**This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.**

## DISCLAIMER

WSRC-TR-2004-00147
Revision 0

# $^{235}$U and TRU Holdup Measurements on the 321-M Sawbenches

**Raymond A. Dewberry**

Unclassified
Does Not Contain Unclassified Controlled Nuclear Information (UCNI)

July 2, 2004

**Westinghouse Savannah River Company**
**Savannah River Site**
**Aiken, SC 29808**

WSRC-TR-2004-00147
Revision 0

Keywords:
NDA, Multichannel,
Far field, Assay, Holdup,
Close Field, Area Source

Classification: U

Authorized Derivative Classifier

# $^{235}$U and TRU Holdup Measurements on the 321-M Sawbenches

Raymond A. Dewberry,
Publication Date: July 2, 2004

| | |
|---|---|
| Raymond A. Dewberry, Author | Date |
| Raymond A. Sigg, Technical Reviewer | Date |
| S. R. Salaymeh, Manager, S&ATG-ADS | Date |
| Malcolm Smith, Management, Excess Facilities | Date |

**Westinghouse Savannah River Company**
**Savannah River Site**
**Aiken, SC 29808**

DISCLAIMER

**ABSTRACT**

The Analytical Development Section of Savannah River Technology Center (SRTC) was requested by the Facilities Disposition Projects (FDP) 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 on two sawbench components that were used for cutting U-Al fuel rods. The benches contained U-Al residue scattered on the bench tops and bagged up in plastic containers. It was very important to obtain an estimate of the HEU content of this residue in order to remove criticality concerns before vacuuming it into a much smaller volume. A portable high purity germanium (HPGe) detection system and a portable sodium iodide (NaI) detection system were used to determine highly enriched uranium (HEU) holdup and to determine holdup Np-237 and Am-241 that were observed in these components. The measured Np-237 and Am-241 contents were especially important in these components because their presence is unusual and unexpected in 321-M. It was important to obtain a measured value of these two components to disposition the saw benches and to determine whether a separate waste stream was necessary for release of these contaminated components to the E-Area Solid Waste Vault. The reported values for Np-237 are < 0.8 mg on each of the two bench tops and < 3 μg in the bag of chips. The reported values for Am-241 are < 500 ng on the two bench tops and < 15 ng in the bag of chips. Our results demonstrate an upper limit of $^{235}U$ content of 0.4 g on the two benchtops and of 0.1 mg in the bag of chips. These reported values completely eliminated the criticality concerns. This report discusses the methodology, non-destructive assay (NDA) measurements, and results of the holdup measured for each of the three actinide species in these saw benches.

This page intentionally left blank.

## TABLE OF CONTENTS



## LIST OF FIGURES



## LIST OF TABLES

# $^{235}$U and TRU Holdup Measurements on the 321-M Sawbenches

**Raymond A. Dewberry**

**Westinghouse Savannah River Company**
**Savannah River Site**
**Aiken SC 29808**

## 1. INTRODUCTION

The Savannah River Site 321-M Reactor Fuel Production facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the Savannah River production reactors. The facility operated for 25 years. During this time thousands of uranium-aluminum-alloy (U-Al) fuel tubes were produced. After the facility terminated 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. The Analytical Development Section (ADS) of Savannah River Technology Center was tasked to perform holdup and material control and accountability assays to determine content of highly enriched uranium (HEU) in the deactivation and decommissioning (D&D) activities of the 321-M facility.[1]

$^{235}$U holdup measurements were performed in the 321-M facility in 1995 and documented in technical report WSRC-TR-95-0492.[2] The holdup values reported in reference 2 did not address all of the process components in the facility and specifically did not address the HEU and TRU content on the sawbenches. Therefore the Facilities Disposition Program (FDP) 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 deactivation 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 $^{235}$U 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 $^{235}$U 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.

These D&D and assay activities have been on-going from FY00 through FY04. In the course of the FY03 D&D activities, Radiation Control Operations (RCO) surveys found Np-237/Pa-233 and Am-241 activity in the C and D outgassing ovens of the facility and

on two saw bench components.[5] The saw benches were used to cut process components including the U-Al product fuel tubes. Radioactivity from sources other than U-235 was completely unexpected in the facility. In the 25 years of operation the facility performed process and mechanical work on uranium fuel and target rods for the production reactors. While recycled material was run through the facility, and therefore uranium material that contained the isotopes 232, 233, 234, and 236 were observed, at no time did the facility expect to observe trans-plutonium activation products. Observation of Np-237 and Am-241 activity in the RCO surveys of 2003 surprised the Facility Disposition Program (FDP) D&D operations management. Along with radiation control concerns, appearance of Np-237 and Am-241 radioactivity was not consistent with the designated solid waste stream for the facility. Observation of these two species appeared to violate the Waste Acceptance Criteria for the facility.[3,4]

To quickly respond to these concerns, ADS derived point-, line-, and area-source efficiency calibration constants from first principles of the HPGe detector to measure the Np-237/Pa-233 equilibrium content and Am-241 content of the C and D ovens. The Np-237/Pa-233 efficiency calibrations were subsequently confirmed by an experimental measurement.[6] Upon experimental confirmation of the technique of first principles, we used the same technique to calculate area and point source calibration constants for both species using the portable NaI detection system.[7] In this report we describe the detector systems, the data acquisitions, and the techniques used to model the data from two different configurations.

This report describes holdup measurements using both the NaI and HPGe detection systems on the two sawbenches and on residue, filings, and chips observed on the bench-tops of these two components. We describe the detector systems, the data acquisitions, and the techniques used to model the data from two different configurations with both detectors. All three species U-235, Np-237/Pa-233, and Am-241 are reported. FDP management was concerned that vacuuming the residue on the bench tops would reduced the volume of the chips and residue and thereby create a criticality hazard. The report also describes measurement of all three species in a bag that contained bulk quantities of chips. This bag of bulk residue also represented a criticality concern.

## 2. EXPERIMENTAL

A 2” x 2” sodium iodide (NaI) detection system and a portable high purity germanium (HPGe) detection system were used to conduct NDA measurements of HEU and TRU holdup on the sawbenches. The NaI detection system uses a 2” x 2” 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.[8] For the NaI acquisitions and analyses we used Canberra Genie-2000 software. For some of the HPGe acquisitions we used 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 $\gamma$-ray spectra. This system is described in reference 9 and has been used extensively in HEU holdup measurements for FDD. At one point the Dart system failed, and we switched to

a Canberra Inspector MCA package with Genie-2000 acquisition software. Both HPGe systems run very similarly and yield multichannel analyzer spectra that are indistinguishable in appearance and in treatment of the data.

To accomplish the sawbench assays, we obtained twelve HPGe acquisitions in the far field point source configuration and five NaI acquisitions in the near field area source configuration. The NaI acquisitions include an area source count taken on the bag of chips and filings found on sawbench two. A sketch of one sawbench is shown in Figure 1. The data obtained in the seventeen acquisitions are shown in Table 1. For each acquisition date we also obtained QC check spectra with the Cs-137 source as prescribed in references 10 and 11. In Table 1 HPGe spectrum T0(65) represents an additional QC check obtained with the 4.20-g HEU transmission source from a distance of 65 inches. This QC spectrum is shown in Figure 2.

From spectrum T0(65) we can calculate an HEU content of

$$[HEU]_{source} = (2.36x10^{-5})(cps)(d)^2, \quad (1)$$

where the first factor is the point source calibration constant of reference 9 in units of g-sec/cm$^2$, *cps* is the detection rate of the 185-keV γ-ray, and *d* is the source to detector distance in cm.

$$[HEU]_{source} = (2.36x10^{-5})(6.73)(65x2.54)^2 = 4.33(10)\ g,$$

which is in excellent agreement with the known content .[12]

Acquisition bench-1-1 was obtained with the detector viewing the left-hand draw of bench one at a distance of 63 inches. Acquisition bench-1-2 was obtained with the detector viewing the right hand drawer of bench one at a distance of 45 inches, and acquisition bench-1-0 was obtained viewing the entire saw bench at a distance of 127 inches. Spectrum bench-1-0 is shown in Figure 3. Acquisitions bench-2-1 and bench-2-0 are the analogous views of bench two from distances of 66 inches and 127 inches respectively. Note for each of these five HPGe views we observed essentially no content in the 186-keV γ-ray photopeak. Each has a measured count rate less than five times the uncertainty of the background measurement BKG528, which is $LLD_{cps}$ = 5x(289x0.47/18000) = 0.038 cps. We do not calculate a formal limit of detection for the drawers of the two benches. We do that below for the bench tops and for the bag of chips.

The next four HPGe acquisitions were obtained to yield far field point source transmission corrected assays of the two cutting blades of the sawbenches. For each of the two saw blades we obtained an experimental measure of the transmission correction value for the 186-keV peak. To obtain a conservative estimate (that is, to overestimate) we used the same transmission correction value for the 312-keV peak from Np-237. These calculations and observed limits of content are reported in the next section.

The five NaI spectra are listed in the lower portion of Table 1. Acquisitions *QC610NaI* and *QC611NaI* are QC check spectra with the Cs-137 source. Acquisitions *NaI-A-1-1* and *NaI-A-2-1* represent area source acquisitions with the NaI looking down onto the two separate bench tops. Spectrum *chipbags* is an area source acquisition of the bag of chips. Spectrum NaI-A-2-1 is shown in Figure 4. In each of these we observed an area of zero counts under the 186-keV and 312-keV photopeaks. Using the integrated counts in these regions of interest we perform formal calculations below of the limit of content for U-235 and Np-237 on the bench tops and in the bag of chips. Figure 5 is a NaI spectrum obtained from the horizontal vent line of outgas oven C. We have included this spectrum to demonstrate the capability to observe activity from U-235 and Np-237/Pa-233 with a NaI detector.

**Table 1. Summary of HPGe and NaI Acquisitions Obtained of the Two Sawbenches and of the Bag of Chips**

| HPGe Acquisition | Count time (sec) | d (in) | Count Config | 186 keV Area | σ | 312 keV Area | σ | 662 keV Area | σ | Field of view |
|---|---|---|---|---|---|---|---|---|---|---|
| QC528 | 60 | Fixed | point | | | | | 1218 | 37 | Check source |
| Bench-1-1 | 600 | 63 | point | 56 | 25 | | | | | Left drawer |
| Bench-1-2 | 300 | 45 | point | 1 | 38 | | | | | Right drawer |
| Bench-1-0 | 300 | 122 | point | 10 | 20 | | | | | whole |
| T0(65) | 300 | 65 | point | 2019 | 48 | | | | | T-Source |
| BKG528 | 18000 | N/A | N/A | 289 | 136 | | | | | Region BG |
| Bench-2-1 | 700 | 66 | point | 47 | 27 | | | | | Left drawer |
| Bench-2-0 | 800 | 127 | point | 22 | 32 | | | | | whole |
| Saw-1 | 700 | 29 | point | 0 | 17 | | | | | Saw blade |
| Saw-1-T | 403.38 | 30 to source | point | 6997 | 91 | | | | | Saw blade & T-source |
| Saw-2 | 700 | 35 | point | 81 | 24 | | | | | Saw blade |
| Saw-2-T | 300 | 37" to source | point | 3597 | 65 | | | | | Saw blade & T-source |
| | | | | | | | | | | |
| **NaI Acquisition** | **Count time (sec)** | **d (in)** | **Count Config** | **186 keV Area** | **σ** | **312 keV Area** | **σ** | **662 keV Area** | **σ** | **Field of view** |
| QC610NaI | 60 | N/A | point | | | | | 1351 | 41 | Check source |
| NaI-A-1-1 | 300 | N/A | Area | 778 | 14 | 0 | 42 | | | Bench top |
| QC611NaI | 60 | N/A | point | | | | | 1505 | 45 | Check source |
| NaI-A-2-1 | 300 | N/A | Area | 0 | 75 | 0 | 43 | | | Bench top |
| Chip bags | 1000 | N/A | Area | 0 | 70 | 0 | 103 | | | Bag of chips & filings |

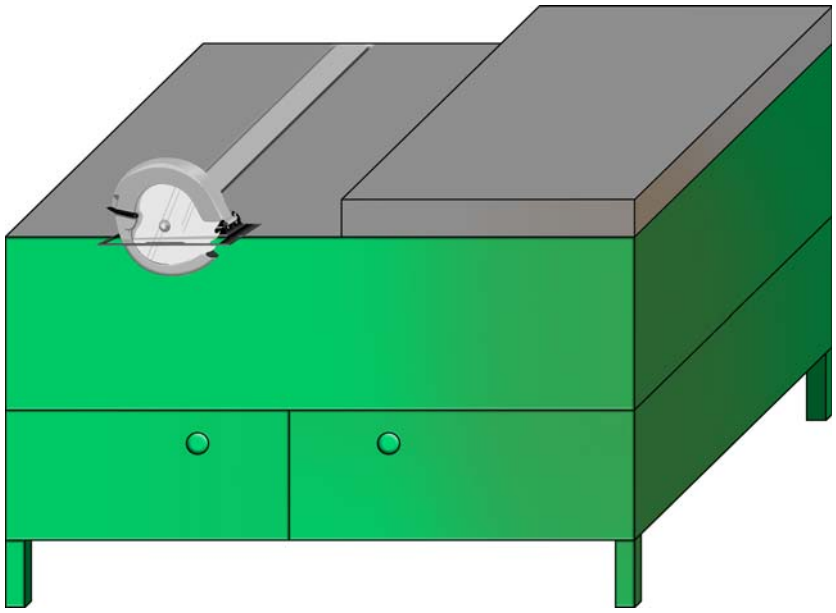

**Figure 1. A sketch of one of the sawbenches showing the saw blade, drawers, and top surface that was contaminated with U-Al residue.**

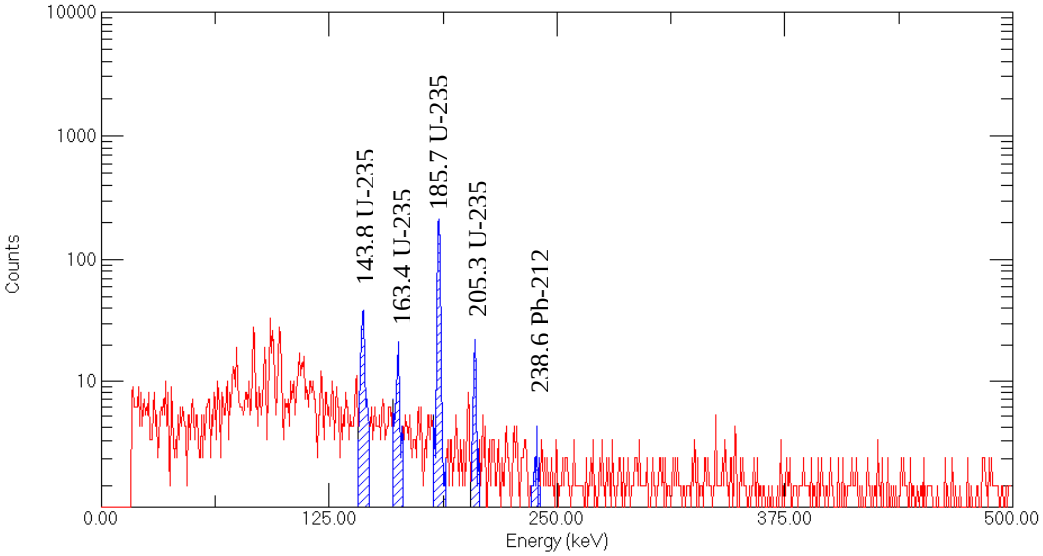


**Figure 2. Spectrum of $^{235}$U source check T0(65).**

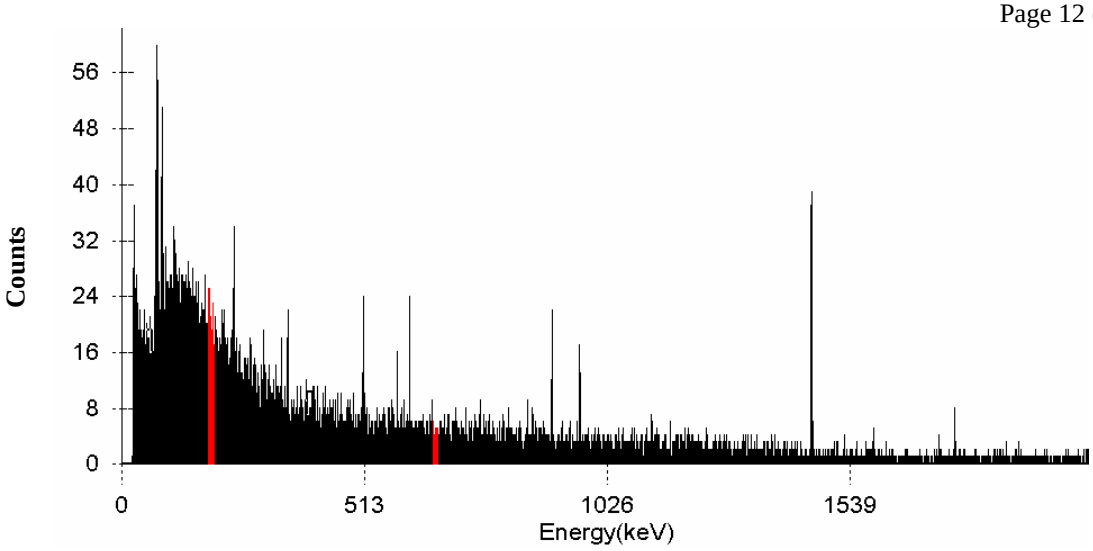


**Figure 3. HPGe spectrum Bench-1-1.**

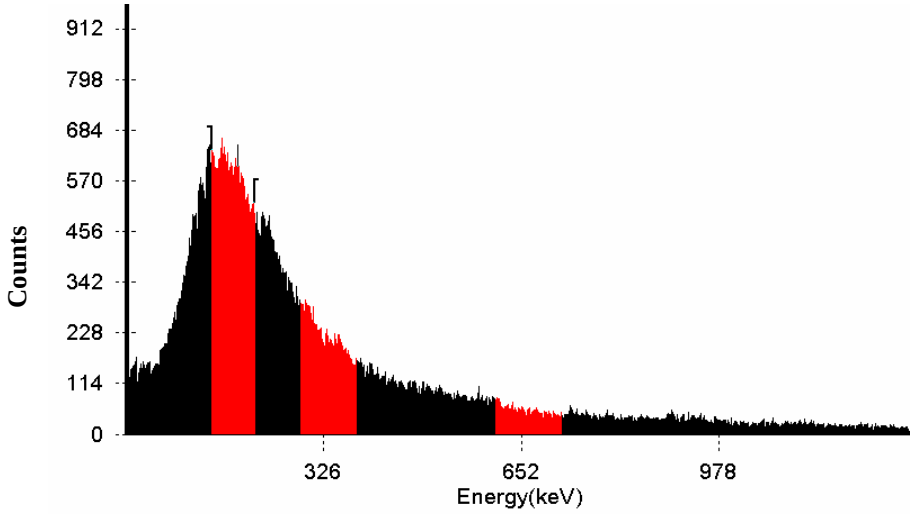


**Figure 4. NaI spectrum NaI-A-2-1.**

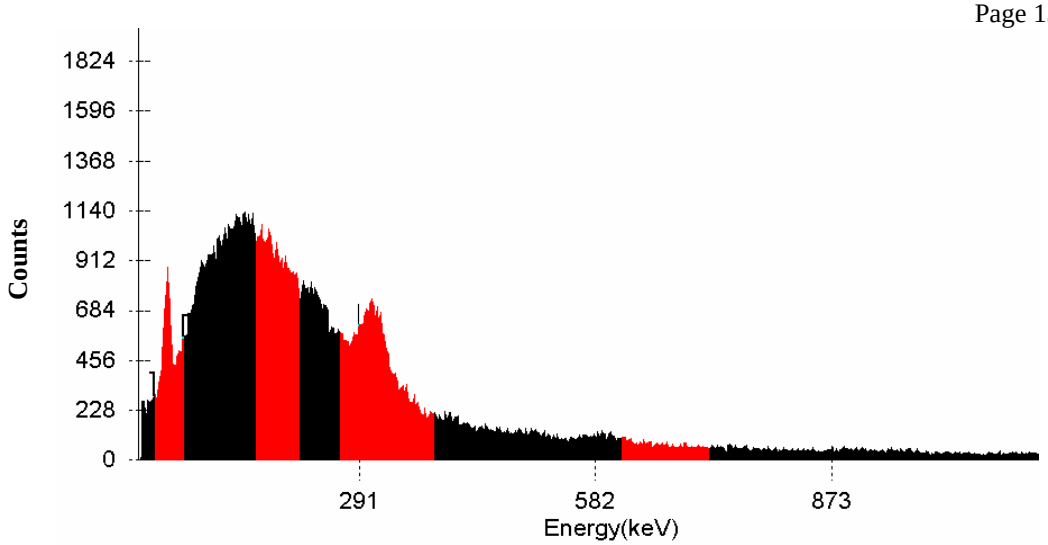


**Figure 5. NaI spectrum showing activity from Am-241 and Np-237/Pa-233.**

## 3. CALCULATIONS

### 3.1 HPGe Spectra

The HEU holdup of each individual component observed in the HPGe measurements were determined using the point source acquisition configuration of equation (1).

$$[\text{HEU}] = (2.36\text{x}10^{-5})(\text{cps})(\text{d})^2(\text{T}_{\text{corr}}), \qquad (1)$$

where the first factor is the point source calibration constant of reference 9 in units of g-sec/cm$^2$, cps is the detection rate of the 185-keV γ-ray, d is the source to detector distance, and $\text{T}_{\text{corr}}$ is the transmission correction factor. This yields a measured mass of

$$[\text{HEU}]_{\text{T0(65)}} = (2.36\text{x}10^{-5})(2109/300)(65\text{x}2.54)^2(1.00) = 4.33(10)$$

for the QC check spectrum of the 4.21-g U-235 transmission source.

The transmission correction factor for the saw blade on bench one was calculated from the experimental spectra *Saw-1 and Saw-1-T*. For the T0(30) detection rate we used the calculated unabsorbed rate that we would observe at a distance of 30 inches. We can use this calculated rate because the HPGe detection system has been qc checked with both the U-235 transmission source (equation 1) and with the Cs-137 check source. Transmission of the source 185 keV γ-rays through saw blade one is measured by

$$\text{T} = \{\text{cps}(\textit{Saw-1-T}) - \text{cps}(\textit{Saw-1})\}/\text{cps}(\textit{T0(30)}) \qquad (2)$$

$$= \{6997/403.38 - 0/700\}/30.723 = 0.5646.$$

The correction factor is then taken to be $\sqrt{\frac{1}{T}} = 1.331$. For the blade on sawbench two we obtain a transmission correction factor of 1.304. Since all of the components of Table 1 have very low HEU content, we arbitrarily used this transmission correction factor in all of the point source HPGe calculations of equation (1). For saw blade two we obtain a measured content of

$$[HEU]_{Saw\text{-}2} = (2.36\text{x}10^{-5})(cps)(d)^2(T_{corr}), \quad (1)$$

$$= (2.36\text{x}10^{-5})(81/700)(37\text{x}2.54)^2(1.304)$$

$$= 0.031(9)\ g.$$

The one-sigma uncertainty in parentheses was obtained using the uncertainty in the area under the 185-keV peak in the spectrum.

Using the identical point source technique we report the measured HEU holdup in each component of sawbenches one and two that are listed in Table 2. The upper limits of content were determined using (1) with the count rate determined from

$$[HEU]_{LLD} = (2.36\text{x}10^{-5})(cps)_{LLD}(d)^2(T_{corr})SQRT(18000/t) \quad (3)$$

$$= (2.36\text{x}10^{-5})5\text{x}\sigma_{BKG}(d)^2(T_{corr})SQRT(18000/t).$$

$\sigma_{BKG}$ is the standard deviation of the background rate, which is 5x136/18000 as shown in Table 1, and t is the count time for each acquisition.

**Table 2. List of reported $^{235}$U gram values.**

| HPGe Acquisition | HEU (g) | Uncertainty (g) |
|---|---|---|
| Bench-1-1 | < 0.2 | N/A |
| Bench-1-2 | < 0.1 | N/A |
| Bench-1-0 | < 0.9 | N/A |
| T0(65) | 4.33 | 0.10 |
| Bench-2-1 | < 0.2 | N/A |
| Bench-2-0 | < 0.7 | N/A |
| Saw-1 | < 0.03 | N/A |
| Saw-2 | 0.031 | 0.009 |
| | | |

### 3.2 NaI Spectra

The HEU contents from the NaI area source acquisitions were calculated using equation (4)

$$[HEU] = K_a(cpm)(A), \qquad (4)$$

where $K_a$ is the area source calibration constant of $9.78 \times 10^{-7}$ g-min/in$^2$ for U-235.[7] For acquisition *NaI-A-1-1* we obtain

$$[HEU] = K_a(cpm)(A) = (9.78 \times 10^{-7})(778/5)(24 \times 45)$$

$$= 0.164(3) \text{ g},$$

where the last term is the surface area of the bench top of sawbench one. For the Np-237 content we used the standard deviation of the region of interest of the 312 keV peak and the area source calibration constant of $3.81 \times 10^{-9}$ g-min/in$^2$.[7] Applying all of the NaI area source acquisitions and calculations we obtain the measured values and limits of detection for the two bench tops and for the bag of chips listed in Table 3.[13] The area source calibration constant for Am-241 is $2.43 \times 10^{-12}$ g-min/in$^2$.[7]

**Table 3. List of reported gram values obtained from the NaI area source acquisitions.**

| Component | $^{235}$U Reported (g) | $^{237}$Np Reported (g) | $^{241}$Am Reported (g) |
|---|---|---|---|
| NaI-A-1-1 | 0.164(3) | < 0.0004 | Not observed |
| NaI-A-2-1 | < 0.4 | < 0.0008 | $< 5 \times 10^{-7}$ |
| Chipbag | < 0.001 | < 0.000003 | $< 1.5 \times 10^{-8}$ |
| | | | |

## 4. CONCLUSION

We have performed transmission corrected point source and area source γ-PHA acquisitions on two sawbenches in Building 321-M. These two components are pieces of process equipment that performed cutting operations for the Reactor Fuel Fabrication Facility for its 25 years of production. We have performed a thorough search for U-235, Np-237, and Am-241 on each of these components. The acquisitions in this report include NaI and HPGe γ-ray analyses of the benchtop surfaces, the drawers, and of the cutting blades of both of the sawbenches. We also assayed a bag of residue and chips that were assumed to be U-Al with measurable contents of perhaps all three species. The thorough searches were required because of Radiological Control Operations observations of non-transferable activity that indicated radioactive contributions from each of the three species. Evaluation of the content was important for disposal of the components as solid waste and for criticality control.

The U-235 contents we measured on the bench top of sawbench one is

[U-235] = 0.164(3) g. On the bench top of sawbench one we observed less than 0.2 mg of Np-237 and less than 100 ng of Am-241. On sawbench two we observed [U-235] < 0.4 g, [Np-237] < 0.8 mg, and [Am-241] < 500 ng. Our observations indicate the bag of filings and chips was empty of U-Al material and of transuranic material. The content of U-235 in the bag of chips is less than 1 mg. The content of Np-237 in the bag of chips is less than 3 μg, and the content of Am-241 is less than 15 ng.

These measurements demonstrated that the sawbenches and the bag of chips did not represent a potential criticality hazard. The residue on the benchtops of both sawbenches could be safely vacuumed up for disposal as solid waste. Likewise the sawbenches themselves were classified as safe for disposal as low level solid waste.

## 5. REFERENCES

1. D. L. Honkonen, "Nuclear Criticality Safety Evaluation (NCSE): Enriched Uranium Removal From Building 321-M," N-NCS-G-00051, February 2000.
2. R. S. Thomason, "235U Holdup in Building 321-M Contamination Areas and Associated Exhaust Systems," WSRC-TR-95-0492, December 1995.
3. Deactivation Project Plan 321-M Fuel Fabrication Facility, V-PMP-M-00004, January 2000.
4. WSRC 1S Manual, Procedure 3.17, Section A.3.a, "E-Area Vaults Nuclear Safety," July 2000.
5. Ron Smith, In-Situ Object Counting System (ISOCS) Radiation Control Operations Survey, March 2003.
6. R. A. Dewberry, "Calculation of Np-237 and Am-241 Detector Calibration Constants From First Principles", WSRC-RP-2003-01113, March 2004.
7. R. A. Dewberry, Laboratory Notebook, WSRC-NB-2003-00086, page 159 – 171.
8. R. A. Dewberry and S. R Salaymeh, "Efficiency Calibration Using HEU Standards of 2" x 2" NaI Detector," WSRC-TR-2000-00269, July 2000.
9. S. R Salaymeh and R. A. Dewberry, "Efficiency Calibration of the Portable High Purity Germanium Detection System for use in the M-Area Deactivation Assays," WSRC-TR-2000-00317, September 2000.
10. R. A. Dewberry, "Revision to M-Area High Purity Germanium Detector System 14Q302 Quality Control", SRT-ADS-2003-0093, February 2003.
11. R. A. Dewberry, "Method Development Report for use of the Portable NaI Detection System for Assay of Highly Enriched Uranium", SRT-ADS-2003-0124, May 2003.
12. R. D. McElroy to P. E. Filpus-Luyckx, "PHA Standards Values", NMP-RMT-920277, October 1992.
13. R. A. Dewberry, Laboratory Notebook, WSRC-NB-2003-00074, page 6.