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U.S. SUPPORT PROGRAM TASKS

U.S. Objectives

To inform the IAEA of the status of work on Support Program tasks.

Background

There are presently five U.S. Support Program tasks at work. Three are complete, and two are underway. Reports are available from two of the completed SP-1s; a draft is in preparation for the third. These tasks are:

1. Plutonium scrap multiplicity counter at Hanford
2. Calorimeter authentication at Hanford
3. Large neutron multiplicity counter at Rocky Flats
4. Calorimeter authentication at Rocky Flats
5. Safeguards Approach Support at the APSF, SRS

Status

The status of the first four tasks above is described below. Information on the work at Savannah River is contained in a separate paper.

3. Large neutron multiplicity counter at Rocky Flats

Summary

In the fall of 1993, President Clinton announced before the United Nations General Assembly, that the United States would voluntarily offer excess fissile material of weapons origin to International Atomic Energy Agency (IAEA) safeguards. The first US facilities accepted under this offer were a storage vault containing highly enriched uranium at Oak Ridge National Laboratory and a second vault containing plutonium oxide and rich plutonium residue at the Hanford Plutonium Finishing Plant. The third facility was a vault at the Rocky Flats Environmental Technology Site (RFETS).

Experience during the Initial Physical Inventory Verification (IPIV) at Hanford led the IAEA to conclude that US excess weapons plutonium was not pure enough to be verified successfully using traditional coincidence counting techniques. At Hanford, neutron multiplicity counting was necessary to successfully complete the IPIV¹. At RFETS, however, the excess plutonium is stored in ten-gallon drums. At the time that the RFETS vault was accepted for IAEA safeguards verification, no multiplicity counter existed that could accommodate this size container. Further, there was no possibility of repackaging the RFETS oxide to fit existing instruments.

Concurrently, an experimental prototype neutron multiplicity counter (NMC) was

being designed at Los Alamos National Laboratory to investigate the measurement of bulk plutonium and weapons components stored in 30-gal drums². Because of the pressure to meet the Presidential Directive, this instrument was re-directed to RFETS for use in their IPIV in December of 1995.

Because of a tight schedule, it was not possible to test this instrument using actual plutonium standards before its delivery to RFETS. The instrument's performance parameters were characterized at Los Alamos using National Institute of Standards (NIST) certified ²⁵²Cf sources. The calibration plan called for final acceptance of the instrument using RFETS plutonium standards. However, it proved impossible for RFETS to deliver these neutron standards in an unmoderated condition to the instrument prior to the IPIV. As such, the LNMC, is the first NMC to ever be characterized and used in an inspection without calibration with plutonium standards. Instead the californium based characterization was "tweaked" to plutonium using cross calibration measurements in the LNMC's identical twin instrument currently at the Lawrence Livermore National Laboratory (LLNL)³. Calibration was completed in November of 1997 with the IAEA's working standards derived from the RFETS inventory by destructive analysis (DA).

The design and performance of the LNMC has been published in four papers. Reference 2 reports on the design of the instrument. Reference 3 reports on the characterization measurements for both the RFETS and LLNL instruments. Reference 4 reports on the performance of the RFETS LNMC during the IPIV. Finally, Reference 5 provides conclusions on the instrument's performance during both the IPIV and the first annual Physical Inventory Verification (PIV) and compares destructive analysis results from 25 drums with the LNMC's assays of these drums. This last report also discusses the one drum that the LNMC has not been able to measure successfully, Drum 012.

Since the publication of these reports, the second annual PIV has occurred adding 13 measurements to the database of sample measurements made with the LNMC. These additional measurements include the measurement of two of the three IAEA working standards, bringing the number of measurements of these standards to six. With these additional data, the investigation of the calibration of the instrument has now been completed.

Timeline of Events for the LNMC Authentication

A chronology of the milestones and achievement dates for the authentication of the LNMC is given below:

November 1994 : RFETS requests LANL to fabricate a 30-gallon Drum Neutron Multiplicity Counter or LNMC for use in the Initial Physical Inventory Verification of excess weapons plutonium by the IAEA then scheduled for March of 1995.

February 1995: The LNMC was characterized using LANL ²⁵²Cf sources including BB-996 and BB-998, National Institute of Standards (NIST) certified sources. The LNMC

was delivered to RFETS and installed in room 3541 of building 371.

March 1995: A large uncertified ^{252}Cf source belonging to RFETS was measured in the LNMC. Its doubles to singles ratio and its triples to singles ratio confirmed that the LNMC was operating nominally. (Note these ratios are independent of source strength and depend only on detector parameters.) The IPIV was delayed due to a delay in vault modifications.

April 1995: RFETS brings standards to the LNMC. Unfortunately they were packed in moderator and could not be used. RFETS was unable to repackage the standards in an unmoderated container.

April 1995: LLNL's twin 30-gallon Drum Neutron Multiplicity Counter was characterized at LANL using LANL ^{252}Cf sources including NIST certified source, CR-6.

May 1995: LLNL's 30-gal Drum NMC was further characterized at LANL by measuring LANL's LAO and PEO standards. Its preliminary calibration parameters were adjusted based on these measurements. The same adjustment was applied to the calibration parameters for the LNMC at RFETS.

June 1995 - November 1995: RFETS measured 8, 10-gal drums that were representative of potential inventory to be offered. The results were analyzed using the adjusted calibration parameters based on measurements in the LLNL counter. These assays agreed well with RFETS book values that were based on CAL-ISO measurements.

September 1995: A multiplicity workshop and hands-on training of the operation LNMC were provided to IAEA inspectors and RFETS personnel by a LANL technical expert.

September 1995: A paper, "The Application of Neutron Multiplicity Counting to the Assay of Bulk Plutonium Bearing Materials at RFETSS and LLNL," was presented at the American Nuclear Society "Fifth International Conference on Facility Operations-Safeguards Interface," Jackson Hole, Wyoming. This paper summarized the characterization of both the LLNL and RFETS instruments.

December 1995: The Initial Physical Inventory Verification was performed. Over 100 drums were tested for partial or gross defect in the LNMC. The latter were of lead-lined drums that could not be tested using gamma-ray measurements. Only one drum, 012, failed to be verified due to a neutron emission rate so high that the multiplicity shift register could not measure it. The oxide in this drum was destructively analyzed. During the IPIV an IAEA owned ^{252}Cf source normally used during Hanford inspections, K-869, was measured to confirm the counter's correct operation. During the IPIV inspectors received further training on the LNMC from a LANL technical expert and preliminary operation procedures were drafted and tested.

February 1996: A meeting was held with the IAEA at LANL to review results and to

plan for cross-calibration of an IAEA ^{252}Cf source for RFETS with LANL's NIST certified sources.

June 1996: A new IAEA ^{252}Cf source, ff-292, was cross-calibrated at LANL with LANL's NIST certified sources and IAEA personnel present. It was measured in the Three Ring Multiplicity Counter used in the Hanford inspections to also compare it to K-869. The source was sealed and shipped to RFETS.

July 1996: A paper, "The Performance of the 30-gallon Drum Neutron Multiplicity Counter at Rocky Flats Environmental Technology Site," was presented at the 37th Annual Meeting of the Institute of Nuclear Materials Management in Naples, Florida. This paper reported on the measurements made with the LNMC both prior to and during the IPIV.

October 1996: The first annual PIV was successfully performed. The instrument was authenticated with the new source, ff-292, and procedures were updated by a LANL technical expert to include this new source. A new feature in the software, the ability to assay to a desired precision, was also tested. A subset of the destructive analysis (DA) results performed by the IAEA on a samples of the RFETS inventory was delivered to LANL personnel. At this time all of the DA had not been completed due to transportation difficulties.

June 1997: The remaining DA results were delivered to LANL.

July 1997: A paper, "Assay of Impure Plutonium Oxide with the Large Neutron Multiplicity Counter for IAEA Verification of Excess Weapons Material at Rocky Flats Environmental Technology Site," was presented at the 38th Annual Meeting of the Institute of Nuclear Materials Management in Phoenix, AZ. This paper summarized the measurements made to this date and included a comparison of the instrument's performance to DA results. A conclusion made by this paper was that the LNMC might have a very small bias in its calibration parameters. Plans were made to investigate this further at the PIV in November 1997.

November 1997: The second annual PIV was successfully conducted. In addition two of the IAEA's working standards were measured over-night to complete an investigation of calibration bias. A LANL technical expert trained inspectors in the use of the new neutron coincidence software, INCC, with the LNMC.

February 1998: The analysis of the LNMC's calibration parameters based on the IAEA's working standards was completed by LANL. This final report was written.

Measurements to Date

In the course of the IPIV and two annual PIV's, 131 measurements of 113 drums from the RFETS inventory have been successfully performed. On average the LNMC has agreed to within 0.6% of the declared mass for these measurements with an

average standard deviation of 3.4%. For the conglomerate of measurements, summarized in Table 1, the assay bias compared with declared drum plutonium mass has been within two standard deviations of the mean

Table 1. Summary of assay performance (A) compared to operator declared mass (D) for the LNMC for 131 measurements of 113 drums at RFETS.

Average (D-A) / D	0.58%
Standard Deviation	3.35%
Standard Deviation of the Mean	0.32%
# Sigmas	1.9

Comparison of the LNMC assay results for a total of six measurements of the three IAEA working standards gives a bias of 0.97 % between the mass obtained by destructive analysis and the assay mass with a standard deviation in the mean of 0.40 %. The bias is more than 2 standard deviations from the mean and thus the probability that this bias is real is about 99%. These results are summarized in Table 2.

Table 2. Comparison of LNMC assay with plutonium mass for three IAEA working standards from the RFETS inventory.

Standard Number	Destructive Analysis / Operator Declared Mass (%)	(Destructive Analysis Mass - Assay) / Assay (%)	Sigma
1	1.0034	-0.336	2.221
		-2.547	1.661
2	0.9952	0.980	0.016
		-1.671	0.265
3	0.9908	1.009	0.045
		4.280	5.490
	Average	0.974	
	Standard Deviation of the Mean	0.403	
	# Sigmas	2.419	

Final Suggested Detector Parameters

The small, statistically significant bias observed in the measurements of the IAEA working standards suggests that the detector's efficiency should be adjusted slightly to remove this bias. The present value being used is 0.4171. To correct for the bias in the standards measurements, this should be changed slightly to 0.4136. The most likely source of the bias is the difference in the packaging between the standards used to calibrate the LNMCC's LLNL twin and those at RFETS. The IAEA working standards have significantly more iron in their packages than the standards used to calibrate LLNL's LNMCC, and there is additionally small amounts of polyethylene surrounding the inner oxide cans. Both of these factors would account for this very slight decrease in detector efficiency relative to the calibration of the LLNL instrument. No other changes in detector parameters are suggested. The remaining detector parameters are summarized in Table 3 below. Table 4 below summarizes the corrected assay results.

Table 3. Summary of suggested final LNMCC calibration parameters as they should appear in the Measurement Parameters screen under the Setup menu of the INCC code.

Shift Register type	MSR4 or 2150	
Serial Port	COM1	
Predelay	3	
Gate Length	32	
High Voltage	N/A	
Die away time	55.4	
Efficiency	0.4136	
Multiplicity Deadtime	25.02	
Deadtime coefficient	A	0.05739
	B	0.03405
	C	0.0
	D	0.0
Doubles gate fraction	0.4100	
Triples gate fraction	0.1794	

Table 4. Summary of corrected assay performance (A) compared to operator declared mass (D) for the LNMC for 131 measurements of 113 drums at RFETS.

Average (D-A) / D	-0.259 %
Standard Deviation	3.375 %
Standard Deviation of the Mean	0.318 %

Next Steps - LANL will provide technical support for this instrument upon request of the IAEA.

Talking Points -- Calibration and authentication of the LNMC is complete. The LNMC is the first multiplicity instrument to be used in an IAEA inspection without calibration based on direct measurement of primary plutonium standards. As such, its success in measuring over 100 drums from the RFETS inventory to an average agreement of 0.6% with operator declarations is excellent!

Drum 012, unscreened by RFETS prior to the IPIV, is the only drum that has been unmeasurable by the LNMC due to its excessive neutron emissions. These emissions are estimated to be in excess of 6.3×10^6 n/s based on measurements made with half of the LNMC detection banks turned off. Thus, handling of this drum will result in significant radiation exposure to both IAEA inspectors and RFETS personnel and should be avoided if at all possible. The only measurement technique that can assay the materials in this drum, other than destructive analysis, is calorimetry, and this is not an option as long as these materials are packaged in the ten-gallon drum. Once these materials are stabilized and re-packaged, NDA by calorimetry will be possible. However, the radiation hazard will likely remain.

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