Conf-950787--64

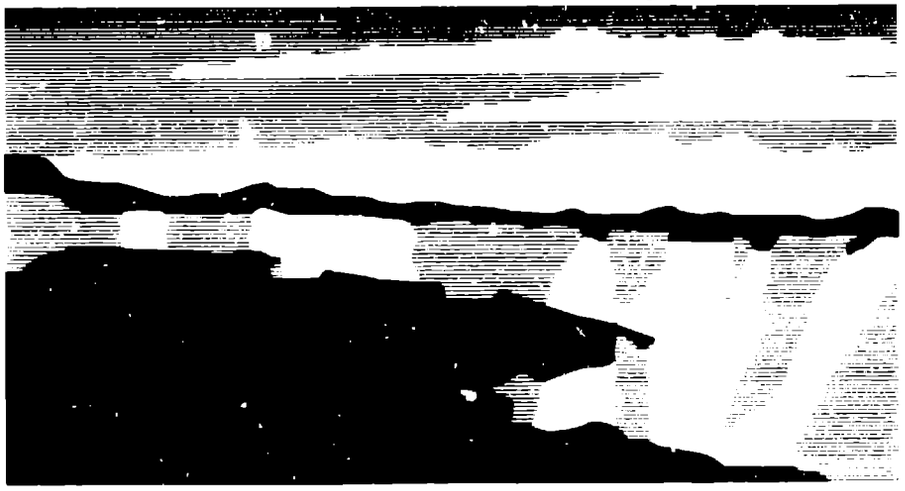
LA-UR- 95 -2329

Title: **IMPLEMENTATION OF NEUTRON COUNTING TECHNIQUES AT U.S. FACILITIES FOR IAEA VERIFICATION OF EXCESS MATERIALS FROM NUCLEAR WEAPONS PRODUCTION**

Author(s): **J. E. Stewart, M. S. Krick, D. G. Langner, T. D. Reilly, W. Theis, R. J. Lemaire, and J. Xiao**

Submitted to: **36th Annual INMM Meeting
Palm Desert, California USA
July 9-12, 1995
(FULL PAPER)**

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED



Los Alamos
NATIONAL LABORATORY

IMPLEMENTATION OF NEUTRON COUNTING TECHNIQUES AT U. S. FACILITIES FOR IAEA VERIFICATION OF EXCESS MATERIALS FROM NUCLEAR WEAPONS PRODUCTION*

J. E. Stewart, M. S. Krick, D. G. Langner, and T. D. Reilly
Los Alamos National Laboratory
Los Alamos, NM 87545

W. Theis, R. J. Lemaire, and J. Xiao
International Atomic Energy Agency
Vienna, Austria

ABSTRACT

The U.S. Nonproliferation and Export Control Policy, announced by President Clinton before the United Nations General Assembly on September 27, 1993, commits the U.S. to placing under International Atomic Energy Agency (IAEA) Safeguards excess nuclear materials no longer needed for the U.S. nuclear deterrent.

As of July 1, 1995, the IAEA had completed Initial Physical Inventory Verification (IPIV) at two facilities: a storage vault in the Oak Ridge Y-12 plant containing highly enriched uranium (HEU) metal and another storage vault in the Hanford Plutonium Finishing Plant (PFP) containing plutonium oxide and plutonium-bearing residues. Another plutonium-storage vault, located at Rocky Flats, is scheduled for the IPIV in the fall of 1995.

Conventional neutron coincidence counting is one of the routinely applied IAEA nondestructive assay (NDA) methods for verification of uranium and plutonium. However, at all three facilities mentioned above, neutron NDA equipment had to be modified or developed for specific facility needs such as the type and configuration of material placed under safeguards.

At Y-12, the size and uranium mass of items to be verified required modification of the Active Well Coincidence Counter (AWCC).[1,2] The facility prepared a set of calibration standards representative of the items to be measured. The IAEA certified these standards by destructive analysis (DA). Compared with operator declarations for ^{235}U mass (weighing and isotopic analysis), the IAEA AWCC measurement values agreed to within 0.5% for randomly selected items.

At Hanford, the IAEA used the standard High-Level Neutron Coincidence Counter (HLNC)[3] for verification of pure PuO_2. For verification of plutonium material containing unknown impurity concentrations, the IAEA used a 3-Ring Multiplicity Counter (3RMC) provided by LANL. The 3RMC gave better results for the impure material than could have been achieved using the HLNC. Also, the 3RMC showed an improvement in measurement performance for pure PuO_2 because of higher efficiency than the HLNC.

At Rocky Flats, a new neutron multiplicity counter designed for multiple-can plutonium oxide containers will be used for the IPIV. This will enable measurement of multiple-can items and thereby reduce radiation exposure to plant personnel as well as inspectors. Also, this counter is expected to be used for facility as well as the IAEA's verification purposes for a variety of nuclear materials present at this facility.

INTRODUCTION

Table I displays the routinely used NDA instruments for the International Atomic Energy Agency (IAEA) inspections of uranium and plutonium at storage facilities.

These standard NDA methods were used by the IAEA for the IPIVs at Y-12 and Hanford, along with other methods, some of which are described below.

OAK RIDGE Y-12 VAULT 16

The letter adding Y-12 Vault 16 to the list of US facilities eligible for IAEA inspections was received in Vienna by the IAEA on Tuesday, September 6, 1994. The vault contains HEU items, determined to be unclassified and excess to U.S. national security interests. Metric tonne quantities of HEU were included in the initial offer.

A description of the individual HEU items appears in Table II.

The IAEA used all of their standard measurement methods for HEU storage facilities at the Y-12 IPIV. These are shown in Table III.

* This work is supported by the US Department of Energy, Office of Nonproliferation and National Security, Office of Safeguards and Security and International Safeguards Division.

Table I. Standard IAEA NDA Instruments Used at Storage Facilities		
Measurement	Method	Instrument (acronym)
U, Pu radiation	Low-resolution gamma spectrometry	Portable MCA - NaI (PMCN)
^{235}U mass	Active neutron coincidence counting	Active Well Coincidence Counter (AWCC)
^{235}U enrichment	Low-resolution gamma spectrometry High-resolution gamma spectrometry	Portable MCA - NaI (PMCN) Portable MCA - HPGe (PMCG)
^{240}Pu-effective mass	Passive neutron coincidence counting	High Level Neutron Coincidence Counter (HLNC)
^{240}Pu-effective fraction	High-resolution gamma spectrometry	Medium Count Rate System (MCRS)

Table II. HEU Material and Storage Container	
Items:	
Material	HEU metal
	93% enriched
Form	Homogeneous annular castings
	Uniform size
Mass	~ 18 kg ^{235}U
Storage Canister:	
Height	~23 cm (9 in.)
Diameter	~18 cm (7 in.)

Table III. IAEA Verification Measurements at HEU Storage Facilities			
Defect Type	Defect Description	Measurements Required	Instruments Used
Gross	No U or HEU replaced by LEU	U radiation	PMCN
Partial	Part of HEU Missing	^{235}U mass ^{235}U enrichment, weight	AWCC PMCN + EBAL*
Bias	^{235}U content bias	^{235}U content	DA + EBAL

*operator's electronic balance validated by IAEA standards

The AWCC is shown in Fig. 1.

The Y-12 facility prepared seven AWCC calibration standards ranging in mass from 14 to 18 kg. Each standard is identical in density and geometry to the annular castings in the inventory offer, except for height, which was varied for mass variation. These standards were used to generate the calibration curves for two AWCCs, one shipped by the IAEA from its regional office in Toronto and one loaned by LANL. AWCC end plugs were configured for a measurement cavity height of 27.9 cm (11 in.) in fast mode (cadmium liners in), without the nickel reflector. AWCC calibration was performed by the IAEA on September 9, 1994. Each of the AWCC standards

was certified by the IAEA by means of sampling for subsequent DA in Vienna.

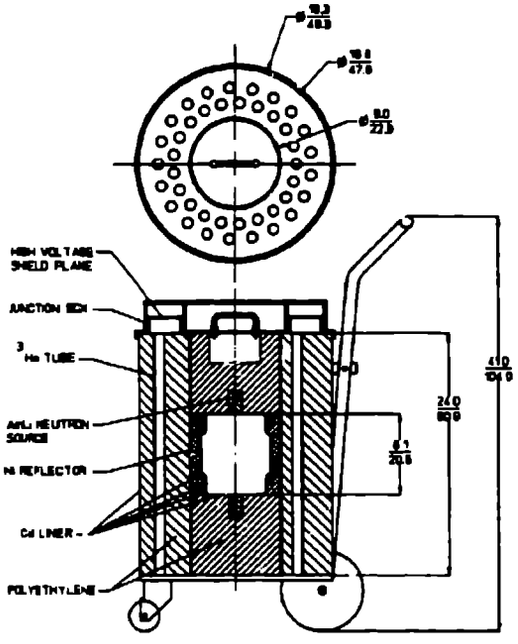


Fig. 1. Active Well Coincidence Counter (AWCC).

Table IV summarizes results of the AWCC verification measurements made by the IAEA.

The IAEA planned to use the AWCC for partial defects measurements only at Y-12. However, the excellent agreement between operator and IAEA values shown in Table IV qualifies the AWCC for bias-defect measurements, thus potentially eliminating the need for DA, except for standards. These excellent results were obtained because of the uniformity of material (both in mass and geometry), representative standards, high mass, and good measurement control.

HANFORD PFP VAULT ROOM 3

IAEA Inspectors arrived at the Hanford Plutonium Finishing Plant (PFP) on November 29, 1994. The initial offer at PFP consisted of over 500 items and included both pure plutonium oxide and plutonium residue items, each packaged in three nested metal cans. The outer cans were typically 4 in. in diameter and 6 in. high. The pure plutonium oxide items ranged in mass from approximately 500 to 1600 g Pu. The plutonium residue items ranged in mass from approximately 300 to 1700 g Pu. Items are stored in Vault Room 3 at PFP on pedestals instrumented for both security and monitoring. A separate measurement room was reserved for the IAEA.

The IAEA used all of their standard measurement methods for plutonium storage facilities at the Hanford IPIV. These are shown in Table V.

In addition to the instruments shown in Table V, the IAEA also brought an Inventory Sample coincidence counter (INVS),[4] which was used to measure few-gram samples taken from the inventory items.

Because the impurities bias conventional neutron assays as a result of (alpha, neutron) [or (α,n)] reactions with the impurities, the IAEA required a neutron multiplicity counter for verification of the plutonium residue items. This requirement was based on IAEA experience in Japan with the Plutonium Scrap Multiplicity Counter (PSMC).[5]

There were no multiplicity counters available anywhere at that time for use at Westinghouse Hanford Corporation (WHC), so one was improvised at Los Alamos. The prototype Active Well Coincidence Counter, which was built in the 1970's, was quickly upgraded to serve as a multiplicity counter. This counter has sixty ^{3}He tubes in three rings and therefore has high-neutron detection efficiency; the

Table IV AWCC Verification of HEU Metal Castings at Y-12: Summary Results

	AWCC #1 (IAEA)	AWCC #2 (LANL)
number of measurements	~100	~100
mean operator-IAEA difference in ^{235}U mass [(O-I)/O]	-0.29%	-0.11%
1 standard deviation in (O-I)/O	0.64%	0.59%

Table V. IAEA Verification Measurements at Plutonium Storage Facilities

Defect Type	Defect Description	Measurements Required	Recommended Instruments
Gross	No Pu	Pu radiation	HLNC or MCRS
Partial	Part of Pu Missing	Pu content	HLNC plus MCRS
Bias	Pu content bias	Pu content	DA + ELVAL

three rings also provide a reasonably constant efficiency as a function of neutron energy—a consideration in multiplicity counter design. The high-voltage junction box was rebuilt to accommodate twelve Amptek amplifier boards and a derandomizer[6] to reduce electronic deadtime. Graphite end plugs were fabricated to flatten the vertical detection-efficiency profile. Multiplicity electronics and power supplies in a NIM bin and a portable IBM-compatible PC with multiplicity-counting software completed the system, which was designated the Three-Ring Multiplicity Counter (3RMC) and was loaned to WHC for the IAEA inspections. With a neutron detection efficiency of 45% and an electronic deadtime of 85 ns, the counter makes a fairly good multiplicity counter for the small sample sizes in the WHC inventory.

Twenty-one of the WHC inventory samples that were measured with the multiplicity counter before the IAEA inspection were studied with four analysis techniques:

(1) Calibration curve

(2) Known α

(3) Known multiplication (known-M)

(4) Multiplicity

The calibration curve technique is the conventional one of constructing a calibration curve of coincidence rate vs effective ^{240}Pu mass. This method works well if the samples to be verified are very similar in geometry, density, uniformity, and impurity level to each other and to the standards used to construct the calibration curve. The coincidence rate is the only measured quantity needed for the analysis.

The α value for a sample is defined as the ratio of neutrons from (α,n) reactions to neutrons from spontaneous fissions.

The known-α technique works well if the verification samples have known impurity levels (from which α can be calculated), but have varying geometries, densities, and uniformities. The known-α analysis requires both the total and coincidence count rates.

The known-M method uses a calibration curve of neutron multiplication vs effective ^{239}Pu mass. It is useful when the samples are very similar in geometry, density, and uniformity, but have varying impurity levels. The known-multiplication technique requires both the total and coincidence rates for the analysis.

The multiplicity technique is useful when the sample densities, geometries, and impurity levels are all varying, but the samples are fairly uniform. Because the multiplicity analysis determines sample mass, neutron multiplication, and α value, three measured quantities are needed: these are the single, double, and triple count rates. The single and double count rates are identical to the total and coincidence count rates used for the other three analysis methods discussed above. The triple count rate is a measure of a higher level of correlation and is obtained from a multiplicity shift register coincidence circuit.

The purpose of studying the four analysis techniques was to determine how well each technique can deal with the sample variations present in the WHC inventory of plutonium oxides. The counter had not been calibrated with plutonium samples before the Hanford exercise, so calibration curves were constructed from the Hanford data on a series of pure-oxide standards. The multiplicity analysis method does not use a calibration curve, but requires three detector parameters for the analysis; two of these were obtained from a ^{252}Cf source with known neutron yield, and the third was obtained from a pure plutonium oxide sample.

The results of the analyses for the four techniques are shown in Figs. 2-5, where the assay masses are plotted vs the declared (facility) masses. The error bars represent standard deviations from counting statistics only; these errors are large enough to be significant only for the multiplicity analysis of samples with high α values and are thus not shown for the other analysis techniques.

As expected for impure oxides, the calibration-curve and known-α techniques do not work well because of the enhanced coincidence rates that result from induced fissions caused by an unknown number of (α,n) neutrons. The multiplicity technique accounts for these (α,n) neutrons, so there are no large outliers in the multiplicity plot—the assays are accurate either to within 3% or three standard deviations (for the high-α samples). The quality of the known-M results largely depends on the density variations in the sample set (the can diameters were similar); these results are somewhat better than expected because the plutonium concentration in the scrap samples is not constant.

The outlier in the known-α plot at a declared mass of 102 g effective ^{240}Pu is caused by a dense, pure-oxide sample with a multiplication (M) much higher than given by the multiplication calibration curve (M = 1.20 vs M = 1.11); note that this sample assays well with the known-alpha and multiplicity techniques.

Table VI shows the percent assay errors for each of the samples and each analysis technique. The multiplicity technique gives the best overall assay results, but requires long measurement times for samples with high α values. Most of the measurements were for one-half hour per sample (including the four samples shown with large error bars); a few high-α samples were measured overnight.

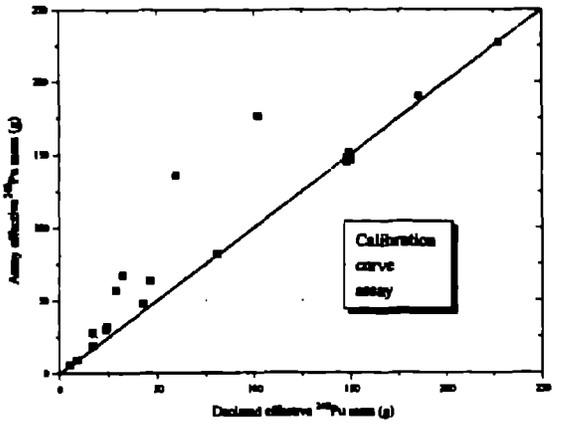


Fig. 2. Assay vs declared effective ^{240}Pu mass for calibration-curve assay. The straight line shows assay mass equal to declared mass.

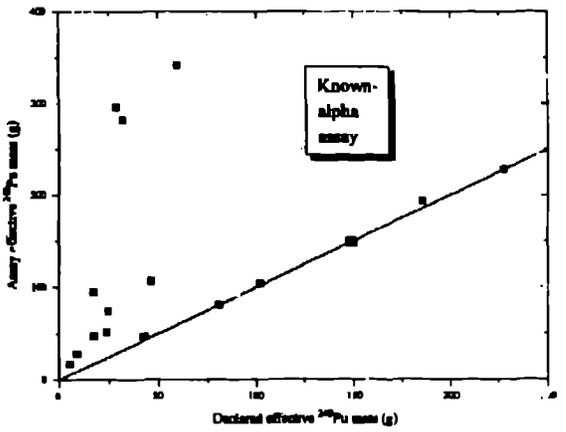


Fig. 3. Assay vs declared effective ^{240}Pu mass for known-α assay. The straight line shows assay mass equal to declared mass.

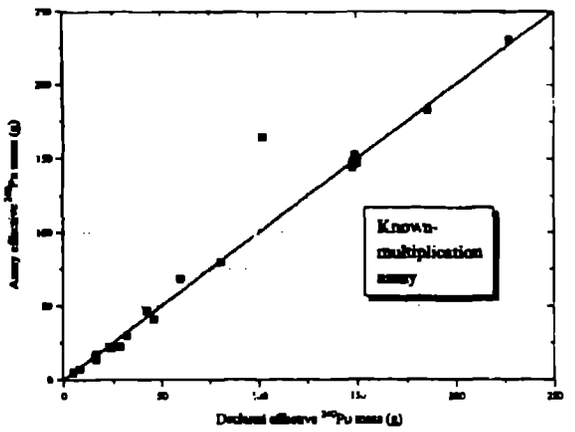


Fig. 4. Assay vs declared effective ^{240}Pu mass for known-M assay. The straight line shows assay mass equal to declared mass.

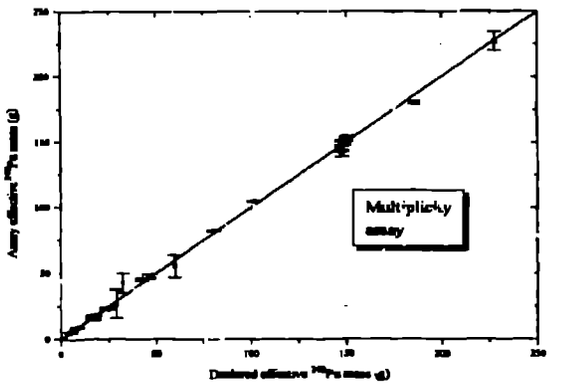


Fig. 5. Assay vs declared effective ^{240}Pu mass for multiplicity assay. The straight line shows assay mass equal to declared mass.

The known-M technique gives generally poorer results than the multiplicity technique, but is very fast—a few minutes measurement time per sample is adequate, even for high-α samples. The known-M and multiplicity techniques can be used together effectively; samples that are outliers in known-M assay can be measured longer and analyzed with the multiplicity technique. For example, the sample with the declared mass of 102 g effective ^{240}Pu is an outlier in known-M analysis. However, even a short measurement of this sample—when analyzed with the multiplicity—shows that the multiplication of this sample is too high for it to be assayed correctly with the known-M technique. A half-hour measurement of this sample results in a multiplicity assay error of 2.8% ± 1.1% compared to a known-M assay error of 61%. For routine use of the known-M technique, a method must be developed to allow error estimates to be made for the known-M assay of individual samples.

ROCKY FLATS ENVIRONMENTAL TEST SITE (RFETS)

The IPIV at RFETS is tentatively scheduled for the fall of 1995. The offering will consist of impure plutonium oxide items packaged in 8801 cans with a 8802 can over-pack. Items are stacked inside a steel tube that is centered in a 10-gal. drum. Typically, there are two items to a drum; however, a few drums contain only a single item. Each 8801 can contains up to 2 kg of oxide. The 10-gal. drums contain from 1 to 4 kg of oxide.

Table VI. Assay Errors for Hanford Plutonium Samples						
			Assay error (%)			
Sample	Eff. ^{240}Pu (g)	Purity	Calibration curve	Known α	Known *M*	Multiplicity
1	5.42	impure	4.5	219.1	-6.2	?.4 ± 5.6
2	8.89	impure	2.2	209.5	-14.7	2.6 ± 4.1
3	16.78	very impure	66.5	464.8	0.6	-5.0 ± 8.7
4	16.97	impure	11.7	177.5	-17.4	6.4 ± 3.2
5	23.64	impure	25.5	118.8	-6.6	-1.2 ± 3.3
6	24.29	impure	29.7	207.3	-10.4	-4.4 ± 4.5
7	28.85	very impure	97.5	926.1	-22.9	-7.5 ± 35.2
8	32.4	very impure	107.9	771	-9.2	31.2 ± 12.6
9	42.76	pure	11.1	9.3	9.6	5.9 ± 2.0
10	46.19	impure	37	130.1	-11.6	2.6 ± 3.0
11	59.62	very impure	127.5	472.3	14.9	-7.3 ± 13.8
12	80.71	pure	1	-0.04	-1.3	2.0 ± 0.4
13	101.9	pure	73.3	2.1	61.2	2.8 ± 1.1
14	147.59	pure	-1.2	0.3	-1.5	-1.8 ± 1.8
15	147.63	pure	0.2	0.8	0.2	0.8 ± 1.6
16	147.7	pure	1.7	1.4	-2.7	-4.3 ± 1.6
17	149	pure	1.0	0.6	2.2	-0.2 ± 0.9
18	149.6	pure	-1.2	0.4	-1.4	2.4 ± 0.3
19	150.1	pure	-2.2	-1.3	-2.2	1.7 ± 1.2
20	185.7	pure	2.2	4.2	-1.6	-2.9 ± 0.6
21	227.9	pure	-0.3	0.08	1.2	-0.4 ± 2.9

Because the oxide samples contain a variety of impurities, neutron multiplicity counting was identified as the NDA measurement technique of choice. However, when the original offer was proposed, there was no multiplicity counter in existence that could accommodate a 10-gal. container. During fiscal 1994, Los Alamos, was tasked by DOE NN-20 to design a multiplicity counter that could accommodate up to 30-gal. drum sized items for the measurement of weapons components.[7] Because this design was complete and because of the original short time frame to acquire instrumentation for an inspection at RFETS, Los Alamos was asked in November of 1994 to produce a counter of this design to be delivered to RFETS by mid-February of 1995. Los Alamos was also tasked at this same time to advise RFETS concerning state-of-the-art NDA instrumentation, including the 30-gal. multiplicity counter, that could be used to upgrade domestic safeguards techniques.

The 30-gal. Drum Neutron Multiplicity Counter, shown in Fig. 6, consists of 126 ^{3}He tubes arranged in three rows in a hexagonal polyethylene moderator. Each sixth of the counter is an independent unit that has its own electronics junction box. Each junction box contains a high-voltage circuit board and a 5-V circuit board to drive and collect signals from the

Fig. 6. Photograph of 30-Gal. Drum Multiplicity Counter System.

tubes. The signals from each sixth are brought together in a derandomizer[6] to produce the total signal and three auxiliary signals from the inner, middle, and outer rows of tubes in the counter. These signals are then processed by a Canberra 2150 multiplicity electronics module, and the data are analyzed using the new NCC Windows code. The measured efficiency of this counter for ^{252}Cf is 42.3%. Its die-away time is 55.4 μs, and its deadtime is 22.5 ns. From calculations, this counter should provide an assay precision

of from 1 to 3% in 30 min. for 1.5 to 5 kg oxide samples that have small to moderate impurities.

The instrument parameters will be adjusted by measuring a set of certified plutonium oxide standards at RFETS and adjusting the efficiency of the counter from plutonium. It is anticipated that this adjustment will be from 1 to 3%, compared with ^{252}Cf measurements. No adjustment for die-away time is expected to be necessary. These standards will also be measured in configurations similar to the samples in the offering to ascertain if any corrections will be needed because of the unusual geometry. Currently, this calibration is expected to take place in late August or early September 1995.

CONCLUSIONS

The excellent quality and uniformity of material at Y-12 enabled unprecedented accuracy and precision for IAEA verification of HEU metal castings. Also, the quantity of material verified during the IPIV was unsurpassed in IAEA experience.

Because of the high impurity levels in some of the Hanford plutonium items, conventional neutron coincidence counting is unsatisfactory; the assay masses are biased high beyond the limit of acceptance for partial defects detection. Multiplicity counting is the best neutron assay method for most of the inventory because it provides the lowest bias, but the technique requires long counting times for items with very high (α,n)-to-spontaneous-fission neutron ratios. For those samples, studies in progress suggest that the multiplicity technique can be augmented by a known-multiplication analysis to provide a rapid verification of impure oxides for partial defects detection, in most cases. The joint use of multiplicity counters and calorimeters combined with germanium isotopic systems is very promising, in principle, for IAEA verification: the neutron counter with the isotopics system quickly verifies the authenticity of the item and determines the plutonium mass, in most cases, at the partial defects level; the calorimeter and the isotopic system then determine the most accurate plutonium mass, in most cases.

The multi-can, impure plutonium oxide items to be verified by the IAEA at Rocky Flats, as well as a variety of process residues, present new challenges and opportunities for implementation and further development of neutron counting.

Experience gained by the IAEA in U.S. facilities will be applied in other nations that also offer excess nuclear weapons materials to international inspection.

REFERENCES

1. H. O. Menlove, "Description and Operation Manual for the Active Well Coincidence Counter," Los Alamos Scientific Laboratory report LA-7823-M (May 1979)

2. H. O. Menlove and J. E. Stewart, "A New Method of Calibration and Normalization for Neutron Detector Families," Los Alamos National Laboratory report LA-11229-MS (April 1988).

3. H. O. Menlove and J. E. Swansen, "A High-Performance Neutron Time-Correlation Counter," *Nucl. Technol.* **71**, 497-505 (1985).

4. H. O. Menlove, D. Davidson, J. Yerplancke, P. Vermuelen, H. G. Wagner, R. Wellum, B. Brandelise, and K. Mayer, "Design and Performance of a New High Accuracy Combined Small Sample Neutron-Gamma Detector," *Nucl. Mat. Manage.* **XXII** (Proc. Issue), 872-880 (1993).

5. H. O. Menlove, J. Baca, M. S. Krick, K. E. Kroncke, D. G. Langner, "Plutonium Scrap Multiplicity Counter Operation Manual," Los Alamos National Laboratory report LA-12479-M (January 1993).

6. S. C. Bourret and M. S. Krick, " A Deadtime Reduction Circuit for Thermal Neutron Coincidence," *Nucl. Mater. Manage*, **XXIII** (Proc. Issue) 646-650 (1994).

7. D. G. Langner, M. S. Krick, and K. E. Kroncke, "A Large Multiplicity Counter for the Measurement of Bulk Plutonium," *Nucl. Mater. Manage*, **XXIII** (Proc. Issue) 474-479 (1994).