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Neutron Multiplicity and Active Well Neutron Coincidence Verification Measurements Performed for March 2009 Semi-Annual DOE Inventory

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Introduction

The Analytical Development (AD) Section field nuclear measurement group performed six "best available technique" verification measurements to satisfy a DOE requirement instituted for the March 2009 semi-annual inventory.[1] The requirement of (1) yielded the need for SRNL Research Operations Department Material Control & Accountability (MC&A) group to measure the Pu content of five items and the highly enrich uranium (HEU) content of two. No 14Q-qualified measurement equipment was available to satisfy the requirement. The AD field nuclear group has routinely performed the required Confirmatory Measurements for the semi-annual inventories for fifteen years using sodium iodide and high purity germanium (HpGe) γ-ray pulse height analysis nondestructive assay (NDA) instruments.[2] With appropriate γ-ray acquisition modeling, the HpGe spectrometers can be used to perform verification-type quantitative assay for Pu-isotopics and HEU content. The AD nuclear NDA group is widely experienced with this type of measurement and reports content for these species in requested process control, MC&A booking, and holdup measurements assays Site-wide.[3-6] However none of the AD HpGe γ-ray spectrometers have been 14Q-qualified, and the requirement of reference 1 specifically excluded a γ-ray PHA measurement from those it would accept for the required verification measurements.

The requirement of reference 1 was a new requirement for which the Savannah River National Laboratory (SRNL) Research Operations Department (ROD) MC&A group was unprepared. The criteria for exemption from verification were[7]

- isotope content below 50 grams
- intrinsically tamper indicating or TID sealed items which contain a Category IV quantity of material
- assembled components
- laboratory samples.

Therefore all (SRNL) Material Balance Area (MBA) items with greater than 50 grams total Pu or greater than 50 grams HEU were subject to a verification measurement. The pass/fail criteria of reference 7 stated "The facility will report measured values, book values, and statistical control limits for the selected items to DOE SR…", and "The site/facility operator must develop, document, and maintain measurement methods for all nuclear material on inventory". These new requirements exceeded SRNL's experience with prior semi-annual inventory expectations, but allowed the AD nuclear field measurement group to demonstrate its excellent adaptability and superior flexibility to respond to unpredicted expectations from the DOE customer. The requirements yielded five SRNL items subject to Pu verification and two SRNL items subject to HEU verification. These items are listed and described in Table 1.

Analytical Development Neutron Multiplicity Capabilities

AD offered to the ROD MBA custodian a possible "best available technique" solution to the unexpected verification requirements. The multiple Director's-Award-winning nuclear nondestructive assay facility was in possession of three passive neutron multiplicity assay instruments that were acquired specifically for determination of Pu items. Two of the instruments had been procured from the Deactivation and Decommissioning of the Rocky Flats Environmental Test Site (RFETS), and the third was obtained to perform active well neutron coincidence analyses of U-Al fuel product and was also capable of passive neutron multiplicity assay of Pu items.[8,9] AD suggested to the SRNL MBA custodian that we bring the capabilities of these instruments to bear on the required Pu and HEU verification measurements.

The Measurement Control Program (MCP) for each of the three instruments [RFETS 3013 can counter, Large Neutron Multiplicity drum counter, and Aquila Active Well Coincidence Counter (AWCC)] are documented in SRNL-ADS-2008-00171[(10)] and SRNL-ADS-2008-00197[(11)]. Quality control (QC) checks are described in reference 11 and are presented in the maintenance logbooks and Laboratory Notebooks referenced therein. While none of the three instruments is 14Q-qualified for neutron multiplicity counter (NMC) verification measurements of Pu content, both passive and active QC checks are prescribed and conducted, and the passive QC checks on all three instruments are extremely thorough – testing five distinct measurement parameters. The Aquila (AWCC) is 14Q-qualified for HEU verification in the range 247 – 1198 g HEU, and has been used to conduct multiple required verifications of U-Al HEU content in that range in K-Assembly inventories in FY00 – FY05.[9,12,13] The customer agreed to have the measurements conducted, and AD and the Actinide Chemical Technology (ACT) group collaborated with considerable effort to move the five 50-material items to the AD nuclear NDA laboratory.

Table 1. SRNL SNM inventory items meeting requirements for March 2009 Verification Measurements.

Item Number	Declared Content SNM	Description
50014668	101.4 gram Pu	Pu metal from FB-Line 3/99
50015480	60.29 gram Pu	Pu oxide from FB-Line 1/99
50015426	59.25 gram PU	Pu oxide from FB-Line 1/99
50015514	64.0 gram Pu	Pu oxide from LANL 11/07
2002026A	102 gram HEU	Pillow-like assimilation of U-Al solid
FI001	99 gram HEU	U-Al slab with no flexibility

The accuracy and precision of all three instruments to perform passive NMC measurements of items of total Pu mass 0.78 g and 7.2 g with known isotopics is documented in reference 8. The passive NMC technique measures neutron coincidence events to determine effective Pu-240 content, and total Pu is predicted from the known isotopic content. Thus, the uncertainty of the total Pu measurement is directly related to the uncertainty in the known isotopic distribution of the sample. Such isotopics is established generally with a γ-ray acquisition, but had not been established for four of the five 50-material items of Table 1. The isotopic information available for each is shown in Table 2.

Table 2. Isotopic Information available for the SNM items of Table 1.

Item Number	Pu-238 Fraction	Pu-239 Fraction	Pu-240 Fraction	Pu-241 Fraction	Unaccounted Pu Fraction	U-235 Enrichment
50014668		0.927	0.059		0.014	
50015480		0.94	0.0153	0.0002	0.0445	
50015426		0.932	0.0174	0.00152	0.049	
50015514		0.9372	0.0628		0	
2002026A						0.6581
FI001						0.5823

Pu Measurement Results

AD analyzed the five 50-material items in each of the three NMC instruments in its possession in the passive NMC mode for determination of Pu-240 and total Pu content. The enriched uranium item 2002026A was analyzed in the Aquila AWCC instrument in the active mode for determination of HEU content. For both passive and active neutron coincidence analyses, the instrument acquisition parameters are extremely important. Coincidence gate width, deadtime correction equations, instrument counting efficiency, and doubles- and triples-gate fractions directly affect the analytical results for passive NMC analysis. The coincidence gate width and correction for accidental coincidence directly affect the analytical results for active coincidence analyses. All of these had been previously accurately determined experimentally for all three instruments. The acquisition parameters are not listed in this report but are documented in Laboratory Notebook WSRC-NB-98-00064[(14)] and appear in the hard copy printed output of all results.[15] Individual measured values for each Pu content from the three instruments appear in Table 3. We discuss results thoroughly below.

Table 3. Passive NMC measurement results from all three instruments. The last column lists the average and 1-σ standard deviation.

Item Number	Declared Mass (g)	Can NMC Result (g)	LNMC Result (g)	AWCC Result(g)	Average
50014668	101.4	68.87±0.6	61.7±1.6	86.4±0.5	72.3±12.9
50015480	60.29	32.1±0.6	27.1±0.9	44.2±1.0	34.5±8.8
50015426	59.25	24.4±0.3	21.1±0.6	31.5±0.5	25.7±5.3
50015514	64.0	56.6±2.1	47.5±2.4	74.0±1.9	59.4±13.5

Discussion of Pu Results

The Pu results for items 50015480, 50015426, 50014668, and 50015514 used declared Pu-240 in the analyses. For item 0015428 we assumed all the unaccounted Pu fraction was Pu-240. For items 50015480 and 50015426 we assumed all the unaccounted Pu fraction was Pu-238. This approach seemed far more rational than making an assumption regarding the Pu-241 fraction. Therefore for each NMC measurement (excluding item 50015514) our largest uncertainty was in the assumed Pu-240 fraction. Further, we had no reliable technique to determine that uncertainty other than an arbitrary declaration of it. Our declaration of uncertainty in the Pu-240 fraction is listed in column 10 of the results table (Table 4).

The overall results and uncertainty for the Pu analyses are reported in columns 11 and 13 of Table 4. The reported measured mass is the average of the three measurements for each item. The overall uncertainty is a quadratic sum of the individual 1- σ uncertainties of columns 4, 6, 8, 10, and 12 where the uncertainty in column ten is converted to a mass uncertainty as a fraction of Pu-240 content times the average. That is, for the mass uncertainty component arising from the uncertainty in the Pu-240 fraction of item 50015428

$$(\sigma_{\text{mass}})_{\text{Pu-240 uncertainty}} = [(\sigma \text{ Pu-240 fraction})/(\text{Assumed Pu-240 fraction})][\text{Average measured mass}]. \quad (1)$$

$$= (3.8/14.7)(73.4) = 19.0 \text{ g.}$$

And the overall uncertainty is the quadratic sum of that component and those of columns 4, 6, 8, and 12. That is

$$\text{Overall } (\sigma_{\text{mass}})_{\text{Pu-240 uncertainty}} = \text{SQRT}[(2.9)^2 + (1.5)^2 + (1.7)^2 + (19.0)^2 + (13.9)^2] = 23.8 \text{ g}, \quad (2)$$

as denoted in column 13.

The combined limit of error (CLOE) is determined with a two-σ uncertainty. Since the declared mass has no designated uncertainty, we used simply √(2) times the two- σ uncertainty of the measured values of equation (2). Then the CLOE for item 50015428 becomes

$$\text{CLOE} = 2(\text{Overall } (\sigma_{\text{mass}})_{\text{Pu-240 uncertainty}})\sqrt{(2)} = 2(23.8)\sqrt{(2)} = 67 \text{ g.} \quad (3)$$

The CLOE's for each item are listed in the last column of Table 4. While the resulting CLOE's are all quite large, note all six verification measurements are successful. Even if we eliminate the component of uncertainty from Pu-240 isotopics (column 10), three of the six items were successfully verified.

HEU Measurements
The Aquila active well neutron coincidence counter is approved for verification analyses of HEU content in U-Al material in the range 247 – 1198 g HEU. While that range does not include the declared masses of the two 20-material HEU items in Table 1, the measured doubles rates can be fit onto the calibration curve of the INCC active well. That calibration curve is

$$D(eps) = 0.190375m - 3.67363x10^{-5}m^2, \qquad (4)$$

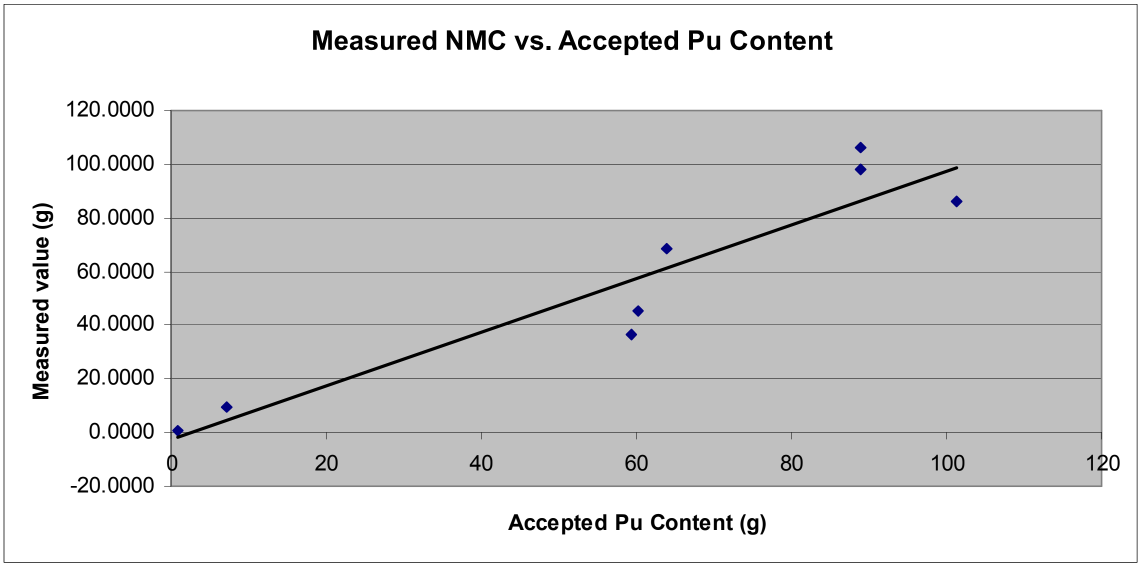
where D is the measured neutron coincidence doubles rate in events/sec, and m is the mass in grams of U-235 content in the item. The measured doubles rate for item 2002026A is 24.79±2.5 eps. Including the variance of the coefficients in (4) yields a measured value of [HEU] = 133±14 g for item 2002026A. As shown in the last row of Table 4, this uncertainty places the measurement well within our equation (3) determination of CLOE and represents a successful verification.

Item FL001 was inflexible and would not fit into the assay chamber of the AWCC. It could not be measured.

Quality Control Checks
As required by the neutron multiplicity and active well Measurement Control Plans[9,10] each instrument was rigorously QC checked both before and after the verification measurements. All three satisfied all five of the prescribed QC checks. Because the SNM items themselves significantly raised the neutron background in the AD nuclear non-destructive assay facility, it was necessary to remove the samples in order for the LNMC to pass the after checks. All QC checks are stored in the instrument data bases.

Qualification of the AWCC for Portable NMC Pu Verification Measurements.

SRNL was subsequently requested to qualify the portable AWCC for passive NMC determination of Pu in the range 50 g – 100 g for the March 2010 Semi-Annual DOE inventory.[16] To accomplish that, we obtained the data listed in Table 5 with eight Pu standards. These eight had Pu-$240_{effective}$ isotopic contents that varied in the range 0.036 – 0.108[(17)]. The results are shown in Table 5 and are plotted in the Figure.



Verification Measurements with eight standards used for qualification of the portable active well coincidence counter for passive neutron multiplicity measurements of Pu in the range 0.78 – 101 g.

Conclusion

The Analytical Development field nuclear measurement group was able to devise a "best available technique" to obtain successful verification measurements to six items of SNM. These neutron multiplicity and neutron activation measurements satisfied the DOE requirement for verification to which SRNL was otherwise unable to comply on short notice.

References

1. PT-31JUL08-SR-3800-SSPS-NMCA.2-003.
2. V. R. Casella, "Gamma Assays Using Ortec's ISOTOPIC Program", ADS 2460, L16.1, February 2006.
3. R. A. Dewberry, V. R. Casella, R. A. Sigg, and N. N. Bhatt, "Benchmarking Ortec ISOTOPIC Measurements and Calculations", SRNS-STI-2008-00053, accepted for publication in **J. Radioanalytical and Nuclear Chemistry**, March 2009.
4. R. A. Dewberry, V. R. Casella, R. A. Sigg, S. R. Salaymeh, F. S. Moore, D. J. Pak, "Holdup measurements for three visual examination and TRU remediation glovebox facilities at the Savannah River Site", **J. Radioanalytical and Nuclear Chemistry**, 275, 3 (December 2008).
5. R. A. Dewberry, K. P. Klapper, and S. R. Salaymeh, "Measured SNM Np-237 Content in ATS Legacy Material", WSRC-TR-2007-00270, October 2007.
6. R. A. Dewberry and C. C. DiPrete, "Measured SNM U-235 Content in ATS Confirmatory Observations to Benchmark ISOTOPIC Application", WSRC-TR-2008-00123, July 2008.
7. DOE M 470.4-6, II.3.a.(1).
8. S. R. Salaymeh, R. A. Dewberry, and W. H. Geist, "Results of a Performance Demonstration Study to Evaluate the KAMS Neutron Multiplicity Counter for Receipt Measurements of Rocky Flats Environmental Technology Site Pu Material", WSRC-MS-2004-00653, September 2004.
9. R. A. Dewberry and S. R. Salaymeh, "Diverse Active Well Neutron Coincidence Counter Utility at the Savannah River National Laboratory, J. Radioanalytical and Nuclear Chemistry, Vol. 277, No. 2, (2008).
10. R. A. Dewberry, "Neutron Nuclear Non-Destructive Assay Instrument Components, SRNL-ADS-2008-00171, April 2009.
11. R. A. Dewberry, "MCP FOR Nuclear Non-Destructive Assay Neutron Multiplicity Counting Systems", SRNL-ADS-2008-00197, October 2008.
12. R. A. Dewberry and S. R. Salaymeh, "Method Development Report for Assay of U-235 Samples in K-Area Storage Facility", SRNL-ADS-2006-00286, May 2006.
13. J. R. Ludwick, "Request for Approval of the AWCC for Uranium Verification Measurements", SEO-MCA-2006-00154, May 2006.
14. R. A. Dewberry, Laboratory Notebook, NB-98-00064, February 1998.
15. B. Harker and M. Krick, INCC 5.0 software, USDOE Office of Safeguards and Security, (1997).
16. Savannah River National Laboratory STAR Item 2009-CTS-002746 - #003, November 2009.
17. R. A. Dewberry, "Method Development Report for Assay of SRNL Pu Samples by the Aquila Active Well Coincidence Counter", SRNL-L4000-2009-00056, January 2010.
18. R. A. Baker, "Qualification Data Analysis for the Savannah River National Laboratory (SRNL) Aquila Active Well Coincidence Counter (AWCC) (U)", SEO-MCA-2010-00023, February 2010.

Table 4. Summary of the passive NMC and active AWCC verification measurements.

		AWCC Measurements		LNMC Measurements		RFETS NMC Measurements		%Pu-240	Assumed Pu-240% uncertainty	average Pu mass	St dev average Pu mass	overall 1-σ uncertainty	Declared - Measured	CLOE
Item#	Book Value	Measured Pu Mass (g)	Uncertainty 1-σ	Measured Pu Mass (g)	Uncertainty 1-σ	Measured Pu Mass (g)	Uncertainty 1-σ							
50014668	101.4	69.8	0.4	49.9	1.3	55.5	0.5	7.3 assumed	3.3	**58.4**	10.3	**28.4**	43.0	**80.2**
50015426	59.25	31.5	0.5	21.1	0.6	24.4	0.3	**1.74** declared	1.5	**25.7**	5.3	**22.8**	33.6	**64.4**
50015428	58.08	88.5	2.9	61	1.5	70.7	1.7	14.7 assumed	3.8	**73.4**	13.9	**23.8**	-15.3	**67.4**
50015480	60.29	44.2	1	27.1	0.8	32.1	0.6	**2.53** declared	1.8	**34.5**	8.8	**26.1**	25.8	**73.8**
50015514	64.0	74	1.9	47.5	2.4	56.6	2.1	6.28 declared	0	**59.4**	13.5	**14.0**	4.6	**39.5**
		AWCC Measurements		LNMC Measurements		RFETS NMC Measurements						**overall 1-σ uncertainty**	**Declared - Measured**	**CLOE**
		Measured HEU Mass (g)	Uncertainty 1-σ	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A			
2002026A	102	133	14									**14**	-32	**40**