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

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ABSTRACT

The Combined Thermal Epithermal Neutron (CTEN) system was developed by the Los Alamos National Laboratory to perform active and passive neutron interrogation of waste. The higher energy epithermal neutrons are able to penetrate further into the matrix and active material, thus reducing matrix attenuation and self-shielding effects compared to a thermal neutron pulse alone. The developmental unit was installed in 2001 at the Los Alamos Non-Destructive Assay (NDA) facility to characterize waste for the TRU Waste Characterization Project (TWCP). This paper summarizes the active mode certification results. National Institute of Standards and Technology (NIST) traceable standards were used to determine the system response as a function of mass. Finally, NIST-traceable verification standards were used to verify the calibration in the range 30 milligrams to 25 g of weapons grade plutonium although self-shielding limits the upper active interrogation to 10 g.

1. INTRODUCTION

The Combined Thermal/Epithermal Neutron (CTEN) non-destructive assay (NDA) system (Figure 1) was designed to assay transuranic (TRU) waste by employing either (or both) an induced active neutron interrogation or a spontaneous passive neutron measurement. In the active mode, the higher energy epithermal neutrons are able to penetrate further into the matrix and active material, thus reducing matrix attenuation and self-shielding effects compared to a thermal neutron pulse alone. 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. This paper summarizes the active mode calibration results.

In an active interrogation, the CTEN employs a Zetatron neutron generator producing pulses of 14-MeV neutrons at repetition rates of 50-100 Hz to interrogate the waste[1]. A total of 83 shielded and bare detectors are used to measure the fissile signal utilizing a differential dieaway technique (DDT). The detectors are located so as to provide four-pi coverage of a 208-L waste drum. Figure 2 shows some of the detectors located in the assay chamber; the remaining detectors are located in the four sides, top, and bottom of the instrument.

Figure 1. The CTEN Assay System

2. NEUTRON MEASUREMENTS

A waste assay with the CTEN depends either on neutron emissions induced in the waste by an external source (active mode) or on the spontaneous emission of neutrons (passive mode). In the active assay, an external neutron source, such as a Zetatron, is used to pulse the waste and induce neutrons primarily from ^{239}Pu. Neutrons are also induced from ^{241}Pu, but because of its much smaller cross section, the number induced is significantly less than from ^{239}Pu. In the passive assay, spontaneous emission is from the even plutonium isotopes (typically ^{238}Pu, ^{240}Pu, and ^{242}Pu) using either single or coincident neutron measurements. In this paper, the focus is on measurement of the induced neutrons from an active assay.

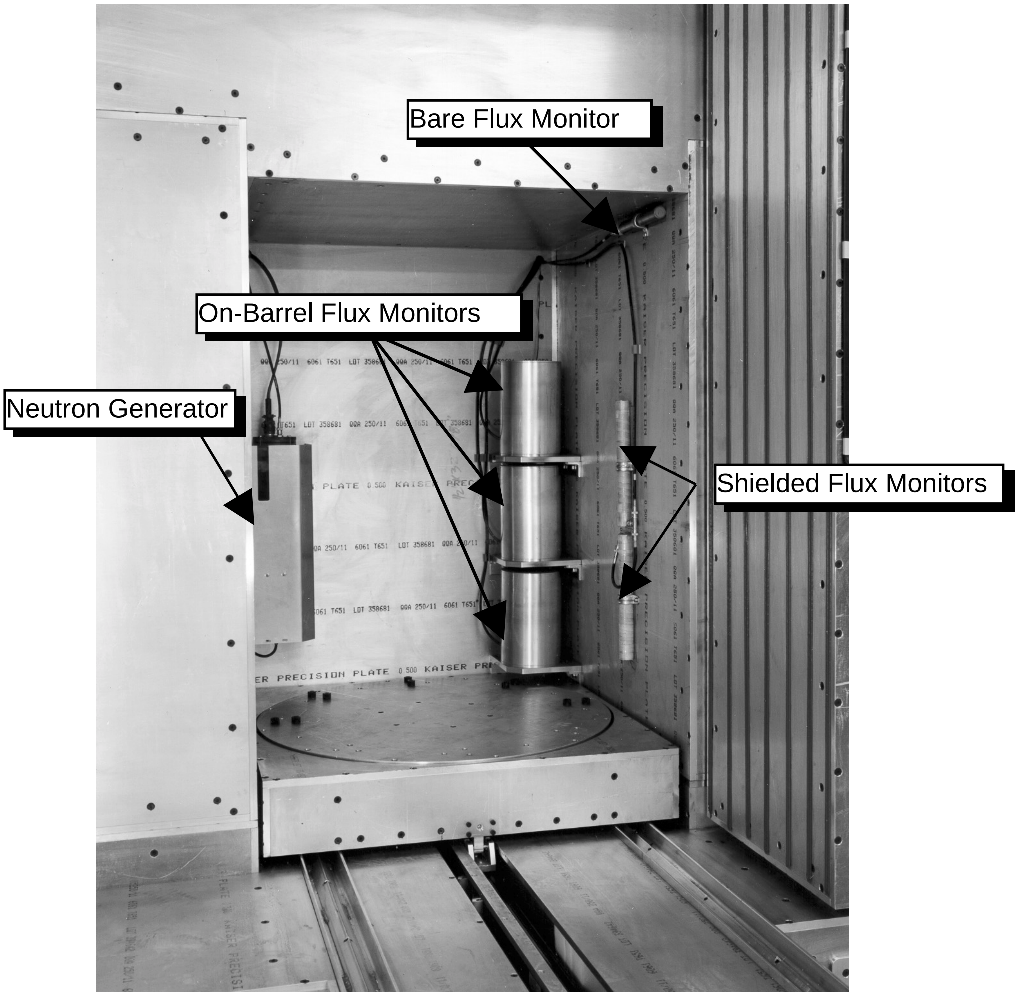

Figure 2. CTEN Assay Chamber Depicting Flux Monitors

In the CTEN active assay, pulses of neutrons are used to interrogate the waste and this is repeated at a 100 Hz rate. During the assay, the waste drum rotates through 360 degree to help remove any bias due to geometric variability. Each pulse, approximately 20 μs long, generates approximately one million neutrons. Typically, 20000 pulses are generated per assay. The incident neutron must interact with the nucleus in order to induce secondary neutrons. The number of induced neutrons produced depends on the cross-section for the interaction and the mean number of induced neutrons emitted per incident neutron. The primary source of induced neutrons in plutonium waste is from the ^{239}Pu isotope with a minor contribution from ^{241}Pu. Consequently, an effective ^{239}Pu mass, m_{Pu239E}, is defined by equation (1) where ICF incorporates both the ^{239}Pu and the ^{241}Pu contributions. The ICF equation is defined by the F-values which are determined from the cross-sections, σ; the average induced neutrons emitted per incident neutron, $\overline{\upsilon}$; the atomic mass, A, and the isotopic mass ratios to total plutonium, f. The subscripts refer to ^{239}Pu or ^{241}Pu, respectively. The values of $\sigma, \overline{\upsilon}$, and A are available from the literature and the mass ratios, f, are available from the calibration data.

$$m_{Pu239E} = ICF * m_{Pu239} \tag{1}$$

$$ICF = \frac{F_{239}}{F_{239} + \frac{f_{Pu241}}{f_{Pu239}} F_{241}} \tag{2}$$

$$F_{239} = \frac{\sigma_{Pu239}\overline{\upsilon}_{Pu239}}{A_{Pu239}}; \quad F_{241} = \frac{\sigma_{Pu241}\overline{\upsilon}_{Pu241}}{A_{Pu241}} \tag{3}$$

Self-shielding occurs when the interrogating neutron pulse cannot reach the atoms to induce fission and when induced or spontaneous neutron emissions cannot reach the detector due to absorption. Monte Carol experiment and analysis with the calibration sources used in this study have shown that self-shielding in an active assay is important, contributing to over 15% even for the smallest source[2]. Consequently, an apparent ^{239}Pu effective mass is defined as shown in equation (4); this equation makes the CTEN response linear with the apparent effective ^{239}Pu mass.

Consequently, CTEN measures the apparent effective mass of plutonium, Pu239EA. The mass fraction must be determined from another instrument, typically an isotopic gamma measurement, while ICF is available from the literature. In typical waste, the plutonium content is assumed to be homogenous and, with this assumption, SSF = 1. However, for the calibration standards used, this is not the case and SSF was determined by Monte Carlo measurements. With the apparent effective mass known, the total plutonium mass is determined by applying equation (5).

$$m_{Pu239EA} = SSF * m_{Pu239E} \tag{4}$$

$$m_{Pu} = \frac{m_{Pu239}}{f_{Pu239}} = \frac{m_{Pu239E}}{f_{Pu239} * ICF} = \frac{m_{Pu239EA}}{f_{Pu239} * ICF * SSF} \tag{5}$$

3. PLUTONIUM CALIBRATION STANDARDS

The calibration standards (Table 1) used are NIST traceable plutonium standards in the range from 0.1 to 25 g. Each source has a certificate of content and traceability. Also used to verify that the calibration produced correct results were NIST traceable verification standards. The verification standards ranged from 0.03 to 9.0 g of total plutonium and were not used to generate the calibration curve.

Decay correction 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 0.07% in the effective ^{239}Pu mass (Figure 3).

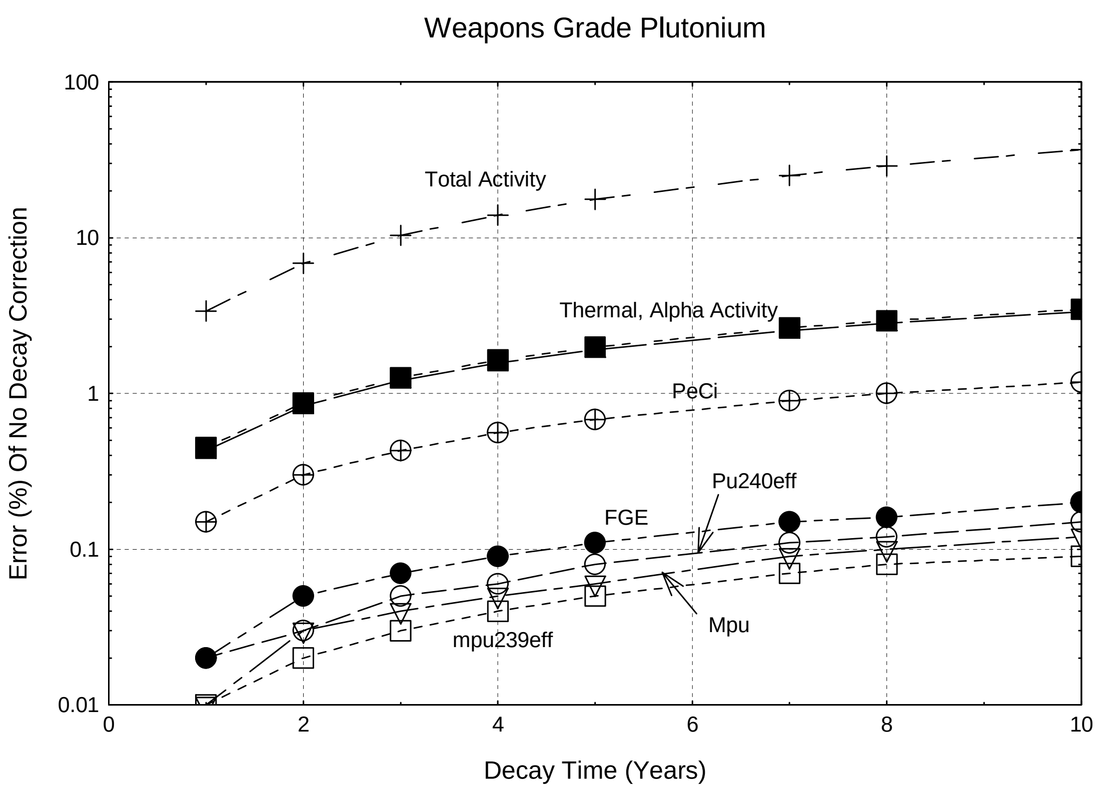

Figure 3. Decay Correction Errors

Table 1. Calibration and Verification Plutonium Standards

Source	Mass Pu (g)	Mass Fraction Pu239	Mass Fraction Pu241	ICF	Self Shielding Factor	Effective Mass Pu239 (g)	Effective Apparent Mass Pu239 (g)	α Activity (MBq)
Calibration Sources								
PDP1-0.1	0.1003	0.937614	0.002237	0.9967	0.8666	0.093721	0.081219	286.2
PDP1-0.5	0.5020	0.937614	0.002237	0.9967	0.8588	0.469073	0.402840	1432.3
PDP1-3.0	3.0176	0.937614	0.002237	0.9967	0.8109	2.819952	2.286699	8609.9
PDP1-10	10.0056	0.937614	0.002237	0.9967	0.7069	9.350248	6.609690	28549.2
RANT25-1	26.9210	0.929661	0.002327	0.9965	0.5296	24.940254	13.208359	78995.0
Verification Sources								
NTP-0071	0.0286	0.937614	0.002237	0.9967	0.8666	0.026689	0.023129	81.5
NTP-0064	0.0315	0.937614	0.002237	0.9967	0.8666	0.029427	0.025502	
NTP-0071	0.0286	0.937614	0.002237	0.9967	0.8666	0.026689	0.023129	
NTP-0078	0.0307	0.937614	0.002237	0.9967	0.8666	0.028699	0.024870	
TOTALS	0.0908					0.084815	0.073501	259.0
NTP-0085	0.3067	0.937614	0.002237	0.9967	0.8588	0.286574	0.246110	
NTP-0092	0.3057	0.937614	0.002237	0.9967	0.8588	0.285677	0.245339	
NTP-0099	0.2927	0.937614	0.002237	0.9967	0.8588	0.273529	0.234906	
TOTALS	0.9051					0.845780	0.726356	2582.2
NTP-0106	3.0553	0.937614	0.002237	0.9967	0.8109	2.855182	2.315267	
NTP-0113	2.9221	0.937614	0.002237	0.9967	0.8109	2.730707	2.214330	
NTP-0120	3.0121	0.937614	0.002237	0.9967	0.8109	2.814812	2.282531	
TOTALS	8.9895					8.400701	6.812128	25648.4

4. CTEN RESPONSE

The Differential Dieaway Technique (DDT) takes neutron measurements in two successive windows, termed the thermal and background windows, to obtain a response (Figure 4). For a 100 Hz pulse used in CTEN, the total length of the window is 10 ms and the pulse is "ON" for only 20 μs. Within this window, two regions are used for the assay. In the thermal region, 0.8 to 2.8 ms, the induced neutrons, $C_{thermal}$, have slowed to thermal energies and are counted and summed over all pulses from the assay. After an elapsed time of 5.6 ms, only background neutrons, C_{bkg}, should remain and these neutrons are counted from 5.6 to 9.6 ms following the pulse. Again, these are summed over all pulses from the assay. The response function, R, is then the difference in the thermal and gate-weighted background neutron counts normalized to the sum of the three on-barrel drum flux monitors, $F_{thermal}$, in the thermal region. The gate width, g, is the ratio of the thermal to background gate widths in milliseconds and is used to weigh the background counts. In addition to the response function, R, for a waste drum, an empty drum response function is needed. This is the blank response, R_o, and is obtained with the same relationship shown in equation (6) using an empty drum without radioactive sources.

$$R = \frac{C_{thermal} - gC_{bkg}}{F_{thermal}} \tag{6}$$

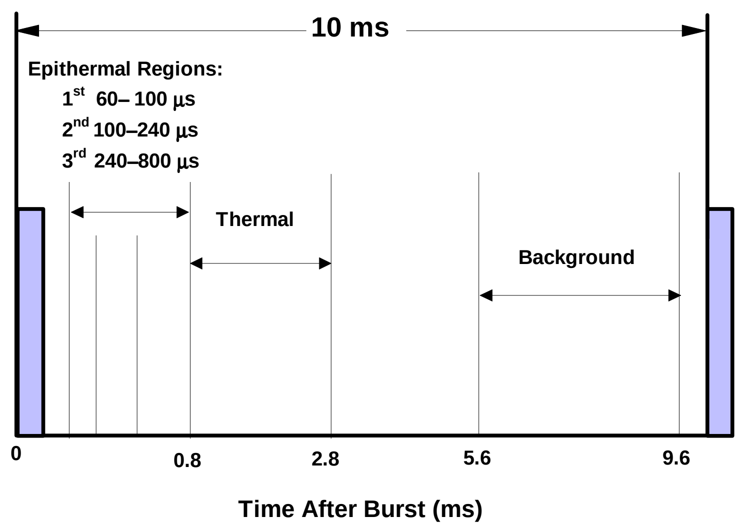

Figure 4. The CTEN DDT Window From A Single Neutron Pulse At 100 Hz

5. RESULTS

Two ways of obtaining the calibration constant, K, were examined. The one used to determine the calibration constant for CTEN takes a plot of the apparent ^{239}Pu effective mass as defined by equation (7). With this option, the self-shielding factor for the calibration sources must be known and these were determined by Monte Carlo techniques[2] and plotted as shown in Figure 5. The matrix correction factor[3], MAT_0, is representative of a low moderator, low absorber matrix—i.e., an empty drum. It was calculated from ten replicate measurements with the PDP1-0.1 source centered in the drum, and found to equal 0.9918 ± 0.0033. With this method, a plot of the response function, $R\text{-}R_o$, versus the apparent ^{239}Pu effective mass is linear.

$$m_{Pu239EA} = K * (R - R_0) * MAT_0 \tag{7}$$

A plot of R – R_0 versus the apparent mass will be a function which, if linear, will be have as solution the form y = ax + b. Since the function goes through zero, then b = 0. Relating this function to equation (7) gives K as a function of the slope, a, and MAT_0:

$$K = \frac{1}{a * MAT_0} \tag{8}$$

An alternative method to determine the calibration constant makes use of Equation (9). This equation relates the effective ^{239}Pu isotopic mass as a function of the calibration constant, the net response function (R-R_o), the matrix correction factor for an empty drum, MAT_o, and the self-shielding factor. A plot of (R-R_o) versus the mass of ^{239}Pu yields a non-linear curve and the derivative of the curve at zero mass defines the slope, a. For this method to work the zero mass value of SSF_0 = 0.8740 is determined by extending the plot of SSF versus response to the zero value (Figure 5). The MAT_0 value may be determined with the same source as the other calibration method.

$$m_{Pu239E} = \frac{K * (R - R_0) * MAT_0}{SSF_0} \tag{9}$$

$$K = \frac{SSF_0}{a * MAT_o} \tag{10}$$

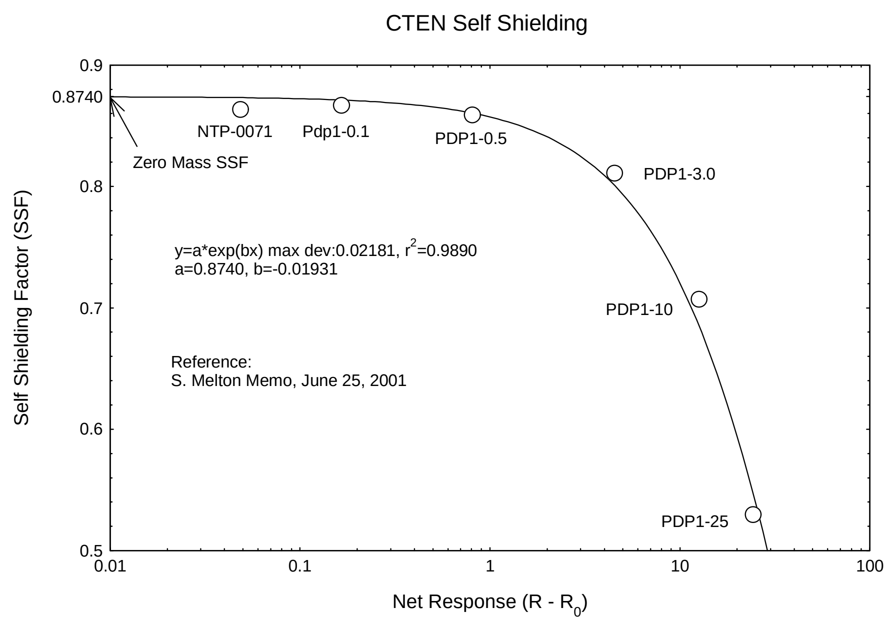

Figure 5. Self-Shielding of Calibration and Verification Sources

The CTEN response for each calibration source is shown in Table 2. The total number of replicates used to determine the uncertainty in the net response function, $R - R_0$, varied with source used. In all cases, the percentage error (%RDS) was less than 2%. For the blank measurement (R_0), twenty replicates were taken resulting in an error of 5.6%.

Table 2. CTEN Response Function Values

Source	# of Replicates	m_{Pu} (g)	m_{Pu239E} (g)	$\sigma_{mPu239E}$ (g)	$m_{Pu239EA}$ (g)	$R - R_o$	σ_{R-Ro}	%RDS
PDP1-0.1	10	0.1003	0.0937	0.0003	0.0812	0.1654	0.0033	2.00%
PDP1-0.5	5	0.5020	0.4691	0.0013	0.4028	0.8087	0.0048	0.59%
PDP1-3.0	10	3.0176	2.8200	0.0076	2.2867	4.5405	0.0274	0.60%
PDP1-10	10	10.0056	9.3502	0.0252	6.6097	12.6114	0.1050	0.83%
PDP1-25	5	26.9210	24.9403	0.0743	13.2084	24.3233	0.1941	0.80%
R_0 (Empty Drum)	20	N/A	N/A	N/A	N/A	0.0419	0.0023	5.59%

A plot of the net response, $R - R_0$, versus mass is shown in Figure 6. The two curves shown are plots of the response function versus the mass of ^{239}Pu effective and apparent. The apparent mass curve is linear, as predicted while the effective mass curve is non-linear. Both curves go through zero. The slope, a, of the apparent ^{239}Pu effective curve is 1.858 and MAT_0 = 0.9918 for the empty drum. Using equation (8) gives a value of K = 0.54. The alternative approach is determined by taking the derivative of the second order polynomial fit in Figure 6 at zero mass giving a slope a = 1.57 and yields a similar value of K = 0.56. However, for purposes of calibration, the calibration constant K = 0.54 is used.

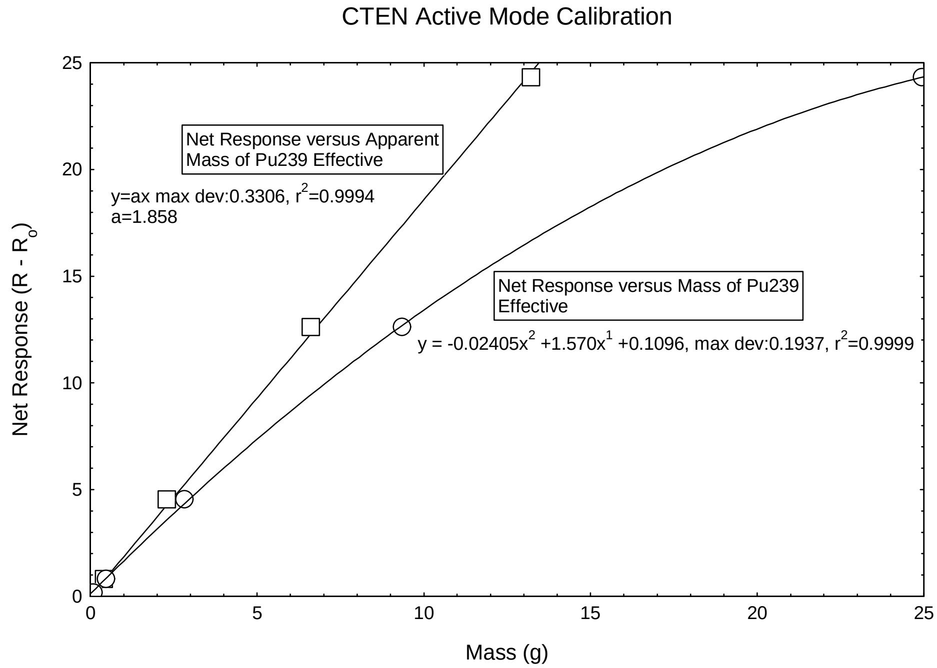

Figure 6. CTEN Response versus Plutonium Mass

6. VERIFICATION

The calibration was verified with NIST-traceable verification standards (Figure 7) in the range 0.03 to 9 g of weapons grade plutonium. These verification results are based on at least fifteen replicate measurements at each standard mass in compliance with the WIPP waste acceptance criteria[4] and are distinct from the standards used for calibration. These results show that the system response is linear through at least 10 g of weapons grade Pu. The error bars were quite small, less than 4% at 0.03 g and decreasing to less than 1% at the higher masses.

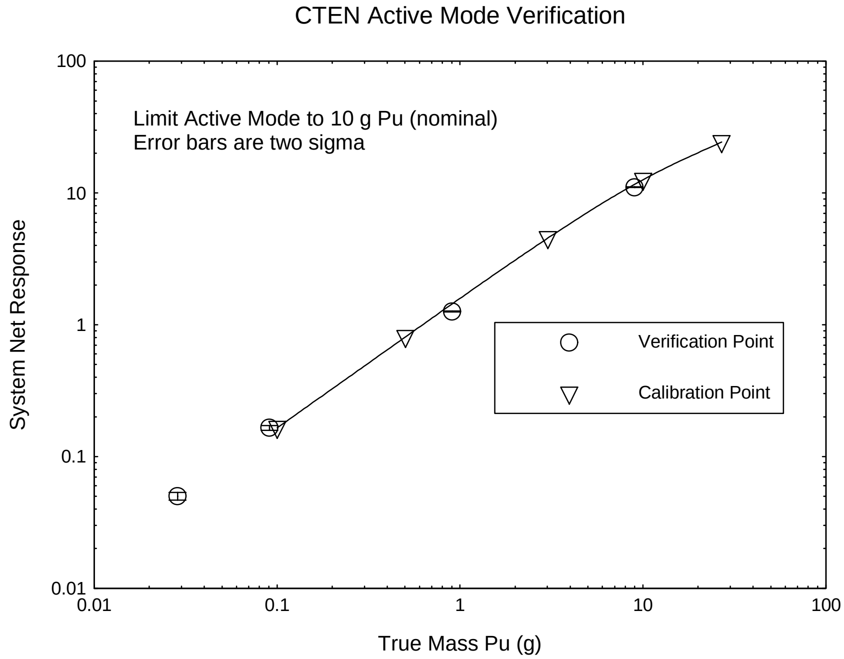

Figure 7. CTEN Active Verification Results

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[1] S. Melton, *Determination Of The Fissile Content Of Radioactive Waste Containers Using The Differential Dieaway Technique*, Los Alamos National Laboratory (CTEN dissertation by S.G. Melton, to be published)
[2] Memorandum, S. Melton, *Calculation of Self Shielding in Calibration Sources*, NIS-6-01:101, June 25, 2001.
[3] Memorandum, S. Melton, *Matrix Correction Factors for CTEN—6-21-01*, NIS-6-00: 106, July 11, 2001.
[4] Waste Acceptance Criteria for the Waste Isolation Pilot Plant, Revision7, Change 2, DOE/WIPP-069, Carlsbad, NM, January 24, 2001.