LA-13743-M: Manual

The Epithermal Neutron Multiplicity Counter Design and Performance Manual: More Rapid Plutonium and Uranium Inventory Verifications by Factors of 5–20

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

This work was sponsored by the US Department of Energy, Office of Nonproliferation and International Security, Office of Safeguards and Security, and the Los Alamos Directed Research Development program.

Edited by Amy C. Ferguson, Group CIC-1
Prepared by Celina Ortiz, Group NIS-5

LA-13743-M
Manual

Issued: August 2000

The Epithermal Neutron Multiplicity Counter Design and Performance Manual: More Rapid Plutonium and Uranium Inventory Verifications by Factors of 5–20

J.E. Stewart
H.O. Menlove
D.R. Mayo
W.H. Geist
L.A. Carrillo
G.D. Herrera

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545

CONTENTS

THE EPITHERMAL NEUTRON MULTIPLICITY COUNTER DESIGN AND PERFORMANCE MANUAL: MORE RAPID PLUTONIUM AND URANIUM INVENTORY VERIFICATIONS BY FACTORS OF 5–20

by

J. E. Stewart, H. O. Menlove, D. R. Mayo, W. H. Geist,
L. A. Carrillo, and G. D. Herrera

ABSTRACT

Thermal neutron multiplicity counters (TNMCs) assay ^{240}Pu-effective mass, isolating spontaneous-fission (SF), induced-fission, and (α,n) neutrons emitted from plutonium metal, oxide, scrap, and residue items. Three independent parameters are measured: single, double, and triple neutron-pulse-coincidence count rates. TNMC assays can become precision limited by high (α,n) neutron rates arising from low-Z impurities and ^{241}Am. TNMCs capture thermal neutrons in 4-atm ^{3}He tubes after fast-source-neutron moderation by polyethylene. TNMCs are ~50% efficient with ~50-µs die-away times. Simultaneously increasing efficiency and reducing die-away time dramatically improve assay precision. Using 10-atm ^{3}He tubes, we've developed and performance-tested the first of a new generation of neutron assay counters for a wide range of plutonium items. The Epithermal Neutron Multiplicity Counter (ENMC) has an efficiency of 65% and a 22-µs die-away time. The ENMC detects neutrons before thermalization using higher ^{3}He pressure counters and less moderator than TNMCs. A special insert raises efficiency to 80% for small samples. For five bulk samples containing 50 to 875 g of ^{240}Pu-effective, ENMC assay times are reduced by factors of 5 to 21, compared with prior state-of-the-art TNMCs. The largest relative gains are for the most impure items, where gains are needed most. In active mode, the ENMC assay times are reduced by factors of 5 to 11, compared with the Active Well Coincidence Counter (AWCC). The ENMC, with high precision and low multiplicity dead time (37 ns), can be used in standards verification mode to precisely and accurately characterize plutonium standards and isotopic sources. The ENMC's performance is very competitive with calorimetry. This report describes the ENMC; presents results of characterization, calibration, and verification measurements; and shows the clear performance and economic advantages of implementing the ENMC for nuclear materials control and accountability.

I. INTRODUCTION

Thermal neutron multiplicity counters (TNMCs) are used in several US Department of Energy (DOE) facilities, for domestic material control and accountability (MC&A), and for International Atomic Energy Agency (IAEA) inspections. The same is true in Japan. TNMCs are also being implemented in Russian facilities for MC&A. A comprehensive review of US experience is Ref. 1.

Occasionally, an item will be encountered where the ratio of (α,n) to spontaneous fission neutrons (the quantity "α") will be so high that TNMC multiplicity assay precision is degraded to the point that long count times are required for acceptable results. For a 1-kg plutonium item, precision worsens by a factor of ~20, for an α of 10, compared to an α of 1. Currently, the IAEA uses the Plutonium Scrap Multiplicity Counter (PSMC)[2] for inventory verifications at the Hanford Plutonium Finishing Plant (PFP). While the majority of PFP items are verified satisfactorily with the PSMC, the occasional item presents difficulties. For example, one item with 1311 g of plutonium and an α of 4.5 requires 2 h to reach a precision of 3%, or 18 h to reach 1% using the PSMC. Another 1350-g plutonium item with an α of 0.74 requires only 30 m to reach 1% in the PSMC. Another item with 879 g plutonium and an α of ~30 would require 30 h to reach 3% using the PSMC. This item would be best assayed by the ENMC (~1.5-h measurement time) or calorimetry (~8 h-measurement time). See, for example, Ref. 3.

For active measurements of uranium, precision is limited by the accidental coincidences produced by the AmLi interrogation sources. Hence, the ENMC plays an important role for improving precision of active measurements, as well as passive.

As more US plutonium is placed under IAEA safeguards, more examples of precision limitations of TNMCs will occur. Also, more such examples will occur in Japan, Russia, and other nations.

II. PHYSICS DESIGN PRINCIPLE

It is well known that high detector efficiency (ε) and low-neutron die-away time (τ, the average time from neutron birth to detection) both quite significantly improve precision of multiplicity assays.[4,5] Figure 1 shows results of Monte Carlo[6] simulations for two neutron multiplicity counter (NMC) designs, one using 4-atm ^{3}He counters (the standard, for many years) and one using 10-atm counters. These results led to a distinction between TNMCs, which detect primarily thermal neutrons after moderation by polyethylene (PE), and ENMCs, which detect, on the average, neutrons with higher energies than thermal (epithermal). ENMCs also use PE moderator, but less than TNMCs.

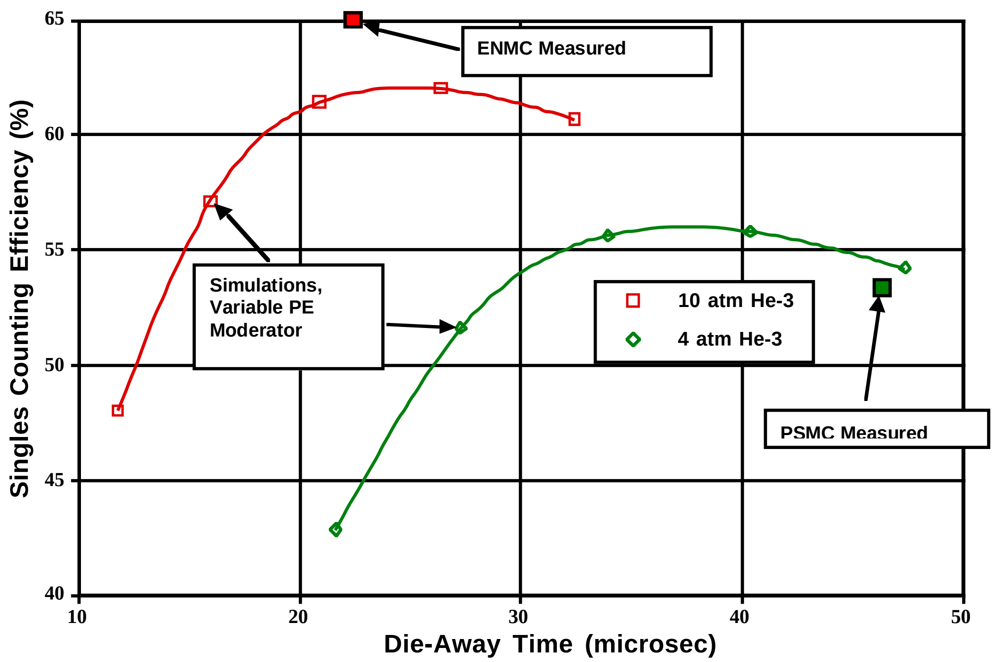

Fig. 1. Efficiency vs die-away time for 10-atm (ENMC) and 4-atm (PSMC) ^{3}He designs, with variable PE moderator thickness between ^{3}He counters.

III. ENMC AND ENMC/INVS DESIGN FEATURES

From the MCNP simulations, we built the ENMC using 10-atm ^{3}He proportional counters with optimum spacing in PE moderator. The basic ENMC design is shown in Fig. 2a.

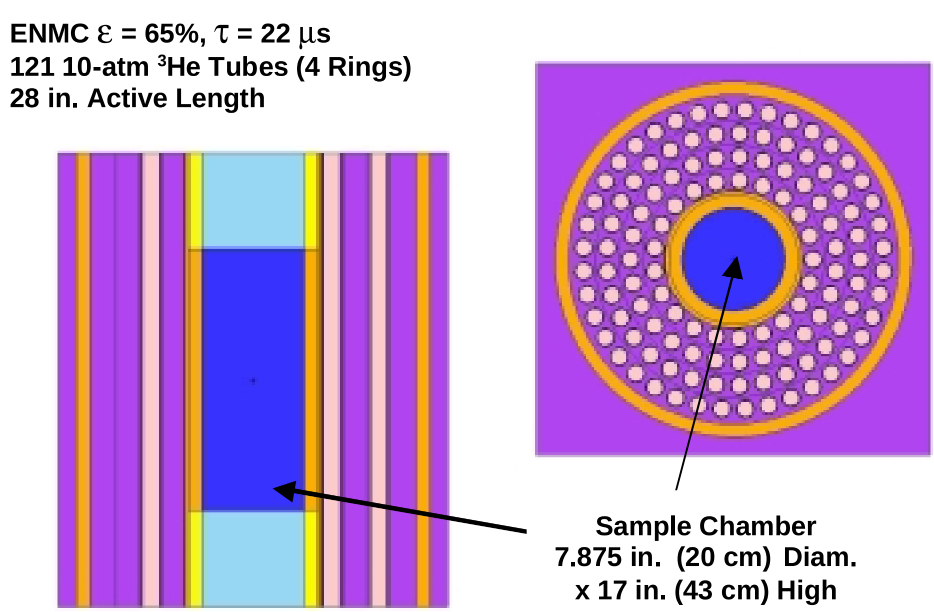

Fig. 2a. ENMC design features, including large graphite endplugs and steel cavity liner.

The ENMC groups 121 ^{3}He counters into 27 channels, each with an AMPTEK preamplifier/amplifier/discriminator. A derandomizing buffer is also included in the detector electronics. The resulting multiplicity deadtime is only 36.8 ns.

Another, separate counter, the Inventory Sample counter (INVS) was designed and constructed to fit inside the ENMC sample chamber for assay of small inventory samples.[7] Figure 2b. is a schematic of the ENMC/INVS combination.

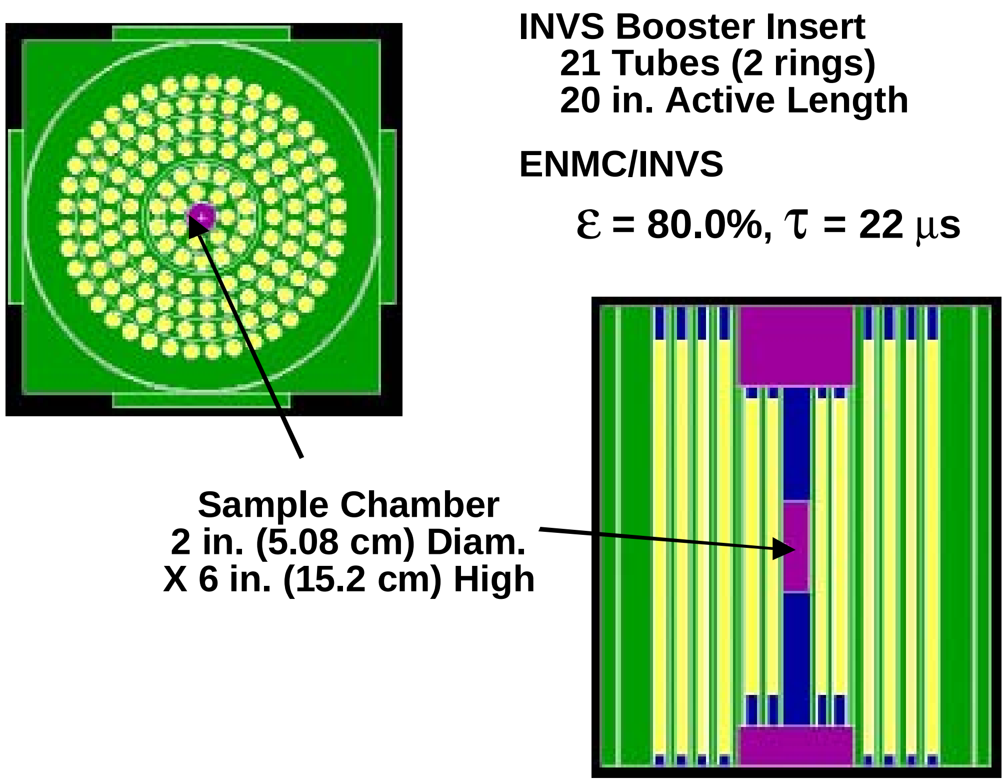

Fig. 2b. ENMC/INVS design features, including small graphite endplugs and removable sample chamber.

The INVS insert groups 21 ^{3}He counters into 3 channels, each with an AMPTEK preamplifier/amplifier/discriminator. A derandomizing buffer is not included in the INVS detector electronics. The resulting ENMC/INVS multiplicity deadtime is ~100 ns. This is quite acceptable for the low count rate applications of the ENMC/INVS.

Table I gives the General Electric Reuter-Stokes ^{3}He proportional counter specifications for the present detector systems.

Table I. ^{3}He tube and amplifier specifications.

Parameter	Value
ENMC tubes @ 28" (RS-P4-0828-105)	121
INVS tubes @ 20" (RS-P4-0820-118)	21
Diameter	2.54 cm
He pressure	10 atm
Cathode	Al
Operating bias	1720 V
AMPTEK A111 – ENMC/INVS	27/3

IV. ENMC AND ENMC/INVS CHARACTERIZATION AND CALIBRATION PARAMETERS

Initial ENMC parameters were measured using NIST-calibrated ^{252}Cf sources.[8] Monte Carlo simulations were used to adjust the measured ^{252}Cf spontaneous-fission neutron-detection efficiency to that for ^{240}Pu. A final, small adjustment (to the triples gate fraction) was made using a single Los Alamos pure plutonium oxide working reference material (WRM). This WRM is ~1 kg of oxide. Table II gives the measured ENMC and ENMC/INVS parameters along with those for the high-level neutron coincidence counter (HLNCC)[9], the HLNCC10 (HLNCC with 10-atm ^{3}He tubes), the AWCC,[10] the 3-ring multiplicity counter (3RMC),[11,12] and the PSMC.

Table II. Multiplicity detector parameters* (preset or measured).

Detector	Efficiency (%)	Pre-delay (µs)	Gate Width (µs)	Doubles Gate Fraction	Triples Gate Fraction	Die-Away Time (µs)	Deadtime Coefficient A (1E-6)	Deadtime Coefficient B (1E-12)	Multiplicity Deadtime (ns)
HLNCC	17	3	64	0.701	0.492	43	0.768	0.248	215
HLNCC10	20	1.5	64	0.792	0.649	37	0.780	0.025	215
AWCC	33	3	64	0.658	0.445	51	0.826	0.267	224
3RMC	43	3	64	0.631	0.414	55	0.315	0.102	90
PSMC	53	3	64	0.651	0.441	47	0.383	0.128	118
ENMC	65	1.5	24	0.621	0.404	21.8	0.0954	0.0289	36.8
ENMC/ INVS	80	1.5	24	0.605	0.399	18.8	0.341	0.017	100

*sufficient for assay – calibration curves are not required.

V. ENMC VERIFICATION RESULTS – PRECISION

We chose five plutonium oxide items (WRMs) for comparing the passive-neutron-multiplicity assay (PNMA) precision performance of the first six detectors in Table II. These WRMs span a wide range of plutonium mass and α. They are described in Table III.

Table III. Description of WRMs for ENMC precision comparisons.

Item	Description	Plutonium mass, g	Multiplication	Alpha
STDISO3,6,9,12	4 LANL Pu-isotopic WRMs; small impure oxides	51.4	1.025	1.036
STD-11	1 LANL NDA WRM; small, very impure oxide	59.8	1.004	4.9
STDSRP12-1	1 LANL NDA WRM; large, impure oxide	874.4	1.12	1.04
41-86-03-240	1 Hanford IAEA NDA WRM; medium, very impure oxide	268.8	1.038	7.84
LAO261C10	1 LANL NDA WRM; large, pure oxide	842.7	1.085	0.486

For each of the first six detectors in Table II, and each WRM in Table III, count times (minutes) required for 1% PNMA precision are reported in Table IV.

Table IV. Count times (min.) required for 1% PNMA precision for six detectors and five WRMs.

Detector	Working Reference Material				
	LANL STD-ISO3,6,9,12	LANL STD-11	LANL STDSRP12-1	Hanford PFP 41-86-03-240	LANL LAO261C10
HLNCC	84	5563	384	15,578	212
HLNCC10	42	2782	203	8,235	110
AWCC	16	672	95	4,500	41
3RMC	11	400	37	2,035	30
PSMC	7.3	170	34	1,352	27
ENMC	1.6	36	5.6	64	2.7
COUNT TIME RATIO: PSMC/ENMC	4.6	4.7	6.1	21	10
COUNT TIME RATIO: AWCC/ENMC	10	19	17	70	15

Table IV data result from a combination of measurements, using multiple runs and sample standard deviations for precision, and a figure-of-merit (FOM) code.[5] Only relative FOM values were used. Results are better than expected compared with preliminary FOM estimates. PSMC/ENMC count time ratios vary from 4.6 to 21 for the WRMs chosen. The PSMC is commercially available, as is the AWCC, which is used as a passive multiplicity counter in some facilities (e.g., in Russia). AWCC/ENMC passive count time ratios vary from 10 to 70 for the WRMs chosen.

Count time to a fixed precision for a range of items is only one measure of the merit of a nondestructive assay (NDA) system. NDA system cost, useful life, load factor, facility-count-room operating cost, productive time per shift, and time required for item movement should also be considered. Table V gives an item throughput/cost index for the following detector systems: ENMC, PSMC, 3RMC, AWCC, HLNCC10, and HLNCC. We define the item throughput index as the number of items measurable in one eight-hour shift, divided by the sum of the system cost (prorated over the useful life, times the load factor) and the count-room operating cost per shift, for each of the WRMs defined in Table III.

Table V. Throughput/total cost Index; 1% PNMA precision for six detectors and five WRMs.

Detector	System Cost ($K)	Working Reference Material: LANL STD-ISO3,6,9,12	LANL STD-11	LANL STDSRP12-1	Hanford PFP 41-86-03-240	LANL LAO261C10
HLNCC	60	31.3	0.5	7.5	0.2	13.2
HLNCC10	65	56.5	1.1	13.8	0.4	24.5
AWCC	110	111.3	4.2	27.6	0.6	56.7
3RMC	165	135.4	6.9	60.5	1.4	71.1
PSMC	215	161.8	15.6	63.6	2.1	75.7
ENMC	290	235.8	59.5	175.4	37.0	215.4
INDEX RATIO:	ENMC/ PSMC	1.5	3.8	2.8	18.0	2.8
INDEX RATIO:	ENMC/ AWCC	2.1	14.0	6.4	57.6	3.8

*Assumptions:

(1) detector system useful life – 15 yr.;
(2) detector load factor – 20 shifts/yr.;
(3) plutonium facility count room operating cost - $10K/shift;
(4) Item transfer time – 10 min.; and
(5) Productive time per 8-h shift – 5 h.

Table V shows the clear superiority of the ENMC for a wide range of plutonium items including the count time for 1% precision, item transfer time, detector system, and facility operating costs as compared with five other detectors. The rank order of the throughput/cost index is the reverse of the system-cost order. The maximum throughput per unit cost is obtained from the system with the best performance – the ENMC. ENMC/PSMC and ENMC/AWCC index ratios are given because the PSMC and AWCC are available commercially, and used often as multiplicity counters.

Note the ENMC yields the largest gain for the most difficult-to-measure item (Hanford PFP # 41-86-03-240), exactly where the gain is needed most. For this item, a six-hour run becomes a 17-min run.

From February to July 1999, The ENMC and the ENMC/INVS were used for verification assays of a wide variety of Los Alamos plutonium WRMs. ENMC/INVS results will be reported in a later section.

Results for ENMC PNMA precisions are shown in Fig. 3. Ninety-four measurements of 45 Los Alamos WRMs are represented.

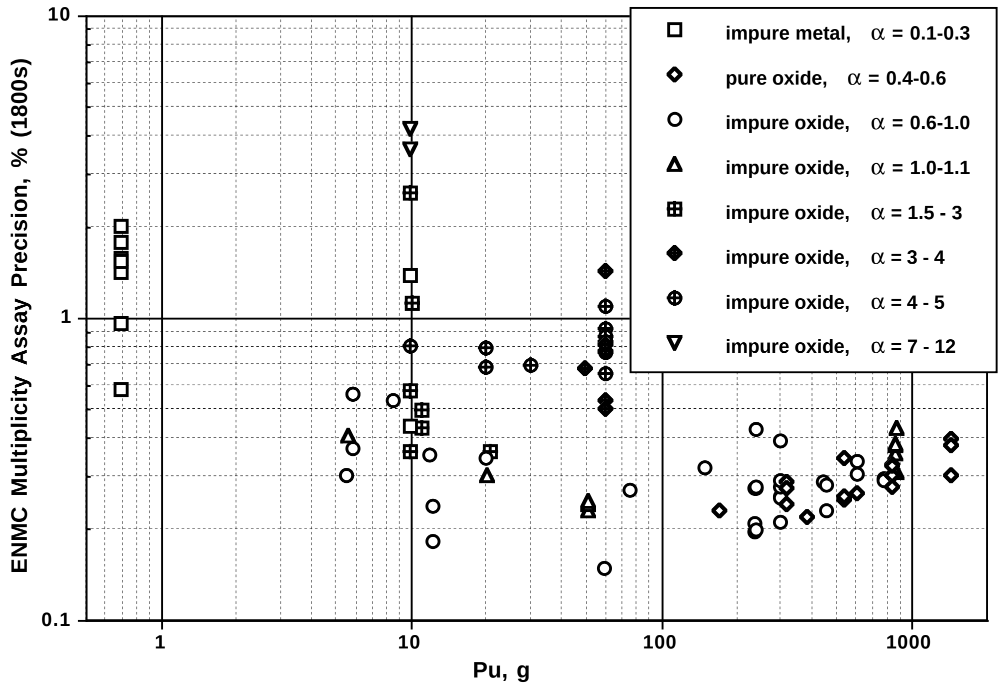

Fig. 3. ENMC PNMA precision for 94 measurements of 45 Los Alamos WRMs; count time is fixed at 1800 s.

Figure 3 shows that for most measurements, the ENMC half-hour measurement precision is less than 0.5%. The complete range is from 0.15% for a 60-g plutonium LAO WRM (pure oxide), to ~4% for a 10-g WRM (very impure oxide). The plutonium mass range in Fig. 3 is 0.7 g to 1451 g, with α varying from 0.1 to 12.

Another series of measurements was carried out to further evaluate the performance of the ENMC. These measurements were made on a set of WRMs prepared for the NDA system designed for the product end of the Advanced Recovery and Integrated Extraction System (ARIES).[13] These standards are all pure plutonium oxide except the first two, which have diatomaceous earth added. These WRMs are described in Table VI.

Table VI. Description of ARIES WRMs for ENMC precision determinations.

	Wt% (relative to total Pu mass)									
WRM ID	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Date	g Pu	g $^{240}Pu_{eff}$	$alpha_{pure}$
LARI-MC-1	0.0134	93.8144	5.9389	0.1795	0.0538	0.1118	8/10/99	20.01	1.213	0.791
LARI-MC-2	0.0134	93.8143	5.9389	0.1797	0.0538	0.1118	8/4/99	100	6.063	0.791
LARI-MC-3	0.0135	93.8122	5.9389	0.1817	0.0538	0.1118	5/9/99	749.49	45.442	0.791
LARI-MC-4	0.0134	93.8139	5.9389	0.1800	0.0538	0.1118	5/21/99	1499.72	90.929	0.791
LARI-MC-5	0.0134	93.8139	5.9389	0.1800	0.0538	0.1118	7/20/99	2999.49	186.861	0.791

This set of standards were measured in the ENMC and the ARIES counter, whose performance characteristics are very similar to the PSMC. Results of these measurements are given in Table VII. The ENMC precisions are all 0.25%. The ARIES measurements were nearly all 1800 s. As Table VII shows the count-time reduction factor (ARIES/ENMC) varies between 5.5 and 12.6. ENMC measurements were also made on ten ARIES product cans and the count-time reduction factor determined was 10.9, compared to the present ARIES counter, for multiplicity assays. These results include the factor-of-two reduction in count time given by the Advanced Multiplicity Shift Register (AMSR).[14,15] These results have important implications for enhancing the throughput of the ARIES facility.

Table VII. Comparison of ENMC and ARIES counter standards verification, pure and lightly impure ARIES oxides (α from 0.8 to 2.1).

				Passive Multiplicity Assay Results						
WRM ID	Counter	Count time, s	Declared Pu, g	Pu, g	1 σ, Pu, g	(D-A),%	1 σ, (D-A),%	Multi-plication	α	Count-time factor
LARI-MC-1	ENMC	4020	20.009	19.591	0.052	2.090	0.260	0.999	1.383	1
LARI-MC-1	ENMC	3300	20.009	19.758	0.052	1.251	0.257	0.998	1.436	1
LARI-MC-2	ENMC	1500	99.994	98.586	0.241	1.408	0.241	1.006	1.389	1
LARI-MC-2	ENMC	2010	99.993	98.713	0.246	1.280	0.246	1.005	1.398	1
LARI-MC-3	ENMC	1350	749.4	733.2	1.8	2.169	0.242	1.140	0.859	1
LARI-MC-3	ENMC	1440	749.4	731.3	1.8	2.414	0.244	1.141	0.863	1
LARI-MC-4	ENMC	1650	1499.6	1468.8	3.8	2.058	0.245	1.213	0.855	1
LARI-MC-4	ENMC	1590	1499.6	1471.0	3.6	1.908	0.242	1.205	0.856	1
LARI-MC-5	ENMC	2370	2999.3	2850.3	7.1	4.967	0.237	1.352	0.912	1
LARI-MC-5	ENMC	1680	2999.2	2846.8	7.0	5.084	0.233	1.359	0.915	1
LARI-MC-1	ARIES	1770	20.007	19.469	0.187	2.688	0.936	1.002	2.144	5.8
LARI-MC-1	ARIES	1770	20.007	19.366	0.163	3.206	0.816	1.002	1.909	5.5
LARI-MC-2	ARIES	3500	99.975	97.172	0.549	2.804	0.549	1.010	1.427	12.6
LARI-MC-2	ARIES	1860	99.975	97.501	0.717	2.475	0.717	1.009	1.559	8.1
LARI-MC-3	ARIES	1860	749.4	741.9	4.3	0.997	0.57	1.143	0.961	7.7
LARI-MC-3	ARIES	1830	749.4	738.1	4.7	1.508	0.631	1.144	0.980	8.5
LARI-MC-4	ARIES	1760	1499.5	1503.4	11.1	-0.261	0.738	1.216	0.980	8.7
LARI-MC-4	ARIES	1800	1499.5	1511.8	10.6	-0.817	0.704	1.215	0.977	9.3
LARI-MC-5	ARIES	1800	2999.3	3079.4	28.6	-2.672	0.955	1.347	1.014	10.6
LARI-MC-5	ARIES	1800	2999.3	3027.7	24.7	-0.948	0.824	1.348	1.049	11.8

VI. ENMC VERIFICATION RESULTS – ACCURACY

Results of ENMC PNMAs to date are shown in Figs. 4a and 4b.

From the data of Figs. 4a and 4b, the mean bias, 1-σ standard deviation, and 1-σ precision for these measurements are +0.6, 1.7, and 0.5% respectively, compared with book WRM values. The plutonium mass range represented in Fig. 4 is 0.7 g to 1451 g, five orders of magnitude using the same set of calibration parameters given in Table II. The impurity parameter α varies from 0.1 to 12. Two mixed uranium-plutonium oxide (MOX) items are included in this data set. These data have not been corrected for neutron energy variations, although, for most measurements, the ratio of singles counts from the inner and outer rings of ^{3}He counters was measured. This ring ratio can be used in the dual-energy model for passive neutron multiplicity counting.[16] This correction will be evaluated in the future for the ENMC and ENMC/INVS. In all cases, weighing, sampling, and chemical analysis are the bases for plutonium/americium isotopics and plutonium-mass declarations.

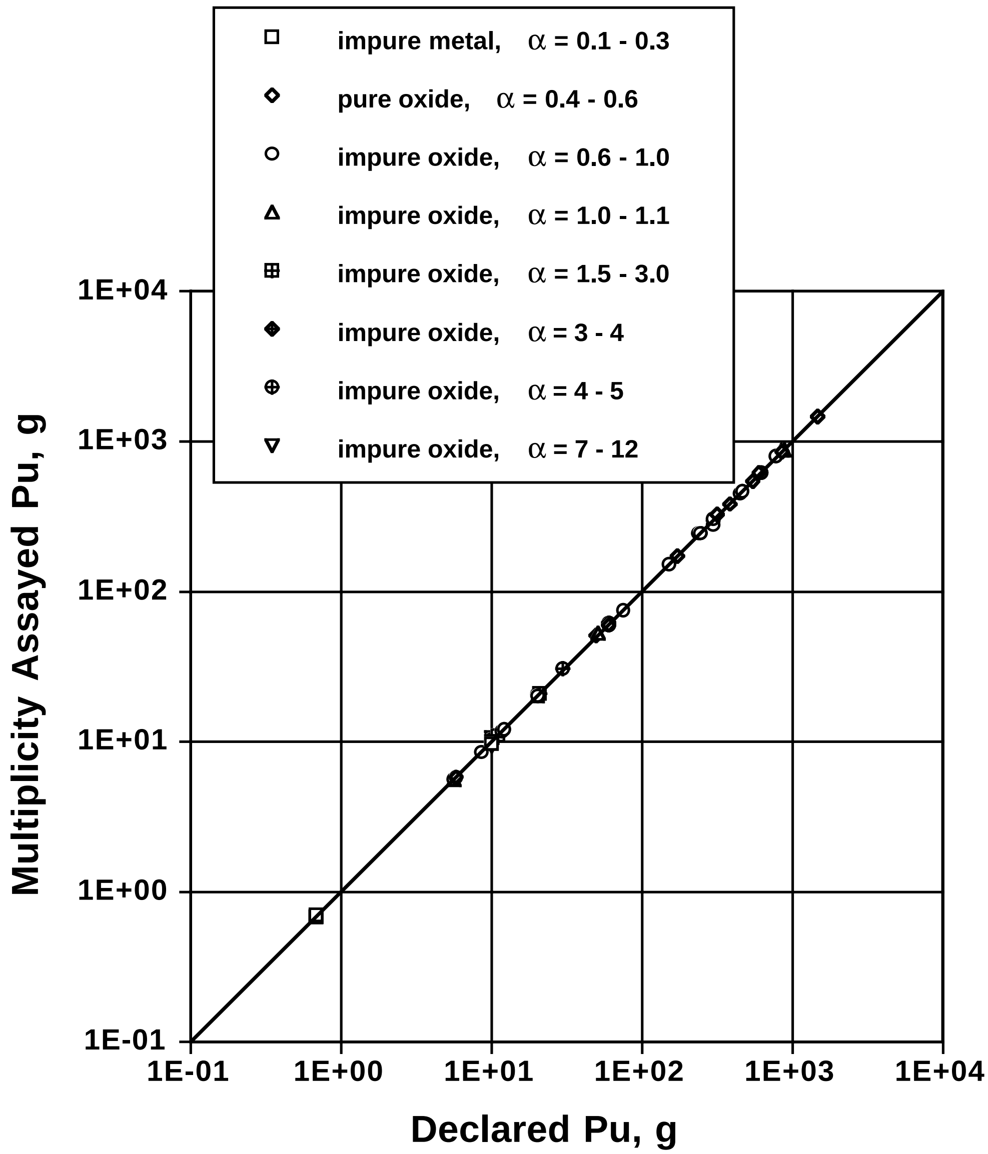

Fig. 4a. ENMC passive neutron multiplicity assay versus book values for 94 measurements of 45 LANL WRMs; count times were variable.

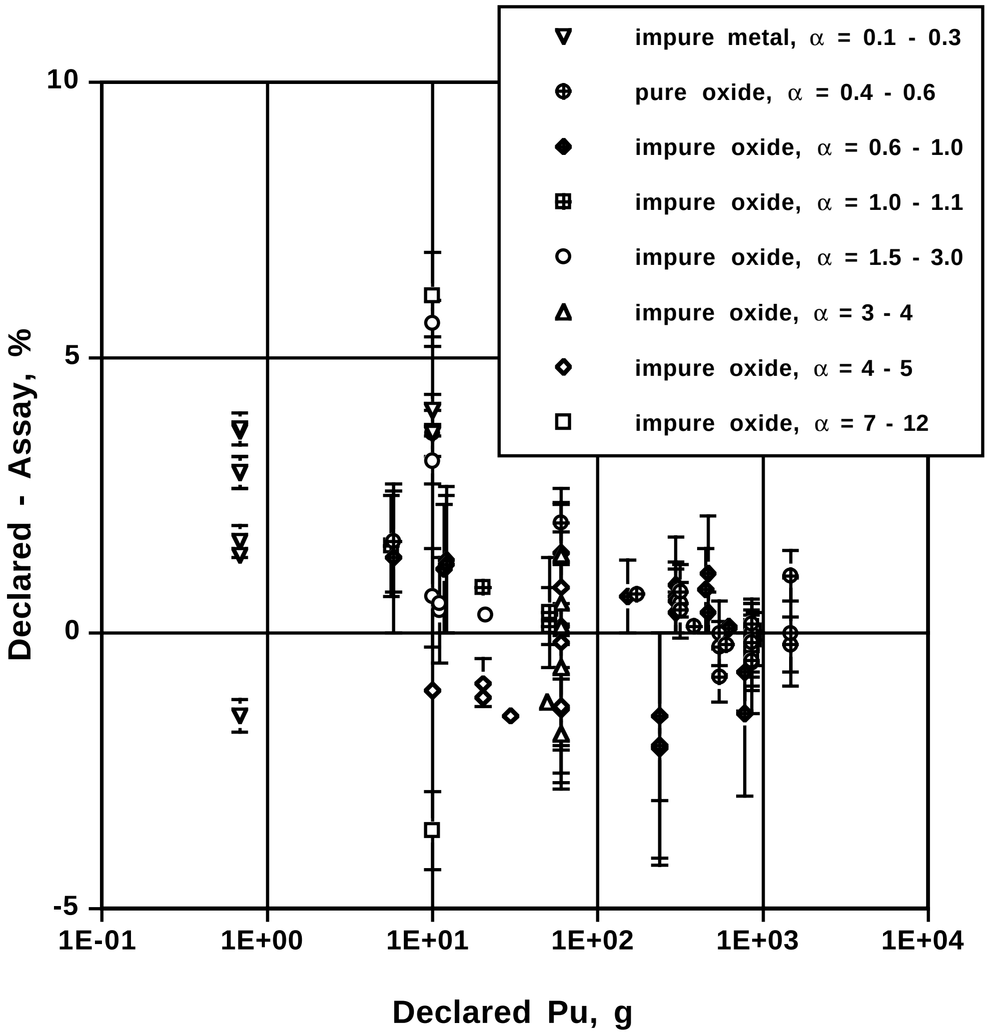

Fig. 4b. ENMC passive neutron multiplicity assay minus book values versus book values for 94 measurements of 45 LANL WRMs; count times were variable.

VII. ENMC STANDARDS VERIFICATION MODE

The performance of the ENMC is good enough to be used in standards verification mode. To illustrate this, Table VIII gives PNMA results of 33 assays of 19 pure and lightly impure plutonium oxides.

Table VIII. ENMC standards verification, pure and lightly impure oxides (α from 0.4 to 1.1)

			Passive Multiplicity Assay Results						
WRM ID	Count time, s	Declared Pu, g	Pu, g	1 σ, Pu, g	(D-A),%	1 σ, (D-A),%	Multi-plication	α	Comment
PEO-382A	3500	19.91	20.04	0.05	-0.676	0.246	1.004	0.941	Impure PEO oxide
STDISO12	14900	20.12	19.96	0.02	0.81	0.104	1.027	1.084	Impure SRP Pu oxide
ISO3,6,9,12	4455	51.37	51.33	0.08	0.086	0.146	1.025	1.036	Impure SRP Pu oxide
ISO3,6,9,12	375	51.37	51.21	0.27	0.303	0.526	1.025	1.044	Impure SRP Pu oxide
LAO250C10	780	59.57	58.39	0.14	1.971	0.227	1.019	0.584	Pure LAO oxide
PEO-382B	600	74.64	74.36	0.35	0.375	0.468	1.015	0.888	Impure PEO oxide
PEO-382C	780	149.3	148.3	0.7	0.649	0.484	1.024	0.868	Impure PEO oxide
LAO251C10	1700	170.7	169.6	0.4	0.686	0.237	1.03	0.516	Pure LAO oxide
PEO-382D	600	297.4	296.4	1.3	0.357	0.444	1.037	0.869	Impure PEO oxide
PEO-382D	300	298.5	295.9	1.5	0.873	0.518	1.037	0.863	Impure PEO oxide
PEO-382D	1500	298.5	296.8	1.3	0.567	0.428	1.034	0.873	Impure PEO oxide
LAO252C10	12000	319.7	318.0	0.3	0.514	0.094	1.049	0.499	Pure LAO oxide
LAO252C10	630	319.7	317.3	1.5	0.75	0.482	1.045	0.502	Pure LAO oxide
LAO252C10	570	319.7	318.4	1.6	0.391	0.485	1.049	0.497	Pure LAO oxide
LAO256C10	1500	382.2	381.8	0.9	0.122	0.241	1.051	0.49	Pure LAO oxide
PEO-385	56600	457.2	455.5	0.2	0.368	0.050	1.05	0.774	Impure PEO oxide
LAO255C10	480	540.0	544.3	2.6	-0.795	0.485	1.056	0.476	LAO shaken
LAO255C10	510	540.0	541.5	2.6	-0.278	0.481	1.062	0.482	Pure LAO oxide
LAO255C10	600	540.0	540.2	3.2	-0.023	0.596	1.061	0.486	LAO on bottom
LAO253C10	2160	608.5	609.9	1.5	-0.228	0.240	1.072	0.494	Pure LAO oxide
PEO-381	14700	611.6	610.9	0.7	0.117	0.117	1.058	0.786	Impure PEO oxide
PEO-381	48000	611.6	611.2	0.4	0.066	0.059	1.058	0.786	Impure PEO oxide
PEO-447	70600	774.5	780.0	0.4	-0.71	0.046	1.071	0.716	Impure PEO oxide
LAO261	17400	842.7	842.7	0.8	-0.005	0.097	1.085	0.483	Pure LAO oxide
LAO261C10	720	842.7	847.2	4.4	-0.533	0.519	1.081	0.475	Pure LAO oxide
LAO261C10	2460	842.7	841.6	2.0	0.136	0.236	1.085	0.486	Pure LAO oxide
STDSRP12-2	720	859.2	860.7	4.8	-0.182	0.561	1.118	1.045	Impure SRS Pu oxide
STDSRP12-2	1500	859.2	858.2	3.6	0.112	0.414	1.117	1.054	Impure SRS Pu oxide
STDSRP12-2	4700	859.2	862.2	2.0	-0.347	0.237	1.122	1.047	Impure SRS Pu oxide
STDSRP12-1	720	874.4	875.6	4.2	-0.137	0.484	1.124	1.037	Impure SRS Pu oxide
STDSRP12-1	5700	874.4	874.2	2.1	0.025	0.241	1.119	1.042	Impure SRS Pu oxide
261+253	690	1451.3	1454.5	7.0	-0.225	0.484	1.079	0.476	2 big LAOs stacked
261+253	1200	1451.3	1436.6	6.7	1.018	0.463	1.088	0.49	2 big LAOs stacked

Mean (D-A), %	0.19
1 standard dev., %	0.56
Precision, %	0.37

In Table VIII, plutonium masses range from 20 to 1451 g. Alpha varies from ~0.4 to ~1.1. These WRMs have been used for NDA calibration and training for many years. They have been studied extensively, and their pedigrees are very good. Their book values were all determined originally by weighing, sampling and chemical analyses. Their documentation is very good. Some of the measurements were taken in training courses, where we made off-normal measurements, e.g., shaking an item to change density, stacking items, and putting an item at the bottom of the sample well. From the data of Table VIII, the mean bias, 1-σ standard deviation, and 1-σ precision for these WRM measurements are +0.19, 0.56, and 0.37 %, respectively, compared with book WRM values. These results compare quite favorably with calorimetry. Various count times were used for these measurements. If all items had been counted long enough to reach 0.2% precision on the plutonium mass assay, count times would have varied from only ~20 to ~100 min. This set of measurements would satisfy the IAEA criterion for "bias-defect" detection (≤1% combined systematic and random error) , which is usually satisfied only by weighing, sampling, and chemical analyses.

VIII. ENMC/INVS VERIFICATION RESULTS – PRECISION

We chose one metal and seven plutonium oxide items (WRMs) for determining the passive-neutron-multiplicity assay (PNMA) precision performance of the ENMC/INVS. They are described in Table IX.

Table IX. Description of WRMs for ENMC/INVS precision determinations

	Wt% (relative to total Pu mass)									
WRM ID	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Date	g Pu	g $^{240}Pu_{eff}$	alpha
FZC158	0.016	0.955	93.77	0.699	4.53	0.001	12/15/78	0.6952	0.7054	0.180
CBNM Pu61	1.1969	62.5255	25.4058	6.6793	4.1925	1.4452	6/20/86	5.745	2.088	0.950
CBNM Pu93	0.0117	93.4123	6.3131	.02235	0.0395	0.1047	6/20/86	5.833	0.374	0.871
STDISO3	0.0065	96.302	3.5622	0.11107	0.01826	0.0172	7/1/86	10.972	0.395	1.51
STDISO6	0.0141	93.501	6.1285	0.29995	0.0569	0.0224	7/1/86	8.448	0.529	0.911
STDISO9	0.0213	92.606	6.8881	0.41144	0.0732	0.0201	7/1/86	11.883	0.840	0.845
STDISO12	0.0584	86.973	11.808	0.93961	0.2214	1.3861	7/1/86	20.220	2.498	1.094
STDISO15	0.1693	82.111	15.406	1.6035	0.7107	0.0682	7/1/86	12.318	2.107	0.691

PNMA assays were measured for all the WRMs in Table IX and for the STDISOs in combination. These assays had variable count times to achieve precisions from 0.1 to 1%. These data were converted to relative standard deviation (RSD) precisions that would be obtained in a 1-hour count time. The measured results are plotted in Fig. 5 along with those predicted by FOM.[5] Precisions predicted by the FOM calculations are uniformly lower than those of the measurements, but agree to within ~40–50%.

Figure 5 shows that 0.2% precision is achievable for a wide range of $^{240}Pu_{eff}$ mass in only 1 hour of counting. This precision performance is comparable to calorimetry, in a much shorter time.

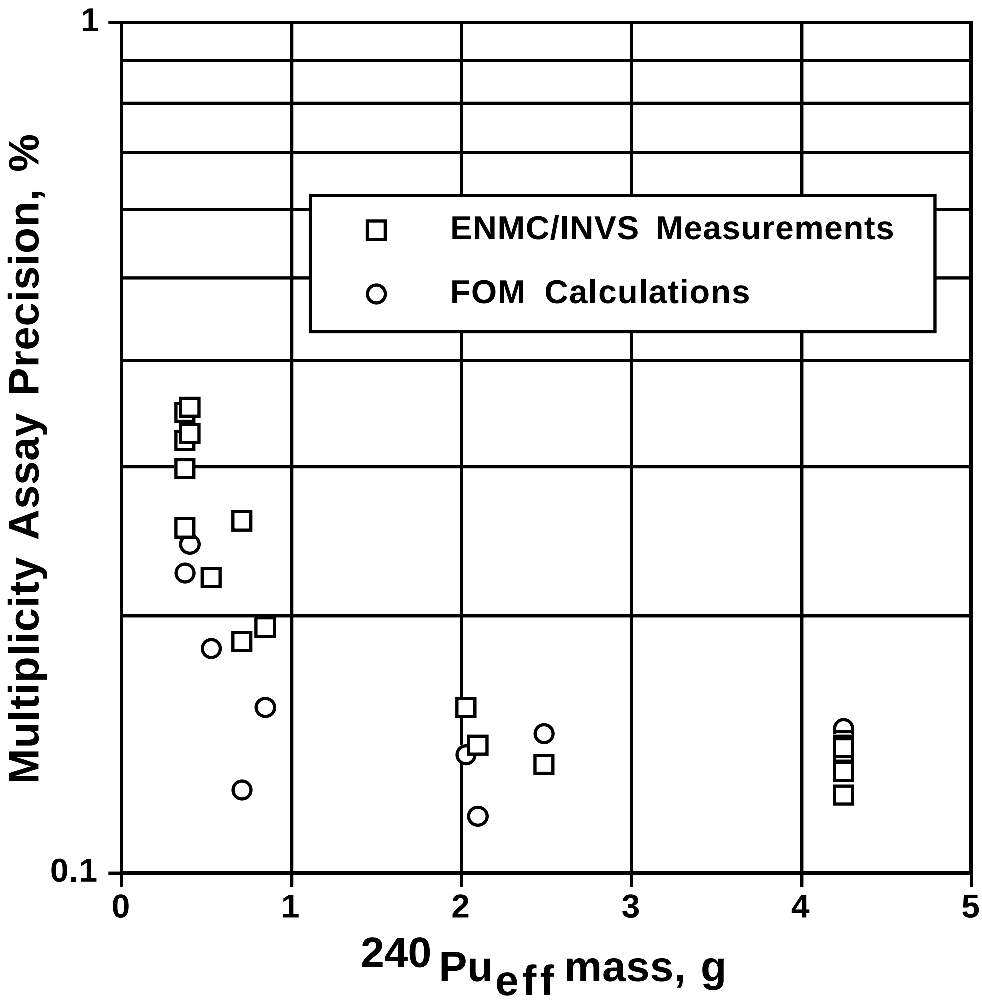

Fig. 5. Precision of ENMC/INVS PNMAs for small plutonium WRMs typical of those collected for on-site laboratories. Count time is 3600 s.

IX. ENMC/INVS VERIFICATION RESULTS – ACCURACY

Results of ENMC/INVS PNMA measurements are shown in Table X and Figs. 6a and 6b. Twenty measurements of the eight WRMs described in Table IX are represented. Count times are variable. The calibration parameters in Table II were used for all assays. These results are excellent, and compare quite favorably with calorimetry. They indicate that the ENMC/INVS can play a pivotal role for on-site laboratories, reducing measurement times and the considerable expense of item sampling and chemical analyses.

Table X. ENMC/INVS small-sample standards verification: pure and impure oxides (α from 0.7 to 1.5), and impure metal (α = 0.2).

WRM ID	Count time, s	Declared Pu, g	Declared $^{240}Pu_{eff}$, g	Passive Multiplicity Assay Results							
				Pu, g	1 σ, Pu, g	(D-A), g	(D-A),%	1 σ, (D-A),%	Multi-plication	α	Comment
FZC-158	375	0.691	0.704	0.681	0.004	0.01	1.429	0.579	0.996	0.183	run to 0.6%
FZC-158	11500	0.691	0.704	0.687	0.001	0.004	0.522	0.145	0.996	0.179	run to 0.15%
CBNM Pu61	29910	5.556	2.019	5.616	0.003	-0.06	-1.078	0.054	1.021	0.968	1000 x 30s
CBNM Pu61	29880	5.556	2.019	5.625	0.003	-0.069	-1.244	0.054	1.021	0.932	1000 x 30s
CBNM Pu93	300	5.824	0.373	5.823	0.065	0.002	0.031	1.116	1.024	0.859	run to 1.1%
CBNM Pu93	29850	5.824	0.373	5.856	0.006	-0.032	-0.551	0.103	1.022	0.846	1000 x 30s
CBNM Pu93	300	5.824	0.373	5.811	0.051	0.013	0.227	0.876	1.023	0.911	run to 0.9%
CBNM Pu93	29910	5.824	0.373	5.844	0.007	-0.02	-0.346	0.120	1.022	0.895	1000 x 30s
STDISO6	19600	8.432	0.528	8.425	0.008	0.007	0.083	0.095	1.008	0.911	run to 1%
STDISO3	375	10.962	0.395	10.981	0.119	-0.019	-0.178	1.086	1.011	1.525	run to 1.1%
STDISO3	38700	10.962	0.395	11.071	0.011	-0.109	-0.992	0.100	1.01	1.509	run to 0.1%
STDISO9	11160	11.855	0.838	11.813	0.013	0.042	0.35	0.110	1.012	0.845	run to 0.1%
STDISO15	7500	12.218	2.09	12.107	0.012	0.111	0.912	0.098	1.011	0.691	run to 0.1%
STDISO12	6600	20.121	2.486	20.212	0.02	-0.091	-0.454	0.099	1.023	1.094	run to 0.1%
ISO3;6;9;12	66	51.37	4.247	50.324	0.497	1.046	2.036	0.967	1.018	1.095	run to 1%
ISO3;6;9;12	72	51.37	4.247	50.708	0.501	0.662	1.289	0.975	1.022	1.074	run to 1%
ISO3;6;9;12	76	51.37	4.247	51.497	0.504	-0.127	-0.247	0.981	1.016	1.05	run to 1%
ISO3;6;9;12	74	51.37	4.247	50.675	0.506	0.695	1.353	0.985	1.02	1.078	run to 1%
ISO3;6;9;12	1410	51.37	4.247	50.892	0.101	0.478	0.931	0.197	1.019	1.073	run to 0.2%
ISO3;6;9;12	8100	51.37	4.247	50.784	0.048	0.586	1.141	0.093	1.019	1.098	run to 0.1%

Mean (D-A), %	0.26
1 standard dev., %	0.92
Precision, %	0.61

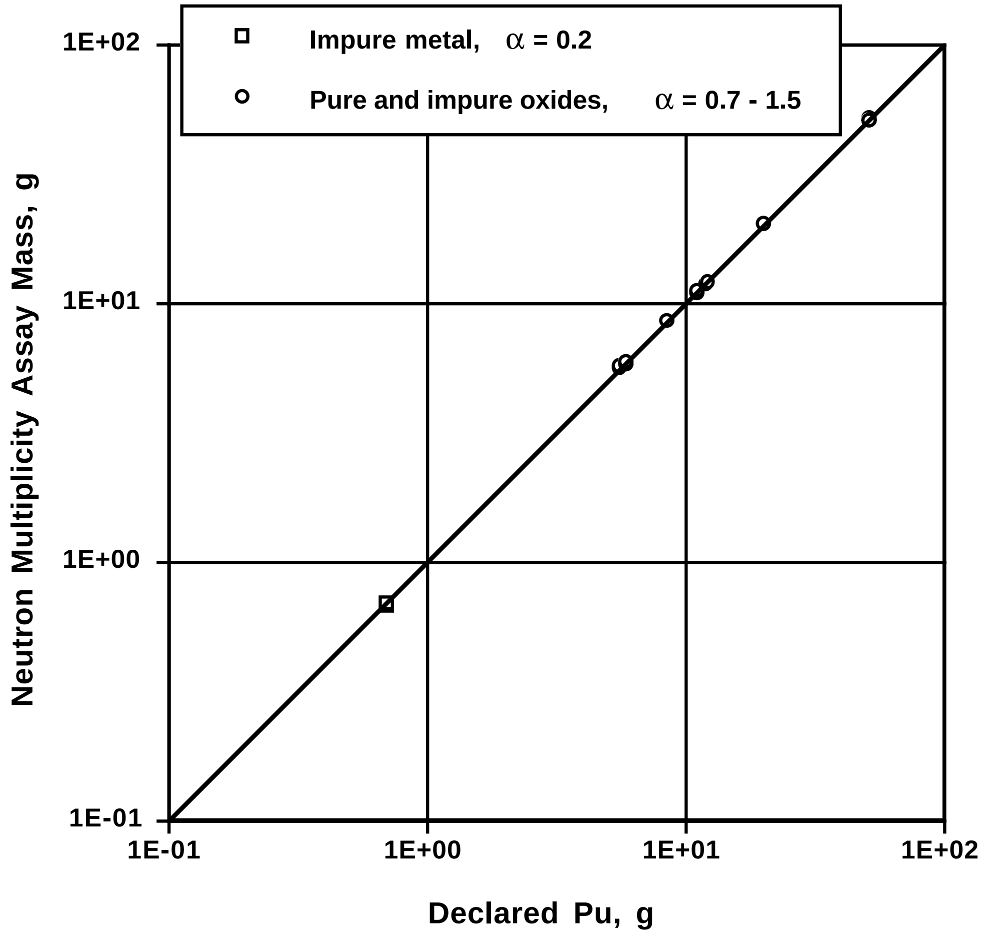

Fig. 6a. ENMC/INVS PNMAs versus book values for 20 measurements of 8 small plutonium WRMs typical of those collected for on-site laboratories. Count time is variable.

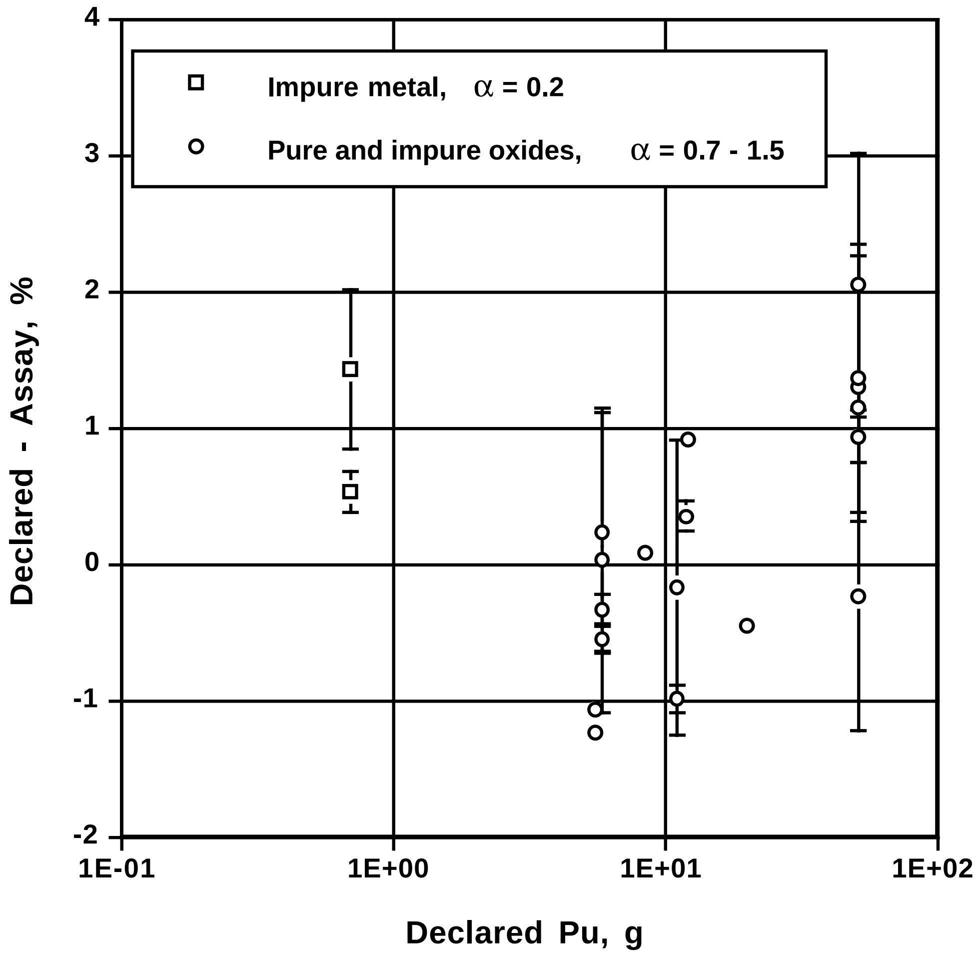

Fig. 6b. Book values minus ENMC/INVS PNMAs for 20 measurements of 8 small plutonium WRMs typical of those collected for on-site laboratories. Count time is variable.

X. ACTIVE ENMC

Monte Carlo simulations[6] were conducted to optimize performance of the ENMC for active measurements of fissile mass, primarily ^{235}U. Combinations of polyethylene, graphite, nickel and aluminum were considered, but the best performance was indicated for pure polyethylene. We fabricated polyethylene end plugs for the sample cavity based on the Monte Carlo simulations. The nominal configuration of the end plugs is shown in Fig. 7. The figure shows an active cavity 8 in. (20.3 cm) tall. The endplugs are adjustable for taller items.

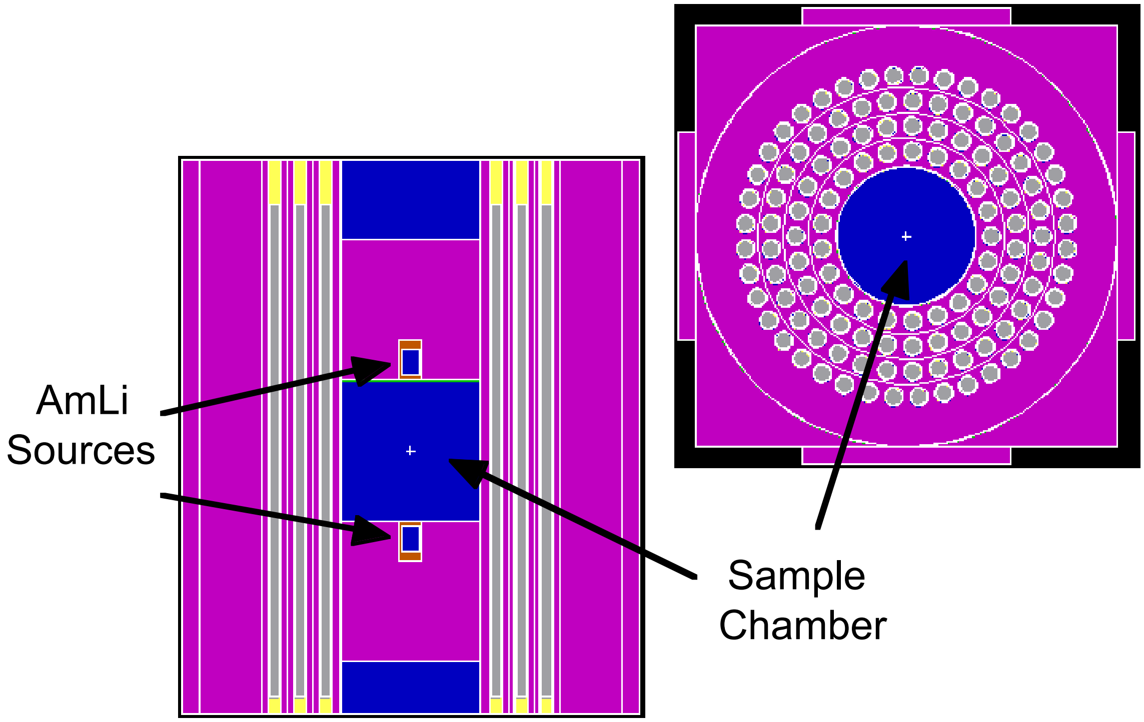

Fig. 7. Configuration of the active-mode ENMC

Series of active-fast-mode measurements were carried out to compare performance of the ENMC with the AWCC.[10] The AmLi sources MRC-115 and MRC-116[17] were used for these measurements. The UISO series of uranium oxide enrichment standards were counted in both systems, using the same cavity height. The data are shown in Table XI.

Table XI. Active-mode measurements of the UISO standards in the ENMC and AWCC.

Standard	U, g	^{235}U, g	AWCC Count time	AWCC Doubles, s^{-1}	ENMC Count time	ENMC Doubles, s^{-1}	Count-time factor
UISO-12	990	116.8	88 x 600 s	31.97 ± 0.32	274 x 30 s	150.2 ± 1.5	6.4 ± 1.1
UISO-13	991	128.4	79 x 600 s	35.77 ± 0.36	218 x 30 s	159.4 ± 1.6	7.3 ± 1.4
UISO-17	989	170.5	282 x 120 s	44.40 ± 0.43	105 x 30 s	198.3 ± 1.9	10.7 ± 1.7
UISO-27	991	265	125 x 90 s	64.41 ± 0.69	99 x 20 s	276.3 ± 2.7	5.7 ± 1.1
UISO-38	991	372.1	203 x 60 s	81.16 ± 0.75	148 x 10 s	351.0 ± 3.5	8.2 ± 1.3
UISO-52	989	515.4	197 x 30 s	102.8 ± 1.1	166 x 5 s	441.5 ± 4.6	7.1 ± 1.1
UISO-66	990	658.8	123 x 30 s	125.5 ± 1.3	144 x 5 s	521.5 ± 5.0	5.1 ± 0.9
UISO-91	990	904	175 x 20 s	165.7 ± 1.5	71 x 5 s	685.5 ± 6.7	9.9 ± 2.0

Each standard was counted to 1% precision using the sample standard deviation method. Note that the ENMC produces 4–5 times the doubles coincidence rate per gram ^{235}U as the AWCC. Table XI also shows the count-time reduction factor of the ENMC compared to the AWCC. This is defined as the relative time (AWCC/ENMC) required to reach a fixed precision for a given standard. This factor ranges from 5.1 to 10.7.

Reference 18 gives results of predictions for % relative standard deviations (%RSDs) of several assay systems, including the AWCC and ENMC. These results are compared with measured values in Table XII. These results agree to within a factor of two. However, the ENMC measurements were made with an AMSR[14,15] while the AWCC measurements were made with a conventional shift register (CSR). The AMSR gives a factor of 1.41 lower precision than the CSR for these measurements. Therefore, the count-time factors for the FOM code and measured values are not directly comparable. The FOM factors should be increased by a factor of two. This leaves the RSD values for the AWCC still within a factor of two and the RSD values for the ENMC in good agreement. The relative count-time reduction factors are overpredicted by the FOM code by more than a factor of two, compared with the measured values.

Table XII. 1000-s active assay precisions from the FOM code compared with measured Values.

Precision Determination	^{235}U, g	%RSD, AWCC	%RSD, ENMC	Count-time factor
FOM code	200	10.2	3.0 (2.1)	11.6 (23.2)
measured	170.5 (UISO-17)	5.8	1.8	10.7
FOM code	1000	3.6	1.0 (0.7)	13.0 (26.0)
measured	904 (UISO-91)	1.9	0.6	9.9

The FOM-code calculations used an ENMC fission-neutron-detection efficiency of 60%, whereas the MCNP simulations predict a value of 55%. Also, FOM used an ENMC AmLi-neutron-detection efficiency of 16% compared with a measured value of 29%. These differences can explain the differences in ENMC relative to AWCC precision performance.

Figure 8 shows the calibration curves for the ENMC and AWCC measurements described above. Error bars are smaller than the plotted points.

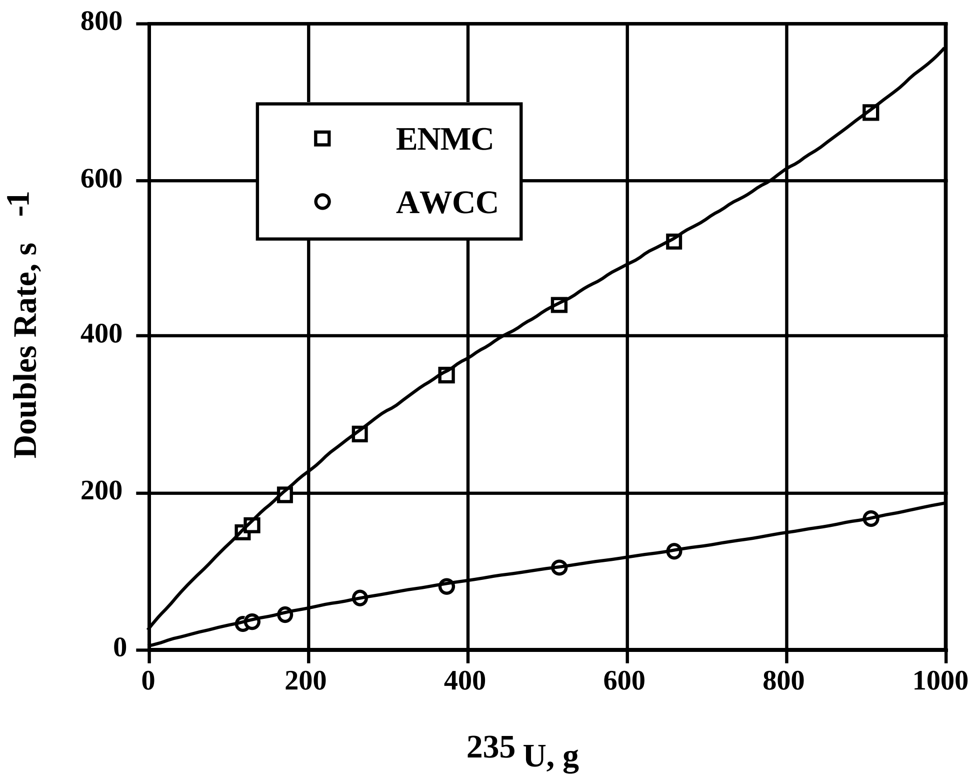

Fig. 8. Calibration curves for the UISO standards measured in the ENMC and AWCC.

XI. CONCLUSIONS

The ENMC is the first of a new generation of neutron coincidence and multiplicity counters. It has unprecedented performance compared with previous designs. The underlying physics principles apply to all existing neutron coincidence and multiplicity counters, both active for uranium and passive for plutonium. For the first time, it may be possible to perform passive neutron multiplicity measurements of some large uranium items, e.g., light-water reactor fresh-fuel assemblies (the neutron coincidence collar[17]). Passive measurements of these large uranium items (especially LEU), would not require as many physical standards as active measurements and be much less susceptible to item bulk density, ^{235}U enrichment variations and burnable poisons. The new SuperHENC[19] standard-waste-box counter for Waste Isolation Pilot Plant certification uses the same physics principles, and will improve assay sensitivity by an order of magnitude. Commercial variants of the ENMC concept are already being developed.

We've built active endplugs (with AmLi sources) for the ENMC to assay ^{235}U. Large gains in precision are obtained because all active neutron coincidence assays are dominated by accidental coincidences, and the ENMC is much less affected by accidental coincidences than TNMCs.

We are interested in working with industrial partners to commercialize this technology.

And finally, fitted with an AMSR[14,15] count times for the ENMC and all other neutron coincidence and multiplicity counters will be reduced by <u>an additional factor of two</u>, except for some small items.

ACKNOWLEDGEMENT

This work was sponsored by the US Department of Energy, Office of Nonproliferation and International Security, Office of Safeguards and Security, and the Los Alamos Directed Research Development (LDRD) program.

REFERENCES

1. N. Ensslin, W. C. Harker, M. S. Krick, D. G. Langner, M. M. Pickrell, and J. E. Stewart, "Application Guide to Neutron Multiplicity Counting," Los Alamos National Laboratory report LA-13422-M (November 1998).
2. H. O. Menlove, J. Baca, M. S. Krick, K. E. Kroncke, and D. G. Langner, "Plutonium Scrap Multiplicity Counter Operation Manual," Los Alamos National Laboratory report LA-12479-M (January 1993).
3. J. E. Stewart, M. S. Krick, J. Xiao, R. J. Lemaire, and V. Fotin, "Performance of the Plutonium Scrap Multiplicity Counter (PSMC) for Verification of Excess Scrap Plutonium Oxide from Nuclear Weapons Production," Los Alamos National Laboratory document LA-UR-97-2070 (January 1993).
4. J. E. Stewart, R. R. Ferran, and M. S. Krick, "Measurement Performance Optimization for Thermal Neutron Multiplicity Counters," in Safeguards and Security Progress Report: January-December, 1989, Los Alamos National Laboratory report LA-11914-PR (November 1990).
5. N. Ensslin, M. S. Krick, and N. Dytlewski, "Assay Variance as a Figure-of-Merit for Neutron Multiplicity Counting," *Nucl. Instr. and Meth.* **A290**, 197–207 (1990).
6. J. F. Briesmeister, Ed., "MCNP – A General Purpose Monte Carlo Code for Neutron and Photon Transport," Ver. 4A, Los Alamos National Laboratory report LA-12625-M (November 1993).
7. H. O. Menlove, J. E. Stewart, and D. R. Mayo, "Advanced Performance Epithermal Neutron Detector for Measurement of Plutonium Inventory Samples," presented at the 6th International Meeting on Facility Operations – Safeguards Interface, sponsored by the American Nuclear Society Isotopes and Radiation Division, Jackson Hole, Wyoming, September 20–24, 1999, Los Alamos National Laboratory document LA-UR-99-2861.
8. L. Carrillo, N. Ensslin, M. S. Krick, and J. E. Stewart, "A Reference ^{252}Cf Source Set for Neutron Coincidence and Multiplicity Counting Applications," *Nucl. Mater. Manage.* **XXVIII,** CD-ROM (Proc. Issue—1999).
9. H. O. Menlove and J. E. Swansen, "A High-Performance Neutron Time Correlation Counter," *Nucl. Technol.* **71**, 497–505 (November 1985).
10. H. O. Menlove, "Description and Operation Manual for the Active Well Coincidence Counter," Los Alamos Scientific Laboratory report LA-7823-M (May 1979).

11. J. E. Stewart, M. S. Krick, T. D. Reilly, D. G. Langner, W. Theis, R. Lemaire, and J. Xiao, "Implementation of Neutron Counting Techniques at U. S. Facilities for IAEA Verification of Excess Materials from Nuclear Weapons Production," *Nucl. Mater. Manage*. **XXIV** (Proc. Issue), 548–554 (1995).
12. J. E. Stewart, M. S. Krick, J. Xiao, R. J. Lemaire, V. Fotin, L. McRae, D. Scott, and G. Westsik, "Assay of Scrap Plutonium Oxide by Thermal Neutron Multiplicity Counting for IAEA Verification of Excess Materials from Nuclear Weapons Production," *Nucl. Mater. Manage*. **XXV** (CD-ROM Proc. Issue), 263–267 (1996).
13. T. L. Cremers, T. E. Sampson, N. L. Scheer, and M. K. McDonald, "ARIES Nondestructive Assay (NDA) System," Los Alamos National Laboratory document LA-UR-98-5337 (November 1998).
14. M. E. Abhold, M. R. Sweet, S. C. Bourret, W. J. Hansen, J. Audia, M. S. Krick, S. D. Salazar, and J. E. Stewart, "Advanced Multiplicity Shift Register with Networking Capability," *Nucl. Mater. Manage*. **XXVIII,** CD-ROM (Proc. Issue—1999).
15. J. E. Stewart, S. C. Bourret, M. S. Krick, M. R. Sweet, W. C. Harker, and A. Gorobets, "New Shift-Register Circuits for Improving Precision of Neutron Coincidence and Multiplicity Assays," paper presented at the 6th International Meeting on Facility Operations – Safeguards Interface, sponsored by the American Nuclear Society Isotopes and Radiation Division, Jackson Hole, Wyoming, September 20-24, 1999, Los Alamos National Laboratory document LA-UR-99-4927.
16. M. S. Krick, D. G. Langner, and J. E. Stewart, "Energy-Dependent Bias in Plutonium Verification Measurements Using Thermal Neutron Multiplicity Counters," presented at the IAEA Symposium on International Safeguards, Vienna, Austria, October 13—17, 1997, Los Alamos National Laboratory document LA-UR-97-3427.
17. H. O. Menlove, J. E. Stewart, S. Z. Qiao, T. R. Wenz, and G. P. D. Verrecchia, "Neutron Collar Calibration and Evaluation for Assay of LWR Fuel Assemblies Containing Burnable Neutron Absorbers," Los Alamos National Laboratory report LA-11965-MS (November 1990).
18. N. Ensslin, W. H. Geist, M. S. Krick, D. R. Mayo, and J. E. Stewart, "Performance Calculations for Fast Active Neutron Assay," *Nucl. Mater. Manage*. **XXVIII,** CD-ROM (Proc. Issue—1999).
19. H. O. Menlove, J. M. Boak, M. L. Collins, M. S. Krick, D. R. Mayo, C. D. Rael, C. M. Brown, J. B. Franco, and D. J. Santi, "The SuperHENC Mobile Passive Neutron Measurement System for Counting Plutonium in Standard Waste Boxes," presented at the 7th Nondestructive Assay Waste Characterization Conference, Salt Lake City, Utah, May 23-25,2000, Los Alamos National Laboratory document LA-UR-00-1687.

This report has been reproduced directly from the best available copy. It is available electronically on the Web (http://www.doe.gov/bridge).

Copies are available for sale to U.S. Department of Energy employees and contractors from—

Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831
(423) 576-8401

Copies are available for sale to the public from—

National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22616
(800) 553-6847

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545