate. This is perhaps due to the fact that in both cases, the detector is seeing a pipe full of uranyl nitrate of the same concentration; in the absence of voiding, it behaves like a static configuration. For the Falcon, the 185.7 keV peak area decreases by 0.27% decreasing from 1070 RPM to 500 RPM, whereas the Osprey peak area decreases by 9.77% and the Inspector by 6.96%. Although these changes are statistically significant with respect to the peak area uncertainty, further study is warranted in intermediary flow rates to control for systematic error (i.e. detector placement, ambient room temperature, vibration correction, etc.)

Figure 8. Comparison of UNCLE Peak Areas at 1070 rpm and 500 rpm.

The pipe specifications and source-detector geometry employed in the dilution experiments were emulated in the UNCLE facility. In addition, the uranyl nitrate employed for the dilution experiment was extracted from UNCLE. However, despite keeping the aforementioned parameters consistent, variables such as changes in temperature and pressure from dynamic fluid flow, in addition to voiding effects, deviate from the measured gamma-ray static dilution signatures.

If peak area is indicative of overall uranium content, the UNCLE data indicate that operating conditions reduce the “effective” concentration of circulating uranyl nitrate to that of the 75–85 g

U/L range of the static measurements (Figure 9). Even though the uranyl nitrate circulating in UNCLE is the same uranyl nitrate employed in the 90 g U/L dilution measurements, vibrations, environmental temperature, geometry variations, and possible voiding effects due to dynamic fluid flow contribute to an overall effective density lower than that of static measurements. Source-detector geometry, ROI selection, and peak discrimination methods employed by GENIE [40], as well as localized voiding, background, and environmental conditions contribute to interpretations of these results.

Figure 9. Comparison of UNCLE Spectra at 1070 RPM and 500 RPM with Dilution Spectra for Falcon BEGe at 185.7 keV ^{235}U Energy.

4.2.2. UNCLE ^{137}Cs Transmission Measurements

As with the static dilution experiments, transmission measurements were taken with ^{137}Cs to assess the 661.7 keV emission sensitivity to uranium content. Spectra were collected at the same location and set up as the flow rate measurements for 3600 s live time at the 1070 rpm flow rate. The ^{137}Cs transmission measurements were conducted at 1070 rpm, emulating the dilution transmission measurements, to determine the effectiveness of ^{137}Cs in assaying uranium content. A comparison of the full energy transmission spectra for the 661.7 keV peak is plotted in Figure 10 for the Falcon BEGe detector. As seen with the static transmission dilution measurements in Figure 7, ^{137}Cs is relatively insensitive to uranium content over the concentration range of 75–90 g U/L, and shown in comparison to the dynamic UNCLE measurements at 90g U/L in Figure 10.

Furthermore, compared to the passive measurements in Figure 9, which indicates that the “effective” concentration of the uranyl nitrate as given by the 185.7 keV peak is lower than the 90 g U/L of the static measurements, Figure 10 would indicate the opposite (i.e. UNCLE uranyl nitrate had a higher “effective” concentration than 90 g U/L than static). They both demonstrate how variables in operating conditions can alter the spectrum, as well as other factors, such as transmission source placement, collimation, etc.

Figure 10. Comparison of UNCLE Transmission Spectra at 1070 rpm with Dilution Transmission Spectra for Falcon BEGe.

4.3. MCNPX Simulation Results

Computational models were created in order to simulate and test the detection response to various concentration and densitometry variables in the static pipe measurement scenario. Passive and ^{137}Cs transmission models were compared with experimental data and coincided in good agreement with experimental measurements. Both ^{133}Ba and ^{57}Co as densitometry transmission sources were additionally simulated to determine the optimal densitometry source for monitoring uranyl nitrate with respect to the highest sensitivity to uranium content.

4.3.1. Simulations of ^{235}U

Based on the computational models, the simulations are well validated for the ^{235}U content, as shown in Figure 11. For the 185.7 keV peak, the 90 g U/L peak area values for the simulated and measured values were within 5% of each other, where the 85, 75 and 50 g U/L converged within <0.76%, showing the model is well validated for this peak signature. At 10 g U/L, the high level of dilution was less well matched in the absence of uranium, from which specific activity, self-shielding and solution inhomogeneity becomes increasingly prevalent.

Figure 11. Comparison of ^{235}U Peak Area for Measured and Simulated Dilution Measurements using Falcon BEGe.

4.3.2. Transmission Simulations of ^{133}Ba , ^{57}Co , and ^{137}Cs

Since measurements were not possible to test these sources' sensitivities to uranium content, use of the validated simulations determined the transmission sensitivity for PM signature determination. The attenuation analysis proposed lower energy photon sources from ^{133}Ba and ^{57}Co would be more sensitive to uranium content. Since experimental measurements were not possible to test these sources' sensitivities to uranium content, use of the simulations provide transmission sensitivities for PM signature determination.

Gross peak areas for ^{137}Cs showed a small approximate linear correlation but were not highly sensitive to dilution concentrations to uranyl nitrate, as shown in Figure 12. Diluting from 90 to 10 g U/L, there is a relative peak area increase 10.3% of transmitted ^{137}Cs 661.7 keV photons

(compared to an 8.1% increase in the static experimental transmission measurements). The peak area for ^{133}Ba was only slightly more sensitive to uranium content, whereas ^{57}Co was much more sensitive to changes in uranium content. The 356 keV transmission peak for ^{133}Ba was assessed with the Falcon BEGe detector response for various concentrations of uranyl nitrate. Since the half-life of ^{133}Ba source is approximately 10.5 years, it is favorable from an operational monitoring perspective, as source replacement would need to be less frequent than ^{57}Co (half-life ~271 days). Since ^{57}Co has such a short half-life, a very high activity source would be required to sufficiently penetrate the uranyl nitrate-filled pipe and provide a statistically confident detection signal. Peak area values of external transmission source for 356 keV and 122.1 keV photons from ^{133}Ba and ^{57}Co are provided in Figure 12. The insensitivity of ^{137}Cs shows common behavior in both the MCNP simulations (Figure 12) and the experimental data (Figure 7) for the Falcon BEGe.

Figure 12. Net Peak Area Comparing ^{133}Ba , ^{57}Co , and ^{137}Cs Transmission MCNPX Simulations as a Function of Dilution Concentration.

5. DISCUSSION

Experimental measurements were conducted using the Canberra 2×2-in. NAIS with the Osprey digital tube base, the Canberra Inspector 1000 with 1.5×1.5 in. LaBr_3 Probe, and the Canberra Falcon BEGe detectors for dilution concentrations of 10–90 g U/L of uranyl nitrate (0.76 wt.% enrichment), as well as in an operational environment in UNCLE at 90 g U/L at 1070 rpm and

500 rpm pump speeds (7963.4 kg/h and 3528.9 kg/h, respectively). Both passive and transmission ^{137}Cs measurements were conducted to correlate the signature peak areas with the uranyl nitrate solution concentration, density, and mass.

In the operational measurements in UNCLE at 90 g U/L, at a varying pump flow rate speed from 1070 to 500 rpm, each of the detectors was insensitive to flow rate. At each flow rate, the response was essentially unchanged, due to the detector seeing a full pipe of uranyl nitrate in its source-detector geometry, independent of pump speed.

In the dilution measurements, a pipe segment was fabricated based on the UNCLE specifications, where the detector response sensitivity was determined for assaying 185.7 keV emissions from ^{235}U , as well as the sensitivity of the 661.7 keV emission from ^{137}Cs as a transmission densitometry source. For all detectors, the 185.7 keV peak correlated with the uranyl nitrate concentration, whereas the 661.7 keV transmission source was very insensitive to changes in uranium concentration and solution density.

The relative insensitivity of ^{137}Cs to uranium content is supported by the convergence of the mass attenuation coefficient at higher energies for uranyl nitrate. Lower energy densitometry sources, for instance ^{57}Co (122 keV) and ^{133}Ba (356 keV), were theorized to be better able to discriminate uranium content. The results of the Monte Carlo simulations for the Falcon BEGe concluded that although ^{57}Co was most sensitive to uranium content at dilution concentrations, it may perhaps be too sensitive because of Compton continuum contributions from ^{235}U emissions between 140–205 keV, contributing higher peak area uncertainty in the 122 keV signature. In addition, from a practical standpoint, due to the short half-life of ^{57}Co and the low penetrability of 122 keV photons, a high-intensity ^{57}Co source would be required and would need to be replaced often in an operational monitoring setting. Transmission densitometry employing ^{133}Ba 356 keV emissions were viable for discriminating among uranyl nitrate dilution concentrations but were not as sensitive as ^{57}Co . Also, ^{133}Ba densitometry measurements using the 356 keV peak would prove problematic with low-resolution detectors, as ^{133}Ba has multiple lines that would interfere with the 185.7 keV signal unless high-resolution gamma spectroscopy was used.

Conversion activities occur in outdoor industrial-scale chemical plants with multiple transfer pipes and chemical storage tanks. The effect of these adjacent tanks and background measurements influences the peak area uncertainty, and hence the confidence of the assay peak area signatures. Sufficient shielding helps mitigate these background effects and must be considered in the installation of any radiation monitoring system in such facilities. In addition, peak area uncertainty due to Compton continuum from ^{137}Cs emissions during transmission measurements, as well as from higher-energy ^{238}U in secular equilibrium, increased the continuum, and consequently, the peak area uncertainty for the ^{235}U emissions in the 100–200 keV monitoring range. A dual detector option – a thin (planar) detector for the 185.7 keV and a large-volume (coaxial) detector for the transmission measurement – could be possible but would add to the cost and complexity for a field setting.

5.1. Diversion Stream Modeling

Making verification assessments requires converting NDA detector measurement data (185.7 keV peak area count rate) into a mass flow rate.

- **Solution Concentration ($cpm \rightarrow g\ U/L$):** The concentration of the uranyl nitrate flowing in the transfer pipes can first be determined by an in-line collimated densitometry source and gamma-ray detector, which will determine uranyl nitrate concentration and density in the detector field of view (pending previous system calibration) from the signature peak area.
- **Flowrate ($rpm \rightarrow kg\ or\ L\ uranyl\ nitrate/hr.$):** A combination of external/inline flow rate meters (Doppler, acoustic, Coriolis) and/or operator declarations provides the flow rate (rpm) and mass flow rate (kg or L uranyl nitrate /h) of uranyl nitrate.
- **Mass Flow rate of U ($kg/h \rightarrow MTU/yr$):** From Equation 1, the concentration and flow rate data can be combined to determine the mass flow rate of U (kg U/h). Conversion of kg U/h can be converted to NUCP throughput of MTU/yr for comparison with IAEA requirements.

At a flow rate of 1070 rpm, representative of Springfields' NUCP, throughput translates into a mass flow rate of 7963.4 kg uranyl nitrate/h. At 90 g U/L, this equates to 653 kg U/h. If UNCLE was run continuously for a whole year, this would correspond to ~5724t U/yr, with 1 SQ representing 0.17% of the annual throughput.

Gradually diverting 10 MTU/yr requires that 1.14 kg U be diverted daily for a 24 h cycle, or 3.42 kg daily in an 8 h cycle. These represent 0.17% (24 h) and 0.52% (8 h) of throughput for a flow rate of 1070 rpm. At a lower flow rate of 500 rpm, daily diversion values represent 0.39% and 1.18% for the 24 and 8 h cycles, respectively.

UNCLE is capable of reaching a mass flow rate of 12,800 kg/h if run at full capacity. Since uranium comprises 8.02% (Table 1), the maximal uranium throughput for UNCLE is ~9200 MTU/yr, potentially classifying UNCLE as a large-scale throughput facility. At the tested pump speeds of 500 rpm and 1070 rpm, where the latter speed emulated the throughput of the Springfields conversion facility at 90 g U/L, these translate into an annual throughput of 2537 MTU and 5724 MTU at full capacity. Detection of 10 MTU for 1070 rpm equates to a measurement uncertainty not less than 0.17%.

From the experimental dilution measurements at 90 g U/L, the 185.7 keV peak uncertainty calculated from the peak area [41] for the detector measurements spans 0.20%–0.55%. If the peak area is correlated with the density and concentration of uranyl nitrate, the uncertainty can be propagated to consider uncertainty in annual throughput. In UNCLE, this uncertainty range translates to 11.6–31.4 MTU, which is in excess of the 10 MTU IAEA criterion. In addition, the dilution experiments demonstrated that it is statistically indiscernible to differentiate a concentration change of ~5 g U/L, especially in the case of low-resolution measurements, in the presence of high Compton continuum contributions, or the use of insensitive transmission sources such as ^{137}Cs . As the concentration changes from 90 g U/L to 85g U/L, the 185.7 keV

peak area decreases by as little as 1.34% for the Falcon detector, in the most conservative detector measurement.

At 90 g U/L, operational measurements in UNCLE were taken for 3600 s, with a conservative statistical uncertainty of 0.4% for the Inspector, 0.17% for the Osprey, and 0.22% for the Falcon detector, respectively. To measure a 1 SQ variation in UNCLE due to changes in uranyl nitrate concentration, the overall uncertainty of the measurement must be below 0.17%. Consequently, optimized background shielding and increased counting time may enhance statistical confidence in gamma-ray spectroscopic data.

6. CONCLUSIONS

In the context of recent IAEA policy recommendations for monitoring purified compounds in uranium conversion facilities, the scope of this project addressed instrumentation testing in dilution measurements and at ORNL's UNCLE facility. The KMP reinterpreted as first satisfying INFCIRC/153 Para. 34(C) materials occurred following solvent extraction, producing uranyl nitrate during the conversion process. Based on in-depth experimental measurements, simulations, and uncertainty/sensitivity quantification, the abilities of COTS gamma-ray detectors were evaluated to determine their role in an overall safeguards system to determine if IAEA safeguards criteria could be met.

Variation in uranium content remains a challenge using stand-alone NDA methods. Without verifiable declarations or further data – i.e., density/densitometry, mass flow rate data, or ^{238}U signatures in secular equilibrium – interpretation of the 185.7 keV peak area alone is incomplete. However, the introduction of a transmission densitometry source does provide information regarding solution concentration, and analysis of the 185.7 keV ^{235}U high-intensity emission provides information regarding uranium enrichment. Together, these two signatures provide invaluable information regarding the uranyl nitrate content.

Although the IAEA requirement of 1 SQ in a period of 1 year cannot be detected using stand-alone NDA measurements, consideration must be given to estimations of how 5 MTU is all that is required to produce 1 SQ of centrifuge material, as stipulated by Nusbaum *et al.* [42]. Nusbaum *et al.* determine that a rogue party would have a 50% probability of getting away with an SQ (10 MT of natural uranium) from a small plant that processes only 100 MTU/year; this consequences are further augmented when a rogue diverter with a clandestine enrichment capability requires only roughly 5 MTU of diverted natural uranium to produce 1 SQ (25kgU) of highly-enriched uranium (HEU) [42].

For NUCPs classified as small (~100 MTU/yr.), 1 SQ represents ~ 10% of the plant throughput [5] [6] [7] [8] [9]. At these levels, it is possible to detect misuse. However, considering the implementation of this criterion in large facilities throughputs higher by levels of magnitude, diversion of 1 SQ becomes essentially undetectable given the systematic uncertainty for the detector scenarios presented in the paper. Given that the UNCLE facility is capable of circulating uranyl nitrate at the level of a large throughput facility, and given its extremely modest size, consideration must be given to how easily operational design (such as a diversion stream) can easily produce undeclared uranyl nitrate. Consequently, consideration of the feasibility of

independent safeguards verification must be further considered if safeguards monitoring is to occur earlier in the fuel cycle.

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