

# Active and Passive Mode Calibration of the Combined Thermal Epithermal Neutron (CTEN) System

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## ABSTRACT

The Combined Thermal/Epithermal Neutron (CTEN) non-destructive assay (NDA) system was designed to assay transuranic waste by employing an induced active neutron interrogation and/or a spontaneous passive neutron measurement. This is the second of two papers, and focuses on the passive mode, relating the net double neutron coincidence measurement to the plutonium mass via the calibration constant. National Institute of Standards and Technology (NIST) calibration standards were used and the results verified with NIST-traceable verification standards.

Performance demonstration program (PDP) "empty" 208-L matrix drum was used for the calibration. The experimentally derived calibration constant was found to be  $0.0735 \pm 0.0059$  g  $^{240}\text{Pu}$  effective per unit response. Using this calibration constant, the Waste Isolation Pilot Plant (WIPP) criteria was satisfied with five minute waste assays in the range from 3 to 177g Pu. CTEN also participated in the PDP Cycle 8A blind assay with organic sludge and metal matrices and passed the criteria for accuracy and precision in both assay modes. The WIPP and EPA audit was completed March 1, 2002 and full certification is awaiting the closeout of one finding during the audit. With the successful closeout of the audit, the CTEN system will have shown that it can provide very fast assays (five minutes or less) of waste in the range from the minimum detection limit (about 2 mg Pu) to 177 g Pu.

## INTRODUCTION

The Combined Thermal/Epithermal Neutron (CTEN) non-destructive assay (NDA) system (Figure 1) was designed to assay transuranic (TRU) waste by employing an induced active neutron interrogation and/or a spontaneous passive neutron measurement. In the active mode, a 14-MeV neutron generator combined with moderating material produces a thermal and epithermal interrogating flux of neutrons to induce fission in waste isotopes and the resultant prompt neutrons are measured. In the passive mode, prompt spontaneous neutron emissions from waste isotopes are measured to produce the assay. The CTEN was installed at the Los Alamos Non-Destructive Assay (NDA) facility to characterize waste for the TRU Waste Characterization Project (TWCP). During early 2001, the unit underwent checkout and calibration in preparation for certification.

The CTEN employs a total of 42 cadmium-shielded and 41 bare (non-cadmium shielded) detectors to measure the fission neutron signal. Nine of the bare detectors are  $^4\text{He}$  types for measuring the response in the epithermal region and the remainders are  $^3\text{He}$  types. For WIPP certification purposes, only the  $^3\text{He}$  detectors are employed. While the passive mode employs both the shielded and bare detectors, the active mode employs the shielded detectors. The CTEN also employs a matrix correction technique derived from correlations of the flux and drum monitor responses (both shielded and bare detectors) to the active interrogation signal to correct the passive or active assayed

mass for absorption and moderation of the neutron signal in the waste matrix. The detectors are located so as to provide four-pi coverage of a 208-L waste drum. Figure 2 shows the flux and drum monitors located in the assay chamber; the neutron signal detectors are imbedded in the four sides, top, and bottom of the instrument. The CTEN electronic modules record the arrival time of each neutron pulse as well as the total neutron counts as a function of one of twelve drum sectors. The passive neutron count data is subsequently processed to determine the coincidence rates using techniques<sup>1</sup> developed at Los Alamos. The coincidence rates are the data used to develop the passive calibration.

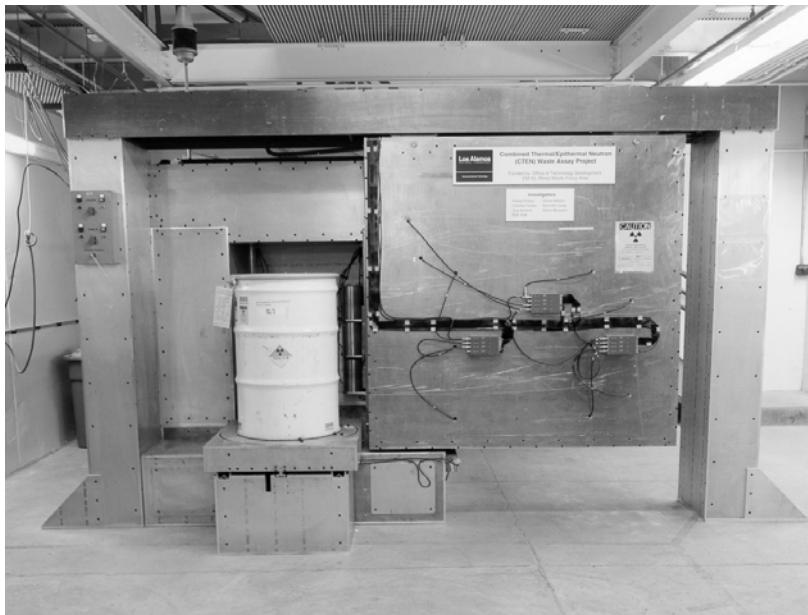


Figure 1. The CTEN Assay System

In the active assay, an external neutron source, a Zetatron, is used to pulse the waste and induce neutrons from fissile material. Neutron counts are normalized to  $^{239}\text{Pu}$  with a correction factor for the  $^{241}\text{Pu}$  contribution, because the former isotope is generally present in much larger concentrations. In the passive assay, spontaneous emission is from the even plutonium isotopes (typically  $^{238}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{242}\text{Pu}$ ) using prompt neutron coincidence measurements with the neutron counts normalized to the  $^{240}\text{Pu}$  mass. Matrix correction techniques employ the active interrogation results to correct the assay in both the active and the passive measurements.

This is the second paper produced to describe the calibration process. The first paper<sup>2</sup> described the active mode calibration and was presented at the Environmental Management NDA Conference<sup>3</sup>. This paper summarizes the passive mode calibration results.

## NEUTRON MEASUREMENTS

In the active calibration, the neutron counts were normalized to the effective  $^{239}\text{Pu}$  by incorporating the  $^{241}\text{Pu}$  contribution via the parameter, ICF. A correction to the ICF definition of the previous paper<sup>2</sup> should have shown it as the reciprocal to the one in the paper, that is, the correct form is depicted in equation (1), with all remaining equations remaining the same.

$$ICF = \frac{F_{239} + \frac{f_{Pu241}}{f_{Pu239}} F_{241}}{F_{239}} \quad (1)$$

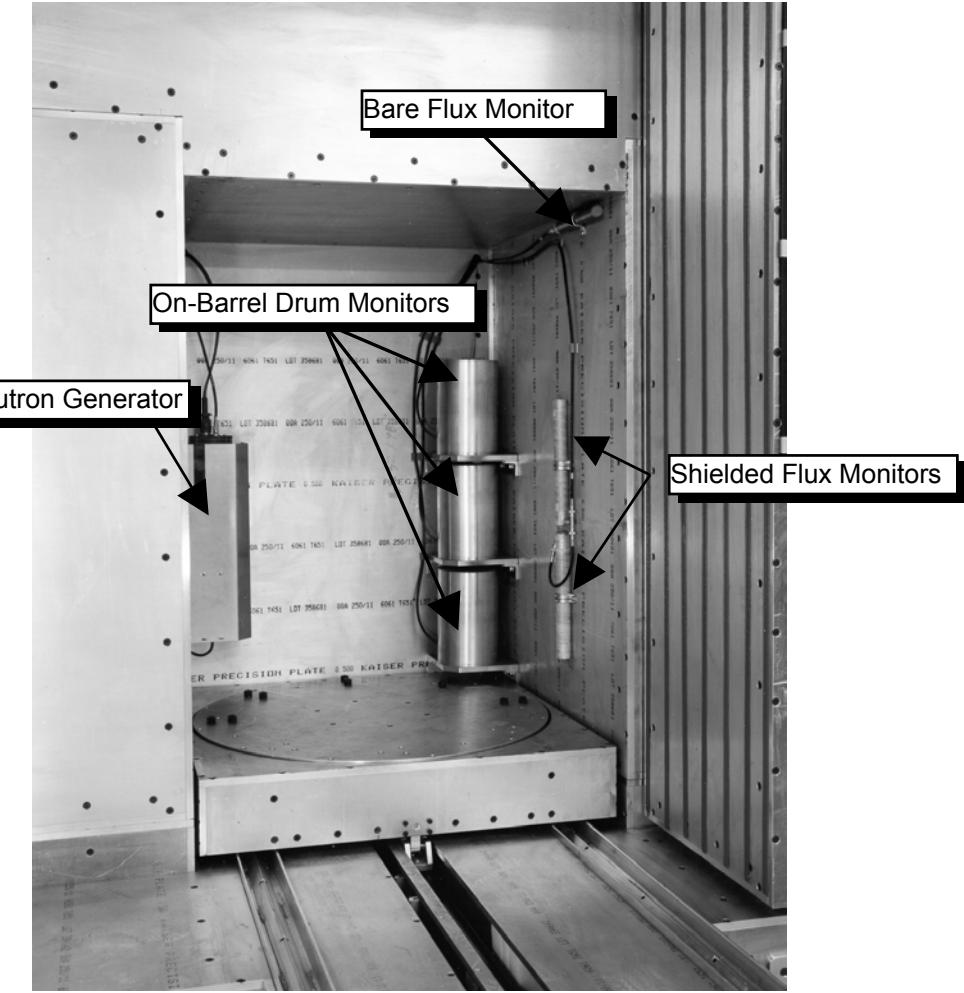


Figure 2. CTEN Assay Chamber Depicting Flux Monitors

In the passive mode, it is the effective mass of  $^{240}\text{Pu}$  that is measured. Assuming that there are no other sources of spontaneous neutron emitters other than the Pu isotopes, the effective  $^{240}\text{Pu}$  mass,  $m_{Pu240\text{Eff}}$ , is given by the correlation<sup>4</sup> shown in equation (2)

$$m_{Pu240\text{Eff}} = 2.52m_{Pu238} + 1.0m_{Pu240} + 1.68m_{Pu242} \quad (2)$$

The passive system response is directly related to the effective  $^{240}\text{Pu}$  mass shown in equation (3) which is in turn related to the net neutron coincidences,  $R - R_0$ , the matrix correction factor, MAT, and the calibration constant, K. For calibration measurements, the MAT value is that of an empty drum, i.e., the matrix is comprised of air with the minimum structure needed to hold the sources.

The blank drum response,  $R_0$ , is the coincidence counts obtained from the empty drum with no radioactive source while  $R$  is the coincidence counts obtained with a radioactive source in the geometrical center of the drum.

$$m_{Pu240Eff} = K(R - R_0)MAT \quad (3)$$

The plutonium mass,  $m_{Pu}$ , is defined via the mass fraction for each isotope, equation (4), and employs the  $^{240}\text{Pu}$  correlation as shown by equation (5).

$$f_j = \frac{m_j}{m_{Pu}} \quad (4)$$

$$m_{Pu} = \frac{m_{Pu240Eff}}{f_{Pu240Eff}} = \frac{K(R - R_0)MAT}{2.52f_{Pu238} + 1.0f_{Pu240} + 1.68f_{Pu242}} \quad (5)$$

A plot of  $(R - R_0) * MAT$  versus  $m_{Pu240Eff}$  can be used to determine the calibration factor,  $K$ , and this is the method used in developing the passive calibration coefficient for CTEN.

## EXPERIMENTAL SETUP

The calibration and verification source standards are National Institute of Standards and Technology (NIST) traceable plutonium standards in the range from 0.5 to 50 g. Each source has a certificate of content and traceability. The calibration standards were grouped to produce nominal mass calibration points at 0.5, 3.0, 25, 50, 100, 125, 150, and 175 g. The verification sources were selected to verify the calibration results and to comply with the quality assurance objectives (QAO) of the Waste Isolation Pilot Plant Waste (WIPP) Acceptance Criteria<sup>5</sup>. Verification was performed at 0.1, 1.0, 3.0, 10, and 160 grams.

Decay correction of the isotope mass fraction to the time of the assay (approximately seven years) was not necessary since the maximum error of neglecting the correction was approximately 2.54% in alpha activity and 0.11% in the effective  $^{240}\text{Pu}$  mass (Figure 3).

The container used for the assay was a 208-L zero matrix (empty) drum developed for the Program Demonstration Program<sup>6</sup> (PDP). These containers have a support structure to securely hold one or more sources as shown in Figure 4. One or more sources were positioned as close to the geometrical center of the drum as possible. In cases where four sources were needed (at 175 g) two sources were positioned at R0 (25 & 50g) and two at R5.5 (14 cm from the center)\*.

The data acquisition software, WIN-CTEN<sup>7</sup>, was used to acquire the data and the data analysis software, CTEN-FIT<sup>8</sup>, was used to determine the neutron coincidence rates from the neutron event binary history files.

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\* For the verification sources at 160g, three sources were centered on the axis and a single source was located at R5.5

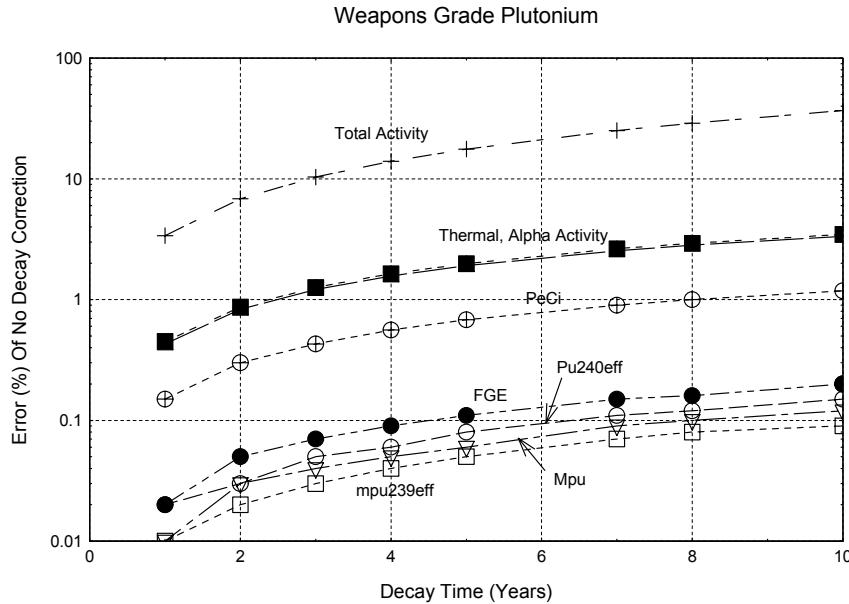


Figure 3. Decay Correction Errors

Each passive measurement was for five minutes while the active matrix correction measurements were taken with 20,000 pulses (about 3:20 minutes each assay). The same measurement times applied for the calibration measurements are also used for all WIPP certification assays.

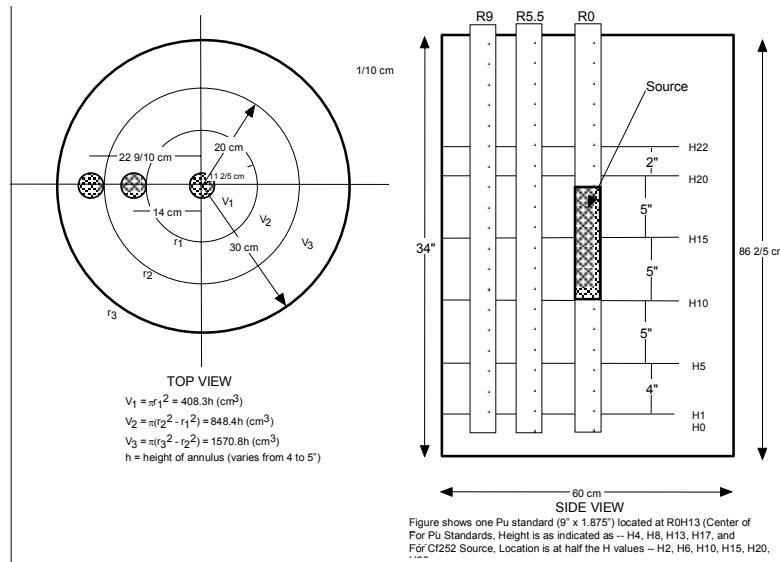


Figure 4. Waste Drum Coordinate System

## RESULTS

The calibration data plotted in Figure 5 can be fitted by a linear response through the zero point with slope  $13.610 \pm 1.09$  net doubles coincidence rate per unit mass of  $^{240}\text{Pu}$  effective (g). According to equation (3), the inverse of this value is the calibration factor, K. That is,

$$K = 0.0735 \pm 0.0059 \text{ grams } ^{240}\text{Pu effective per net unit double coincidence rate} \quad (6)$$

In this expression, the uncertainty in K is the one standard deviation of the propagated error due to the uncertainty in the standard mass, the response, the blank response, and the passive matrix transmission coefficient, MAT.

$$\sigma_K = \sqrt{\sigma_{Pu240Eff}^2 + \sigma_R^2 + \sigma_{R_0}^2 + \sigma_{MAT}^2} \quad (7)$$

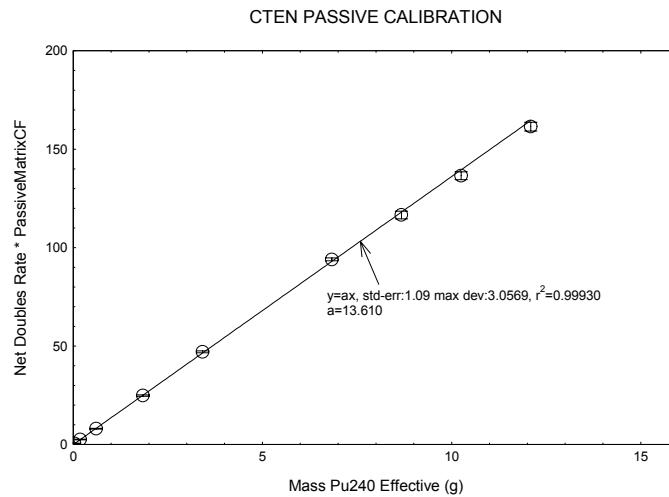


Figure 5. Passive Neutron Coincidence Calibration

The calibration measurements were compared to the verification measurements in developing the WIPP QAO. In this instance, the net response is compared to the total mass showing good agreement above 3 g between the net double neutron coincidence measurements for the calibration and verification standards, as shown in Figure 6. The range of useable passive assays was restricted to a  $^{240}\text{Pu}$  effective mass between 0.1845 g and 12.0913 g (approximately 3 g to 177 g of weapons grade Pu) because the QAO acceptance criteria failed below 0.1845 g and the larger limit represents the highest calibration mass used.

The range of certifiable assays with CTEN (Table 1) varies from the minimum detectable concentration (MDC) to 177 g of weapons grade plutonium. The MDC can be as low as 0.002 grams of Pu, depending on the background uncertainty in the facility. At the Radioassay and Nondestructive Testing (RANT) facility where CTEN is located, the MDC is typically around 0.005 g. In a typical waste assay, both an active and passive measurement is performed with active assays usually limited to about 3 g of Pu due to self-shielding concerns. The passive assay is typically used above 3 g.

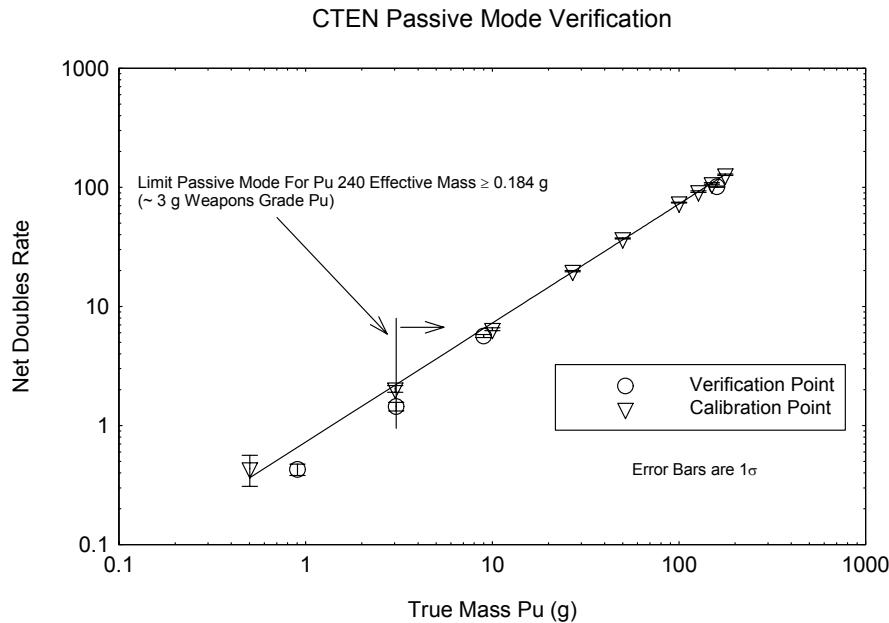


Figure 6. Verification and Calibration Results Compared

CTEN participated in the PDP Cycle 8A blind assay<sup>9</sup> and passed all the criteria, including both precision and accuracy requirements. Two matrices were evaluated: an organic sludge matrix and a metals matrix. The sludge matrix contained 20,035 MBq (0.55 Ci) of plutonium standards and was assayed with the active mode<sup>2</sup>. The metals drum contained 108,780 MBq (2.94 Ci) and was assayed with the passive mode with the passive results summarized in Table 2. The passive assay resulted in an accuracy of 92.86% and an uncertainty of 1.54%, based on the standard deviation of six replicate measurements.

Table 1. CTEN Assay Range For Weapons Grade Plutonium

Assay Method	Equivalent Weapons Grade Pu (grams)
Active	MDC – 9.0
Passive	3.06 – 177.0

The CTEN WIPP/EPA audit was conducted February 25 through March 1, 2002. All elements were reviewed including procedures for data acquisition, data analysis and reporting, calibration, quality, calibration verification, matrix correction, self-shielding, total measurement uncertainty, range of the assay, and problem identification and resolution. There was one condition adverse to quality (CAR) generated for CTEN related to incorporation of the active-mode self-shielding from waste in the total measurement uncertainty for an assay. Prior to the audit, self-shielding in fissile material was assumed to be 1.0 (no self-shielding) with no error. In the post-audit assay, the self-shielding from the plutonium fissile waste was included based on Monte Carlo derived estimates and the error added in quadrature to the propagated error. We are currently awaiting approval of the CAR response prior to certifying waste for WIPP on the CTEN.

Table 2. PDP Cycle 8A Scoring Report For Metal Drum

Site	Method			Acceptance Criteria--Interfering Matrix					
		Measured Parameters		Bias		Precision		Status	
		%R	%RSD	Lower	Upper	%R	%RSD	Bias	Precision
INEEL	PAN-SGRS	120.87%	3.94%	44.14%	155.86%	6.00%	PASS	PASS	
INEEL	PAN-WAGS	119.76%	2.60%	42.73%	157.27%	6.00%	PASS	PASS	
<b>LANL</b>	<b>CTEN</b>	<b>92.86%</b>	<b>1.54%</b>	<b>41.61%</b>	<b>158.39%</b>	<b>6.00%</b>	<b>PASS</b>	<b>PASS</b>	
LANL	TGS	79.62%	0.98%	41.03%	158.97%	6.00%	PASS	PASS	
LANL	HENC	107.40%	0.55%	40.57%	159.43%	6.00%	PASS	PASS	
RFETS	PAN-Bldg 569	83.51%	3.19%	43.35%	156.65%	6.00%	PASS	PASS	
RFETS	SuperHENC	155.90%	2.10%	42.20%	157.80%	6.00%	PASS	PASS	
WRAP	GEA-A	99.52%	0.76%	40.80%	159.20%	6.00%	PASS	PASS	
WRAP	GEA-B	95.41%	1.17%	41.23%	158.77%	6.00%	PASS	PASS	

## CONCLUSION

The passive mode calibration constant is  $0.0735 \pm 0.0059$  grams  $^{240}\text{Pu}$  effective per unit doubles coincidence rate. The passive system response (the doubles coincidence rate) is linear over the assay range of 0.5 to 177g of weapons grade plutonium. The passive assay is restricted to the range 3 – 177 g with an assay time of five minutes to meet WIPP requirements on precision and accuracy. Below approximately 3 g, the active mode is the preferred assay method. The CTEN passed the Program Demonstration Program (PDP) cycle 8A in both the active and passive mode. The WIPP/EPA audit for certification was conducted February 25 – March 1, 2002 that resulted in one condition adverse to quality (CAR) that has been addressed; we are currently waiting closeout of that item before becoming certified with CTEN.

## REFERENCES

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- <sup>3</sup> *8<sup>th</sup> Environmental Management Nondestructive Assay Characterization Conference Proceedings*, December 11-13, 201, Denver, Colorado.
- <sup>4</sup> Ensslin, N., W. Harker, M. Krick, D. Langner, M. Pickrell, and J. Stewart, *Application Guide to Neutron Multiplicity Counting*, Los Alamos National Laboratory, LA-13422-M, November 1998.
- <sup>5</sup> *Waste Acceptance Criteria for the Waste Isolation Pilot Plant*, Revision 7, Change 2, DOE/WIPP-069, Carlsbad, NM, January 24, 2001.
- <sup>6</sup> *Performance Demonstration Program for Nondestructive Assay of Drummed Wastes for the TRU Waste Characterization Program, Cycle 7A Scoring Report*, CAO Technical Assistance Contractor, October 2000.
- <sup>7</sup> Estep, R., *Requirements Document for WIN\_CTN*, Los Alamos National Laboratory, TWCP-06780, July 9, 2001.
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