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DISCLAIMER

HEU Measurements of Holdup and Recovered Residue in the Deactivation and Decommission Activities of the 321-M Reactor Fuel Fabrication Facility at the Savannah River Site

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Abstract
This paper contains a summary of the holdup and material control and accountability (MC&A) assays conducted for the determination of highly enriched uranium (HEU) in the deactivation and decommissioning (D&D) of Building 321-M at the Savannah River Site (SRS). The 321-M facility was the Reactor Fuel Fabrication Facility at SRS and was used to fabricate HEU fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the SRS production reactors. The facility operated for more than 35 years. During this time thousands of uranium-aluminum-alloy (U-Al) production reactor fuel tubes were produced. After the facility ceased operations in 1995, all of the easily accessible U-Al was removed from the building, and only residual amounts remained. The bulk of this residue was located in the equipment that generated and handled small U-Al particles and in the exhaust systems for this equipment (e.g., Chip compactor, casting furnaces, log saw, lathes A & B, cyclone separator, Freon™ cart, riser crusher, ...etc). The D&D project is likely to represent an important example for D&D activities across SRS and across the Department of Energy weapons complex. The Savannah River National Laboratory was tasked to conduct holdup assays to quantify the amount of HEU on all components removed from the facility prior to placing in solid waste containers. The U-235 holdup in any single component of process equipment must not exceed 50 g in order to meet the container limit. This limit was imposed to meet criticality requirements of the low level solid waste storage vaults. Thus the holdup measurements were used as guidance to determine if further decontamination of equipment was needed to ensure that the quantity of U-235 did not exceed the 50 g limit and to ensure that the waste met the Waste Acceptance Criteria (WAC) of the solid waste storage vaults. Since HEU is an accountable nuclear material, the holdup assays and assays of recovered residue were also important for material control and accountability purposes. In summary, the results of the holdup assays were essential for determining compliance with the Waste Acceptance Criteria, Material Control & Accountability, and to ensure that administrative criticality safety controls were not exceeded. This paper discusses the γ-ray assay measurements conducted and the modeling of the acquired data to obtain measured holdup in process equipment, exhaust components, and fixed geometry scrap cans. It also presents development work required to model new acquisition configurations and to adapt available instrumentation to perform the assays.

1.0 INTRODUCTION

This report contains a summary of the holdup and material control and accountability (MC&A) assays conducted for the determination of highly enriched uranium (HEU) in the deactivation and decommissioning (D&D) of Building 321-M at the Savannah River Site (SRS). The 321-M facility was the Reactor Fuel Fabrication Facility at SRS and was used to fabricate highly enriched uranium (HEU) fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the SRS production reactors. The facility operated for more than 35 years. During this time thousands of uranium-aluminum-alloy (U-Al) production reactor fuel tubes were produced. After the facility ceased operations in 1995, all of the easily accessible U-Al

was removed from the building, and only residual amounts remained. The bulk of this residue is located in the equipment that generated and handled small U-Al particles and in the exhaust systems for this equipment (e.g., Chip compactor, casting furnaces, log saw, lathes A & B, cyclone separator, Freon™ cart, riser crusher, ...etc). [1]

The D&D activities were conducted by the Facilities Deactivation and Decontamination Program (FDD) at SRS. The D&D project was designated as an award fee item for the Site in fiscal years 2000 and 2001, and we believe it will represent an important example for D&D activities across SRS and across the Department of Energy weapons complex. FDD requested technical assistance from the Savannah River National Laboratory (SRNL) to determine the holdup of enriched uranium in the 321-M facility as part of the overall deactivation project of the facility.[2] This project included the dismantling and removal of all held-up HEU to the extent practical.

SRNL was tasked to conduct holdup assays to quantify the amount of HEU on all components removed from the facility prior to placing in B-25 containers. The U-235 holdup in any single component of process equipment must not exceed 50 g in order to meet the B-25 limit.[3] This limit was imposed to meet criticality requirements of the low level solid waste storage vaults. Thus the holdup measurements were used as guidance to determine if further decontamination of equipment was needed to ensure that the quantity of U-235 did not exceed the 50 g limit and to ensure that the waste met the Waste Acceptance Criteria (WAC) of the solid waste storage vaults. [3]

As each piece of process equipment was decontaminated in the deactivation project, HEU residue was collected into 2-gallon scrap cans for nondestructive γ-PHA assay (NDA). During operation the facility used a large number of scrap cans to store highly enriched uranium chips and filings for reprocessing. The scrap cans were designed to be critically safe, which made them extremely useful during the deactivation of the facility. These cans provided a geometrically safe container for placing the residue, filings, chips, and sweepings of HEU remaining in the building. This recovered residue would be considered attractive material for adversaries seeking to obtain bulk quantities of HEU, and so it was important to obtain accurate assay of it for MC&A measurement purposes.

In summary, the results of the holdup assays were essential for determining compliance with the Waste Acceptance Criteria, Material Control & Accountability, and to ensure that administrative criticality safety controls were not exceeded.

2.0 EXPERIMENTAL

All of the assays for HEU content were conducted using γ-PHA to count the passive 185 keV γ-ray to determine the α-decay rate of U-235 in the sample. Three distinct detection systems were used. They are a portable high purity germanium (HPGe) system, a portable sodium iodide (NaI) system, and the commercial Q^2 waste assay system that uses three HPGe detectors. In each case, use of the HPGe detectors requires periodic fill of liquid nitrogen to maintain the detectors at 77°K to sustain high-resolution operation. The NaI detector does not require cooling. It provides low-resolution γ-ray spectra with better detection efficiency and superior ergonomic acquisitions.

The HPGe and NaI detection systems were energy and efficiency calibrated in three acquisition configurations as described in references 4 and 5. Daily quality control (QC) checks were used to document satisfactory performance of these two detection systems throughout the lifetime of this project. The QC checks are described in references 6 – 9. The Q^2 detection system is QC checked using the 313-M operating procedure. For the adapted Q^2 measurements that we report, each detector was QC checked before and after each shift of operation using a 4.41-g source of HEU.

2.1 Adapted Q^2

To conduct the assays of HEU content in high density 55-gallon drums of solid waste, we adapted the commercial Q^2 assay system to a three-segment segmented γ-ray scanner.[10] The commercial Q^2 system is manufactured by Canberra Industries. It runs on NDA2000 acquisition and analysis software to perform a transmission corrected γ-PHA assay of fixed geometry solid waste. The 313-M Q^2 system is configured to count 55-gallon drums. The commercial system uses a direct measure of drum mass to determine overall counting efficiency of photons ranging in energy from 59 keV up to 1408 keV. A direct measure of sample mass (thus density for a fixed geometry) is used to calculate overall counting efficiency for each photon. Therefore every calculated counting efficiency includes an internal determination of photon transmission through the item.[11]

This technique of determination of transmission correction is very similar to the ingenious method employed by Dewberry in the differential absorption transmission corrected γ-PHA acquisition system of 235-F.[12] The commercial Q^2 direct assay technique assumes that the sample is approximately 100% full, uniformly distributed, low density, and low-Z material. To generalize, the instrument is intended for uniformly packed paper and plastic solid waste. The drums that we were interested in assaying specifically violated all of these four assumptions. They contained high-density solid waste that frequently was made of iron and steel and that was specifically not uniform. An example would be process equipment of unusual geometry like the hoist shown in Figure 1.

Figure 1. A two-ton hoist that represents a typical item of process equipment upon which g-PHA holdup assays were performed in this report.

To obtain a Q^2 γ-PHA assay of this non-uniform equipment, it was necessary of obtain a transmission correction factor for each of the three drum segments. It was also important to recognize that the high-density material frequently exceeded the range of density over which the instrument was efficiency calibrated. To overcome these limitations we used a movable source of HEU to obtain a direct measure of the transmission of the 185 keV γ-ray through each horizontal segment of each drum as it was viewed by the three detectors of the commercial instrument.

The adapted Q^2 technique is described completely in reference 10. Reference 10 contains an Excel speadsheet in which we perform all of the required calculations for each drum segment and for each composite drum. The transmission correction for each segment j is obtained by comparing the detection rate of the source only (T_0) spectrum to the detection rate of the sample only (S_j) spectrum and to the source plus sample (T_j) spectrum. Using this method yields a direct measure of transmission of the 185 keV γ-ray through the segment. The transmission is determined by equation (1).

$$T = [cps(T_j) - cps(S_j)] / cps(T_0), \quad (1)$$

where cps represents the detection rate of the 185 keV γ-ray in the spectrum. In our spreadsheet, the transmission correction value for each segment is then determined by two representations as explain in the text of reference 10. The first representation assumes a point source far field configuration in which the correction factor is simply $Cf_t(far)$= square root (1/T). The second representation assumes a close field point source configuration in which the correction factor is a more complex function.

$$Cf_t(close) = -k\ln(1/T) / [1-(1/T)^k], \quad (2)$$

where $k = \pi/4 = 0.785$.[13] We do not further describe the application of these transmission correction factors in this report. Our results are presented in section 3.1.

2.2 Holdup in Process Components

The holdup assays on the process equipment components represented the most difficult non-destructive assay problems that we encountered in this project. As always, process equipment components do not fit any of the traditional point, line, or area source acquisition configurations. It was necessary to develop approximate models and to obtain multiple, redundant acquisitions in order to determine defensible measured values. Russo provides an excellent treatise on γ-ray holdup analyses of HEU as well as other forms of special nuclear material.[14]

The difficulties with holdup measurements were compounded in these assays because the important U-235 γ-rays are low energy photons that are strongly self-absorbed by the component. The four important photons from decay of U-235 range from 143 keV up to 205 keV. We used the 57% branch 185 keV photon for all of our calculations, but frequently used measured ratios of all four to diagnose the transmission characteristics of the component. The Ortec computer code ISOTOPIC represents a much more general and comprehensive use of this technique.[15]

The process equipment holdup data were acquired in all three of the point, line, and area source acquisition configurations. We discuss specific examples in section 3.2 below. In the point source acquisitions the U-235 contents were related by equation (3).

$$[U\text{-}235] = K_p(cps)d^2(Cf_t), \quad (3)$$

where K_p is the point source calibration factor in units of g-sec/cm^2, d is the source to detector distance, and Cf_t is the transmission correction factor determined by a technique identical to that of equation (1). The calibration factor is specific to each detection system and was derived for each detector in references 4 and 5.

In the line source acquisitions the U-235 contents were related by equation (4).

$$[\text{U-235}] = K_l(\text{cps})(d)(Cf_t), \qquad (4)$$

where K_l is the line source calibration factor in units of g-sec/cm^2, d is the source to detector distance, and Cf_t is the transmission correction factor determined in the same manner. The line source calibration factor is also specific to each detection system and was derived for each detector in references 4 and 5. Line source acquisitions were also used to obtain measures of Np-237 and Am-241 holdup in two out-gassing ovens in Building 321-M. We describe the efficiency calibration of the two detector systems separately for these holdup measurements.

In the area source acquisitions the U-235 contents were related by equation (5).

$$[\text{U-235}] = K_a(\text{cps})(A)(Cf_t), \qquad (5)$$

where K_a is the area source calibration factor in units of g-sec/cm^2, A is the surface area of the component observed, and Cf_t is the transmission correction factor determined in the same manner. The area source calibration factor is also specific to each detection system and was derived for each detector in references 4 and 5. The 321-M fuel rod extrusion lathe is an example of a representative process component where we used multiple redundant area source and point source acquisitions. The lathe is shown in Figures 2 and 3.

We also include two acquisition configurations that we developed for special cases that arose in the D&D holdup measurements. We designated these two configurations as the oven source configuration and the cylindrical field configuration. Both of these acquisition configurations were empirical representations of the special case assay situation encountered. However we believe we have developed the calculus that describes the cylindrical field view, and we present that in a separate paper.

2.3 Permanent Assay Station

To conduct the MC&A assays of the HEU residue collected in 2-gallon scrap cans we assembled a permanent fixed geometry assay station that utilized a portable HPGe detection system that was operated by the same Ortec DART system as described above.[16,17] The assay station was set up in Building 324-M, which was contiguous to 321-M. The station was operated in the point source acquisition configuration and performed transmission corrected assays of the 108 scrap cans that were generated by FDD in the D&D operations. The assay station is described completely in references 16 and 17.

The assay station is shown in the photograph of Figure 4. This photo shows the DART system and three personal computers (PC) that we used in the data acquisition and analysis. Using three PCs allowed us to provide on-the-spot assay results. All of the operations of the assay station could have been performed with a single PC, but having three allowed us to run efficiently without continually switching the desktop view of a single PC.

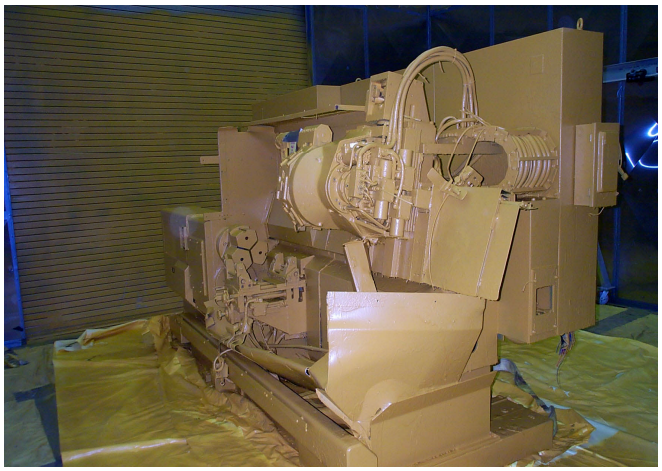

Figure 2. General photo of the 321-M fuel rod lathe, where we obtained point source and area source acquisitions.

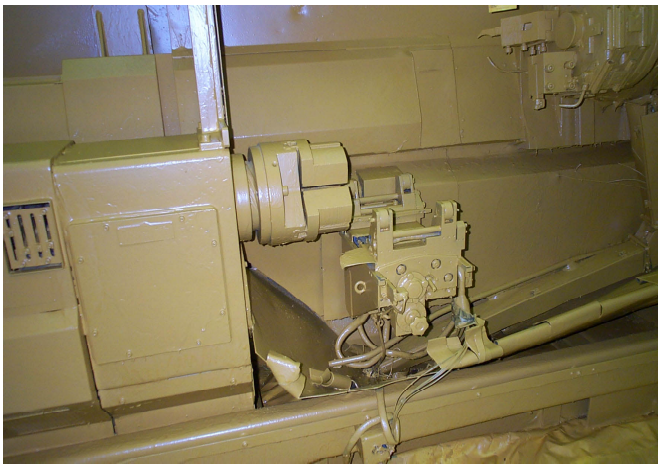

Figure 3. Close-up view of the front face of the lathe. We obtained multiple close coupled area source acquisitions.

Figure 4. A photograph of the permanent assay station in Building 324-M.

The scrap can HEU contents were determined by the same transmission corrected point source equation as above in the holdup measurements. The point source calibration constant of 2.36×10^{-5} g-sec/cm^2 was applied, and the identical technique of shining a source of 4.41 g of U-235 through each sample was applied to determine the transmission correction factor of each can.

Using the permanent assay station in two distinct acquisition configurations, we obtained accountability assays of HEU content in 111 scrap cans, in 170 uranium storage pigs, and in twelve process pressure cookers. We describe these analyses in the RESULTS section below.

3.0 RESULTS

Overall we performed holdup measurements and measurements on recovered residue that yielded a combined value of 1600(300) g of HEU assayed.[18] We also conducted multiple assays that cleared six process components as qualified for free release as non-radioactive. The holdup measurements were on 163 components of the reactor fuel and target processing facility that included the fuel rod lathe, four casting furnaces, two electric motors, three out-gassing ovens, two sawbenches, five truck-sized HEPA filter housing units, and diverse other items of process equipment and exhaust components. In these 163 components we measured a total of 770 g of HEU with an overall uncertainty of approximately 300 g. Using the scrap can assay station we measured 825 g of recovered HEU in the form of filings, sweepings, and residue with an overall uncertainty of 50 g. Finally we have assayed 221 55-gallon drums of high-density solid waste that did not qualify for direct assay by the routine Canberra Q^2 assay instrument. These drums came from waste packaged from the deactivation of 321-M, 313-M, and 323-M. Of the 221, fifteen contained components from depleted uranium processing in 323-M, and seventeen contained low enriched uranium reactor slugs that were assayed to contain a total of 900 g of U-235.[19] These last seventeen drums would have been ideal candidates for assay by thermal active well neutron coincidence counting. The facility would not cooperate with our request to perform those assays.

3.1 Adapted Q^2 Results

Using the adapted Q^2 technique we have assayed or evaluated 221 drums of high-density solid waste that amounts to approximately eighteen metric tons of process components. We do not count the contribution these 221 high density drums made to the total of 1600 g of HEU above. The majority of these drums contained less than 0.50g of HEU and had measurable transmission characteristics. In most cases the measured value by our adapted technique provided a good confirmation of the direct Q^2 assay result. For seventeen drums the adapted technique yielded a measured value near 0.005 g while the direct technique provided an upper limit of content only. The detection limit for the direct technique is generally about 0.01 g of HEU.

Using the adapted technique we identified thirty-one drums that we were unable to assay by either technique. That is, these thirty-one drums had effective infinite thickness for the 185 keV γ-ray in one or more of the three vertical segments. In those cases the direct Q^2 measurement is unable to recognize its own deficiency, and would report an unreliable result. Only by applying the adapted, segmented technique were we able to identify these drums. For each of these drums we informed the waste generator that a neutron activation technique of assay is required. These drums were identified specifically in the technical document in which they were reported.[10] The facility developed an ingenious smear-to-curie technique of reporting HEU holdup for the contents of these drums.[20] This technique of measuring HEU holdup from counting surface smears deserves further review by the radiological control sections of SRS and by the DOE radioactive contamination control community, as its use will simplify all future holdup measurements across the DOE community.

The contents of several of the drums of high density waste received redundant assays using either direct holdup measurements or by using the scrap can assay station. Examples include an electric motor and gearbox from Building 321-M. Both of these items, which had masses of 245 kg and 298 kg respectively, were measured by direct holdup assay using portable NaI and HPGe detection systems. The holdup contents were reported in reference 21. The adapted Q^2 assays of these two items after packaging as high-density waste were reported in reference 22 and were in good agreement with the values reported in 21. Another example where redundant assays

served as excellent confirmation of the adapted Q^2 assays involved components of the fuel rod lathe of Building 321-M.[23] In this example adapted Q^2 results were similarly confirmed by the direct holdup assays using the NaI and HPGe detection systems and by scrap can assays of recovered residue using the Building 324-M HPGe assay station.

The adapted Q^2 technique was also used to assay 110 empty scrap cans for residual HEU content and to assay 170 process pressure cookers for residual HEU content.[24,25] The adapted Q^2 results for these items were tested by assaying individually a selected subgroup of each on the 324-M assay station and comparing the sum with the bulk assay obtained by the Q^2. For these items we were able to compare not only the assay results, but also the measured transmission correction values. Agreement between the two techniques yielded excellent mutual support.

3.2 Holdup Assay Results

Using a combination of γ-ray assays with the portable NaI and portable HPGe detection systems we provided off-line assay results for 163 process components and building exhaust components. The holdup results for these components were reported in references 24 – 33 and resulted in a total of 770(300) g of process HEU identified. A summary of the holdup analyses is provided in Table 2, where we describe some of the interesting components and provide a matrix of γ-ray assay configurations employed for them.

Many of the holdup components were very massive items that had effective infinite thickness for the 185 keV γ-ray that we counted in the assays. For these items we were able to measure surface contamination only. However, as noted above in the Adapted Q^2 Results section, we were often able to obtain a redundant analysis to confirm our results.

For most of the holdup components we were able to measure a correction factor for transmission of the 185 keV γ-ray through the item. We applied this correction factor to area source, line source, and point source acquisition configurations to obtain measured values. For multiple components we were able to obtain data and calculated contents in two of these configurations and to provide analytical results from both. Generally these two results agreed very well. Examples where we used both configurations include the Freon™ cart, the 321-M exhaust elbows and HEPA filter housing units, the C and D out-gassing ovens, and the 321-M cooling hut and HEPA filter housing units.[26-28,31-33]

In the holdup measurements on the Freon™ cart and on the 321-M cooling hut HEPA filter housing unit we identified an interference that required evaluation of a separate correction factor. This interference involves acquisition of data in the area source configuration when a secondary area source contributes to the field of view of the detector. For both the Freon™ cart and cooling hut units we were unable to isolate each area source component we wished to view. Therefore it was necessary to modify the calculation of U-235 content from the traditional area source acquisition to include the interference correction factor Cf_{ar} in equation (6).

$$[\text{U-235}] = K_a(\text{cps})(A)(Cf_t)(Cf_{ar}), \qquad (6)$$

where K_a is the area source calibration constant, A is the total surface area of the component observed, Cf_t is the transmission correction factor, and Cf_{ar} is the correction factor to account for contributions from the secondary source.[34] Using an experimental setup of two planar sources placed in parallel and acquiring data from the primary

planar source in the traditional area source configuration, we determined a correction factor of Cf_{ar} = 0.932(136). Clearly this factor is dependent upon the specific geometry of each component observed. We were able to reproduce our experimental correction factor with a Monte Carlo calculation for the Freon™ cart and for the cooling hut unit. We have good confidence in the correction factor that we measured, and we have developed this technique with much more detailed experimental acquisitions and Monte Carlo calculations.[34,35]

3.2.1 Holdup Assays in a HEPA Filter Housing Exhaust Unit

The cooling hut unit and the HEPA filter housing unit 2968 are excellent examples of the use of redundant area source and point source acquisitions along with application of the secondary source correction factor.[27,33] A photograph of unit 2968 is shown in Figure 5. All of the acquisitions and calculations for unit 2968 are summarized in Table 1. In Table 1, each of the point source acquisitions was interpreted to view the entire component, and so each should be a good measure of the complete HEU content of item 2968. For the point source acquisitions the correction factor was measured experimentally through the entire item (two walls). All four point source acquisitions are in good agreement. In reference 33 we discuss why acquisition 2968-9 is the best representation.

Table 1. HPGe Acquisitions of the 2968 HEPA Bank.

Acquisition	Configuration	Distance (cm)	Area (of surface) (cm)	t (sec)	counts	s	^{235}U Grams
Cs-137 QC	Point source	3.81		60	1137	34	Good check
2968-1A	Area source	137	27000	600	221	28	0.14±0.02
2968-1AT	Area source	137	27000	600	1098	45	Cf=(1.33±.08)
T_0	Point source	262	N/A	60	1540	50	Good check
2968-P1	Point source	635	N/A	800	222	35	3.5±0.6
2968-P2	Point source	399	N/A	600	306	31	2.6±0.3
BKG				8000	814	85	LLD =0.05 cps
2968-1	Area source	61	27000	600	137	24	0.09±0.02
2968-2	Area source	61	39000	600	128	25	0.12±0.03
2968-3	Area source	61	39000	600	1039	41	0.92±0.07
2968-4	Area source	61	13000	600	3221	66	0.96±0.14
2968-5	Area source	69	27000	600	60	26	*< 0.07*
2968-6	Area source	69	39000	600	144	27	0.13±0.03
2968-7	Area source	69	39000	600	1128	44	1.00±0.08
2968-8	Area source	69	13000	600	2773	61	0.82±0.06
2968-9	Point source	399	N/A	600	502	35	4.2±0.4
2968-10	Point source	399	N/A	600	355	31	3.0±0.3

The area source acquisitions in Table 1 were close field views interpreted to measure a portion of the surface contamination on separate faces of item 2968. These contents were calculated with equation (6) using a transmission correction measured experimentally through only one wall of item 2968. The secondary source correction factor of 0.92 was applied in each calculation. Note the sum of the area source measurements should then represent the total HEU content in item 2968. The sum of the ten area source contents in Table 1 yields a total of (4.22 ± 0.20), which is in

outstanding agreement with the point source measurements. Both measures are also in good agreement with the predicted range of contents in exhaust components listed in Table 20-3 of reference 36. Further examples that we do not discuss in detail in this summary report involved assay of HEU, Np-237, and Am-241 on two sawbenches.[37] These involved use of both the NaI and HPGe detection systems in both the point and area source acquisition configurations as well as the NaI contact configuration (see below). Results for the two sawbenches are listed in Table 2.

Figure 5. HEPA unit 2968.

3.2.2 Holdup Assays in the C and D Out-gassing Ovens

A second example where we used redundant measurements from line source and point source acquisitions involves measurements of Np-237, U-235, and Am-241 content in the C and D out-gassing ovens. The horizontal vent line at the bottom of each of these two ovens was an excellent representation of a line source of radioactivity. A photograph of oven C is shown in Figure 6.

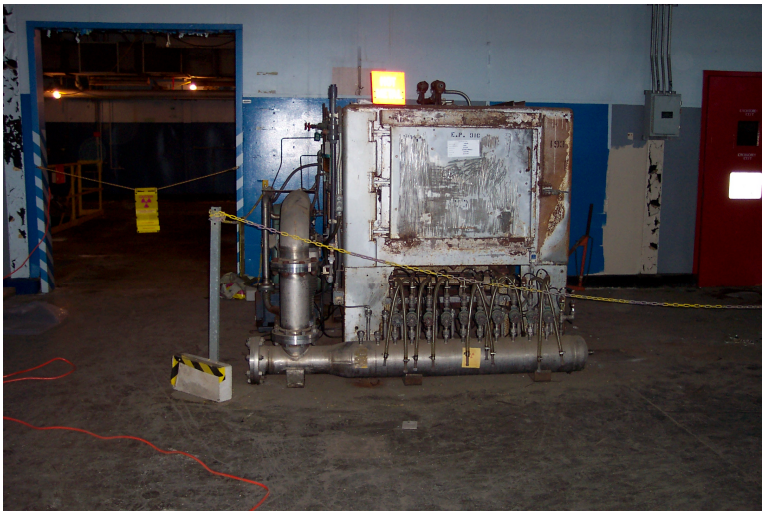

Figure 6. **Photograph of Oven C showing horizontal and vertical vent lines as well as oven chamber with door closed and oil reservoir (left rear).**

For the horizontal vent line of ovens C and D we used a line source configuration to assay for all three species. We obtained four line source acquisitions to confirm the uniformity of the activity from each species. The vertical vent line was short enough that we could obtain close field acquisitions that qualified as line source configurations and a single far field acquisition that qualified as a point source configuration. Results were reported to the facility in reference 32.

Another very important aspect of the assays of out-gassing ovens C and D was that the facility requested the assays and results before we had a chance to efficiency calibrate our detectors for Np-237 or Am-241. We performed a theoretical calculation of the point-, line-, and area-source efficiency calibration factors for both of these species and for both of our detectors from first principles of detection. We described those calculations in a separate paper that received external publication. In that paper we compared our results to experimental measurements we were able to obtain later.[38]

3.2.3 Holdup Assays in Casting Furnaces

We now discuss the holdup assays on four casting furnaces. These four, along with the process extrusion lathe, were the most difficult to assay due to the huge size, unique shapes, and extreme density. For these assay calculations we used a combination of far field point source acquisitions with both detection systems, close field area source acquisitions with both systems, and contact acquisitions with the NaI detection system to yield defensible reported values. For the four casting furnaces we were able to obtain three distinct values of HEU contamination on several of the six faces. These came from viewing the face in a point source and an area source configuration with both detection systems. Referring to references 29 and 30, we obtained measured values with a precision of 40% relative standard deviation in those cases. This is excellent agreement for holdup assay values.

The contact measurements with the NaI system were especially important in the furnace assays, because the well on the top of each furnace represented such a unique shape that no traditional acquisition configuration would fit it. The contact measurements are described in references 29 and 30, and the efficiency calibration of the contact measurement technique is described in reference 5. Figure 7 shows the calibration data that we obtained in a group of contact measurements we made with the NaI detection system. These data were fit with the curve

$$cpm(m) = 1.03 \times 10^5 m/(1 + 0.147m), \quad (7)$$

where m is the mass of the HEU under observation in the contact configuration. The contact measurements formed important components for the assay of the casting furnaces and for the 321-M process lathe.

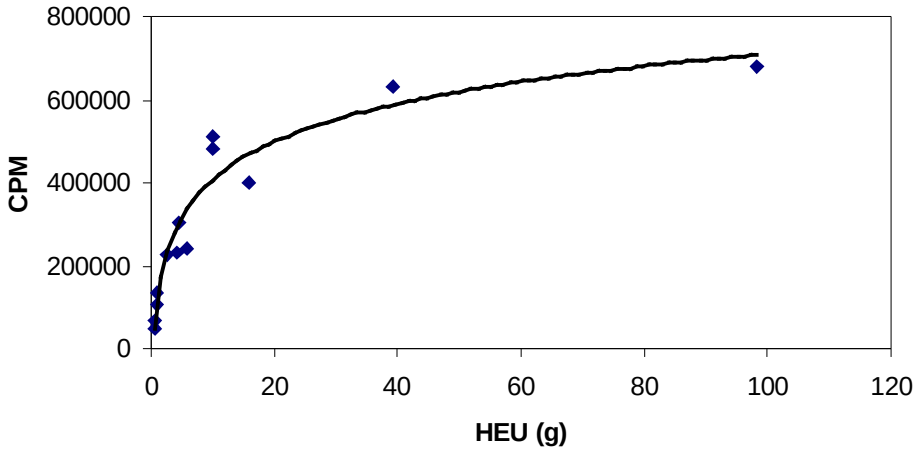

Figure 7. NaI contact configuration calibration curve.

3.2.4 Holdup Assays in F Out-gassing Oven

The holdup assays in the F out-gassing oven presented another unique problem where none of the point, line, or area source configurations could represent the chambers of this oven. A photograph of oven F is shown in Figure 8. Each of the ten chambers in oven F was four feet wide and approximately five feet deep and each was approximately uniformly contaminated with HEU. An extreme advantage for assaying this item was that each oven chamber individually represented an excellent shield of the detector from background radiation and from HEU radiation from an adjacent chamber.

Because of this excellent shielding, we observed that we could acquire the activity from each chamber by inserting a bare NaI detector into the chamber. In this manner we collected the holdup spectrum in each chamber with the detector completely exposed to all five chamber walls. This bare detector acquisition configuration, which had approximately 5/6th's of 4π geometry, we designated the oven source acquisition detector configuration. We describe this acquisition configuration in reference 39 and in Invention Disclosure SRS-04-029.[40]

Figure 8. A photograph of the 321-M F Out-gassing Oven.

We calibrated the bare NaI detector in the oven source configuration using a flexible planar source of dimensions 10" x 20" that we could wrap exactly once around the detector.[41] The source contained activity from Eu-152 that provided useful γ-rays at 121.8 keV and 244.7 keV, which we used to efficiency calibrate for the U-235 γ-ray at 185.7 keV. The equation we developed for using the oven source configuration is

$$[HEU] = K_{ov}(d)(cpm)(L) \quad (8)$$

where K_{ov} is the calibration constant, d is one half the oven chamber width, and L is the oven chamber depth. The NaI oven source calibration factor had a value of 1.132×10^{-6} g-min/in^2, which we compare to the point source value of 1.67×10^{-6}. With the oven source acquisitions we were able to obtain a limit of detection of 0.04 g of HEU with a 10-minute acquisition for each oven chamber. We reported a limit of < 0.4 g HEU in the entire F oven.[39] Using transmission-corrected point source acquisitions would very likely have yielded limits in the 10 – 100 gram range.

3.2.5 Summary of Process Component Holdup Measurements

We believe the redundant values obtained from multiple techniques and from multiple acquisition configurations were very important to lend firm credibility to our holdup results. Development and use of the NaI contact measurement technique and oven source technique and of the secondary area source correction factor will be valuable contributions to future NDA holdup measurements on this site and at other DOE facilities. In almost all cases of reported holdup we were able to compare our results and uncertainties with those predicted for process components in Tables 20-3 and 20-6 of reference 36. Often our redundant measurements and off-line acquisitions allowed us to report defensible uncertainties better than those predicted by Table 20-6. Some examples where we obtained uncertainties better than those of Table 20-6 of reference 36 are the lathe exhaust elbows, the HEPA filter housing units, and the F out-gassing oven.[26-28,39]

3.3 Fixed Geometry Assay Station

The fixed geometry permanent assay station that we set up in Building 324-M and later moved to the billet room of Building 321-M made a very valuable contribution to the material control and accountability measurements. As described in the EXPERIMENTAL section above, the assay station was designed to perform on-the-spot transmission-corrected MC&A assays of recovered HEU residue in the 2-gallon scrap cans. We assayed 108 scrap cans using this device in the point source assay configuration.[16,17] Data were analyzed using the traditional transmission correction technique as well as using the Deming least squares technique that contained the transmission correction within the empirical calibration curve that we described above.

As the decontamination of Building 321-M evolved, we obtained two additional uses for the fixed assay station. One use was to assay 170 uranium storage pigs for residual contamination from U-235.[42] During the production years of 321-M, these pigs were used for storing liquid product HEU. A storage pig is shown in the photograph of Figure 9. The pigs are tall and thin in order to be geometrically critically safe for close packing when full of HEU. Because of their shape we were able to assay them using the transmission corrected line source acquisition configuration and also using an empirical least squares curve derived especially for the pigs. The line source contents used equation (9)

$$[U\text{-}235] = K_l(cps)d(Cf_t), \quad (9)$$

where d is the source to detector distance in cm, and K_l is the line source calibration constant equal to 0.00154 g-sec/cm. K_l is a composite of the line source calibration constant $1.72(6)x10^{-5}$ g-sec/cm^2 and an effective viewing length of 89.30 cm for a line source viewed at 25.5 inches.[42]

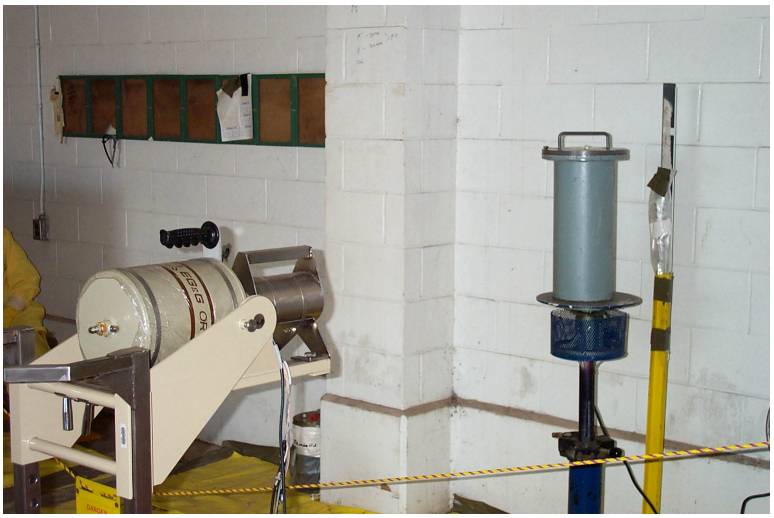

Figure 9. Assay of a uranium storage pig in the point source acquisition configuration. The photograph shows the HPGe detector as well as the transmission source placed behind the pig.

All 170 pigs had been emptied of all but residual contamination, and so most contained quantities very near our detection limit of 0.2 g. In order to improve that limit, we developed a cylindrical shell acquisition configuration that is described in reference 43 and illustrated in Figure 10. By sliding our portable HPGe detector inside of each pig and acquiring data from inside the cylindrical shell, we were able to reduce our detection limit down to 0.0007 g of HEU. We were further able to derive the differential calculus to aptly represent the detection efficiency of the 185 keV γ-ray acquired in this configuration.[43]

In the cylindrical configuration, we derived a U-235 content of

$$[\text{U-235}] = K_{cyl}(\text{cps}) \qquad (10)$$

for each storage pig. K_{cyl} = 0.00222 g-sec was determined empirically for the pigs with the detector inserted an exact and reproducible distance.

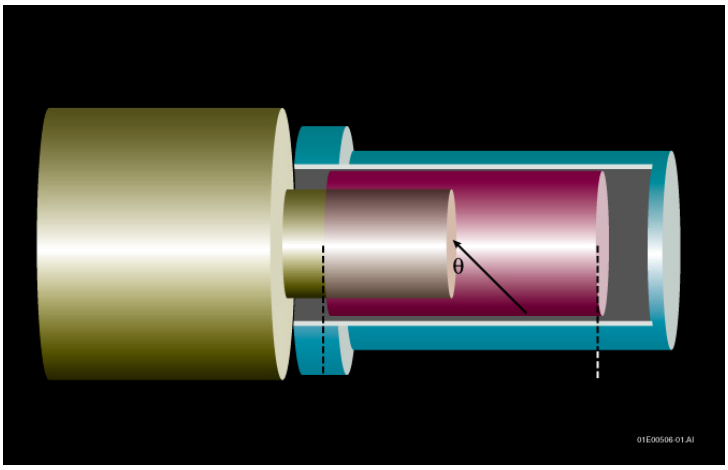

Figure 10. Drawing of the cylindrical shell acquisition as described in text.

Another valuable use of the permanent assay station was for determination of HEU content in 110 process pressure cookers.[25] One of these pressure cookers is shown in the photograph of Figure 11. We assayed twelve of these cookers using the point source transmission corrected acquisition configuration and then stacked nine of those twelve in a close packed arrangement into a 55-gal drum. We assayed the contents of this drum using the adapted Q^2 technique to confirm that the measured value obtained was a reliable measure of the HEU content. Using the assay station each of the twelve pressure cookers was determined to contain residual HEU contents near 0.10 g, and the nine selected for the 55-gallon drum contained 0.94±0.14 g. After stacking the group of nine together the sum was measured by the adapted Q^2 instrument to contain 0.76±0.21 g of HEU. With this acceptable agreement we continued to assay the remaining 98 pressure cookers in batches of nine stacked into 55-gallon drums.

Figure 11. Photograph of a pressure cooker on the 324-M billet assay station turntable.

4.0 Conclusion

We have used three instruments to obtain γ-PHA acquisitions to perform holdup and recovered residue assays of highly enriched uranium content in the deactivation and decommissioning of the 321-M Fuel Fabrication Facility at the Savannah River Site. The assays have supported D&D activities that were designated as award fee milestones by the DOE customer for fiscal years 2000 and 2001, and the assay development work and results have yielded two Site Vice President's awards for the authors. In this paper we report six technical developments and innovations for HEU assays of holdup and recovered residue.

As always, the holdup assays in this project did not fit the traditional point, line, and area source acquisition configurations, and so it was necessary to use creative modeling and to develop new techniques to obtain defensible assay results. Holdup assays were performed on items that ranged in size and density of a kitchen pressure cooker up to the 321-M process lathe that is larger than a pickup truck. In general we were able to obtain transmission corrected acquisitions in both the point and area source configurations with both NaI and HPGe detection systems. Frequently we were also able to obtain contact measurements with the shielded NaI detector. We obtained specific efficiency calibration of the NaI detector for contact measurements that have a field of view that we could measure exactly. Combining the NaI contact measurements with the NaI and HPGe point and area source acquisitions and with Q^2 acquisitions we were able to obtain up to six redundant measurements of each component. When these measurements agreed, we were able to report defensible values of HEU holdup. When they did not agree, we used this as a diagnostic tool for further development or modeling.

Another technical development that we applied to holdup assays involved assays of HEPA filter housing units from the exhaust system of Building 321-M. Transmission corrected assays in the area source configuration of these items required correction for contributions from secondary sources. Since we were not always able to isolate the primary area source that we were counting it was necessary to correct the measured data to remove contributions from secondary area sources. We experimentally measured those contributions using two planar area sources available to us, and then modeled those contributions using the MCNP Monte Carlo computer code.

A third technical development involved adaptation of the commercial Q^2 solid waste assay instrument to a three-segment segmented gamma scanner for assay of high density solid waste. A fourth technical contribution was to develop a cylindrical shell acquisition configuration for assays of uranium storage pigs. This acquisition configuration allowed us to obtain holdup measurements with a minimum detection limit as low as 0.0007 g U-235. A fifth technical contribution was to develop an oven source acquisition configuration to allow holdup measurements with a minimum detection limit as low as 0.04 g of U-235 in a massive outgassing oven. In separate outgassing oven measurements we demonstrated calculation of our detector calibration constants using first principles of detector calibration. These calculations obtained external publication in the Journal of Radioanalytical and Nuclear Chemisty.

We implemented two significant innovations in our material control and accountability assays with a fixed geometry assay station. One innovation involved use of a least squares fit model to obtain a separate measure of recovered HEU residue in scrap cans. Since the geometry and recovered material were both fairly constant, a measure of the transmission factor of a sample was directly related to the HEU content. Therefore we were able to predict content directly from a least squares fit to the measured counts per second. This provided a second measure redundant to the transmission-corrected measure obtained in the traditional point and line source configuration that we used.

Finally we implemented a technique to provide on the spot reporting of HEU content using our two redundant measurements. By using three personal computers that contained separately the acquisition software, the least squares fit curve, and an Excel spreadsheet to perform the transmission corrected content calculations, we were able to report measured HEU content and uncertainty immediately to the customer.

Table 2. A list of some of the 163 holdup components assayed, plus a matrix of configurations used (e.g. point source, area source, contact, surface only, transmission corrected), and assay result.

Process Component	item #	NaI area	NaI point	NaI contact	HPGe area	HPGe point	Q2 Assay	Surface Activity Only?	HEU Content
Wrightway hoist EP20609	1806		X						< 0.2
Water Cooler Capacitor	1807		X						< 0.2
Process water-cooled blue cylinder	1936		X						< 0.2
Yellow Transformer	1935					X			< 0.2
Black Transformer	1937		X						< 0.2
Red Transformer	1938					X			< 0.2
HEPA filter	1939		X		X				< 0.2
Black Transformer	1940					X			< 0.2
2-ton CM hoist	1808					X			< 0.2
Electrical box 20172A	1941					X			< 0.2
Electrical box 20172B	1942	X							< 0.2
Casting Guard	1943		X						< 0.2
Crucible Heating Coil (1)	1944					X			< 0.2
Crucible Heating Coil (2)	1985					X			< 0.2
Crucible Heating Coil (3)	1986		X						< 0.2
Crucible Heating Coil (4)	1987					X			< 0.2
Crop Shear	1888					X		X	0.3 < ^{235}U < 1.2
Oil reservoir	1889		X						0.2 < ^{235}U < 0.46
Motor M28619	1890				X		X	X	15 < ^{235}U < 61
Gear box	1891		X			X	X	X	6 < ^{235}U < 25
Furnace A		X		X	X	X		X	3 < ^{235}U < 13
Furnace B	1893	X	X	X	X	X		X	13 < ^{235}U < 51
Furnace C		X		X	X	X		X	3 < ^{235}U < 10
Spare Furnace	1894	X	X	X	X	X		X	17 < ^{235}U < 67
Lathe		X	X	X	X	X	X	X	129(31) gram found
Freon Cart		X				X			77(20)
Sawbench 1		X	X	X					0.164(3)
Sawbench 2		X	X	X					< 0.4

Process Component	item #	NaI area	NaI point	NaI contact	HPGe area	HPGe point	Q^2 Assay	HPGe Line	HEU Content
Exhaust HEPA filter component	2624	X				X			16(3)
Exhaust HEPA filter component	2629	X	X			X			10(2)
Exhaust HEPA filter component	2630		X			X			13(2)
Exhaust HEPA filter component	2637	X				X			13(5)
Exhaust HEPA filter component	2638					X			6(1)
Exhaust HEPA filter component	2639					X			8(1)
cooling hut HEPA filter component	20906		X		X	X			17(12)
Exhaust elbow	2612	X				X			1.00(12)
Exhaust elbow	2613	X				X			0.91(16)
Exhaust elbow	2614	X				X			1.98(42)
Exhaust elbow	2615	X				X			1.66(31)
Exhaust elbow	2620	X				X			1.56(27)
Exhaust elbow	2621	X				X			1.17(13)
Exhaust elbow	2622	X				X			2.24(17)
Exhaust elbow	2623	X				X			3.50(26)
Drawbench						X			< 5
Straightener						X			< 5
Fluoroscope						X			< 5
C outgassing oven						X		X	< 0.2
D outgassing oven	2966	X	X			X		X	0.105(48)
Sawbenches		X	X	X					
Cooling hut		X			X	X			17(12)
HEPA filter hut	2968				X	X			4.18(14)
HEPA filter hut	2969				X	X			5.0(17)
HEPA filter hut	2970				X	X			4.72(17)
HEPA filter hut	2911				X	X			6.84(10)
F outgassing oven	**Unshielded NaI Oven Source Acquisitions**								< 0.4
Gate Valve	2961				X	X			< 0.4

Process Component	item #	NaI area	NaI point	NaI contact	HPGe area	HPGe point	Q^2 Assay	HPGe Line	HEU Content
Gate Valve	2984				X	X			0.54(19)
Gate Valve	2985				X	X			0.85(28)
Gate Valve	2986				X	X			0.78(6)
Gate Valve	2990				X	X			< 0.04
Uranium Storage Pigs		**Cylindrical Shell Acquisitions**				X	X	X	All < 0.1
Pressure cookers						X	X	X	10.8(6)
Exhaust Component	2945				X	X			< 0.4
Exhaust Component	2946				X	X			1.42(42)
Riser Crusher					X	X			18(18)

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