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^{235}U Holdup Measurements in Three 321-M Exhaust HEPA Banks

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ABSTRACT

The Analytical Development Section of Savannah River National Laboratory (SRNL) was requested by the Facilities Disposition Division to determine the holdup of enriched uranium in the 321-M facility as part of an overall deactivation project of the facility. The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. The results of the holdup assays are essential for determining compliance with the Waste Acceptance Criteria, Material Control & Accountability, and to meet criticality safety controls. This report covers holdup measurements of uranium residue in three HEPA filter exhaust banks of the 321-M facility. Each of the exhaust banks has dimensions near 7' x 14' x 4' and represents a complex holdup problem. A portable HPGe detector and EG&G Dart system that contains the high voltage power supply and signal processing electronics were used to determine highly enriched uranium (HEU) holdup. A personal computer with Gamma-Vision software was used to control the Dart MCA and to provide space to store and manipulate multiple 4096-channel γ-ray spectra. Some acquisitions were performed with the portable detector configured to a Canberra Inspector using NDA2000 acquisition and analysis software. Our results for each component uses a mixture of redundant point source and area source acquisitions that yielded HEU contents in the range of 2 – 10 grams. This report discusses the methodology, non-destructive assay (NDA) measurements, assumptions, and results of the uranium holdup in these items. This report includes use of transmission-corrected assay as well as correction for contributions from secondary area sources.

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

^{235}U Holdup Measurements in Three 321-M Exhaust HEPA Banks

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1. INTRODUCTION

The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and other components for the production reactors. It operated for over 35 years. During this time thousands of uranium-aluminum-alloy (U-Al) 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). [1]

^{235}U holdup measurements were performed in 1995 and documented in technical report WSRC-TR-95-0492.[2] The holdup values reported in WSRC-TR-95-0492 were best estimates only, due to lack of time for conducting the measurements and analyses. Therefore, Facility Decontamination Division (FDD) requested technical assistance from the Analytical Development Section (ADS) of the Savannah River National Laboratory (SRNL) to determine the holdup of enriched uranium in the 321-M facility as part of an overall deactivation project of the facility.[3] This project includes the dismantling and removal of all held-up highly enriched uranium (HEU) to the extent practical. ADS was tasked to conduct holdup assays to quantify the amount of HEU on all components removed from the facility prior to placement 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.[4] This limit was imposed to meet criticality requirements of the E-Area Low Level Vaults. Thus the holdup measurements are used as guidance to determine if further decontamination of equipment is needed to ensure that the quantity of U-235 does not exceed the 50 g limit. In summary, the results of the holdup assays are essential for determining compliance with the Waste Acceptance Criteria, Material Control & Accountability, and to ensure that criticality safety controls are not exceeded.

This report covers holdup measurements of uranium residue in three 321- M exhaust high efficiency particulate air (HEPA) filter housing units. Because holdup values are extremely difficult to determine, conservative assumptions are usually made to report the U-235 gram values. Relative uncertainties for this kind of measurements are generally quoted as +100% and –50%.[2] Our results indicated that each of the HEPA banks contains 2 - 10 g of enriched uranium. We discuss the acquisitions, calculations, and uncertainties for each unit in the **EXPERIMENTAL** and **CALCULATIONS** sections below. For each of the three exhaust banks we use a combination of point source and

area source γ-ray acquisitions that were developed and taught by the Los Alamos Safeguards Technology Training Program.[5] For the area source acquisitions we include an additional correction factor of our own that is unique to γ-ray acquisitions for bulk material of these types.[6]

2. EXPERIMENTAL

A portable high purity germanium (HPGe) detector system was used to conduct nondestructive assay (NDA) measurements of HEU holdup on the three HEPA filter housing units 2968, 2970, and 2911. The detector system uses an EG&G Dart package that contains a high voltage power supply and signal processing electronics. A personal computer with Gamma-Vision acquisition software was used to provide space to store and manipulate multiple 4096-channel γ-ray spectra. This system is described in reference 7 and has been used extensively in HEU holdup measurements for FDP. Some of the HPGe spectra were acquired using the Canberra Inspector system and NDA2000 software. The Dart and Canberra acquisition systems yield identical spectra that are analyzed in almost identical manners. While the two systems are not interchangeable, the results are in excellent mutual agreement.

Photographs of HEPA filter housing unit 2968 are shown in Figures 1 and 2. Units 2970 and 2911 look very similar. We do not include photos of them. Unit 2968 is approximately a rectangular prism with dimensions 7' x 12' x 4'. The acquisitions for HEPA hut 2968 are summarized in Table 1. We obtained thirteen HPGe acquisitions. Five acquisitions were in the far-field configuration from a range of 13 feet or more. One of the acquisitions was a Cs-137 source check acquisition,[8] one was a U-235 source check acquisition, and one (BKG) was a background. A U-235 component spectrum of item 2968 is shown in Figure 3, and a component spectrum of item 2970 is shown in Figure 4. The acquisitions obtained from 24 – 54 inches were treated as area source configurations in which the detector field of view was always less than the entire surface area of the component observed. The detector field of view is documented in reference 7. When the effective field of view is less than the total surface area of the uniformly contaminated component observed, we have a valid area source acquisition configuration.

The acquisitions of unit 2968 are sketched on page 164 of NB-2000-00086.(9) We do not reproduce that sketch here, but note that acquisitions 2968-1 and 2968-5 approximately represent the same component of surface area taken at two different spots on that component. Likewise acquisitions 2968-2 and 2968-6 view the same component, as well as acquisitions (2968-3 and 2968-7) and (2968-4 and 2968-8). These four pairs of area source acquisitions demonstrate that each of the components was individually fairly uniformly contaminated with HEU. The four pairs also demonstrated to us where the bulk of the HEU contamination was on HEPA 2968. Almost all of the observed content was uniformly distributed near the top of the unit in acquisitions 2968-3, 2968-4, 2968-7 and 2968-8. The eleven area source measurements in Table 1 should then be summed to approximately represent *two times* the total HEU content on the front face of unit 2968.

Figure 1. The front face of the HEPA filter housing unit 2968. This photo also shows an HPGe acquisition in progress.

Spectrum BKG is the background spectrum associated with all of the component spectra. We used spectrum BKG to determine the limit of detection in units of cps for each of the subsequent measurements. The limit of detection is defined as 4.65 times the standard deviation of the background.[10] We apply this LLD in the **CALCULATIONS** section.

$$LLD = \{2.71 + 4.65 \times \sigma(BKG)\}/T = \{2.71 + 4.65(85)\}/8000 = 0.05 \text{ cps.} \quad (1)$$

We discuss application of all of the point source and area source measurements for item 2968 in the **CALCULATIONS** section. For items 2970 and 2911 we present a synopsis of the results. All of the calculations are very similar.

Figure 2. One side of HEPA filter housing unit 2968.

Table 1. HPGe Acquisitions of the 2968 HEPA Bank.

Acquisition	Configuration	Distance (inch)	Area (of surface) (cm)	t (sec)	counts	σ	^{235}U Grams
Cs-137 QC	Point source	1.5		60	1137	34	Good check
2968-1A	Area source	54	27000	600	221	28	0.14±0.02
2968-1AT	Area source	54	27000	600	1098	45	Cf=(1.33±.08)
T_0	Point source	103	N/A	600	1540	50	Good check
2968-P1	Point source	250	N/A	800	222	35	3.5±0.6
2968-P2	Point source	157	N/A	600	306	31	2.6±0.3
BKG				8000	814	85	LLD =0.05 cps
2968-1	Area source	24	27000	600	137	24	0.08±0.02
2968-2	Area source	24	39000	600	128	25	0.12±0.03
2968-3	Area source	24	39000	600	1039	41	0.92±0.07
2968-4	Area source	24	13000	600	3221	66	0.96±0.14
2968-5	Area source	27	27000	600	60	26	< 0.07
2968-6	Area source	27	39000	600	144	27	0.13±0.03
2968-7	Area source	27	39000	600	1128	44	1.00±0.08
2968-8	Area source	27	13000	600	2773	61	0.82±0.06
2968-9	Point source	157	N/A	600	502	35	4.2±0.4

2968-10	Point source	157	N/A	600	355	31	3.0±0.3

Table 2. HPGe Acquisitions of the 2970 HEPA Bank.

Acquisition	Configuration	Distance (inch)	Area (of surface) (cm)	t (sec)	counts	σ	^{235}U Grams
Cs-137 QC0506	point source	1.5		60	1205	36	Good check
2970-1	point source	191		1000	471	42	3.50±0.31
2970-2A	Area source	31	27000	600	64	25	0.03±0.02
2970-3A	Area source	20	6000	400	980	39	0.21±0.01
2970-4A	Area source	19	27000	400	2026	61	1.85±0.08
BG0506	N/A	N/A	N/A	1080	0	12.25	LLD =0.056 cps
2970-5	point source	109	N/A	600	1281	51	5.16±0.21
2970-6	point source	109	N/A	600	1033	41	4.16±0.17
2970-7	point source	109	N/A	180.8	73	16	0.98±0.21
T_0	point source	39	N/A	152.88	2701	54	4.09±0.08
					Twice Summed Area Source HEU Content		**4.72±0.17**

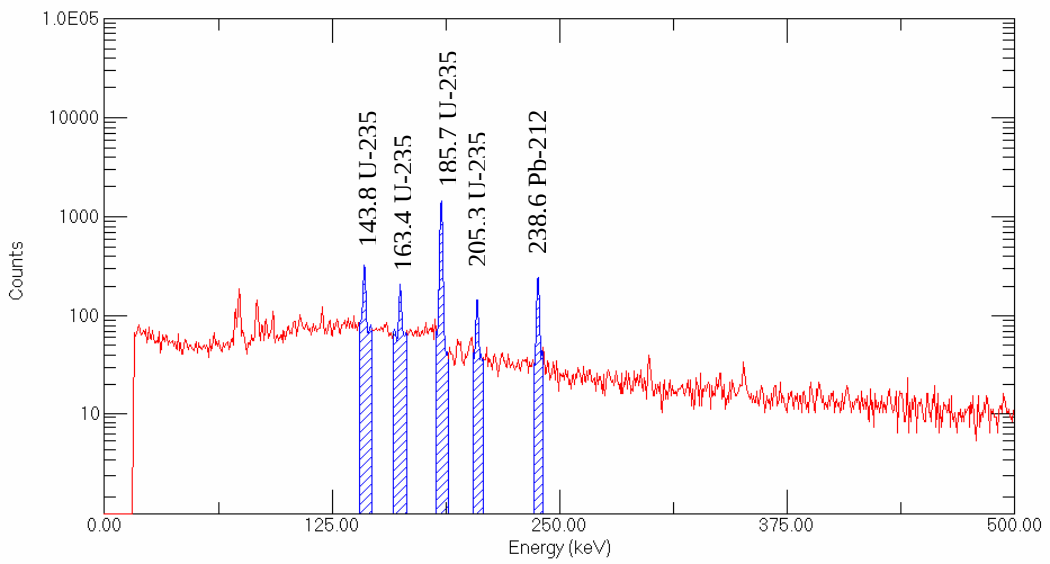

Figure 3. HPGe spectrum 2968-3 of Table 1.

Table 3. HPGe Acquisitions of the 2911 HEPA Bank.

Acquisition	Configuration	Distance (cm)	Area (of surface) (cm)	t (sec)	counts	σ	^{235}U Grams
Cs-137 QC0501	point source	1.5		60	1255	36	Good check
2911-1P	point source	41		600	2822	65	1.59±0.04
2911-1A	Area source	12	34000	600	2822	65	2.20±0.09
2970-2A	Area source	12	18000	600	2659	61	1.09±0.04
2970-3A	Area source	12	13000	600	444	34	0.13±0.01
BG0501	N/A	N/A	N/A	3000	95	8	LLD =0.013 cps
T_0	point source	40	N/A	100	1695	44	4.13±0.11
					Twice Summed Area Source HEU Content		**6.84±0.10**

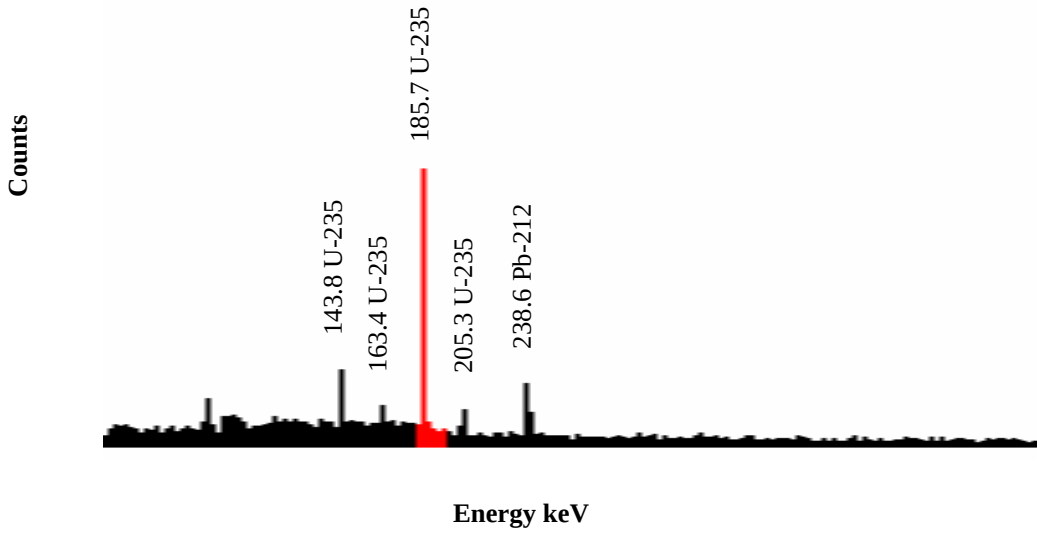

Figure 4. HPGe spectrum 2970-6 of Table 2.

3. CALCULATIONS

Hut 2968

The holdup in each individual point source measurement observed in the HPGe measurements was determined from equation (2) and is listed in Table 1.

$$[HEU] = (2.36\times10^{-5})(cps)(d)^2(Cf_T), \quad (2)$$

where the first factor is the point source calibration constant of reference *7* in units of

g-sec/cm^2, cps is the measured detection rate in the 185-keV peak, d is the source-to-detector distance to the center of the HEPA hut in cm, and Cf_T is the transmission correction factor. Each of the four point source acquisitions was obtained at a distance great enough to assume the entire HEPA component was in full view. Each was taken at a different detector height to the floor, and so each had a slightly different field of view.

The holdup or surface contamination for each of the area source acquisitions of the HEPA filter bank was determined using equation (3) from the HPGe acquisitions in Table 1.

$$[\text{HEU}] = (1.29\text{x}10^{-5})(\text{cps})(Cf_T)(\text{A})(Cf_{sec}), \qquad (3)$$

where the first factor is the area source calibration constant of reference 7 in units of g-sec/cm^2, cps is the measured detection rate in the 185-keV peak, Cf_T is the transmission correction factor, A is the total surface area of each of the components, and Cf_{sec} is the secondary source correction factor that we describe in references 6 and 11. The secondary source correction factor is necessary to correct for the contribution made by the radiation coming from the opposite side of the surface being observed. We describe that calculation below. The secondary source correction factor is defined in Moore and Dewberry[6] and is approximately 0.92(3) for these acquisitions.

In addition to the QC check prescribed in reference 8, the system was QC checked using the U-235 standard source wt2025a that has an HEU content of 4.41 g. Acquisition T_0 in Table 1 was a point source acquisition obtained at a source to detector distance of 103 inches, as documented on page 164 of reference 9. The measured content for the source check yielded

$$[\text{HEU}] = (2.36\text{x}10^{-5})(\text{cps } T_0)(103\text{x}2.54)^2 = 4.15(17)\text{ g}, \qquad (4)$$

which is in satisfactory agreement with the known value.[12] This acquisition represents a second QC check of the system as well as the bare source acquisition required for the transmission correction calculation.

To determine the transmission of the 185 keV γ-ray through hut 2968, we obtained the acquisitions labeled 2968-1A, 2968-1AT, and T_0 in Table 1. Transmission is measured directly by

$$T = \{\text{cps}(2968\text{-}1\text{AT}) - \text{cps}(2968\text{-}1\text{A})\}/\text{cps}(T_0) \qquad (5)$$

$$= \{1.830 - 0.368\}/2.567 = 0.569.$$

The correction factor is then taken to be $(1/T)^{1/2} = 1.325$. We used this value for all of the measurements on hut 2968. Similar transmission measurements were made for huts 2611 and 2970. We do not describe the calculation of the correction factor for them.

The point source acquisition 2968-P1 was obtained from a distance of 250 inches and from a height of 30 inches off of the floor. Using equation (2) and with a Cf_T of 1.325, we obtain a measured content of

$$[HEU] = (2.36\times10^{-5})(cps)(d)^2(Cf_T)$$

$$= (2.36\times10^{-5})(222/800)(635)^2(1.325) = 3.5 \pm 0.6 \text{ g.}$$

The uncertainty represents a propagation of the uncertainty in all four spectra 2968-1P, 2968-1A, 2968-1AT, and T_0. The remaining three point source acquisitions in Table 1 were calculated in the identical manner. Each should be a fairly good representation of the HEU content of unit 2968.

The content of the component of HEPA unit 2968 observed in the area source acquisition 2968-1 was calculated by equation (3).

$$[HEU] = (1.29\times10^{-5})(cps)(Cf_T)(A)(Cf_{sec}), \quad (3)$$

$$= (1.29\times10^{-5})(137/600)(1.15)(27000)(0.92) = 0.08 \pm 0.02.$$

The total uncertainty comes again from propagation of the uncertainty in the three spectra required for the transmission correction as well as the uncertainty in acquisition 2968-1. The uncertainty in the area source calculations also includes the 3/92 uncertainty in the secondary source correction factor.

Note the transmission correction factor is only 1.15 for the area source acquisitions. The transmission calculated in (5) above was for transmission through both walls of the HEPA hut. For the area source acquisitions we assume we are observing the activity through only one exterior wall. Therefore the area source correction factor is then the square root of the point source correction factor. We use $Cf_T = 1.15$ for the area source acquisitions.

The surface area in the fourth term of (3) is determined for each component observed in the acquisition. For acquisition 2968-1 we were observing a 4' x 86" section, thus $A = 4 \times 12 \times 2.54 \times 86 \times 2.54 = 27000$ cm^2. At a distance of 24 inches, the field of view of the detector is approximately 6900 cm^2, and so this acquisition clearly qualifies as an infinite area source configuration.

The remaining ten area source acquisitions were calculated in the identical manner. Taken together they should approximately represent a sum of the total HEU content in HEPA hut 2968. We have acquisition (2968-5) in Table 1 for which we measured less than the detection limit of HEU. Our limit of detection in units of cps was described in equation (1) above and is 0.05 cps for an 8000 second count. For acquisition 2968-5 we obtain a limit of detection of

$$LLD(HEU) = (1.29\times10^{-5})(LLD_{cps})(Cf_T)(A)(Cf_{sec}), \quad (4)$$

where LLD_{cps} for this 600 second acquisition is obtained from

$$LLD(t)_{cps} = (0.05)SQRT(8000/t) = (6)$$

$$= (0.05)SQRT(8000/600) = 0.183 \text{ cps.}$$

Therefore LLD(2968-5) is

$$LLD(HEU) = (1.29x10^{-5})(0.182)(Cf_T)(A)(Cf_{sec}) = 0.07 \text{ g. } (4)$$

Recall we noted in the **EXPERIMENTAL** section that the eleven area source measurements represent twice the sum of the HEU content on the front face. These area source measurements represent only the front face of the hut (unlike the point source measurements, which represent the entire hut). The area source acquisitions represent only the front face, and to be consistent with that assumption, we corrected for transmission through only one exterior wall, and we corrected for contributions from the backside of the hut using Cf_{sec}.

Since we have removed the contributions from the opposite side, it is necessary to add them back in. We do that by assuming the opposite side to be very nearly identical to the front with respect to HEU content. Thus, after dividing the sum of the eleven measurements by two to account for double counting the four components, we go back and double the sum to account for contributions from the opposite face of the hut. Using this result as the whole HEU content of the hut disregards contributions from the two sides. We believe this is a reasonable approach.

Finally we note that two times half the sum of the eleven area source acquisitions equals (4.18 ± 0.20) g of HEU. The uncertainty here comes from the quadratic sum of the uncertainties in the eleven area source measurements in column eight of Table 1. This is in outstanding agreement with the average of the point source acquisitions, which is (3.3 ± 0.7). To be further analytical, we note that the point source acquisition 2968-9 was taken from a height of 104" and clearly is the most representative point source view of where the bulk of the HEU content was observed in the area source acquisitions. The calculated value of (4.2 ± 0.6) for this point source acquisition is in even better agreement with the area source sum. We reported an HEU content of 1.5 – 6 g in HEPA unit 2968.[13]

The γ-ray acquisitions and results for HEPA filter housing unit 2970 are listed in Table 2. Note three of the four point source acquisitions again yield measured HEU contents in good mutual agreement. Excluding acquisition 2970-7 we obtain an average point source content of (4.27 ± 0.84) g. Spectrum 2970-7 was taken of the lowest portion of item 2970. Like the acquisitions for item 2968, the area source configurations demonstrated that the bulk of the HEU contamination was near the top of the item. The summed area source measurements indicate a holdup content of (4.72 ± 0.17) g of HEU. We reported a total content of 3 – 10 g to the facility.[13]

The γ-ray acquisitions and results for HEPA filter housing unit 2911 are listed in Table 3. We obtain only three acquisitions of this item that had dimensions 98" x 82" x 54". Acquisition 2911-1 was treated as both a point source and an area source in two separate calculations. We include that calculation in Table 3, but note that 2911-1P is not an applicable measure of the HEU content for item 2911. The source to detector distance was not sufficiently far field to qualify as a point source acquisition. The three close field area source acquisitions yield a summed content of (6.84± 0.10) g. Excluding the point source measurement, we reported a total content of 2 – 9 g to the facility.[13] All three reported values are in excellent agreement with the expected contents for process exhaust units that is listed in Table 20-3 of reference 5.

4. CONCLUSION

We have performed three distinct γ-PHA measurements of the ^{235}U holdup content in three 321-M exhaust HEPA filter housing units. One measurement was a summed set of eleven area source acquisitions combined with a set of four point source acquisitions. The second measurement was a summed set of four HPGe area source acquisitions. We provide detailed calculations for these measurements on item 2968. We show excellent agreement between the two sets of results. For the second unit, item 2970, we obtained seven measurements in the area source configuration and one measurement in the point source configuration. Again the two sets yielded results in excellent agreement. For the last unit, item 2911, we obtained three area source acquisitions and one confirming point source acquisition. All measurements used transmission correction in the calculations. The reported contents for all three items were obtained with approximately 8% precision and were all approximately 5 g in total content.

Our discussion of results included use of the secondary source correction factor for each of the area source acquisitions. This correction factor derives from the finite width of each of the HEPA filter housing units. Each area source acquisition was set up to observe the surface contamination of one wall of the unit. However each unit then has an opposite face wall that contributes to the observed radiation and that we were not able to shield out of the observation. We removed this "secondary source" contribution using the correction factor derived and measured in a previous technical report.

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