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Revision 0

**Gamma-Ray Holdup Measurements of U-235, Np-237, and Am-241 Content in the
C and D Out-gassing Ovens in the Deactivation and Decommissioning Activities in
321-M**

Raymond A. Dewberry

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September 16, 2004

Westinghouse Savannah River Company
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Aiken, SC 29808



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Abstract

The Analytical Development Section of Savannah River National Laboratory (SRNL) 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 in the C and D out-gassing ovens that were used to remove gas entrained in billet assembly material prior to the billets being extruded into rods by the extrusion press. 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 of Np-237 and Am-241 that were observed in these components. The HPGe detector was run by an EG&G Dart™ system that contains the high voltage power supply and signal processing electronics. A personal computer with Gamma-Vision software was used to control the Dart™ MCA and provide space to store and manipulate multiple 4096-channel γ -ray spectra. The NaI detector was run with a Canberra NaI+ MCA card that converts a personal computer to a full function multichannel analyzer and with Canberra Genie-2000 acquisition and analysis software. 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 species to disposition the out-gassing ovens 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 (17 ± 7) mg in oven C and < 0.5 mg in oven D. The reported values for Am-241 are (1.3 ± 0.2) μ g in oven C and < 400 ng in oven D. Our results indicate an upper limit of U-235 content of 0.2 g for oven C and (0.105 ± 0.048) g in oven D. 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 out-gassing ovens.

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Gamma-Ray Holdup Measurements of U-235, Np-237, and Am-241 Contents in the C and D Out-gassing Ovens in the Deactivation and Decommissioning Activities in 321-M

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1.0 INTRODUCTION

The 321-M 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 more than 30 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 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 the Savannah River National Laboratory 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

These 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 and Am-241 activity in the C and D out-gassing ovens of the facility.² Radioactivity from sources other than U-235 was completely unexpected in these ovens. In the 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 plutonium product or 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 To quickly respond to these concerns the non-destructive assay (NDA) expert scientists of ADS were requested to perform a rapid assessment of the equilibrium Np-237/Pa-233 and Am-241 content of the C and D ovens.

At the time of the request ADS had no detector efficiency calibrated to perform the Np-237/Pa-233 or Am-241 γ -ray assays. We performed a rapid calculation of the necessary point source and line source calibration constants using the first principles described in reference 4. This report describes the acquisitions and holdup measurements for U-235, Np-237, and Am-241 in the C and D out-gassing ovens using the experimental calibration constants for U-235 and using the calculated calibration constants for Np-237 and Am-241. The reported values for U-235 are < 0.2 g in oven C and (0.105 ± 0.048) g in oven D. The reported values for Np-237 are (13 ± 10) mg in oven C and < 5 mg in oven D. The reported values for Am-241 are (1.3 ± 0.2) μ g in oven C and < 400 ng in oven D.⁽⁵⁻⁹⁾ We show in the **Calculations and Results** section below that the relative values of Am-241 and Np-237 clearly demonstrate these two parent-daughter species are not in a parent/daughter radioactive relationship, and therefore the two species were introduced separately into the C and D ovens.

2.0 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 on the C and D ovens. 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.¹⁰ For the NaI acquisitions and analyses we used Canberra Genie-2000 software. The HPGe detection system uses an EG&G DartTM 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 11 and has been used extensively in HEU holdup measurements for Facility Decontamination Division (FDD) and FDP.

A photograph of oven C is shown in Figure 1. To accomplish the oven C assays, we obtained four line source acquisitions of the horizontal vent line shown in Figure 1 and one line source acquisition of the vertical vent line. These acquisitions are denoted acquisitions *S1* - *S5*. Acquisition *S1* was obtained at a source to detector distance (*d*) of eighteen inches. It was facing the horizontal vent line at a point 26 inches from the left end. Acquisition *S2* was obtained facing a point 21 inches to the left of *S1*. Acquisition *S3* was obtained facing a point 21 inches to the right of *S1*, and acquisition *S4* was obtained facing a point 21 inches further still to the right of *S3*. Acquisition *S4* was obtained five inches from the right end of the horizontal vent line, which is a total of 80 inches long. All of the source to detector distances for *S1* - *S4* were eighteen inches. Acquisition *S5* was obtained from a distance of 25 inches, and it viewed the vertical vent line as a line source of length 36 inches. All of the data from these line source acquisitions are summarized in Table 1. We then obtained the point source acquisitions *Chamb1* and *Chamb2* of the oven interior and the oil reservoir. These are shown in Table 1 also. The line source acquisition spectrum *S5* is shown in Figure 2.

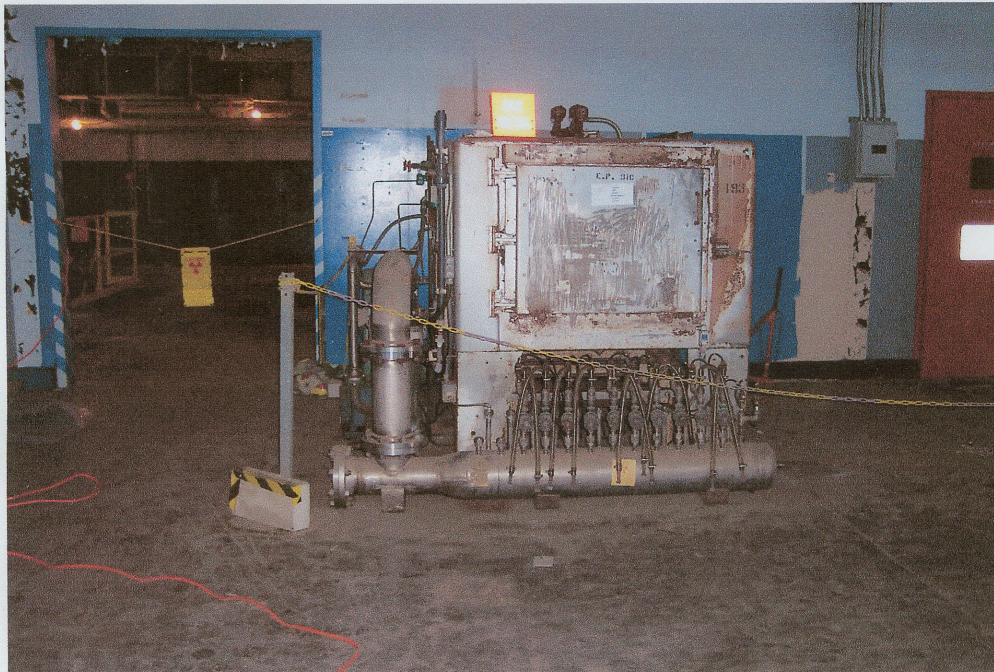


Figure 1. Photograph of Oven C showing horizontal and vertical vent lines as well as oven chamber with door closed and oil reservoir (left rear).

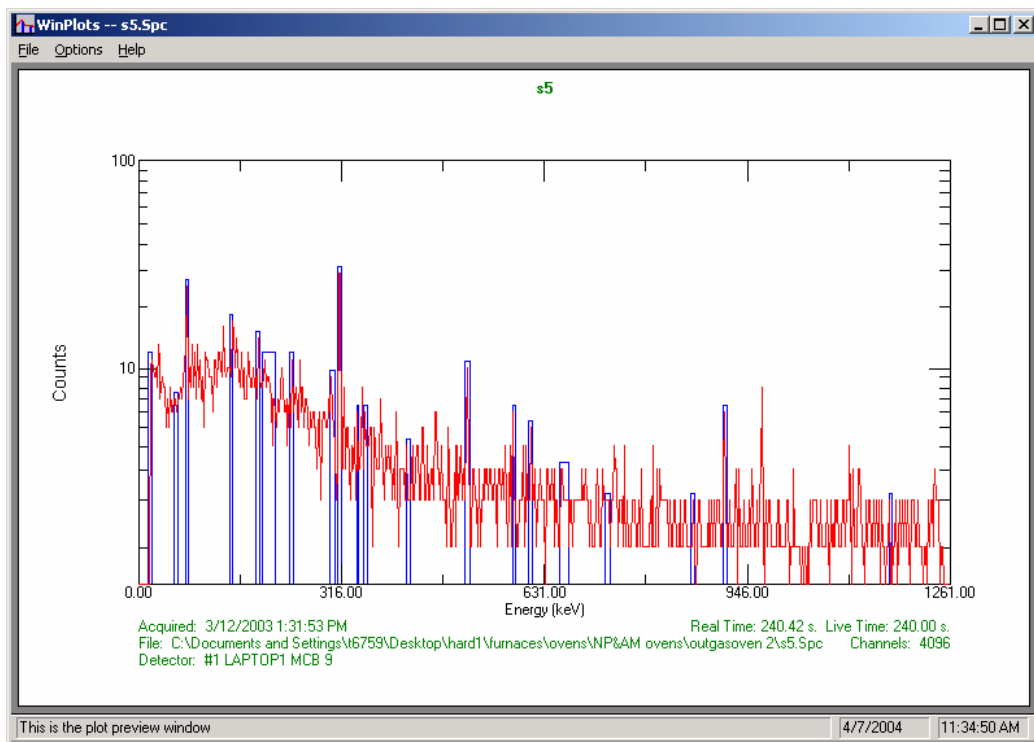


Figure 2. HPGe Spectrum S5.

We next obtained the transmission correction data for the horizontal vent line using the Cs-137 check source and using the U-235 transmission check source. For these transmission correction measurements we obtained acquisition $S4T(Cs)$ and $S4T(U)$ from the same positions as acquisition $S4$. Acquisitions $S4T(Cs)$ and $S4T(U)$ were obtained successively with the Cs-137 check source behind the vent line and 26.5 inches from the detector and with the HEU transmission source behind the vent line and 26.5 inches from the detector. Finally we obtained acquisitions $T0(Cs)$ and $T0(U)$, which were acquisitions with the bare sources placed 26.5 inches from the detector in the point source configuration. We explain the transmission correction measurements completely in the **Calculations and Results** Section.

The calculations of holdup in oven D were performed in a manner identical to those of oven C in sections 3.1 - 3.3. For some of these acquisitions we used the NaI detection system and the calibration constants determined for it.⁴ To use the NaI detector it was first necessary to demonstrate that we could observe and resolve the γ -rays from each of U-235, Np-237/Pa-233, and Am-241. To do that we obtained a NaI spectrum of the oven C horizontal vent line from a distance of 24 inches. This line source spectrum is shown in Figure 3 and very clearly shows the peaks from Am-241 and the 312 - 340 keV doublet from Np-237/Pa-233. The QC check spectrum showing the Cs-137 peak at 662 keV is displayed in Figure 4. We then obtained the same acquisitions from the D oven that we obtained from the C oven. These acquisitions are summarized in Table 2.

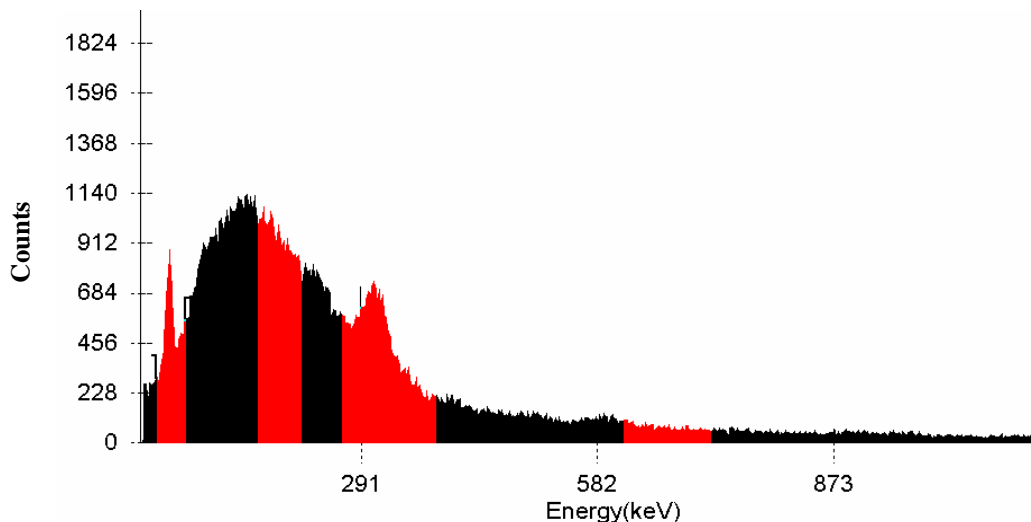


Figure 3. NaI Spectrum Showing Activity from Am-241 and Np-237/Pa-233.

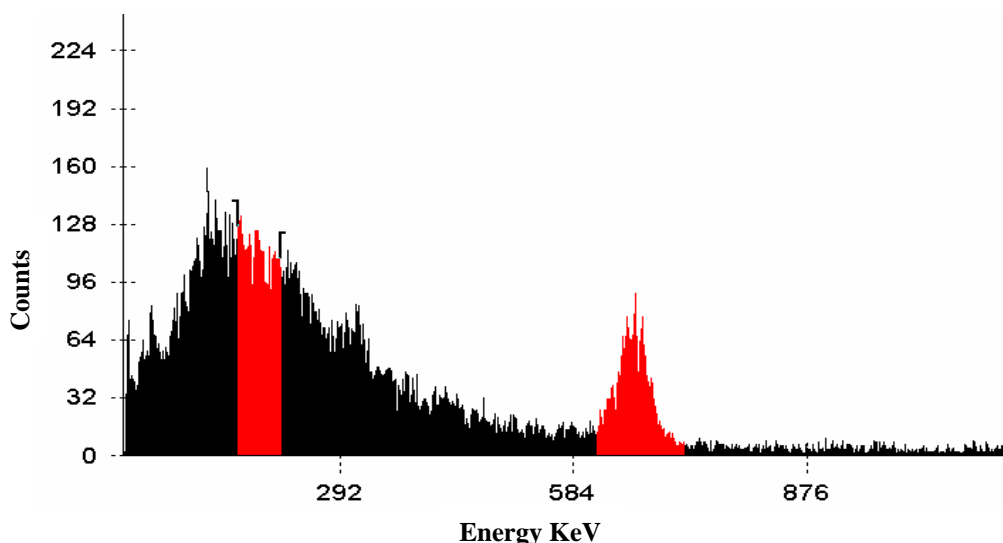


Figure 4. NaI QC Check Spectrum Obtained for Oven D Acquisitions.

3.0 CALCULATIONS AND RESULTS

After the γ -ray acquisitions were obtained in the line source and point source configurations, it was necessary to model them and to obtain transmission correction values for each of the three species. Since our two detection systems had not been experimentally calibrated for the Np-237/Pa-233 and for the Am-241 holdup measurements, it was further necessary to obtain defensible point source and line source efficiency calibration constants for them. These four required constants were derived from first principle arguments that we described and justified in reference 4. From here in this report we describe our calculations separately for each species in each of the two ovens. We first deal with the HPGe data from oven C, where we have individual subsections describing the U-235 calculations, then the Np-237/Pa-233 calculations, and then the Am-241 calculations. We then perform the same treatment with the NaI data from oven D.

3.1 Line Source Holdup in Vent Line of Oven C

In the **EXPERIMENTAL** section and from Table 1, we obtained four line source acquisitions of the horizontal vent line (Figure 1) of oven C, a line source acquisition of the vertical bellows, and two point source acquisitions of the oven chamber and oil reservoir. We first treat the line source acquisitions S1 - S4 and the transmission correction measurements S4T(Cs) and S4T(U) that we obtained on the horizontal vent line. These were obtained at a distance of 18" from the vent line.

In the line source configuration, the HEU content is determined by

$$[U-235] = K_1(\text{cps})(d)(L)Cf_T, \quad (1)$$

where K_1 is the line source calibration constant derived in reference 11. K_1 is 1.72×10^{-5} g-sec/cm². The third term, d , is the acquisition distance (45.7 cm), and the fourth term, L , is the length of the line source. For the horizontal vent line L is 80" (203.2 cm). Cf_T is the transmission correction factor, which we treat below. cps is the measured count rate in each acquisition. For an ideal line source of activity, each acquisition rate S_n ($n = 1, \dots, 4$) would be identical. For our purposes, we take the average measured value for each S_n as the measured (cps) for equation (1).

$$(\text{cps}) = [116/300 + 24/300 + 140/300 + 69/300]/4 = 0.291(61). \quad (2)$$

Substituting, we obtain

$$[U-235]_{\text{vent line}} = (1.72 \times 10^{-5})(0.291)(18 \times 2.54)(80 \times 2.54)(Cf_T[U]) = 0.046Cf_T.$$

For the transmission correction we used the Cs-137 QC check source and the U-235 transmission source along with the line source measurement S4. The four additional measurements required are listed in Table 1 as S4T(Cs), S4T(U), T0(Cs), and T0(U). The first two were acquired with the Cs-137 and U-235 sources placed behind the horizontal vent line directly along the symmetry axis of the detector. The latter two were obtained with the sources only and with no component of the oven in view. All four of these acquisitions were obtained with a source to detector distance of 26.5 inches. The sample to detector distances were 18 inches for acquisitions S4T(Cs) and S4T(U).

The correction factor for the 185-keV γ -ray was determined by

$$Cf_T[U] = \text{SQRT}(\text{cps}[T0(U)]/\{\text{cps}[S4T(U)] - \text{cps}[S4]\}), \quad (3)$$

where each count rate is taken from the 185-keV peak in the spectrum. For equation (3) we obtain

$$Cf_T[U] = \text{SQRT}(37.46/\{5.11 - 0.23\}) = 2.77.$$

Using the Cs-137 transmission source to determine the correction factor for the 661-keV γ -ray we obtain,

$$\begin{aligned} Cf_T[Cs] &= \text{SQRT}(\text{cps}[T0(Cs)]/\{\text{cps}[Cs4T(Cs)] - \text{cps}[S4(Cs)]\}) \\ &= \text{SQRT}(0.208/\{0.062 - 0\}) = 1.83. \end{aligned} \quad (3),$$

To determine $Cf_T[Np]$ for the 312-keV γ -ray we interpolate using $d\{\ln(Cf_T[E])\}/dE$. For $Cf_T[Am-241]$ we extrapolate the same function down to 59 keV. For the 312-keV γ -ray

$$\begin{aligned} d\{\ln(Cf_T[E])\}/dE &= [\ln(1.83) - \ln(2.77)]/(662 - 185) \\ &= [0.604 - 1.019]/477 = -0.000869/\text{keV}. \end{aligned} \quad (4)$$

$$\ln(Cf_T[312]) = -0.000869(312 - 662) + \ln(1.83) = 0.908. \quad (5)$$

$Cf_T[312] = 2.48$, and $Cf_T[59] = 3.09$.

Finally, for $[U-235]_{\text{vent line}}$ using (1) and (3) we obtain $[U-235] = 0.13$ g.

The line source activity for Np-237/Pa-233 in the vent line is determined by analogy to equation (1).

$$[Np-237]_{\text{vent line}} = K_l(\text{cps})(d)(L)Cf_T, \quad (1),$$

where K_l is 1.00×10^{-7} g-sec/cm².⁽⁴⁾ For the line source we again take the average value of all four measurements S_n ($n = 1, \dots, 4$). For the Np-237 content we use the values tabulated for the 312-keV γ -ray in Table 1.

$$(\text{cps}) = [1360/300 + 64/300 + 3067/300 + 2094/300]/4 = 5.49(422). \quad (6)$$

$$[Np-237]_{\text{vent line}} = (1.00 \times 10^{-7})(5.49)(18 \times 2.54)(80 \times 2.54)(2.48) = 0.0126(96) \text{ g.}$$

For Am-241 we obtain

$$[Am-241]_{\text{vent line}} = (6.38 \times 10^{-11})(0.568)(18 \times 2.54)(80 \times 2.54)(3.09) = 1.0(8) \mu\text{g},$$

where the line source calibration constant was calculated in reference 4.

3.2 Line Source Holdup in Acquisition S5 of Oven C

For the vertical vent line of acquisition S5 the line source acquisition distance was 25", and the length of the line source was 36". We use the same correction factors. The single acquisition S5 is used as an ideal line source.

$$[U-235]_{S5} = (1.72 \times 10^{-5})(0.092)(25 \times 2.54)(36 \times 2.54)(2.77) = 0.025(21) \text{ g.}$$

$$[Np-237]_{S5} = (1.00 \times 10^{-7})(0.467)(25 \times 2.54)(36 \times 2.54)(2.49) = 0.67(8) \text{ mg.}$$

$$[Am-241]_{S5} < (6.38 \times 10^{-11})(0.069)(25 \times 2.54)(36 \times 2.54)(3.09) < 80 \text{ ng.}$$

For Am-241 we have calculated an upper limit of content based on 5 times the uncertainty in the background spectrum BG0312. $LLD(\text{cps})_{5\sigma} = 5 \times 97/7000 = 0.069$ cps.

$$LLD(\text{cps})_E = 5(\text{uncertainty}_E \text{ in BG})/7000. \quad (7)$$

3.3 Point Source Holdup in Oven Chamber and in Oil Reservoir

The holdup in the oven chamber (acquisition *Chamb1*) and in the oil reservoir (acquisition *Chamb2*) were calculated for each species using the point source configuration.

$$[U-235] = K_p(d)^2(\text{cps}) Cf_T, \quad (8)$$

where the point source calibration constant is 2.36×10^{-5} , and the acquisition distance was 25 inches.

$$[U-235]_{\text{chamb1}} < (2.36 \times 10^{-5})(0.067)(25 \times 2.54)^2(1.00) < 0.006 \text{ g.}$$

The $LLD(\text{cps})_{186}$ of 0.067 was determined using equation (7). Note the transmission correction factor is 1.00 because there was nothing but air between the source (oven chamber face) and detector.

$$[\text{Np-237}]_{\text{chamb1}} < (1.37 \times 10^{-7})(0.038)(25 \times 2.54)^2(1.00) < 20 \mu\text{g},$$

where 1.37×10^{-7} is the point source calibration constant calculated for Np-237 in reference 4, and $\text{LLD}(\text{cps})_{312} = 0.038$ was calculated using (7).

$$[\text{Am-241}]_{\text{chamb1}} < (8.76 \times 10^{-11})(0.069)(25 \times 2.54)^2(3.09) < 20 \text{ ng}.$$

where 8.76×10^{-11} is the point source calibration constant calculated for Am-241 in reference 4, and $\text{LLD}(\text{cps})_{59} = 0.069$ was calculated using (7).

In the oil reservoir acquisition *Chamb2* we perform the same calculations of upper limit of content using the transmission correction factors measured above for the vent line. The exception here is for the Np-237 content in the oil reservoir, where we observed measurable content.

$$[\text{U-235}]_{\text{chamb2}} < (2.36 \times 10^{-5})(0.067)(25 \times 2.54)^2(2.77) < 0.02 \text{ g}.$$

$$[\text{Np-237}]_{\text{chamb2}} = (1.37 \times 10^{-7})(0.122)(25 \times 2.54)^2(2.49) = 168(47) \mu\text{g}, \text{ and}$$

$$[\text{Am-241}]_{\text{chamb2}} < (8.76 \times 10^{-11})(0.069)(25 \times 2.54)^2(3.09) < 80 \text{ ng}.$$

3.4 Calculation of Relative Am-241/Np-237 Radioactive Content

Note that Np-237 is the radioactive decay daughter of Am-241. Perhaps the observed Am-241 and Np-237 activity were introduced together in a single contamination event. If so, the two would be in a fixed radioactive relationship, and it should be possible to calculate the relative activity of the two species using radioactive equilibrium equations. For a single contamination event the Am-241 activity is defined by

$$A(\text{Am-241}, t) = A_0(\text{Am-241})\exp[-\lambda_1 t], \quad (9)$$

where A_0 is the initial activity at the contamination event, and λ_1 is the radioactive decay constant for Am-241.

The Np-237 activity in radioactive equilibrium is defined by

$$A(\text{Np-237}, t) = [\lambda_2 A_0(\text{Am-241})\exp(-\lambda_1 t) - \lambda_2 A_0(\text{Am-241})\exp(-\lambda_2 t)]/(\lambda_2 - \lambda_1),^{(12)} \quad (10)$$

where λ_2 is the radioactive decay constant for Np-237. Since the half-lives of these two are so vastly different, $\lambda_1 \gg \lambda_2$, and the expression $(\lambda_2 - \lambda_1)$ is approximately equal to $-\lambda_1$. For times less than 100 years $\exp(-\lambda_2 t)$ is very close to 1. Thus the ratio of activities (9)/(10) simplifies to

$$A(\text{Np-237}, t)/A(\text{Am-241}, t) = (\lambda_2/\lambda_1)[1 - \exp(-\lambda_1 t)]. \quad (11)$$

For all times less than 100 years, which is clearly the limit for operations in 321-M, the term in brackets is between 0 and 0.077. At time $t = \text{infinity}$ the expression of equation (11) grows only up to the value λ_2/λ_1 , which is 0.0002. Thus at all times the radioactive daughter Np-237 activity must be much less than the Am-241 activity. Since the observed activities range from $A(\text{Np-237}, t)/A(\text{Am-241}, t) = 0.55$ in chamb1 to 9.67 in the vent line, we see that the Np-237 activity is *not* much less than the Am-241 activity. Thus the observed Np-237 activity could not have been formed only from decay of Am-241, and must have been introduced from two independent sources.

3.5 Holdup in Oven D

The calculations of holdup in oven D were performed in the identical manner as those of oven C in sections 3.1 - 3.3. For these acquisitions we used the NaI detection system, and the calibration constants determined for it.⁴ To use the NaI detector it was first necessary to demonstrate that we could observe and resolve the γ -rays from each of U-235, Np-237/Pa-233, and Am-241. To do that we obtained a NaI spectrum of the oven C horizontal vent line from a distance of 24 inches. This line source spectrum is shown in Figure 3 and very clearly shows the peaks from Am-241 and the 312 - 340 keV doublet from Np-237/Pa-233. The qc check spectrum showing the Cs-137 peak at 662 - keV is displayed in Figure 4.

We then calculated the line source and point source calibration constants for Am-241 and Np-237 for the NaI detection system using the techniques recorded in reference 9 and described in reference 4. The detector had already been efficiency calibrated for U-235.⁽¹³⁾ The point and line source calibration constants we measured for U-235 and calculated for Np-237/Pa-233 and for Am-241 are listed in Table 3. Using the constants of Table 3 and the measured rates of Table 2, (in units of cpm for the NaI acquisitions) we calculated the contents of each species in a manner identical to those of sections 3.1 - 3.3. For each of Np-237 and Am-241 we obtained limits of detection only. It was not necessary to obtain a background for these spectra. Instead we used the square root of the integrated area in the appropriate regions of interest as the one sigma standard deviation of the background in that region of interest. The line source limit of detection is then determined by five times that. For example

$$[\text{Np-237}]_{\text{vent line}} = K_i(\text{cps})(d)(L)\text{Cf}_T, \quad (1),$$

$$[\text{Np-237}]_{\text{S2}} < (7.07 \times 10^{-9})(5 \times 4.44)(15)(80)(2.49) < 0.5 \text{ mg.}$$

Table 3.	NaI Calibration Constants Used for Np-237, U-235, and Am-241 Calculations (g-min/in ²).		
	U-235	Np-237	Am-241
point	1.67×10^{-6}	9.69×10^{-9}	4.15×10^{-12}
line	1.22×10^{-6}	7.07×10^{-9}	4.51×10^{-12}
area	9.77×10^{-7}	3.81×10^{-9}	2.43×10^{-12}

Without showing the calculations, we obtained for oven D

$$[\text{U-235}]_{\text{vent line}} = 0.04(1) \text{ g, and}$$

$$[\text{Am-241}]_{\text{vent line}} < 400 \text{ ng.}$$

$$[\text{U-235}]_{\text{S5}} = 0.065(36) \text{ g.}$$

$$[\text{Np-237}]_{\text{S5}} = 0.3(1) \text{ mg.}$$

$$[\text{Am-241}]_{\text{S5}} < 80 \text{ ng.}$$

$$[\text{U-235}]_{\text{chamb1}} < 0.03 \text{ g.}$$

$$[\text{Np-237}]_{\text{chamb1}} < 20 \text{ }\mu\text{g.}$$

$$[\text{Am-241}]_{\text{chamb1}} < 20 \text{ ng.}$$

$$[\text{U-235}]_{\text{chamb2}} < 0.03 \text{ g.}$$

$$[\text{Np-237}]_{\text{chamb2}} < 200 \text{ }\mu\text{g.}$$

$$[\text{Am-241}]_{\text{chamb2}} < 100 \text{ ng.}$$

4.0 CONCLUSION

We have used γ -ray acquisitions with HPGe and NaI portable detection systems to perform holdup assays for HEU, Np-237/Pa-233, and Am-241 in the C and D out-gassing ovens of Building 321-M. The assays were performed in the line source and point source transmission corrected configurations to report content of these three actinides. The assays were necessary for correct disposition of the process ovens as radioactive solid waste.

After the unexpected Np-237 and Am-241 activity were observed in RCO screening surveys, the Np-237 and Am-241 contents were very important to the Facilities Disposition Program to determine whether a separate solid waste stream was required for disposal of these ovens. The results of the assays were reported in a fast turn around time frame of less than one week. The measured values in oven C were reported in references 5, 6, 7, and 9 and were 0.6 - 2.6 μg Am-241, 9 - 36 mg Np-237, and < 0.2 g HEU. The measured value in oven D were reported in reference 8 and were < 0.4 μg Am-241, < 0.5 mg Np-237, and 0.105 ± 0.048 g HEU.

This report documents the assay acquisitions and the calculations performed to obtain the measured values reported. From the relative detection rates of the γ -rays from Am-241 and Np-237/Pa-233, we were able to demonstrate that the Am and Np activity were not consistent with a parent/daughter radioactive relationship. So the contamination in the ovens came from two separate incidents or from one incident that contained separate sources of Am and Np activity. Since the two were not in radioactive pre-equilibrium, it was necessary to obtain calibration constants for both species. The calibration constants for the calculations of this report were performed from first principles. These calculations are documented separately along with an experimental measurement of the point source and area source calibration constants for Np-237/Pa-233.

The measured values, acquisitions, and calculations reported here represent important first steps in the demonstration of the capability at the Savannah River Site to deactivate and decommission a radioactive DOE weapons production facility.

5.0 References

1. Deactivation Project Plan 321-M Fuel Fabrication Facility, V-PMP-M-00004, January 2000.
2. Ron Smith, In-Situ Object Counting System Radiation Control Operations survey, February 2003.
3. WSRC 1S Manual, Procedure 3.17, Section A.3.a, "E-Area Vaults Nuclear Safety," July 2000.
4. R. A. Dewberry, "Calculation of Np-237 and Am-241 Detector Calibration Constants From First Principles", **Journal of Radioanalytical and Nuclear Chemistry**, Vol. 262, No. 3 (2004) 783 – 787.
5. R. A. Dewberry, "Np-237 Content in Outgas Oven", SRT-ADS-2003-00175, March 2003.
6. R. A. Dewberry, "Am-241 Content in Outgas Oven", SRT-ADS-2003-00176, March 2003.
7. R. A. Dewberry, "HEU Content in C Oven", SRT-ADS-2003-0345, June 2003.
8. R. A. Dewberry, "Actinide Contents in D Oven", SRT-ADS-2003-00272, April 2003.
9. R. A. Dewberry, Laboratory Notebook NB-2000-00086, pages 155 – 161.
10. R. A. Dewberry and S. R Salaymeh, "Efficiency Calibration Using HEU Standards of 2" x 2" NaI Detector," WSRC-TR-2000-00269, July 2000.
11. 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.
12. Gerhart Friedlander, Joseph Kennedy, and Julian Miller, Nuclear and Radiochemistry, (John Wiley & Sons, New York, 1949).
13. R. A. Dewberry, "Method Development Report for Use of the Portable NaI Detection System for Assay of Highly Enriched Uranium", SRT-ADS-2003-00124, May 2003.

Table 1. **Summary of HPGe Acquisitions Obtained on Oven C.**

[illegible]

Table 2. **Summary of NaI and HpGe Acquisitions Obtained on Oven D.**

[illegible]