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HEU Holdup Measurements in 321-M Freon Cart

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ABSTRACT

The Analytical Development Section of SRTC was requested by the Facilities Disposition Division (FDD) 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 facility also includes the 324-M storage building and the passageway connecting it to 321-M. The results of the holdup assays are essential for determining compliance with the solid waste Waste Acceptance Criteria, Material Control & Accountability, and to meet criticality safety controls. Two measurement systems were used to determine highly enriched uranium (HEU) holdup. One is a portable HPGe detector and EG&G Dart system that contains high voltage power supply and signal processing electronics. A personal computer with Gamma-Vision software was used to provide an MCA card, and space to store and manipulate multiple 4096-channel γ-ray spectra. The other is a 2"x 2" NaI crystal with an MCA that uses a portable computer with a Canberra NaI+ card installed. This card converts the PC to a full function MCA and contains the ancillary electronics, high voltage power supply and amplifier, required for data acquisition. This report covers holdup measurements in an ultrasonic cleaning Freon™ cart. Our results indicated a total amount of enriched uranium in the cart, excluding the cart pump, was 77±20 grams. This report will discuss the methodology, Non-Destructive Assay (NDA) measurements, assumptions, and results of the U holdup in the Freon™ cart.

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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 miscellaneous components for the production reactors. The facility also includes the 324-M storage building and the passageway connecting it to 321-M. The facility operated for 25 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 residual is located in the equipment that generated and handled small U-Al particles and the exhaust systems for this equipment (e.g., Chip compactor, log saw, lathes A & B, cyclone separator, Freon™ cart, riser crusher, ...etc). [1]

U-235 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 only best estimates, due to lack of availability of time for conducting the measurements and analysis. Therefore, FDD has requested technical assistance from the Analytical Development Section (ADS) of the Savannah River Technology Center (SRTC) 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 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 a 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 solid waste Waste Acceptance Criteria, Material Control & Accountability, and to ensure that criticality safety controls are not exceeded.

This report discusses the methodology, NDA measurements, assumptions, and results for the holdup in a Freon™ cart. Since the 1995 assay indicated that the cart contained 32±32 g of U-235 holdup, a more precise value was required to determine disposition with respect to the 50 g limit. Freon™ 113 was used to carry out ultrasonic cleaning of the cores (machined hollow cylinders of U-Al) in the Freon™ cart. The cart is made of stainless steel and the dimensions of the main body are 21"x21"x24". The catch pan dimensions are 21"x21"x8".

2. EXPERIMENTAL

We used two measurement systems for determining the quantity and approximate location of the HEU holdup in the Freon™ cart. One is a portable HPGe detector and EG&G Dart system that contains high voltage power supply and signal processing electronics. A personal computer with Gamma-Vision software was used to provide an MCA card, and space to store and manipulate multiple 4096-channel γ-ray spectra. The other is a 2"x 2" NaI crystal with an MCA that uses a portable computer with a Canberra NaI+ card installed. This card converts the PC to a full function MCA and contains the ancillary electronics, high voltage power supply and amplifier, required for data acquisition[4].

NaI System

In order to calibrate the NaI system, we acquired spectra from fourteen U-235 standards, ranging in mass from 0.65 g up to 98.53 g, at a distance of 8" from the detector face. The source to detector distances did not include source thicknesses, which for some sources ranged up to about 1.5". The spectra were acquired using the Canberra NaI+ multichannel analyzer with Genie 2000 software for control of the acquisition parameters and storage of spectra. The spectra were acquired in the energy range 0 – 2 MeV with an ADC gain of 1024 channels and with the detector high voltage set to +800V. Only sample self absorption limited the interaction of the 185 keV γ-ray from U-235 with the detector. For the NaI detector measurements we used a distance of 8" from the cart to the detector face. Measurements were taken at the positions shown in Figure 1. This system was used to determine the approximate location of the HEU holdup.

HPGe Far Field System

The high-resolution, portable, high purity germanium (HPGe) detector was positioned 122 cm from the center of the Freon™ cart. The far-field counting system was previously calibrated for measurement of the U-235 content of material in M-Area[5]. Since our NaI measurements showed that the highest activity readings were obtained from the catch pan on the lower portion of the cart, transmission measurements were done on the catch pan. In order to determine the transmission correction, a standard containing about 100 g of U-235 was counted behind the two sides of the catch pan. Figure 2 shows the HPGe far field arrangement. The catch pan is made of 0.125-inch stainless steel. The cart was also counted in the exact same positions without the source, and the source was counted in the same position without the cart. Transmission measurements were not done on the front or back of the catch pan because of attenuation from the pump, filter and cart handle support. This system was used to determine the quantity of the HEU holdup.

Figure 1. Positions of the Freon™ Cart Measured with NaI Detector

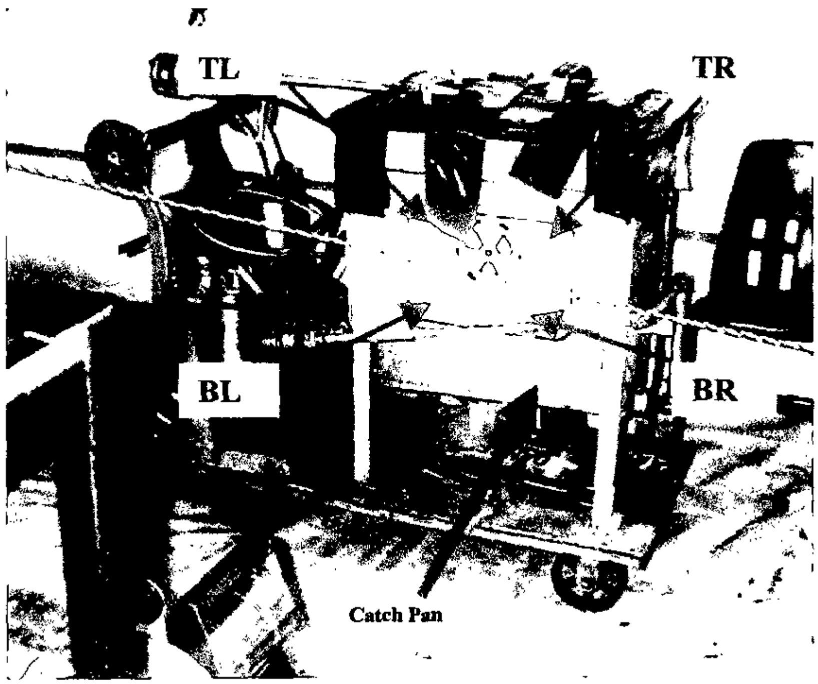
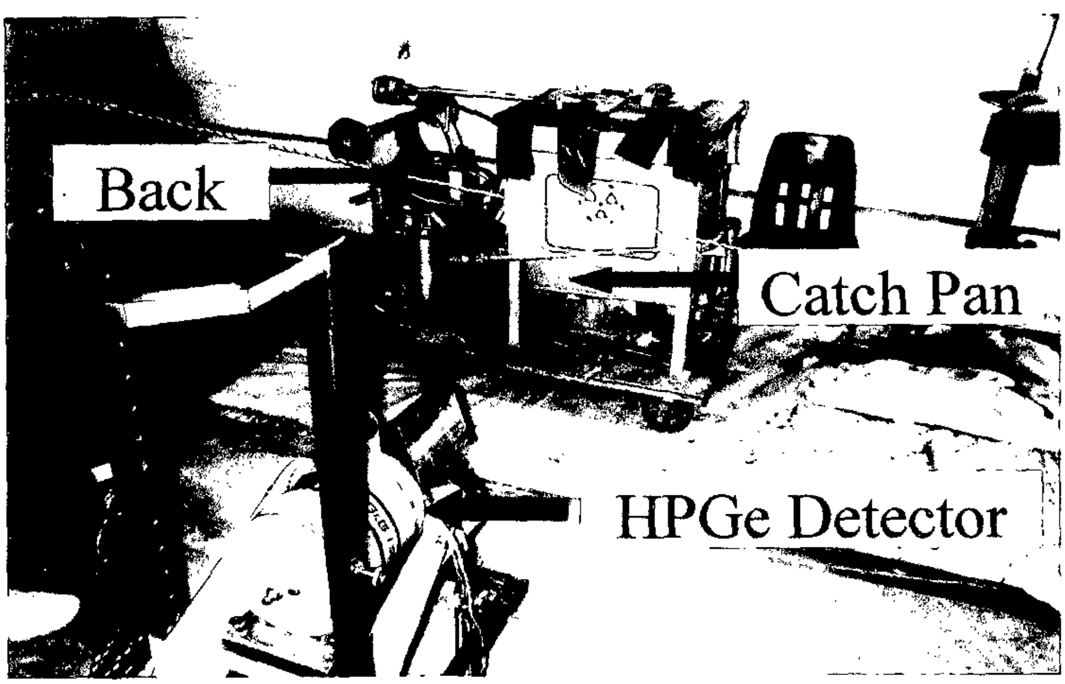


Figure 2. HPGe Far Field Arrangement



3. RESULTS AND DISCUSSION

3.1 Results From the NaI System

The NaI data were acquired in fourteen segments taken from a distance of eight inches. Note that we obtained spectra from the top left, top right, bottom left, and bottom right of the top portion of the cart, and we obtained spectra from the left and right side of the catch pan underneath the top portion of the cart. In this manner, we acquired six spectra with the cart in orientation one, and then we acquired six additional spectra with the cart in orientation two. Orientation two was simply a 180° rotation of orientation one. Therefore the orientation two spectra are identical to the orientation one spectra, except we were viewing the cart from the "other" side. In addition to the twelve spectra above, we acquired spectra from the cart filter and from the cart pump. These two were taken from a single orientation. Each of the fourteen spectra was acquired for a one-minute count time, and each was fit to determine the area under the U-235 γ-ray at 185 keV. The data acquired are listed in Table 1. The NaI data served primarily to locate the bulk of the U-235 holdup. It is clear from Table 1 that most of the HEU is located in the catch pan of the cart.

Table 1. Measured 185-keV γ-ray peak area and U-235 mass obtained with the 2x2 NaI detector in each configuration at 8 inches. All count times are one minute.

Sample Segment	Sample Position	185 KeV Peak Area Far Field
TL,1	Top Left	12540±320
TR,1	Top Right	12490±320
BL,1	Bottom Left	39040±460
BR,1	Bottom Right	26230±400
TL,2	Top Left-180	10140±290
TR,2	Top Right-180	10389±300
BL,2	Bottom Left-180	25206±400
BR,2	Bottom Right-180	25868±410
1,1	Catch Pan Left	64230±590
2,1	Catch Pan Right	86810±660
4,2	Catch Pan Left-180	85080±650
3,2	Catch Pan Right-180	82120±650

3.2 Results From the HPGe Far Field System

The HPGe detector far-field results are given in Table 2. All counting times are one minute, and the transmission standard count rate was 6500 counts per minute. Counts and counting uncertainties for the cart and cart plus transmission standard are given in the table. The grams of uranium-235 were calculated from the equation:

M_U (g U-235) = $(C_C/t_C)(2.36E\text{-}05)(48x2.54)^2(SQRT\ (S_T/Cart_T))$

where, $Cart_T = ((C_T/t_T) - (C_C/t_C))$

C_C – counts for cart position

T_C – count time in seconds for cart position

2.36E-05 – constant (g-sec/cm^2) for far-field setup (Ref. 6)

48x2.54 – distance of cart from detector in cm (48 in x 2.54 cm/in)

SQRT ($S_T/Cart_T$) – transmission correction is square root of the ratio of the standard counts per second with no attenuation divided by the standard counts per second of the cart with attenuation of the catch pan.

$Cart_T$ = counts per second of transmission source through the cart is equal to the counts per second of the transmission source plus the cart minus the counts per second of the cart.

Far field measurements were performed on each side of the Freon™ cart, and the results are given in Table 2.

Table 2. Results for the HPGe Detector Far-Field System.

Cart Position	Counts Cart (min)	σ Counts Cart	Counts Cart+Std (min)	σ Counts Cart+Std	Trans Correction	U-235 Mass (g)	σ U-235 Mass (g)
Side1	6399	92	7975	104	2.03	76	6
Side2	6890	95	8670	108	1.91	77	6

Considering the count rate of the cart sides given in Table 2, the HPGe far-field system measured about 77 grams of U-235. The uncertainty listed only takes into account counting statistics errors and calibration errors for the far field measurement. Uncertainties from the irregular sample shape, nonuniform distribution of U-235 in the cart, and U-235 self-absorption will increase the overall uncertainty. Its apparent that the cart contains more than 50 g of HEU holdup and some must be removed to meet the 50 g criticality limit.

Therefore the cart was further decontaminated to ensure the U-235 content is below the 50 g limit before disposal. The residue from decontamination was placed into scrap cans number 5413 – 5415. These three scrap cans were assayed individually using the two constant geometry transmission correction methods reported in reference 6. Then the decontaminated cart was assayed a third time for U-235 holdup[7].

The values for the point source transmission corrected assays of cans 5413 – 5415 are listed in Table 3. Using the Deming curve in references 6 and 8, we obtain a sum of U-235 content in the three cans of 69.0±3.0 g. The scrap-can-transmission-correction method described in reference 6, is not listed because of the difficulty experienced in making accurate transmission measurements. The assayed value for holdup of material remaining in the decontaminated cart is 10±1 g as reported in reference 7. These measurements sum to a value that is in very good agreement with the holdup value presented above of 77±20 g for the Freon™ cart.

Table 3. Result of the assays of the scrap cans.

Can Number	Can Counts (cps)	Can+Std Counts (cps)	Deming U-235 Mass(g)
5413	36.84±0.50	39.62±0.43	25.6±1.9
5414	37.91±0.66	41.33±0.62	26.7±2.0
5415	26.89±0.50	31.72±0.50	16.7±1.1

4. CONCLUSIONS

Using the data obtained by the HPGe far field system we determined that the original Freon™ cart contained 77± 20 grams of U-235. The uncertainty due to the counting statistics and calibration errors is 6 g. The balance of the reported uncertainty is a conservative estimate of the sample inhomogeneity, self-attenuation due to lumps within the sample, variations in gamma ray transmission for different sample segments, variations in the self-absorption of the standards and cart, and uncertainties from the irregular cart shape.

From the NaI detector system measurements we were able to determine the location of HEU in the cart. This information enabled Operations to decontaminate where the material was concentrated.

The values obtained from assaying the three scrap cans plus the value obtained from assaying the residue are in good agreement with original HPGe assay. The sum of the three scrap cans and the residue is 79±3 and the initial HPGe measurement is 77± 20. These results demonstrate that we have significantly improved upon the precision of the 1995 measurement[2]. This was critical for the proper disposal of the cart

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