LA-UR-05-4296

Comparison of Gamma and Passive Neutron Non-Destructive Assay Total Measurement Uncertainty Using the High Efficiency Neutron Counter

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ABSTRACT

Since 1997, the High Efficiency Neutron Counter (HENC) has been employed by the Los Alamos National Laboratory (LANL) to assay and ship to the Waste Isolation Pilot Plant (WIPP) plutonium waste using passive neutron methods. During 2004 and early 2005, the HENC has undergone a modification and recertification to add a Canberra broad energy germanium detector (BEGe), DSA-1000 digital spectrum analyzer, and JSR-14 neutron analysis shift register so as to conduct simultaneous gamma and passive neutron measurements. The BEGe detector provides both isotopic and quantitative measurement data to complement the neutron detectors. The assay is under the control of Canberra's NDA-2000 software suite that provides acquisition control and analysis.

As part of the recertification effort, the total measurement uncertainty (TMU) is being evaluated. A comparison of the gamma and the passive neutron TMU will be provided and the NDA 2000 settings used to achieve the results will be summarized and reported.

INTRODUCTION

The High Efficiency Neutron Counter (HENC) has been employed by the Los Alamos National Laboratory (LANL) to assay plutonium waste using passive neutron methods since 1997. During 2004 and early 2005, the HENC has undergone a modification and recertification to add a Canberra broad energy germanium detector (BEGe), DSA-1000 digital spectrum analyzer, and JSR-14 neutron analysis shift register so as to conduct simultaneous gamma and passive neutron measurements. The BEGe detector provides both isotopic and quantitative measurement data to complement the neutron detectors. The assay is under the control of Canberra's NDA-2000 software suite that provides acquisition control and analysis.

As part of the recertification effort, the total measurement uncertainty (TMU) is being re-evaluated. Unfortunately, the measurements to support a full TMU determination could not be performed so, in lieu, data from a similar instrument, the MCS HENC, was used to evaluate the effect of the changes in operating methodology[1] LANL intends to use.

The focus of this study is to evaluate how the gamma and neutron measurements, and the associated TMU, should be used for the LANL HENC in light of the operational experience of the MCS HENC. There are significant differences in how the neutron data is to be used, utilizing only the add-a-source corrected doubles coincidences and neutron Ones rates. Further, while the MCS

HENC utilizes a second order calibration curve, the LANL HENC uses a linear calibration constant. These are the changes with which the MCS data are evaluated, and care should be exercised in extending these results to MCS Henc operations.

METHODOLOGY

1. NDA 2000 Data from HENC Measurement of Standards

The MCS HENC has been acquiring assay data at Los Alamos since late 2003. The database for this study ranges from data acquired beginning in January 2003 through April 2005. This data consists of assays acquired with National Institute of Standards and Technology (NIST) traceable standards in three matrices: empty drum, debris drum, and inorganic solid (sludge) drum. Source locations were varied and provide a reasonable estimate of location variation for the TMU estimate.

The data of interest for this study are the neutron and gamma assay results. The neutron data acquired include the doubles coincidence rate, the "Ones" rate, the background doubles and ones rate, the Add-a-Source (AAS) correction factor, and the standard deviations of each of the above values. For the gamma acquisition, the Pu239 mass, Pu239 activity, Pu239 minimum detectable activity (MDA), and the related standard deviations were evaluated.

2. Total Measurement Uncertainty

The TMU developed for the MCS HENC[2] is summarized in Table 1. The individual components of the TMU are shown expressed as relative standard deviation percentages (%RSD). A subtotal %RSD was added because the neutron assay requires an additional isotopic measurement (usually from a gamma detector) to obtain the total Pu mass. The gamma detector in the HENC consists of an energy efficiency calibration and thus a separate isotopic measurement is not required. In the analysis to follow, the isotopic uncertainty is not propagated since true source isotopic values are used. Consequently, the comparison is to the values identified for the Subtotal in Table 1.

Table 1. Total Measurement Uncertainty

	Neutron Detector		Gamma Detector	
	Debris	Sludge	Debris	Sludge
Counting Statistics	1%	1%	0.8%	1.1%
AAS Correction/Matrix Non-Homogeneity	4%	5%	5%	5%
Source Distribution	2.9%	17%	9%	25%
Multiplication Effects	0%	0%	N/A	N/A
Background Effects	0%	0%	N/A	N/A
Calibration Uncertainty	2%	2%	2.5%	2.5%
SUBTOTAL	5%	18%	11%	26%
Isotopic Ratio Uncertainty	12%	12%	N/A	N/A
Nominal TMU (1σ)	**13.2%**	**21.5%**	**10.6%**	**25.6%**

3. Standards

The standards used for this study are summarized in Table 2. All the sources are traceable to the NIST. The sources are either PDP[3] sources or are designed just like the PDP sources. The data needed for this study include the total Pu mass of the source, the Pu240 effective mass fraction, and the Pu239 mass fraction. The source name is included with the table.

The mass fraction of Pu240 effective mass to total Pu, f_{Pu240E}, is defined in terms of the mass fractions of Pu238, Pu240, and Pu242 by the following equation[4]:

$$f_{Pu240E} = 2.52 f_{Pu238} + 1.0 f_{Pu240} + 1.68 f_{Pu242} \tag{1}$$

Table 2. Standards

Nominal Source Strength	Mass Pu (g)	Pu240 Effective Mass Fraction	Pu239 Mass Fraction	Source Name
0.1	0.10029	0.06075	0.93761	PDP1-0.1
0.2	0.22249	0.06545	0.93308	NTP-0001, 0064, 0071, 0078
0.5	0.50195	0.06075	0.93761	PDP1-0.5
0.59	0.59936	0.05945	0.93761	NTP-0085, 0099
0.6	0.60224	0.06075	0.93761	PDP1-0.1, PDP1-0.5
3.01	3.01210	0.06075	0.93761	NTP-0120
3.02	3.01760	0.06075	0.93761	PDP1-3.0
3.05	3.05530	0.06075	0.93761	NTP-0106
3.4	3.40805	0.06075	0.05945	NTP0064, 0071, 0099,0106
10	10.00560	0.06075	0.93761	PDP1-10
13	13.02320	0.05945	0.93761	PDP1-3.0, PDP1-10
15	15.04800	0.05946	0.93772	NTP-0140
25	26.92100	0.06869	0.92966	RANT25-1
30	30.03500	0.06080	0.93772	NTP-0148
50	50.03132	0.06869	0.92985	RANT50-1
95	95.04000	0.06080	0.93772	NTP0140, 0148, 0156
125	126.96264	0.06869	0.92985	RANT25-1, 50-1, 50-2
150	150.07806	0.06869	0.92985	RANT50-1, 50-2, 50-3
160	160.00900	0.06080	0.93772	NTP0140, 0148, 0156, 0164
175	176.99906	0.06869	0.92985	RANT25-1, 50-1, 50-2, 50-3

4. Matrix Drums and Source Positioning

The matrix drums are those built to PDP[5,6] specifications. The matrix drums contain tubes permitting one to place one or more standards at varying positions in the drum. The empty and debris drums each contain three tubes, one along the central axis, one displaced 5.5” from the center, and the third tube displaced 9” from the center in which standards may be placed. The height of the source can be varied from the bottom to the top of the drum. For the sludge drum, four tubes are available starting at the center with the remaining tubes located 3.5”, 5.75”, and 9” respectively from the center.

The frequency of source placement for the given HENC assay data is tabulated in Figure 1. As shown, there is a reasonably good distribution of sources for the empty and debris drums, but the source positions are mostly centered in tubes T3 and T4 for the sludge drum.

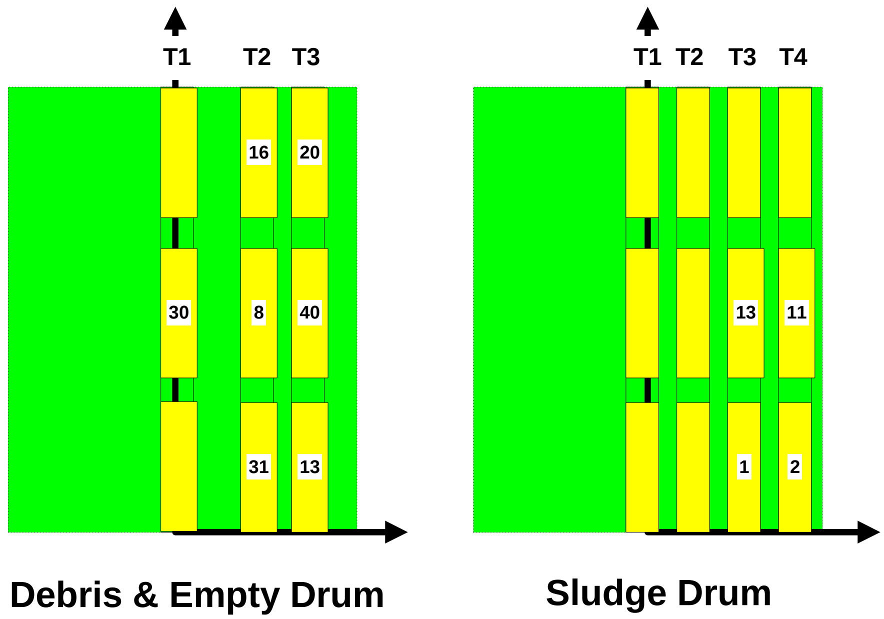

Figure 1. Source Location Frequency in 55 gal PDP Type Drums

5. Operations

The operating conditions include a 4 Pi neutron detector with 113 neutron detectors covering the drum. The gamma detector is a single broad energy germanium detector positioned approximately at the midpoint of the drum. During the count, the drum is rotated and data is acquired by both the neutron and the gamma electronics. A Cf252 add-a-source correction is made to the neutron assay to correct for matrix effects. During the time the Cf252 source is exposed, the gamma acquisition is inhibited. The total assay time for the gamma and neutron measurements is 30 minutes.

6. Calculation of Pu Mass for the Neutron Assay

The plutonium mass estimated from an assay depends on whether neutron or gamma data is to be utilized. For the purpose of this study, mass estimates from both the neutron and gamma measurements are evaluated.

For the neutron assay, either the net neutron doubles rates or the ONES rates is selected depending on the cutoff value. The neutron doubles rate[1] is the number of neutron coincidences measured over the length of the assay divided by the assay time. The coincidences include all neutron multiplicities from 0 to 255. The neutrons ONES rate excludes multiplicities higher than two to minimize the effect of cosmic radiation and are therefore more suited when the Pu mass is small.

The Neutron Doubles cutoff and the Ratio of Doubles to ONES rate was originally calculated for the LANL HENC and reported at the last INMM annual meeting . The MCS HENC utilizes a calibration of the ONES rate rather than a ratio of the doubles to ONES rates that is planned for the LANL HENC. However, for the purpose of this study, the ratio must be recalculated since it will

[1] The singles rate is actually the sum of all shift register triggers from a neutron event divided by the count time. The doubles rate is the singles rate times the mean of the reals plus accidental neutron distribution minus the mean of the accidental neutron distribution. See reference 4 for a complete description.

differ from one instrument to the next. The doubles cutoff for the MCS HENC was re-evaluated based on the neutron data for the empty drum at 3 net doubles coincidences and is comparable to approximately 1 gram (See Figure 2). Thus, at a net doubles below 3 n/s, the ONES rate is used while above that value, the Doubles rate is used.

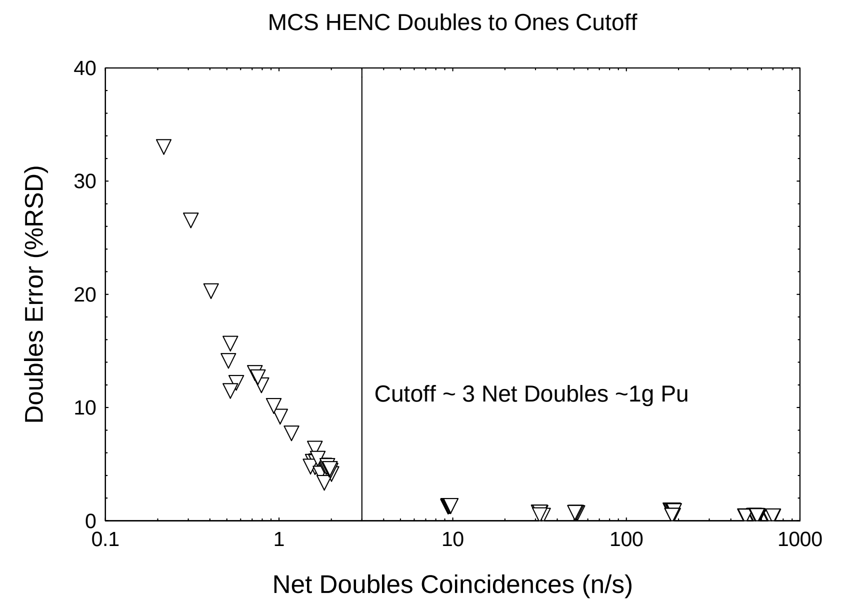

Figure 2. Neutron Doubles to Ones Cutoff

The ratio of the Doubles to Ones rate is determined from the Doubles and Ones calibration factor. The Doubles calibration for the MCS HENC is 51.71 ± 0.17 net doubles per gram of Pu240 effective while the Ones calibration is 41.642 ± 0.818 net doubles per gram of Pu240 effective. Consequently, the ratio of Doubles to Ones is 1.242 ± 0.835. This factor effectively converts the Ones rate to a Doubles rate permitting the use of the doubles calibration with the Ones rate.

The mass of Pu (in grams) is given by the following equations. Here,

k_{AAS} = Add-a-Source (AAS) correction factor,

k_{DO} = Doubles to Ones Ratio,

K_D = Doubles calibration factor,

N_D = Doubles coincidence rate (n/s),

N_O = Ones coincidence rate (n/s).

$$m_{Pu} = \frac{k_{AAS} N_D}{K_D f_{Pu240E}}, \text{ for } N_D > 3 \tag{2}$$

$$m_{Pu} = \frac{k_{AAS} k_{DO} N_O}{K_D f_{Pu240E}}, \text{ for } N_D \leq 3 \tag{3}$$

The lower limit of detection in Pu mass units is calculated from the standard deviations of the daily background and the assay as shown below. These relations are taken from Currie[7] and Makaruk[8] and represent the error at the 95% confidence level with type I and II errors both set at 5%. Here, the following symbols are used:

σ_{BD} = Standard deviation of the background doubles rate

σ_{BO} = Standard deviation of the background ones rate

σ_{SD} = Standard deviation of the assay doubles rate

σ_{SO} = Standard deviation of the assay ones rate

$$m_{LLD} = \frac{1.65 k_{AAS}(\sigma_{BD} + \sigma_{SD})}{K_D f_{Pu240E}}, \text{ for } N_D > 3 \tag{4}$$

$$m_{LLD} = \frac{1.65 k_{AAS} k_{DO}(\sigma_{BO} + \sigma_{SO})}{K_D f_{Pu240E}}, \text{ for } N_D \leq 3 \tag{5}$$

7. Gamma Data

The gamma assay Pu239 mass is taken directly from the NDA 2000 acquisition and converted to total Pu mass from the known Pu239 mass fraction of the standard. The LLD is taken from the NDA 2000 reported Pu239 minimum detectable activity (MDA) and converted to total Pu mass by applying the activity per unit mass of Pu239 (0.0629 Ci/g).

$$m_{Pu} = \frac{m_{Pu239}}{f_{Pu239}} \tag{6}$$

$$m_{LLD} = \frac{MDA_{Pu239}}{0.0629 f_{Pu239}} \tag{7}$$

ASSAY RESULTS

The results of these assays are summarized in Table 3. There are a total of 234 assays in the database. The three matrices are shown along with the true mass range. The total number of data points for the neutron (N) and the gamma (G) assay are shown. The numbers of neutron and gamma assays are not always identical because for some assays either the neutron or gamma assay measurement failed; in these cases, the neutron or gamma data was removed from the analysis. The average ratio of the measured to the true mass (R%), expressed as a percentage is shown next followed by the one percent relative standard deviation (%RSD) of the mass points. Finally, the average LLD mass is shown.

Table 3. Assay Summary

			Neutron			Gamma		
Matrix	Pu Mass (g)	Data Points (N/G)	R%	%RSD	LLD (g)	R%	%RSD	LLD (g)
Empty	0 - 1	37/31	110.7%	14.6%	0.067	85.3%	11.5%	0.027
	1 - 15	16/16	100.1%	1.3%	0.131	86.1%	9.5%	0.048
	15 - 175	48/23	105.5%	4.2%	1.131	85.2%	23.6%	0.205
	Overall	234/174	106.6%	15.6%	0.577	85.4%	15.6%	0.101
Debris	0 - 10	31/28	104.1%	12.7%	0.110	103.2%	16.7%	0.077
	10-25	27/27	108.8%	5.1%	0.430	87.2%	13.9%	0.186
	25-175	42/35	110.5%	11.6%	1.201	94.4%	29.0%	0.322
	Overall	106/93	107.2%	11.6%	0.625	94.7%	22.2%	0.203
Sludge	0.5 - 25	22/27	111.9%	29.2%	0.416	138.9%	37.2%	0.313
All Matrices	0 - 1	41/43	107.5%	17.8%	0.057	93.8%	25.7%	0.038

1-15	62/59	101.0%	11.6%	0.163	107.3%	33.5%	0.109
15-25	36/36	116.9%	16.0%	0.524	94.9%	19.6%	0.247
25-175	84/52	107.9%	9.1%	1.229	89.2%	23.8%	0.292
Overall	229/196	107.3%	13.7%	0.58	97.2%	28.4%	0.178

The accuracy or bias (%R) generally favors the neutron assay. The neutron data exhibits a high bias of less than 7.2% for the empty or debris matrix and 11.9% for the sludge matrix. By comparison, the gamma detector exhibits a low bias of less than 14.6% for the empty or debris matrices, and a high bias of 38.9% for the sludge. The data therefore suggests that the neutron data is preferred IF a good isotopic measurement can be obtained from the gamma detector and IF the assay is above the neutron LLD.

The precision (%RSD) is generally better for the neutron detectors than for the gamma detector. For all matrices, the neutron %RSD is 13.7% compared to 28.4% for the gamma. In all cases except for the range 0-1 g Pu, the precision is better for the neutron detectors than the gamma detectors. Even for the sludge matrix, the neutron detectors perform better in both %RSD (29.2 vs 37.2%) and accuracy (111.9 vs 138.9%). These data also suggest that the neutron measurement should be the measurement of choice for all matrices provided a good gamma isotopic measurement can be obtained and the assay is above the LLD.

The LLD for the gamma detector is generally better than the neutron LLD as shown in the table. The gamma LLD ranges from 25 to 82% better than the neutron LLD. This suggests that for small quantities of Pu (e.g., below 1 g weapons grade Pu or equivalent), the gamma detector will perform better than the neutron detectors.

The TMU must include both the accuracy and precision of the errors added in quadrature. The results of this evaluation are tabulated in Table 4. The accuracy and precision errors are taken from Table 3 and the isotopic error is taken from Table 1 without change. The revised TMU is compared with the reported TMU in Table 1. The sludge TMU may be understated because the source positioning did not cover the entire drum; on the other hand, the basic assumption for sludge is that the nuclear material is homogeneous. The data suggests that the neutron TMU is better (smaller) than the gamma TMU.

Table 4. Total Measurement Uncertainty Comparison

	Accuracy |100-%R|	Precision (%RSD)	Isotopic (%)	Revised TMU (%)	Reported TMU (%)
Neutron					
Debris	7.2%	11.6%	12%	18.2%	13.2%
Sludge	11.9%	29.2%	12%	33.7%	21.5%
Gamma					
Debris	5.3%	22.2%	NA	22.8%	10.6%
Sludge	38.9%	37.2%	NA	53.8%	25.6%

CONCLUSIONS

The neutron assay is the preferred assay when a measurable isotopic value is available and the assay is above the systems lower limit of detection.

The gamma assay generally produces a lower limit of detection that is 25 to 82% better (lower) than available from the neutron assay and should be used whenever the neutron assay fails.

The total measurement uncertainty (TMU) originally reported should be increased for both the neutron and gamma assays.

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