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Use of InSpector$^{\text{TM}\,1}$ 1000 Instrument with $LaBr_3$ for Nuclear Criticality Safety (NCS) Applications at the Westinghouse Hematite Decommissioning Project (HDP) – 13132

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ABSTRACT

The Westinghouse Hematite Decommissioning Project (HDP) is a former nuclear fuel cycle facility that is currently undergoing decommissioning. One aspect of the decommissioning scope is remediation of buried nuclear waste in unlined burial pits. The current Nuclear Criticality Safety program relies on application of criticality controls based on radiological set-points from a 2 x 2 Sodium Iodide (NaI) detector. Because of the nature of the material buried (Low Enriched Uranium (LEU), depleted uranium, thorium, and radium) and the stringent threshold for application of criticality controls based on waste management (0.1 g ^{235}U/L), a better method for ^{235}U identification and quantification has been developed. This paper outlines the early stages of a quick, in-field nuclear material assay and ^{235}U mass estimation process currently being deployed at HDP. Nuclear material initially classified such that NCS controls are necessary can be demonstrated not to require such controls and dispositioned as desired by project operations. Using Monte Carlo techniques and a high resolution Lanthanum Bromide (LaBr) detector with portable Multi-Channel Analyzer (MCA), a bounding ^{235}U mass is assigned to basic geometries of nuclear material as it is excavated. The deployment of these methods and techniques has saved large amounts of time and money in the nuclear material remediation process.

INTRODUCTION

The Westinghouse Electric Company LLC (Westinghouse) Hematite Site, located near Festus, Missouri, is a former nuclear fuel cycle facility that is currently undergoing decommissioning. Throughout its history, operations at the Hematite facility included the manufacture of uranium metal and compounds from natural and enriched uranium for use as nuclear fuel. Specifically, operations included the conversion of uranium hexafluoride gas of various ^{235}U enrichments to uranium oxide, uranium carbide, uranium dioxide pellets, and uranium metal from 1956 until 2001. Cessation of manufacturing operations in 2001 led to an amendment of the facility license to reflect a decommissioning scope.

Decontamination and Decommissioning (D&D) of equipment and surfaces within the process buildings, in addition to the buildings themselves, was accomplished in the first phase of the project. The current phase of the site-wide remediation operations includes clean-up of facility process wastes that were consigned to unlined burial pits.

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[1] Canberra InSpector™ 1000 Digital Hand-Held Multichannel Analyzer. InSpector, Genie, ISOCS and LabSOCS is a trademark of Canberra Industries, Inc.

The InSpector 1000 instrument from Canberra is a hand-held multichannel analyzer with a LaBr probe, designed for easy portability, easy decontamination and rugged conditions. It offers easy nuclide identification for less sophisticated users, or full spectral analysis capabilities for expert users. The LaBr scintillator probe is a sourceless stabilized gamma probe with excellent energy resolution. The sourceless stabilization allows performance throughout extreme temperature variations and limits energy drift that generally degrades isotope identification results. The result is a uniform response over the full temperature range. This feature is useful in HDP remediation, which operates in year round extreme and harsh temperature environments.

Specific to HDP operations, the limits above which materials encountered during HDP remediation activities are designated at Non-NCS Exempt Material are based upon the concentration of fissile nuclides in the material, or the total fissile nuclide mass content of the material. Non-NCS Exempt Material is the classification given to material that has a fissile nuclide concentration greater than the limit established for NCS and requires criticality safety controls to ensure their safe handling, packing, processing, and storage. Specifically, materials determined to satisfy either of the following limits are designated as NCS Exempt Material and require no controls to ensure they remain safely subcritical:

- A fissile nuclide concentration ≤ 0.1 g ^{235}U/L; or
- A total fissile nuclide mass content ≤ 15 g ^{235}U and occupying a volume of at least 5 L; or
- A ^{235}U/total U enrichment ≤ 0.96 wt. %

In order to support decision making regarding when exhumed material may be re-classified as NCS Exempt Material based on low ^{235}U concentration, a calibration analysis is used for conducting in-situ radiological surveys. This calibration analysis determines, for a given set of conditions, the minimum observed detector count rate as a function of the ^{235}U quantity associated with the in-situ materials under investigation.

Traditionally, remediation surveys for fissile material are performed using a windowed 2 x 2 NaI detector, set to detect ^{235}U gammas (> 75 keV). The HDP has implemented a Nuclear Criticality Safety (NCS) Program based on screening response from a NaI detector.

Since ^{235}U is the primary nuclide of concern for NCS at HDP, a method to identify and quantify ^{235}U in the field is beneficial for initial application of NCS controls. It is recognized that depleted uranium (< 0.96 wt % ^{235}U in UO_2), thorium, and radium are abundant in HDP burial pits. They are gamma radioactive and produce high count rates in a windowed NaI. The InSpector 1000 instrument with a LaBr probe allows for a Region of Interest (ROI) to be set around the 186 keV photopeak (57.2% intensity) in order to determine in real-time if ^{235}U is detected and at what activity.

The distinct difference between the traditional NaI used in remediation surveys and the InSpector 1000 instrument with a LaBr probe is the qualitative distinction that can be made using the InSpector 1000 instrument MCA. This allows discrimination between ^{235}U and other nuclides not of concern from an NCS standpoint that may potentially exist in the HDP burial pits. Utilizing these properties of the detector, NCS may be able to exercise exemption from criticality control on items or areas. This proves especially pertinent in the case of locating an in-situ Stop Work point based on NaI detector response.

CALIBRATION ANALYSIS

The calibration analysis is performed by accurately modeling the response of the LaBr detector in two commonly encountered modeling scenarios. The detector response is modeled using the pulse height tally feature of the Monte Carlo N-Particle (MCNP5) code. MCNP5 uses the continuous energy cross section libraries from ENDF/B-VI.

Because the calibration analysis is intended to be used for NCS purposes, the factors predict an upper bound of ^{235}U mass, and a number of conservative assumptions are employed as described below:

- A bounding fixed uranium enrichment of 100 wt.% ^{235}U/U;
- The uranium particulate within the waste is modeled as uranium oxide (UO_2) at its full theoretical density of 10.96 g/cm^3;
- The analysis uses a tap density for UO_2 of 3.5 g/cm^3;
- The reported results are at the 97.7% confidence level to account for statistical uncertainty.

The calibration analysis is based on two waste medium configurations, which are the most commonly encountered for placement under NCS controls:

- Model A: Multiple mass lumps of UO_2 in dry soil with a density of 1.73 g/cm^3 for cut depths of 2, 4, 6, and 8 inches;
- Model B: Homogeneous mixture of UO_2 and wet soil with a density of 2.03 g/cm^3 for fill fractions of 25, 50, 75, and 95 percent.

The following waste material types are used to represent in-situ and containerized waste under evaluation. In the calibration calculations, the waste material type is uniformly mixed with the uranium under evaluation.

- Dry soil at a density of 1.73 g/cm^3;
- Saturated soil at a density of 2.03 g/cm^3;
- UO_2 tap density of 3.5 g/cm^3;
- Overall lump density of 2.78 g/cm^3 for dry soil.

The MCNP model examines a range of parameters in Model A as detailed in Table 1.

Table 1: Input Parameters and Ranges Investigated for Model A

Model Parameter	Parameter Units	Parameter Range Examined
Concentration	g ^{235} U/L	N/A
UO_2 Lump Mass	g	3, 6, 10, 15, 25, 50, 75, 100, 125, 200, 300, 350
Waste Depth Above Lump	in	2, 4, 6, 8
UO_2 Lump Tap Density	g/cm ^{3}	3.5
Soil Density	g/cm ^{3}	1.73
Lump Overall Density	g/cm ^{3}	2.78
Detector-Waste Distance	in	3

In Model B, the container is loaded to various fill heights with a homogeneous mix of UO_2 and wet soil. The LaBr probe is placed on contact at the bottom of the container for response measurements to be applied to the developed ^{235}U mass/count rate relationship. The container used in the calibration analysis is materially synonymous with those currently in rotation at HDP for elevated material isolation. The input parameters for Model B are detailed in Table 2.

Table 2: Input Parameters and Ranges Investigated for Model B

Model Parameter	Parameter Units	Parameter Range Examined
Source	N/A	Homogeneous mix
UO_2 Mass	g	3, 6, 10, 15, 25, 50, 75, 100, 125, 200, 300, 350
Fill Percentages	%	25, 50, 75, 95
Soil Density	g/cm ^{3}	2.03
Detector position	N/A	bottom

Because the fill height fraction inside the containers is known, providing a calibration for the detector in the bottom position provides the most accurate approach for predicting the amount of ^{235}U that may be present inside the assayed container. A previous calibration analysis (Ref. 6) has shown this to be the best position for measurements. Therefore, restricting the assay location to the bottom of the container and selecting a conservative fill height fraction provides a more accurate assessment of the amount of ^{235}U that may be present in the container.

The detector used in both models is detailed in Ref. 1. A graphical representation of the LaBr detector MCNP model is presented in Figure 1. Some features have been simplified and have an insignificant effect on the detector's response.

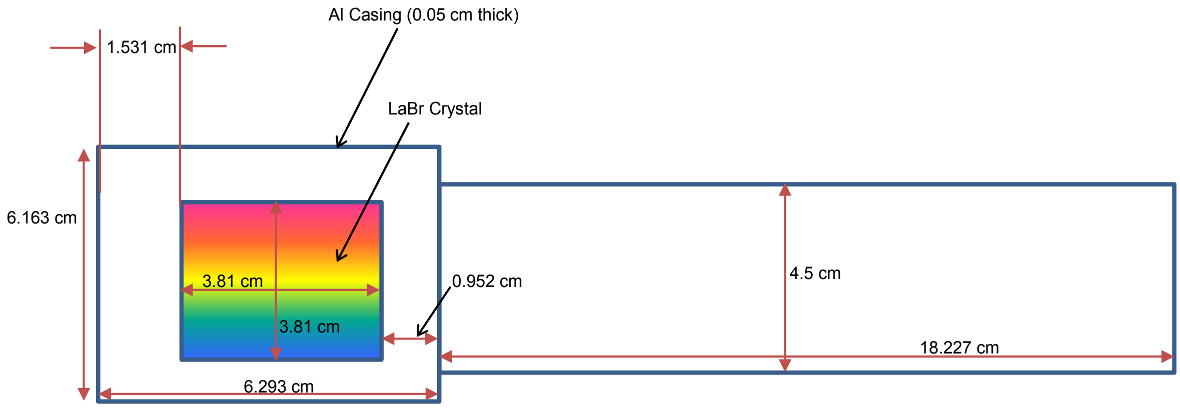

Figure 1: MCNP Representation of the Canberra Industries Model IPROL-1 Intelligent LaBr Probe

In Model A, the LaBr detector is modeled centralized above the centerline of the in-situ waste materials at a fixed height of 3-in above the surface of the waste. The configuration for Model A is illustrated in vertical cross section in Figure 2 for a lump at a 6-in depth.

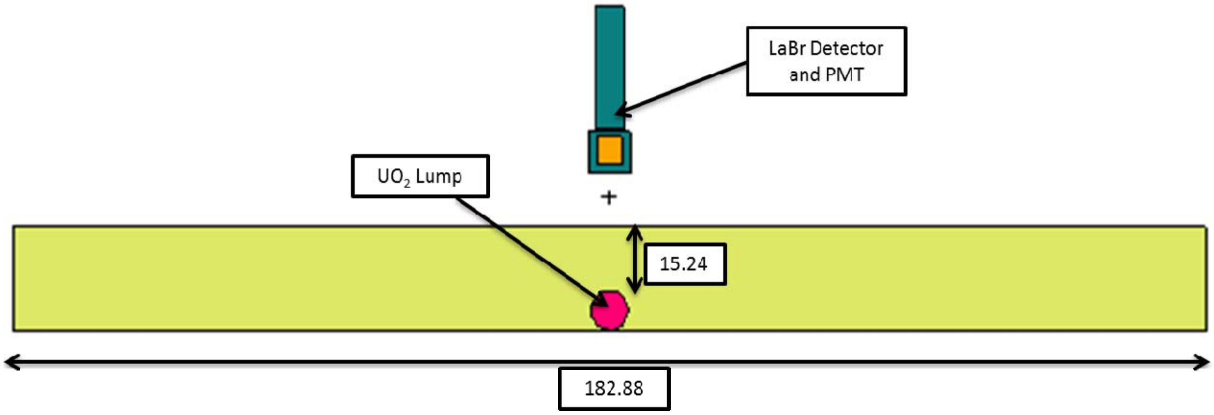

Figure 2: Vertical Cross Section of the MCNP Lumped Contamination Model (Model A) [dimensions in cm]

In Model B, the LaBr detector is placed on contact to the container in the bottom position as shown in Figure 3. The fissile material source is homogeneously mixed in the soil and loaded into the container to the specified fill height.

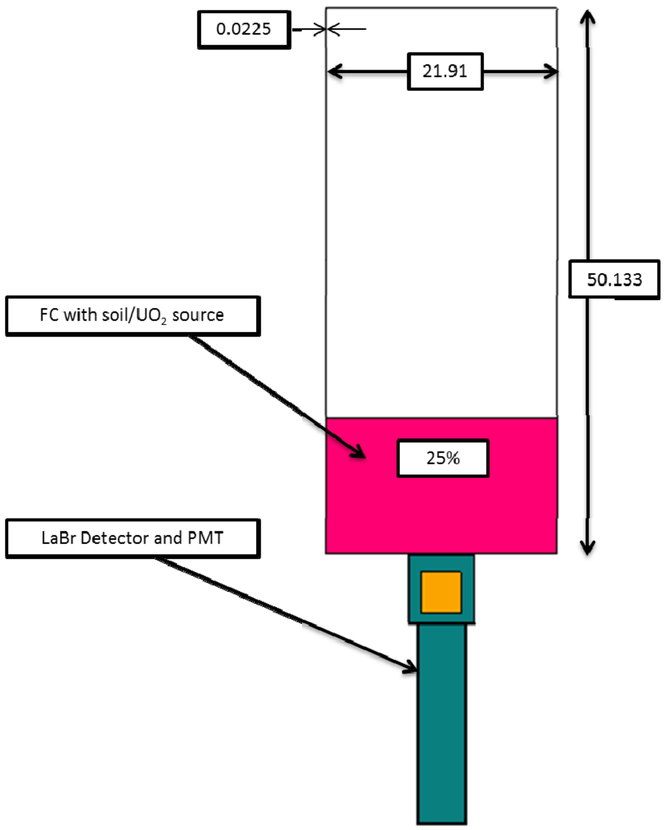

Figure 3: Vertical Cross Section of the MCNP Container Model (Model B) [dimensions in cm]

The source term employed for the calibration analysis is determined by decaying UO_2 with a fixed ^{235}U mass content of 1 g (activity 2.161×10^{-6} Ci) for 50 years using the SCALE ORIGEN-S depeltion code with UO_2 bremsstrahlung photo data libraries.

Since the LaBr detector coupled with an MCA is capable of supplying the detector response specific to the ^{235}U 185 keV photopeak, the calibration is intended to provide guidelines for bounding ^{235}U mass using solely this photopeak. Figure 4 presents the results of the MCNP calculations for Model A, and Figure 5 presents the results for Model B. The figures are presented as mass of ^{235}U as a function of detector count rate for better field implementation. A quadratic regression fit to the data is shown on each figure. The equation and its associated Goodness of Fit parameter are also displayed.

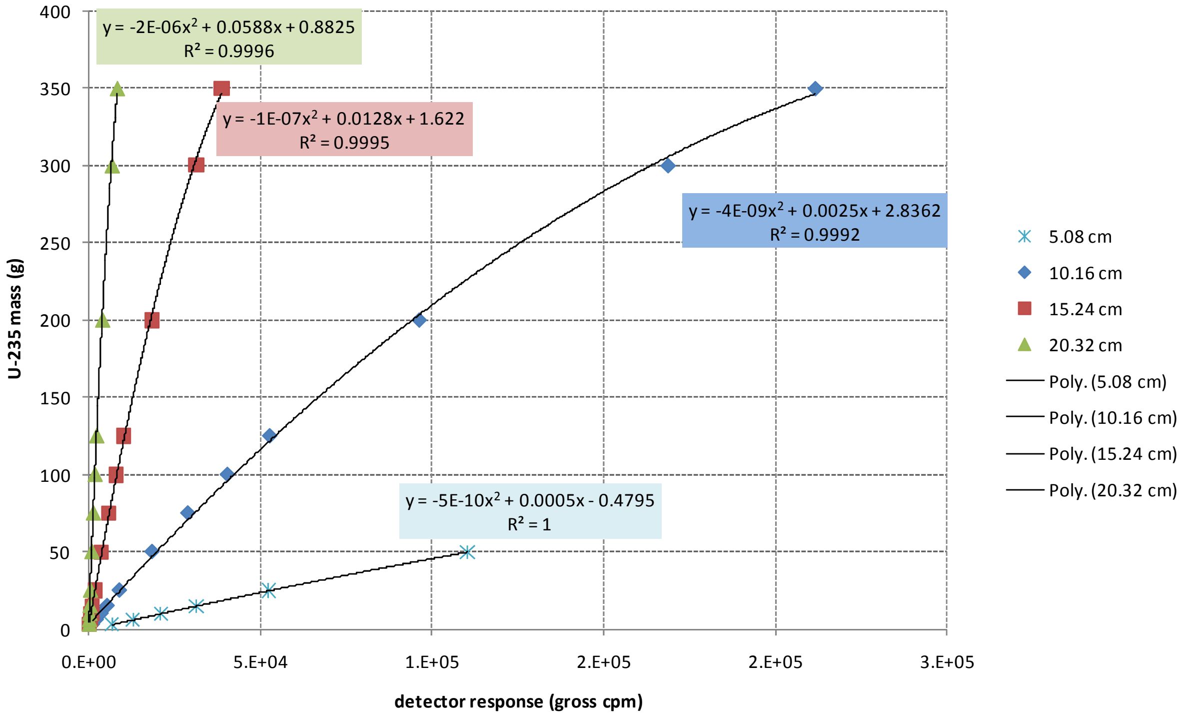

Figure 4: ^{235}U Mass as a Function of 185 keV Detector Response with Trend Line for Model A

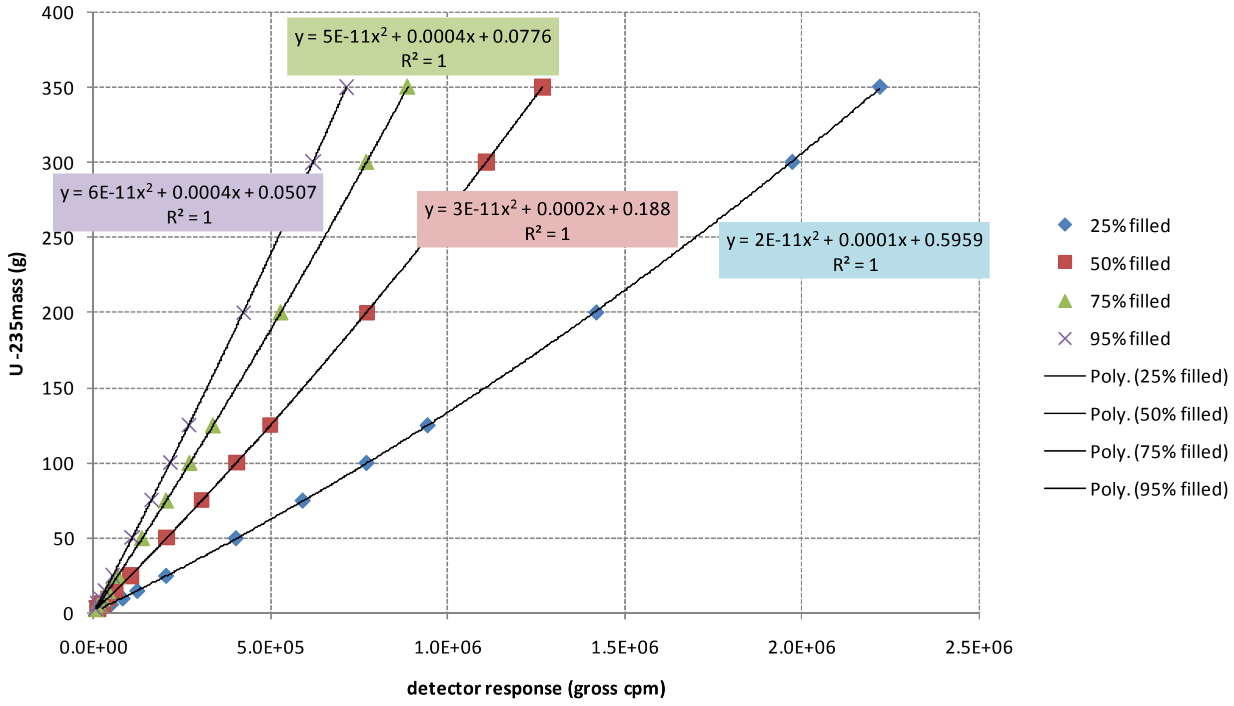

Figure 5: ^{235}U Mass as a Function of 185 keV Detector Response with Trend Line for Model B

Table 3 summarizes the fit equations for each model and variable. The 'cpm' parameter is the gross number of counts from the LaBr detector 185 keV photopeak Region of Interest.

Table 3: Fit Equations for Estimating g ^{235}U

Geometry	Variation	Fit
Lump	5.08 cm	g ^{235}U = -5E-10 * cpm^2 + 0.0005 * cpm - 0.4795
	10.16 cm	g ^{235}U = -4E-09 * cpm^2 + 0.0025 * cpm + 2.8362
	15.24 cm	g ^{235}U = -1E-07 * cpm^2 + 0.0128* cpm + 1.622
	20.32 cm	g ^{235}U = -2E-06 * cpm^2 + 0.0588 * cpm + 0.8825
Field Container	25%	g ^{235}U = 2E-11 * cpm^2 + 0.0001 * cpm + 0.5959
	50%	g ^{235}U = 3E-11 * cpm^2 + 0.0002 * cpm + 0.188
	75%	g ^{235}U = 5E-11 * cpm^2 + 0.0004 * cpm + 0.0776
	95%	g ^{235}U = 6E-11 * cpm^2 + 0.0004 * cpm + 0.0507

FIELD IMPLEMENTATION

During a normal operating day, HDP can produce around 50 containers with material flagged for some aspect of NCS control. Of these 50, about 30% would be initially classified as Non-NCS Exempt and require additional NCS controls including control of interaction (spacing). This is accomplished through use of an over pack drum and an attached collar. The use of such controls in a burial pit remediation can be cumbersome and time consuming.

Through the first 6 months of remediation operations, it became apparent that many of the containers initially classified as Non-NCS Exempt were grossly overestimated when containerized. This is because the NCS program limits are set based on 2 × 2 NaI detector response with a window set at > 75 keV. With the presence of ^{232}Th and ^{226}Ra, the NaI response, when applied to the original calibration analysis, will grossly over-predict the ^{235}U loading of any container.

Given that NCS thresholds and decisions are based on bounding estimates for fissile nuclides, this approach is reasonable. However, the loss of time and additional cost associated with implementing unnecessary control over material that is suitable for release can be eliminated, if there is a more appropriate solution.

The intention of the InSpector 1000 instrument is field use. The detector is rugged and portable with a wide range of feasible conditions for operation. It can easily be held in one hand while the probe is detached for measurement in hard to reach places. An example of in-field use is shown in Figure 6.

Figure 6: In-Field Use of InSpector 1000 for Nuclear Criticality Safety Measurements

Upon the initial determination that a subset of material may fall into Non-NCS Exempt status, the operator of the InSpector 1000 is called to the area. Following survey from Health Physics Technicians, and confirmation that the assayed material will qualify as Non-NCS Exempt, the InSpector 1000 operator assess the situation. This assessment includes a visual inspection of the remediation area or container in question, relayed information from the Health Physics Technicians, and finally a sample measurement. The flow of this process is depicted in Figure 7.

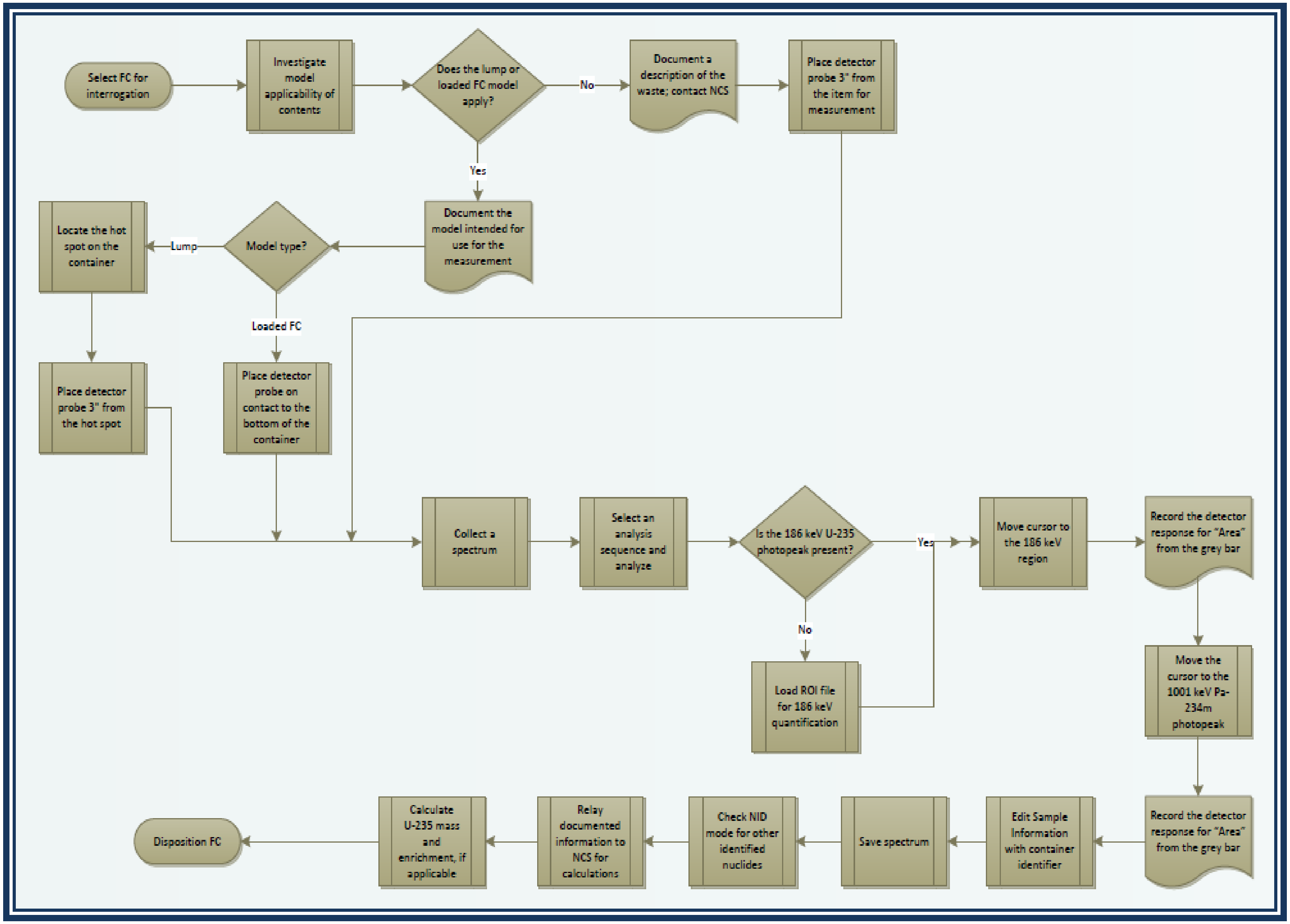

Figure 7: In-Field Non-NCS Exempt Material Assay Process Flow from Onset to Disposition

Once a spectrum is collected, the analysis sequence currently loaded on the InSpector 1000 automatically runs. Using peak search and locate algorithm, the peaks are identified and highlighted, and net and gross peak area are displayed on the screen. If a nuclide library is loaded on the instrument, a nuclide identifier mode identifies and lists the nuclides present.

The gross area under the 185 keV photopeak and the 1001 keV photopeak are recorded and used in the NCS analysis of the material. The operator determines which model to apply to the scenario at hand or whether a model is not applicable, after assessing the conditions. On occasion, two measurement locations or two modeling approaches may be used.

In general, if the material seems relatively homogenously dispersed throughout the container, with no large point source locations, Model B is applied. The fill height of the material is rounded up to the nearest percentage model. If a point source is located within the bucket, a version of Model A is applied. To correctly mimic the MCNP modeling scenario, the probe is located at a distance of 3-in from the container while acquiring a measurement. If the location of the hot spot appears to be along the centerline of the bucket, Model A is applied with 4-in of soil above the lump. If the hot spot appears to be along the outside of the bucket, Model A is applied with 2-in of soil above the lump.

Using the relationships in Table 3, a bounding estimate on ^{235}U grams is established at the time of measurement. This is then used to make a decision on the handling of the container in regards to NCS controls based on ^{235}U gram quantity. In addition, enrichment is estimated using peak ratios from the 185 keV and 1001 keV peak. The container may be released from NCS controls based on its inherent low or depleted enrichment.

Conducting in-field measurements does not have access to software capable of calculating efficiencies for the given geometry, and therefore, an efficiency equal to one is used for most calculations. Using the most common sources (an array of pellets and the 20 L Field Container), variations on input parameters to the geometry were tested to depict the effect on enrichment while still maintaining cases that were applicable to the calibration analysis. A variation to a single parameter was made and the resulting effect on enrichment noted. The parameters varied included:

- Composition (Z)
- Density
- Detector offset
- Geometry thickness
- Geometry dimensions

The count rates used for each photopeak are taken from an actual depleted pellet measurement and a container measurement. These results are shown in Table 4.

Table 4: Efficiency Effect on ^{235}U Enrichment Calculations

	Model	Slab Dimensions (in)	Detector offset (in)	Composition	Density (g/cc)	U-235 Efficiency	U-238 Efficiency	U-235 cpm	U-238 cpm	Percent Enrichment
pellets	no efficiency					1	1	2180	2490	0.20
	2" slab	12x12	3	70% dd; 30% UO2	1.2	3.51E-04	3.55E-04	2180	2490	0.20
	1" slab	12x12	3	70% dd; 30% UO2	1.2	6.92E-04	4.94E-04	2180	2490	0.15
	1" slab	6x6	3	70% dd; 30% UO2	1.2	1.22E-03	7.89E-04	2180	2490	0.14
	1" slab	6x6	0	70% dd; 30% UO2	1.2	4.02E-03	2.97E-03	2180	2490	0.16
	1" slab	6x6	0	100% UO2	3.5	1.29E-03	2.66E-03	2180	2490	0.41
	1" slab	6x6	0	100% UO2	2.5	1.78E-03	2.97E-03	2180	2490	0.34
Field Container	no efficiency					1	1	78735	2296	7.26
	25% full		0	70% dd; 25% poly; 5% UO2	0.8	8.19E-04	2.93E-04	78735	2296	2.72
	50% full		0	70% dd; 25% poly; 5% UO2	0.8	7.87E-04	3.47E-04	78735	2296	3.34
	75% full		0	70% dd; 25% poly; 5% UO2	0.8	9.04E-04	3.90E-04	78735	2296	3.27
	95% full		0	70% dd; 25% poly; 5% UO2	0.8	7.93E-04	3.53E-04	78735	2296	3.37
	95% full		0	70% dd; 20% poly; 10% UO2	0.8	6.37E-04	3.53E-04	78735	2296	4.16
	95% full		0	70% dd; 20% poly; 10% UO2	0.9	5.75E-04	3.34E-04	78735	2296	4.36

*dd-dry dirt; poly-polyethylene

The enrichment results for the pellets all indicated ≤ 0.96 wt.% ^{235}U and would have been dispositioned the same, as NCS Exempt material. In the cases for the Field Container, the largest effect was seen with added high-Z material (uranium). However, the bounding enrichment estimation for the container is shown with dual efficiencies of one. This is considered sufficient for NCS applications.

A beginning-of-day and end-of-day source check is conducted with a UO_2 source (two depleted uranium pellets and two low enriched pellets) because ^{235}U is the nuclide of interest in remediation. The energy calibration and peak magnitudes are verified as accurate to within 20%. Because the instrument is used to make NCS decisions, two independent measurements are required. In order to eliminate instrument error and qualify two independent measurements, a single pellet source check is conducted in between sample measurements with a single UO_2 source pellet. The InSpector 1000 has not failed a source check to date. Elimination of human error is accounted for with a second independent measurement conducted by an additional operator.

CONCLUSIONS

The results presented in Figures 4 and 5 establish conservative 185 keV photopeak count rates for the LaBr gamma detector which correspond to two common modeling scenarios encountered at the Hematite site. The regression fit lines generated for each scenario are applicable to waste remediation operations at the Hematite site in order to obtain a better understanding of the mass of ^{235}U presented in the remediation area of concern.

Through the use of the MCNP calibration analysis coupled with the response of the InSpector 1000 LaBr detector, NCS decisions can be made in real-time for the handling of excavated materials. Within the first three weeks of use, 85% of the containers initially declared Non-NCS Exempt were changed to NCS Exempt.

There are future intended uses for the InSpector 1000 at HDP including in-situ scans of the sanitary waste water treatment system and scans of sub-surface piping. Real-time identification and quantification of ^{235}U is an invaluable NCS tool for a remediation site handing consistent unknown material.

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