PASSIVE ACTIVE NEUTRON RADIOASSAY MEASUREMENT UNCERTAINTY FOR COMBUSTIBLE AND GLASS WASTE MATRICES

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ABSTRACT

Using a modified statistical sampling and verification approach, the total uncertainty of the Idaho National Engineering Laboratory's Passive Active Neutron (PAN) radioassay system was evaluated for combustible and glass content codes. The waste structure and content of 100 randomly selected drums in each of the waste categories were computer modeled based on the review of real-time radiography video tapes. Specific quantities of Pu were added to the drum models according to an experimental design. These drum models were then submitted to the Monte Carlo Neutron Photon code processing and subsequent calculations to produce simulated PAN system measurements. The reported plutonium masses from the simulation runs were compared with the corresponding input masses. Analysis of the measurement errors produced uncertainty estimates. This paper presents the results of the uncertainty calculations and compares them to previous reported results obtained for graphite waste.

INTRODUCTION

At last year's NDA/NDE conference, we reported on the methods and results from a total uncertainty analysis of a Passive Active Neutron (PAN) radioassay system at the Idaho National Engineering Laboratory's (INEL) Stored Waste Examination Pilot Plant (SWEPP)[1]. That analysis was specific to the measurement of graphite molds. Since that time, similar uncertainty analyses have been completed for an additional two waste forms: combustibles and glass.

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The combustible waste form includes Item Description Codes (IDC) 330 (dry paper and rags) and 336 (moist paper and rags). Upon review of real time radiography tapes of combustible filled waste drums it became clear that those drums contained numerous types of combustible material in addition to paper and rags, as well as some noncombustible items. Thus this waste form comprises considerable heterogeneity.

The glass waste analyzed consists of IDC 440, 441, and 442 drums. IDC 440 waste consists primarily of broken glass vials and related products. The code 441 and 442 drums both contain raschig rings, the only difference being that the code 442 waste has been leached while the code 441 waste has not. Compared to the combustible waste form, the glass wastes are considerably more homogeneous.

The particular methods used to perform the uncertainty analysis for the combustible and glass wastes paralleled that previously described for graphite waste. Some changes and enhancements to the methods used for graphite were implemented. Details of the procedures used are available elsewhere[2-3]. This paper provides a brief overview of the methods employed and a description of the major changes in the methods from those applied for graphite waste. The primary purpose of this paper is to present the uncertainty findings for the combustible and glass waste forms. The results for these two waste forms are compared with each other as well as with the previous findings for graphite. These comparisons will point out the broad effects of waste matrix on radioassay performance and re-emphasize the importance of including matrix effects in any system performance assessment.

PASSIVE ACTIVE NEUTRON ASSAY SYSTEM

The SWEPP PAN system is a second generation passive active neutron system developed in the early 1980's by Los Alamos National Laboratory (LANL) for the U. S. Department of Energy (DOE) and delivered to the INEL in 1983. This system was designed to assay drums containing transuranic contaminated waste.

The PAN assay system consists of a shielding housing which surrounds the drum on all four sides, top and bottom. Each side of the housing contains moderator, thermal and low-energy neutron shielding and He-3 neutron detectors. There are two types of detector assemblies contained in each side of the assay system; i.e., bare detectors (sensitive to all neutrons) and shielded detectors (sensitive to fast neutrons and insensitive to thermal and low-energy neutrons).

The assay system operates in two modes, passive and active. In the passive mode the detector assemblies (bare and shielded) are detecting neutrons produced by spontaneous fission and (α,n) interactions in the waste matrix. Differentiation between the fission neutrons and the (α,n) neutrons is accomplished by coincidence event counting. In addition to the coincidence counting, single event counting is also accumulated during the passive mode. The single event counting data are used to derive chance coincidence corrections to the coincidence data and also to arrive at a Moderator Index (MI).

In the active mode the shielded detectors are used to detect neutrons produced by stimulated fission resulting from thermal neutron interrogation. The interrogation neutron source for the active mode is a Zetatron 14 MeV neutron generator located at one corner inside the system shield enclosure. The high energy neutrons are moderated to thermal via the graphite moderator in the enclosure walls and varying amounts of moderator in the waste matrix. Also during the active mode two monitors are used to monitor the interrogation neutron flux (cavity monitor) and the effective transmission of interrogation neutrons through the contents of the drum (barrel monitor). The ratio of the cavity monitor to the barrel monitor count during active mode is referred to as the Absorber Index (AI).

The Moderator Index (from the passive mode count) and the Absorber Index (from the active mode count) are used in the analysis algorithm to arrive at correction factors which are intended to correct for moderator and absorber effects on the measured responses (both active and passive responses). The corrected responses are used to determine the measured plutonium mass. Therefore, both the active and passive counts must be completed to obtain the needed correction factors. Three measured mass values are obtained by the system for each measurement; i.e., a mass value determined from the active mode count, a mass value determined from the passive short-gate coincidence count and a mass value determined from passive long-gate coincidence count. However, not all three values are valid over the mass range and waste forms covered in SWEPP waste. A set of selection algorithms is included in the system software to determine which of the three assay values should be used in the waste certification documentation. For most waste, only the two passive mass values are considered for selection as the reported mass. The selection criterion for the reported mass is based on which of the short-gate and long-gate coincidence mass values has the smaller relative error.

MATRIX EFFECTS AND TOTAL UNCERTAINTY

The PAN system itself reports a counting statistics error based on the detector response values and an assumed Poisson distribution for counts. Beyond the counting statistics errors, there are various other contributors to the total uncertainty of a measurement. These include

- Source isotopic/chemical composition variability
- Non-uniform matrix absorption
- Non-uniform matrix moderation
- Non-uniform source distribution
- Variations in source particle size
- Significant voids in the matrix
- Shadow shielding of one region by high neutron absorption in another region
- Waste elemental composition not addressed by the calibration routine
- (α,n) source interference

Properly designed, a total uncertainty analysis will include the effects of these factors in addition to the counting statistics.

Some waste matrix effects on the system response were studied during the original calibration series and an algorithm for determining correction factors was developed by LANL. The correction factors were determined empirically using simulated waste drums in which generic materials (e.g., vermiculite, boric acid, sand, and metal scraps) were used to simulate the waste matrix. The basic assumption in the development of the simulated waste was that the matrix was uniform, the source distribution was uniform and that each waste drum was filled to near the volume capacity of the drum. Over the years, there have been small changes made to the correction factor algorithm, but the basic premises; i.e., uniform matrix and uniform source distributions, have not changed. Because the PAN reported counting statistics error does not reflect the uncertainty associated with the calibration derived matrix correction factors, and because those correction factors themselves were based on simplified assumptions regarding waste types and configurations, a more thorough analysis is required to assess the system's total uncertainty.

UNCERTAINTY ANALYSIS APPROACH

The large number of parameters with potential significant effects on assay quality precludes evaluating the PAN system's uncertainty by actual physical experimentation. Instead a calculation/simulation method was used. This computer intensive approach involves three main components: an input data set consisting of statistically generated waste drum and related measurement parameters, a computer model for the physical processes associated with each PAN assay measurement (neutron transport and temporal response), and a set of computer programs duplicating the PAN system's data processing routines (from which counting data and final mass values are obtained). The neutron transport and temporal response calculations are performed using a benchmarked neutron transport model based on the Monte Carlo Neutron Photon (MCNP) transport code[4].

The idea behind this approach is to simulate a process in which total uncertainty is established by comparing data from PAN measurements on representative drums with known contents. By analyzing the differences between the known and measured Pu quantities for the sampled drums, both the bias and precision of the system's measurements for the waste form of interest can be established. The basic steps in performing the uncertainty analysis are as follows:

1. Perform a content code specific review of probable causes of error in the PAN measurements to identify those factors which need to be included in the uncertainty analysis.

2. Review real-time radiography (RTR) video tapes, a data base of previous PAN assay results, and information provided by the waste shipper to establish either statistical distributions or appropriate values for factorial design settings for the parameters considered to have an effect on the assay results.

3. Develop a composite statistical model incorporating the variances and correlations of the distribution-based parameters identified in (1) and (2).

4. From the statistical model developed in (3), generate sets of simulated values for the distribution-based parameters by random sampling. Combine these values with assigned values for the factorial design variables to produce a complete set of parameter settings for each of 100 simulated waste drums.

5. For each simulated waste drum in the set from (4), apply the MCNP model of the PAN assay system to determine the basic efficiencies and temporal responses of the assay system.

6. For each case in the set from (4), input the results from the MCNP model produced in (5) and other parameters from the statistical model into the simulation routine of the assay system response to determine the basic counting data that would be produced by the PAN assay system under the conditions specified.

7. Apply the PAN assay system analysis algorithm to the set of counting data produced in (6) to determine the measured plutonium mass. Compare that mass to the mass used as input to the simulation process (i.e., the true plutonium mass).

8. Analyze the results from all the specified cases to arrive at the total uncertainty which can be assigned to the measured plutonium mass for the waste form being studied.

In the previous analysis of graphite waste, the statistical model used to produce the simulated drum parameters (steps 3 and 4 above) was established by sampling strictly from theoretical distributions for all relevant parameters. The distributions assigned to each of these parameters as well as their correlations with one another were based on the statistical analysis of data from previous PAN assay results. That is, the theoretical distributions were obtained from data pertaining to actual waste drums and then used to generate a specific computer simulated representative sample of drum characteristics. For the combustible and glass waste forms a more direct method of creating most of the matrix characteristics for the representative computer simulated drums was used. A random sample of RTR tapes was obtained and the sampled drums were modeled directly (instead of assigning theoretical distributions to each matrix characteristic). This assures a realistic representative sample of each characteristic's values and their correlations with one another without the necessity of fitting theoretical distributions and sampling from them. (In effect we are using an empirical model for the statistical distributions and correlations of parameters). For characteristics not obtained from the RTR tape reviews (e.g. background radiation levels, (α, n) singles counts, etc.), theoretical distributions were still used. This change in methodology is more straightforward and more easily accommodates the more heterogeneous configurations found in combustible and glass wastes as compared to graphite.

As a check on the validity of the simulation approach for a particular content code, the simulation process is benchmarked against actual measurements on a calibration drum. The calibration drum is an actual drum built of known contents to approximate the matrix configuration found in a typical drum of the waste form under study. Measurements of the calibration drum using the PAN system are obtained with Pu sources with known properties placed in the drum. The quantities and locations of the Pu sources are varied from run to run. The calibration runs are also computer modeled

using the same basic approach as that for the simulated waste drums. Comparison of the simulated PAN results to the known PAN results for the calibration runs thus provides a check on the validity of the simulation process.

Other major differences between the approach used for combustible and glass wastes compared to that used previously for graphite include the number of drums simulated and the use of factorial design factors. The graphite analysis was based on 50 simulated waste drums; 100 cases were used for combustibles and glass. Adding cases was made possible by increased calculation efficiencies and has the benefit of giving more accurate results. Factorial design parameter settings were not used in the graphite analysis. For combustibles and glass, Pu quantity, Pu in chunks vs. fines, and moisture content (combustibles only) were determined according to a factorial experimental design. Treating moisture as a factorial effect was necessitated by the inability to detect it in the RTR reviews. The same is true for Pu in fines vs. chunks. Pu quantity was treated as a factorial effect primarily to gain better control over the quality of the uncertainty estimates at particular Pu levels.

While the PAN system operates in both active and passive modes, the current uncertainty analysis considers only the passive system response. Work on assessing the active mode uncertainty is ongoing.

## UNCERTAINTY ANALYSIS RESULTS FOR COMBUSTIBLE WASTE

For brevity, the Pu quantity specified for each simulated drum will generally be referred to in this section as the "actual" or "true" quantity, and the simulated PAN system measurement result as simply the "PAN Pu" quantity. The true vs. PAN measured Pu quantities are plotted in Figure 1. The line of perfect agreement specified in the plot indicates where the data would fall if there were no measurement bias and/or precision error. The second line in the plot is a regression line fit to the data using the method described below. That the regression line falls below the line of perfect agreement indicates a negative bias exists in the PAN system results. The degree of the scatter in the points about the regression line is an indicator of the degree of precision in the measurements.

Bias and precision in the results can be quantified by considering the following basic mathematical model for the data in Figure 1

$$y_i = \alpha + \beta(x_i) + \varepsilon_i \tag{1}$$

where y_i is the PAN system Pu measurement for drum i and x_i is the true Pu quantity in the drum. The parameters α and β relate to the bias in the measurement process. If $\alpha \neq 0$ and $\beta = 1$, then there is a constant bias in the measurement system. If $\alpha = 0$ and $\beta \neq 1$, then there is constant relative bias in the system. It is also possible to have both constant and relative bias terms at the same time. The term ε_i in the model is the random error component of the measurement. Precision of the measurement system is evaluated by considering the variance (or equivalently the standard deviation) of the ε_i values.

Figure 1. PAN Pu measurements vs. true Pu quantity for the
simulated combustible waste drums
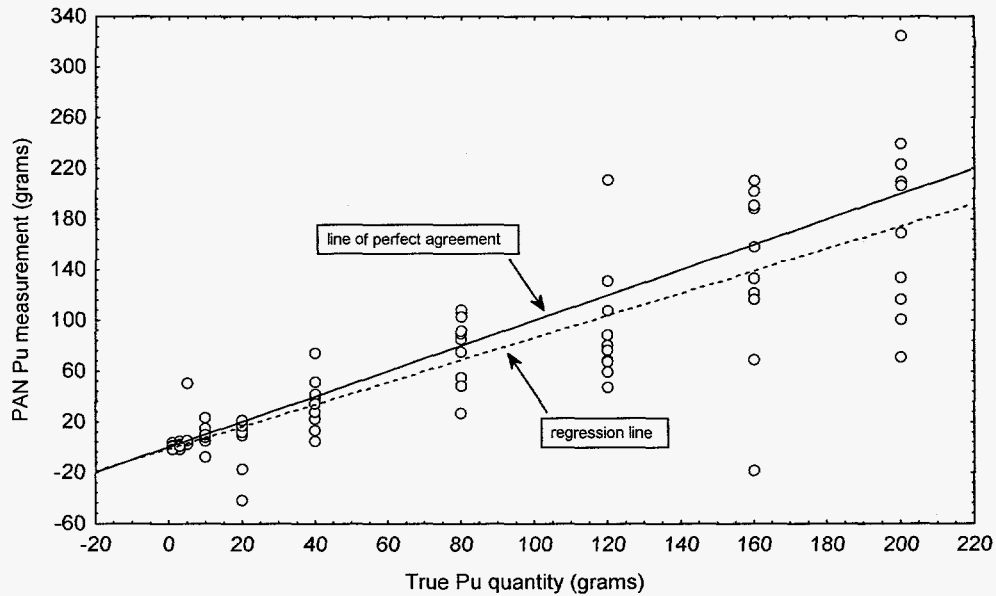
Using this basic measurement model for the PAN system, bias and precision can be evaluated by applying appropriate statistical analysis to the data from the simulations. These calculations are described in detail below.

Bias

Simply defined, bias is the expected difference between the measured and actual Pu quantities. Thus it can be assessed more directly by subtracting the true Pu quantity x_i from both sides of Equation 1 to give:

$$y_i - x_i = \alpha + \beta(x_i) - x_i + \varepsilon_i$$
$$= \alpha + \beta^*(x_i) + \varepsilon_i \qquad\qquad (2)$$

where $\beta^* = \beta - 1$. The meanings of α and ε_i remain the same as before. β^* still relates to relative bias, but now no relative bias results in a value of 0 for β^*. Based on this equation then, the bias in a PAN measurement of x grams Pu is simply $\alpha + \beta^*(x)$, the expected value of Equation 2. Since the parameters α and β^* are not known they must be estimated using regression techniques applied to the simulated waste drum data.

The data for the revised model in Equation 2 are plotted in Figure 2. If there were no bias in the measurements, the data in Figure 2 would be centered around the line of perfect agreement. (Variability about the line represents the random error or precision of the measurements.) That the data points tend to fall below this line indicates once again a negative bias in the PAN measurement system.

Figure 2. Bias in the PAN system measurements for the
simulated combustible waste drums
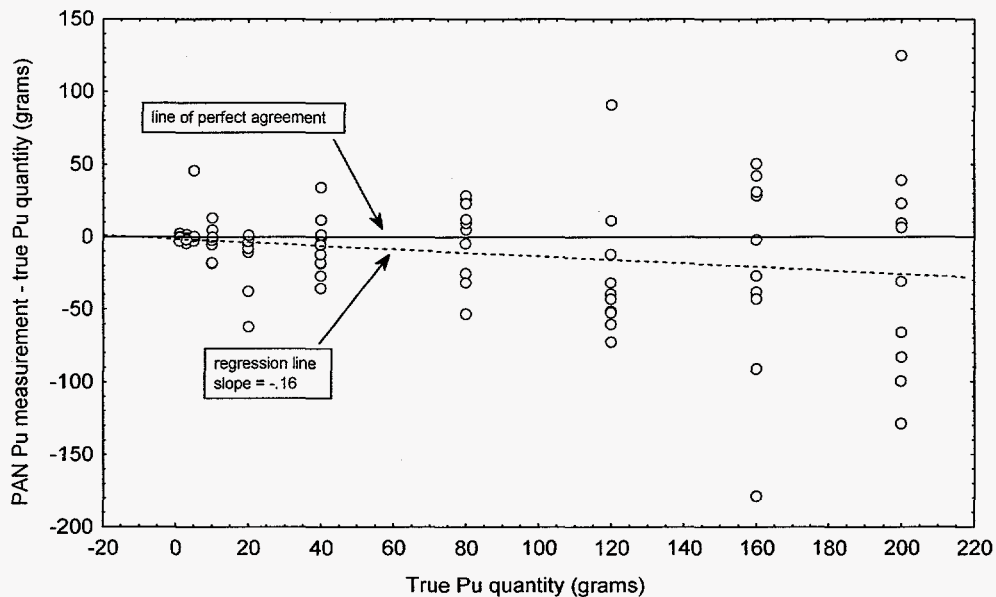
A regression analysis was performed to estimate the parameters α and β^* in Equation 2. Since the variability of the data increases with increasing true Pu quantity, a weighted least squares analysis was performed[5]. Ideally, weights should be $1/\sigma_x^2$ where σ_x^2 is the variance of ε for a given value of x.

Note that these variances are also the desired precision estimates for the PAN system (as defined in Equation 1). Since these values are not known, they must be estimated. The method of estimation is described in the section on precision below.

Weighted Least Squares Results

Using the weights defined by the precision estimates, a weighted least squares analysis of the model in Equation 2 was performed. The first test in the analysis was to consider the presence of a quadratic effect in the bias trend as a means of assessing the validity of the linear model stated in Equation 2. The test for a quadratic effect had a p-value of .45. Hence there is no indication of significant departure from linearity in the bias trend (i.e., Equation 2 is an appropriate model for the data).

Fitting a strictly linear equation to the data produced an estimate of α that was not significantly different from zero (p = .89), hence that parameter was dropped from the model. In the reduced model with α set to zero, the effect of β^* was highly significant (p = .01). Substituting 0 for α and the estimated value of -0.16 for β^* into Equation 2 and taking expectations of both sides gives the following simple model for bias in the PAN system for combustible waste.

$$\text{Bias}_{\text{combustible}} = -0.16(\text{true Pu mass}). \tag{3}$$

In other words, for combustible waste the PAN system produces measured values that are 16% lower than the true Pu quantities. The standard error of the estimated value for β^* is .062, so an approximate 95% confidence interval on the -16% relative error value is (-28%, -4%). The large inaccuracy in the bias estimate is due to the large variability in the data.

Precision

As indicated in Figure 2, variability of the PAN response for the combustible measurements increases with increasing Pu mass. Hence precision was estimated by fitting an appropriate smooth function to standard deviations calculated for each of the 10 sets of 10 replicate Pu mass measurements (1, 3, 5, 10, 20, 40, 80, 120, 160, and 200g). These data are plotted in Figure 3.

Figure 3. Standard deviations of replicate data for the simulated combustible waste drums
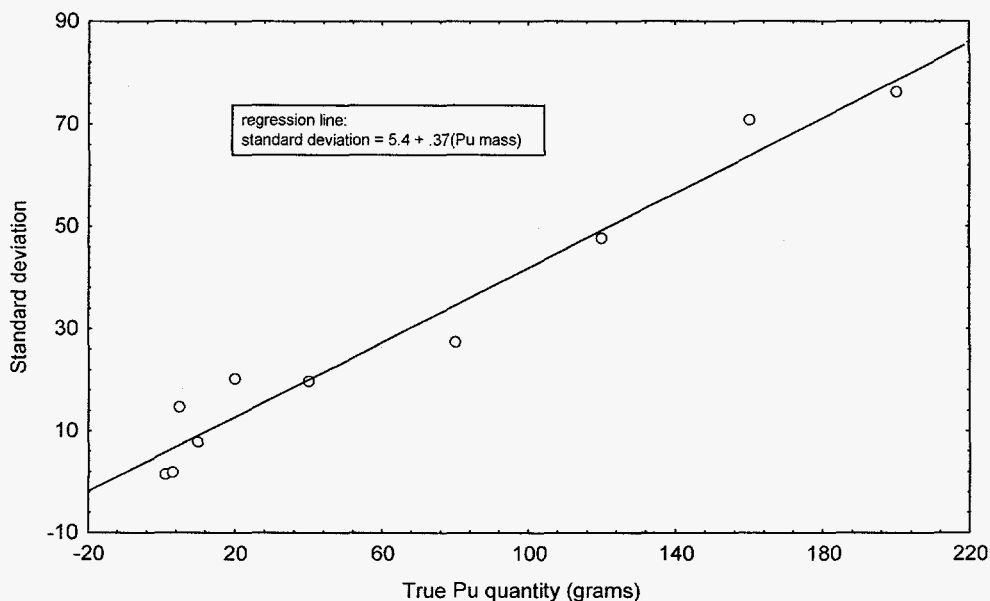
A linear regression line fit to the data produced the following equation for expressing precision of the PAN system for combustible waste drums:

$$\text{standard deviation}_{\text{combustible}} = 5.4 + .37(\text{true Pu mass}). \tag{4}$$

The intercept term in the equation did not quite meet the .05 level of significance ($p = .06$), but was left in because of the known fact that a certain amount of variability will occur even in repeat measurements on an empty drum. (Excluding an intercept term would produce zero variability at zero grams Pu). A test of a quadratic effect was non-significant, indicating the appropriateness of representing the relationship with a linear function. In addition to its use as a representation of precision for combustible drum measurements in the PAN system, Equation 4 was also used to derive the weights used in the weighted least squares regression for bias estimation in the previous section.

Comparison to Counting Statistics Error

It is of interest to compare the precision estimates obtained from Equation 4 to the standard deviations reported automatically by the PAN system. The PAN system estimates of standard deviations include variability due to counting statistics only. Since standard deviations obtained from Equation 4 also include matrix effects etc., they should in general be larger than the PAN reported values. The PAN

reported standard deviations are plotted in Figure 4. A weighted least squares fit to the PAN data is also plotted and compared to the formula in Equation 4. The plot shows the values from Equation 4 exceed the PAN fitted value line over the entire range plotted. The distance between the line increases gradually with Pu quantity. The amount of this increase in standard deviation due to matrix effects varies from approximately 5g at low Pu quantities to 10g at 200g Pu.

Figure 4. Comparison of Equation 4 results and PAN reported standard deviations for the simulated combustible waste drums
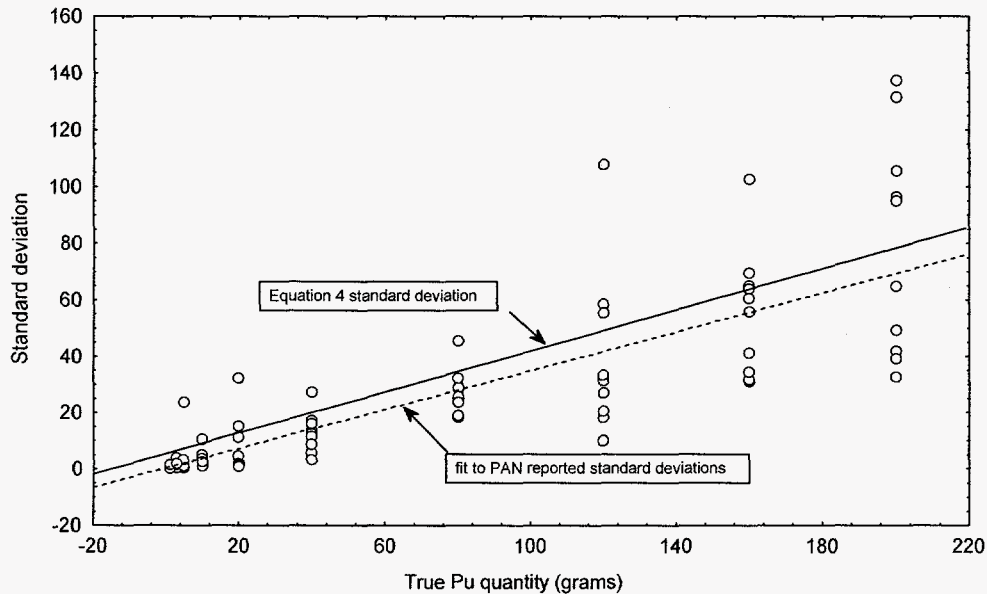
It should be noted that one major matrix-related parameter contributing to the precision error is the (α,n) singles rate. The variability in this rate also affects the counting statistics error reported by the PAN system. So in that regard the reported counting statistics error actually already contains some additional variability (i.e. that due to (α,n) interference).

Matrix Parameter Effects on Uncertainty

The analysis of data related to Equations 1 and 2 established a relationship between the error in a combustible waste measurement and the Pu quantity being measured. It is also of interest to consider whether any of the matrix parameters have consistent effects on the measurement error $y_i - x_i$. This was accomplished by performing a statistical analysis using key matrix parameters determining the simulated combustible waste drum configurations. These include dichotomous design parameters (presence or

absence of added moisture and whether or not the plutonium occurs in fines or chunks) and continuous modeled parameters (absorber index, fill height, the shielded and system (α, n) singles rates, Pu-240 mass fraction, and the mass fractions of the 25 other elements making up the matrices in the drums).

Note that effects of the various parameters, especially the elemental mass fractions are not independent (because of the requirement that the mass fractions sum to unity, when one percentage goes up, the remaining ones must go down). To specify the effects in detail, a complicated mixture model analysis of variance would be needed. But for the purpose of this analysis (i.e., to get a general idea of which parameters have noticeable effects), examination of the individual effects using t-tests, correlations, and other methods, interpreting them in light of the known dependencies, is sufficient.

Parameter Effects on Bias

Because Equations 2 and 3 specifies a known effect of Pu quantity on the error, the quantities $y_i - x_i$ were adjusted prior to the analysis of parameter effects using the formula:

$$\text{adjusted value} = y_i - x_i + .16(x_i). \tag{5}$$

In regression terms the adjusted values are the residual values remaining from the regression analysis of the model in Equation 2. The residuals are essentially measurement errors adjusted for the bias due to Pu quantity. Hence the results below refer to the effects of other matrix parameters after the previously identified effect of Pu quantity has been taken into account. The result is that any significant effects identified will indicate bias effects of the matrix parameters that are in addition to the bias due to Pu quantity.

Neither of the dichotomous design parameters had significant effects on the bias of the PAN measurements. For added moisture vs. no added moisture, the t-test for differences in bias had a p-value of .30. The p-value for the t-test of Pu in chunks or fines was .37.

Effects of the continuous scale parameters were tested using Pearson's r correlation coefficient. Parameters with significant ($p < .05$) or nearly significant ($.05 < p < .10$) are listed in Table 1. The p-values given are for the test of the null hypothesis that the correlation is zero. Thus, low p-values (e.g., $p < .05$) indicate statistically significant correlations.

Table 1. Significant or near significant correlations of matrix parameters with adjusted deviation data (indicating effects on bias) for simulated combustible waste drums.

Matrix parameter	Correlation	p-value
Fill height	-.36	.00
System (α,n) singles rate	.18	.08
Shielded (α,n) singles rate	.18	.07
Mass fraction, lead	-.48	.00
Mass fraction, sulfur	.35	.03

As might be expected due to lead's shielding properties, the strongest correlation with bias was a negative correlation with lead mass fraction. While the correlation of bias with lead mass fraction is strong, it is largely due to one drum with a very high lead content (mass fraction = .54 for a drum containing a large quantity of lead tape). The range of lead mass fractions for the remaining 99 drums (0.0 to .20) had little systematic effect on bias. A similar situation exists for the correlation with sulfur, in that it was also due to a single outlier among the 100 cases, without which the correlation is not significant. Fill height's negative correlation with bias is in contrast to the significant positive correlation found previously with graphite waste.

The equality of the system and shielded (α,n) correlations is due to the high correlation of these values with each other. The average effect on the adjusted bias due to (α, n) count ranges from about -4 grams Pu for smaller (α, n) count values to 30 grams for the largest (α, n) count values.

The (α, n) interference is an inherent problem with coincidence systems as used in the PAN passive mode. In order to get sufficient sensitivity with the He-3 detectors mounted inside polyethylene moderators it is necessary to operate the coincidence units with relatively wide gate widths (i.e., 35μs for short-gate coincidence and 250μs for long-gate coincidence). Normal coincidence systems used in nuclear physics applications operate with gate widths which are much smaller (e.g., 5μs or less). Therefore, the probability for chance coincidence counts are much higher with the wider gate widths. Obtaining the real coincidence rate with the PAN gate widths becomes a problem of taking the difference between two large statistically varying numbers and therefore the difference will be highly uncertain. This effect is expected to be more pronounced on the precision of a coincidence measurement as shown in the next section. However, it does affect the bias as well.

Parameter Effects on Precision

To assess the effects of the categorical experimental design parameters on precision in the combustible measurements, standard deviations of the adjusted difference values given in Equation 5 were calculated and compared using F-tests. Since there were no replicate measurements of PAN results for specific values of the continuous scale parameters, their effects were examined by considering correlations with the absolute values of the adjusted differences calculated in Equation 5. The absolute value of a difference can be interpreted essentially as a standard deviation estimate based on a single observation. Thus, parameters with large effects on precision errors should show significant correlations with the expected magnitude (i.e., absolute value) of the differences between measured and observed values. Because the standard deviation of differences is known to vary by Pu quantity as described earlier, the difference values obtained from Equation 5 were normalized prior to analysis by dividing by the standard deviation for the relevant Pu mass as calculated in Equation 4. Thus the correlations represent effects of the parameters on precision in excess of that attributable to Pu mass.

The difference between standard deviations of the adjusted absolute deviations for drums with and without added moisture was small and not statistically significant ($p = .69$). For drums with Pu in fines vs. chunks, the difference in standard deviations was considerable and highly significant ($p = .000$). For drums with Pu fines, the standard deviation was 25 compared to a value of 47 for drums with chunks.

Significant ($p < .05$) correlations of matrix parameters with the adjusted and normalized absolute deviations are given in Table 2. The (α, n) singles rates show the largest correlation with precision error. (As, with the bias correlations, the values for shielded and system counts are equal due to their high correlation with each other.) All the correlation values, except for copper mass fraction, show positive correlations.

Table 2. Significant correlations of matrix parameters with adjusted and normalized absolute deviations (indicating effects on precision) for simulated combustible waste drums.

Matrix parameter	Correlation	p-value
System (α,n) singles rate	.43	.00
Shielded (α,n) singles rate	.43	.00
Mass fraction, copper	-.26	.02
Mass fraction, iron	.40	.01
Mass fraction, manganese	.24	.04

The (α, n) interference effect was expected as explained in the previous section. However, the effect of the metals (copper, iron, and manganese) are somewhat surprising considering they are minor constituents in combustible waste. This points to the fact that their transport properties (i.e., elastic scattering, inelastic scattering and absorption) must be considered in any simulation of system performance.

Propagation of Total Uncertainty for Measured Mass Values

In the previous sections, bias and precision estimates for the combustible drums are reported relative to the true Pu mass (i.e., the mass specified in the simulation). It is also helpful to convert these estimates so that they are relative to the measured Pu mass. Then, given the measured mass, we can calculate a confidence interval (based on the total uncertainty) within which the true mass is expected to fall. These calculations were obtained as follows.

Based on Equation 3, the expected bias can be restated as

$$\text{(measured Pu mass) - (true Pu mass)} = -.16\text{(true Pu mass)}. \tag{6}$$

Solving for the true Pu mass gives

$$\text{true Pu mass} = 1.19\text{(measured Pu mass)}. \tag{7}$$

The applicable precision for a particular measured Pu mass is the standard deviation specified in Equation 4 applied to the bias corrected mass value:

$$\text{standard deviation} = 5.4 + .37(1.19\text{(measured Pu mass)})$$

$$\tag{8}$$

$$= 5.4 + .435\text{(measured Pu mass)}.$$

To get approximate 95% uncertainty bounds on a measured mass value, consider propagation of error for Equation 7, acknowledging that both the measured Pu mass and the bias parameter have associated precision error. Treating the bias as a parameter, Equation 7 can be restated as

true Pu mass = $(1 + b)^{-1}$(measured Pu mass) (9)

where $b = -.16$ with standard error $s_b = .062$.

Let s_{true} be the standard deviation obtained from Equation 8. Taking partial derivatives of Equation 9 with respect to b and the measured Pu mass, and applying standard propagation of error formulas yields the following equation for the standard deviation of the adjusted measured Pu quantity:

$$s_{adjPu} = [((1+ b)^{-1})^2(s_{true})^2 + ((\text{measured Pu mass})(1 + b)^{-2})^2(s_b)^2]^{1/2}$$ (10)

Substituting the previously estimated values for b, s_{true}, and s_b for combustibles gives

$$s_{adjPu(combustible)} = [((1 - .162)^{-1})^2(5.408 + .365(1.193(\text{measured Pu mass}))^2$$
$$+ ((\text{measured Pu mass})(1 - .162)^{-2})^2(.062)^2]^{1/2}$$

(11)

$$= [41.6 + 6.71(\text{measured Pu mass}) + .278(\text{measured Pu mass})^2]^{1/2}.$$

(Note for precision in calculation, more decimal places than previously shown are given for the parameter estimates in Equation 11.) An approximate 95% confidence interval for the true mass is:

$$1.19(\text{measured Pu mass}) \pm 2s_{adjPu(combustible)}.$$ (12)

Using the results of this evaluation we can make comparisons with performance criteria as established in Table 9-1 of the TRU Waste Characterization Quality Assurance Program Plan (QAPP) (DOE, 1995). The parameter of interest in this table is "Total Uncertainty" which is what has been evaluated in this report. The performance criteria are given in terms of total alpha activity, but we can make limited comparisons based on Pu mass. Shown in Figure 5 is the confidence interval for the true Pu mass plotted against the measured Pu mass. Also shown on this figure are the upper and lower limits as contained in the Table 9-1 of the QAPP. This figure shows that the performance criteria (based on Pu mass only) cannot be met for PAN measurements on drums containing combustible waste.

Figure 5. Uncertainty bounds for combustible drum measurements
(bias adjusted and accounting for uncertainty in bias estimate)
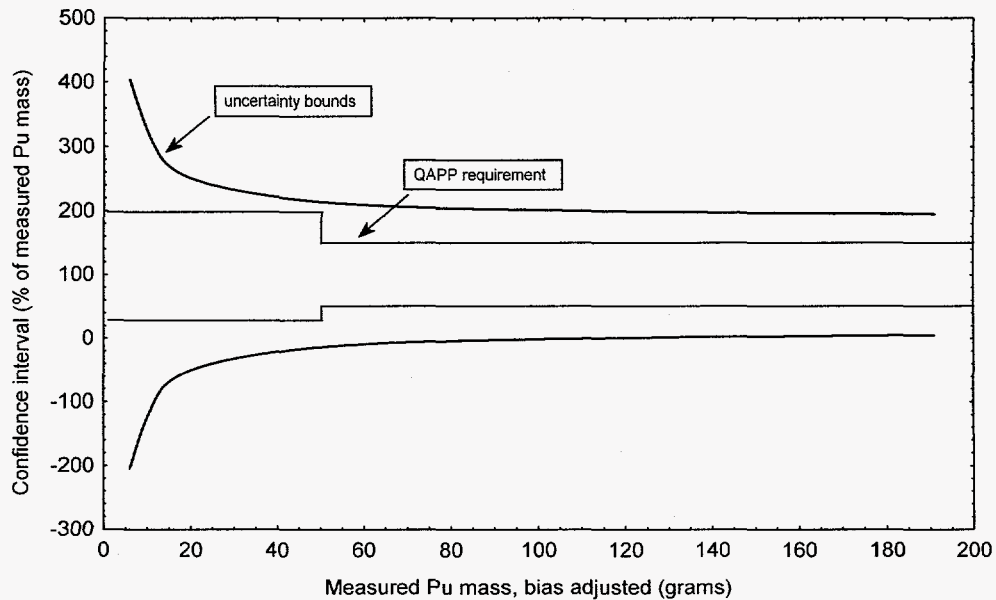
New criteria have recently been proposed for Table 9-1 of the QAPP. These criteria only impose restrictions on the bias component of the total uncertainty. These criteria vary by activity level. The most restrictive of these criteria indicate that the smallest allowable lower limit on the 95% confidence interval for relative bias is -33% and the maximum allowable upper limit is 150%. As stated above, the expected bias for combustibles is -16% with a standard error of 6%. An approximate 95% confidence interval for the true bias is then (-28%, -4%). This interval applies to all mass levels. Hence the new criteria for Table 9-1 can be met.

In order to be critically safe, there is a safety limit requiring that the measured Pu mass plus $2s_\varepsilon$ (i.e., the upper bound in Figure 5) must not exceed 200g. Based on the equations used to derive Figure 5 the measured mass must be below 83g in order to meet this criterion for drums containing combustible waste. This requirement does not present much of a limitation for combustible waste drums as only two of the over 900 drums in the PAN data base used for this study exceeded 83g.

UNCERTAINTY ANALYSIS RESULTS FOR GLASS WASTE

The uncertainty analysis for the simulated glass waste drums followed the same procedures as that for the combustible waste. Thus presentation of the results for the glass drums given below closely

follows that for the combustible drums, except that the development of the modeling and analysis methods is not repeated.

The true vs. PAN measured Pu quantities for the glass waste drums are plotted in Figure 6, and their differences in Figure 7. As observed previously with the combustibles, the regression line for the glass data falls below the line of perfect agreement, indicating a negative bias exists in the PAN system results. The glass data exhibit greater bias but less variability than the combustible data.

Bias

Using the weights defined by the precision estimates given below, a weighted least squares analysis of the model in Equation 2 was performed for the glass data. The first test in the analysis was to consider the presence of a quadratic effect in the bias trend as a means of assessing the validity of the linear model stated in Equation 2. The test for a quadratic effect had a p-value of .90. Hence there is no indication of significant departure from linearity in the bias trend (i.e., Equation 2 is an appropriate model for the glass data).

Fitting a strictly linear equation to the glass data produced an estimate of α that was not significantly different from zero (p = .43), hence that parameter was dropped from the model. In the reduced model with α set to zero, the effect of β^* was highly significant (p < .001). Substituting 0 for α and the estimated value of -0.24 for β^* into Equation 2 and taking expectations of both sides gives the following simple model for bias in the PAN system for glass drums

$$\text{bias}_{\text{glass}} = -0.24(\text{true Pu mass}). \tag{13}$$

In other words, for glass waste the PAN system produces measured values that are 24% lower than the true Pu quantities. The standard error of the estimated value for β^* is .028, so an approximate 95% confidence interval on the -24% relative error value is (-30%,-18%).

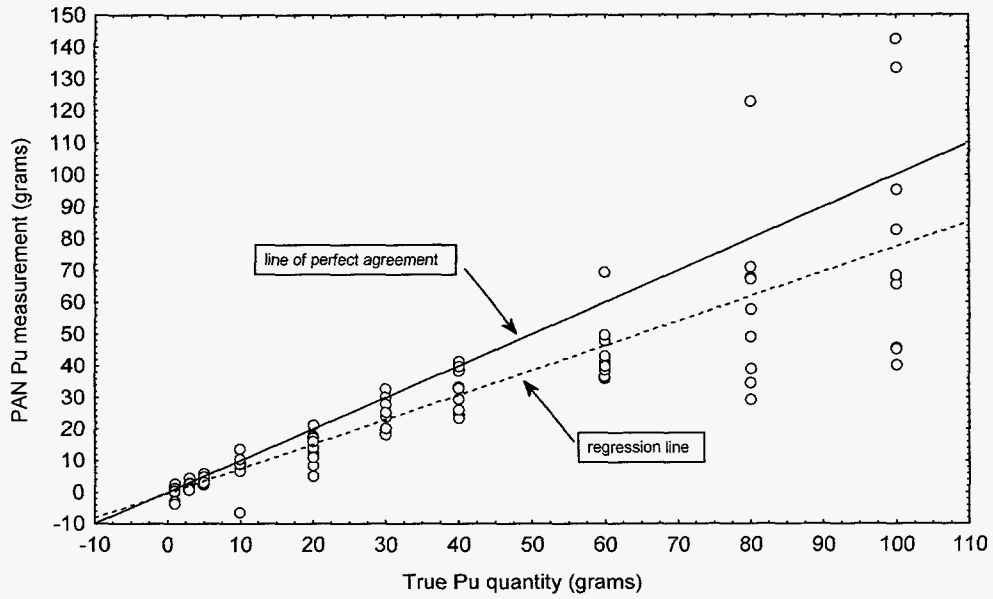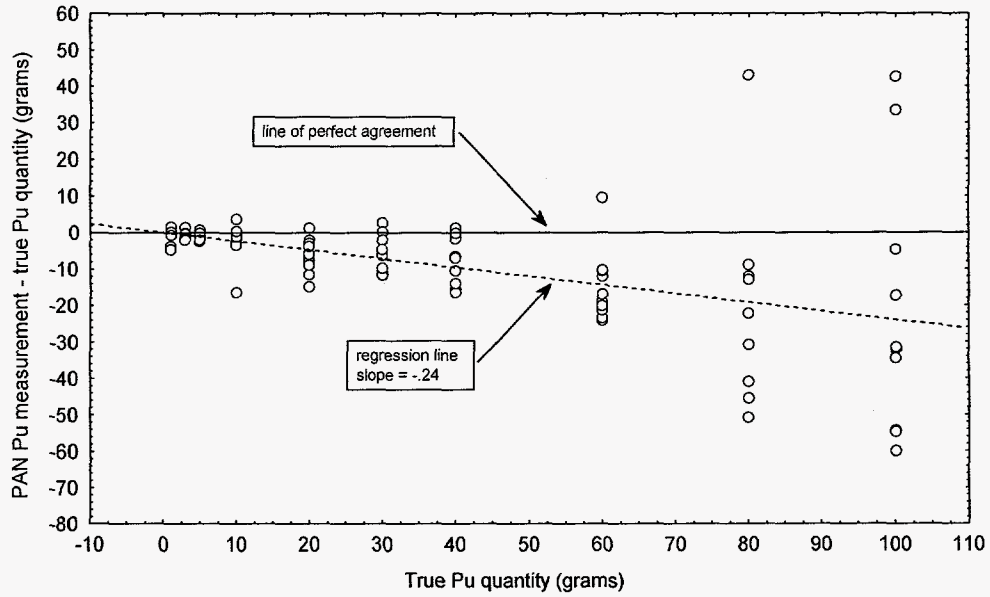Figure 6. PAN Pu measurements vs. true Pu quantity for the
simulated glass waste drums

Figure 7. Bias in the PAN system measurements for the
simulated glass waste drums

Precision

As indicated in Figure 7, variability of the PAN response increases with increasing Pu mass.
Hence precision was estimated by fitting an appropriate smooth function to standard deviations

calculated for each of the 10 sets of 10 replicate Pu mass measurements (1, 3, 5, 10, 20, 30, 40, 60, 80, and 100g). (These gram quantities were different than that used for the combustible waste to better reflect the cover the range of Pu quantities found in glass waste.) These data are plotted in Figure 8.

Figure 8. Standard deviations of replicate data for the simulated glass waste drums
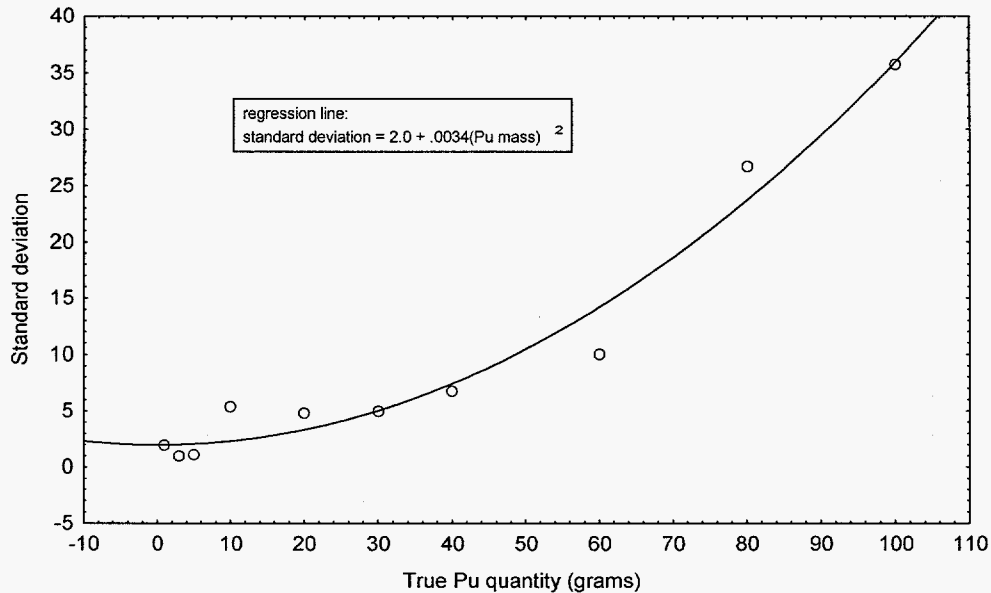
A regression analysis of these produced the following quadratic equation for estimating precision for the glass waste:

$$\text{standard deviation}_{\text{glass}} = 2.0 + .0034(\text{true Pu mass})^2. \tag{14}$$

The linear term in the quadratic regression was clearly not significant ($p = .65$) so was eliminated from the model. The intercept term in the equation did not quite meet the .05 level of significance ($p = .054$), but was left in because of the known fact that a certain amount of variability will occur even in repeat measurements on an empty drum. (Excluding an intercept term would produce zero variability at zero grams Pu.) In addition to its use as a representation of precision for glass waste drum measurements in the PAN system, Equation 14 was also used to derive the weights used in the weighted least squares regression for bias estimation in the previous section.

Comparison to Counting Statistics Error

Figure 9 compares the precision estimates for glass waste obtained from Equation 14 to the standard deviations reported automatically by the PAN system. A weighted least squares fit to the PAN reported data is also plotted to aid in the comparison. The analysis showed that a linear fit to the PAN reported data was appropriate. (The p-value for a quadratic term was .99). The plot shows the values from Equation 14 are actually less than the PAN fitted value line over the intermediate range of Pu quantities. This anomaly is to a large extent due to the amount of variability observed in the PAN data and the fact that the PAN reported values are positively skewed. Other methods of fitting a line to the PAN data produced different results. For example, a fit to the median PAN reported standard deviation values at each Pu quantity produced a line which always falls below the plot for Equation 14, as indicated in Figure 10. Additional work is planned to determine the best method of quantitatively comparing the two sets of results.

Figure 9. Comparison of Equation 14 results and regression fit to individual PAN reported standard deviations for the simulated glass waste drums
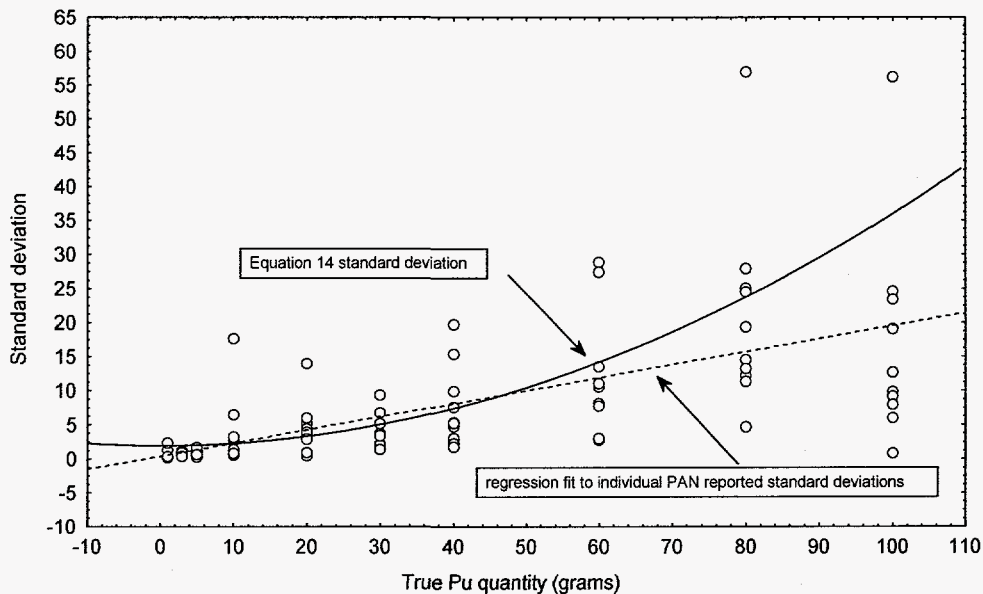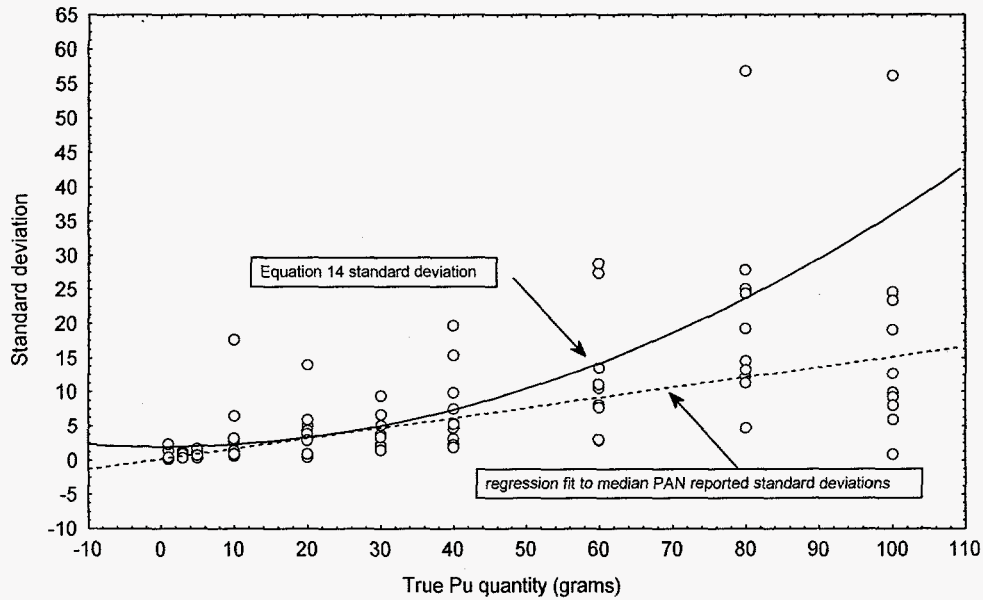
Figure 10. Comparison of Equation 14 results and regression fit to median PAN reported standard deviations for the simulated glass waste drums

Matrix Parameter Effects on Uncertainty

Key matrix parameters for the glass matrix simulated waste drums include categorical parameters (number of fiber packs, content code, and whether or not the plutonium occurs in fines or chunks) and continuous modeled parameters (absorber index, average fill height, mass of the drum contents, the shielded and system (α, n) singles rates, Pu-240 mass fraction, and the mass fractions of the other elements making up the matrices in the drums).

Parameter Effects on Bias

As with the combustible data, the analysis for bias effects was performed on adjusted data to indicate bias effects of the matrix parameters that are in addition to the bias due to Pu quantity. The adjusted values were obtained from the equation

$$\text{adjusted value} = y_i - x_i + .24(x_i) \tag{15}$$

which is the same as Equation 5 except that the bias estimate for the glass rather than combustible data is used.

Effects of the continuous scale parameters were tested using Pearson's r correlation coefficient. Parameters with significant ($p < .05$) or nearly significant ($.05 < p < .10$) are listed in Table 3. The p-values given are for the test of the null hypothesis that the correlation is zero. Thus, low p-values (e.g., $p < .05$) indicate statistically significant correlations.

Table 3. Significant or near significant correlations of matrix parameters with adjusted deviation data (indicating effects on bias) for simulated glass waste drums.

Matrix parameter	Correlation	p-value
Absorber index	.36	.00
Mass fraction of carbon	-.21	.00
System (α,n) singles rate	.17	.09

All three of the correlations in Table 3 are due to two or three outlier values, without which the correlations are not statistically significant. (The correlation with system (α,n) singles rate actually becomes negative without the two highest valued cases.) Thus, substantive conclusions can not be drawn from these data.

The categorical parameters were tested using analysis of variance (i.e., ANOVA) techniques. Both standard ANOVA techniques (which tests for differences between means) and nonparametric ANOVA techniques (which tests for differences between medians) were utilized. In terms of significance of effects the results were consistent between the two methods. Significant effects were found for the effects of Pu in chunks vs. fines, and content code. The mean adjusted difference for cases with Pu in fines was -4.5g while for cases with Pu in chunks it was 5.1g. The difference in medians was -1.4g for fines vs. 1.0g for chunks.

For content codes, the mean adjusted difference values were -1.6 for code 440, 23.1 for code 441, and -1.0 for code 442. The corresponding median values were 0.7, 5.8, and -0.5 respectively. Thus the biggest difference is between the code 441 drums and the others. There are only six code 441 drums in the simulated data. The significant difference appears due to only two of the six drums. These two drums were also two of the outlier cases identified earlier in the correlation analysis. Examination of the characteristics of these two drums did not reveal any single factor differentiating them from other drums.

Parameter Effects on Precision

The effects of the various parameters on precision were examined by considering correlations with the absolute values of the adjusted differences calculated in Equation 15. As with the combustible data, the absolute deviation values were normalized prior to analysis. For the glass data this was achieved by dividing by the standard deviation for the relevant Pu mass as calculated in Equation 14. Thus the correlations represent effects of the parameters on precision in excess of that attributable to Pu mass. To reduce the skewness of the data, the analysis was performed on the logarithms of the normalized deviation data. Correlations with elemental mass fractions were only considered when there were at least ten drums containing a particular element.

None of the categorical variables showed significant effects. Significant ($p < .05$) correlations of matrix parameters with the adjusted and normalized absolute deviations are given in Table 4.

Table 4. Significant correlations of matrix parameters with the logarithms of the adjusted and normalized absolute deviations (indicating effects on precision) for simulated glass waste drums.

Matrix parameter	Correlation	p-value
System (α,n) singles rate	.22	.00
Shielded (α,n) singles rate	.22	.00

Only the (α, n) singles rates showed significant correlations with precision error. The values for shielded and system counts are equal due to their high correlation with each other. The (α, n) interference effect has been seen in uncertainty analysis of other waste types (i.e., graphite and combustibles).

Propagation of Total Uncertainty for Measured Mass Values

Using the same propagation of errors procedure described above for combustibles, the standard deviation for an adjusted measured mass value for glass waste drums is

$$S_{adjPu(glass)} = [6.7 + .042(\text{measured Pu mass})^2 + .000060(\text{measured Pu mass})^4]^{1/2}, \qquad (16)$$

and an approximate 95% confidence interval for the true mass is:

$$1.32(\text{measured Pu mass}) \pm 2s_{adjPu(glass)}. \tag{17}$$

Figure 11 compares the confidence intervals calculated using Equation 17 to performance criteria as established in Table 9-1 of the TRU Waste Characterization Quality Assurance Program Plan (DOE, 1995). This figure shows that the performance criteria (based on Pu mass only) can be met for measured PAN measurements of glass waste in the range from approximately 8g to 50g.

Figure 11. Uncertainty bounds for glass drum measurements (bias adjusted and accounting for uncertainty in bias estimate)
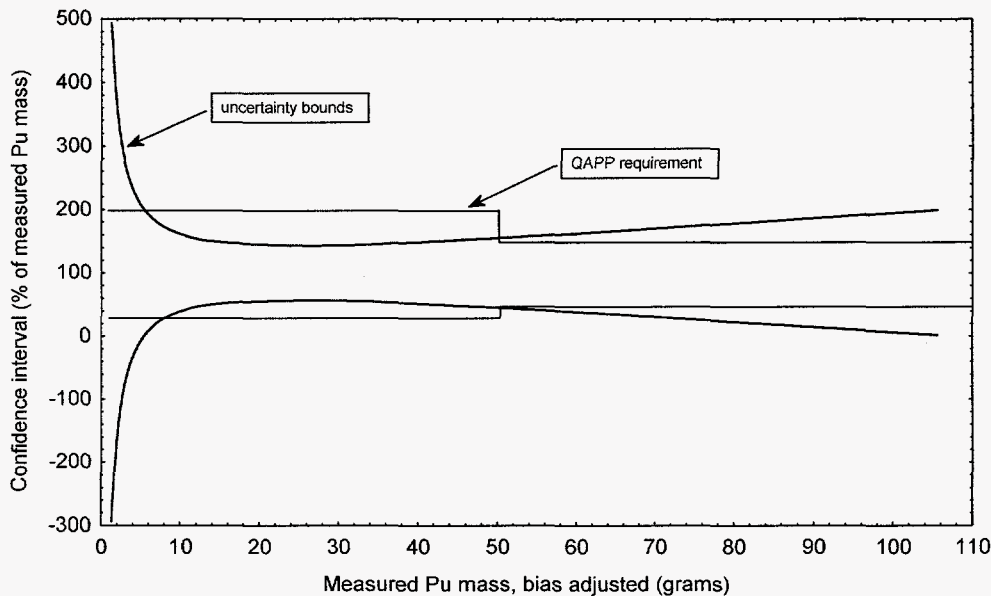
As mentioned before, new criteria have recently been proposed for Table 9-1 of the QAPP. These criteria only impose restrictions on the bias component of the total uncertainty. The expected bias for glass is -24% with a standard error of 2.8%. An approximate 95% confidence interval for the true bias is then (-30%, -18%). This interval applies to all mass levels. Hence, based on uncertainty for the PAN measured Pu mass, the new criteria for Table 9-1 can be met for glass waste, at least when only the uncertainty in the Pu mass is considered. (The final uncertainty must also include the uncertainty in the isotopic ratios used to convert the PAN computed Pu mass to curie content. The additional uncertainty may mean that the bounds are exceeded for some measurements, unless a bias correction is applied.)

In order to be critically safe, there is a safety limit requiring that the measured Pu mass plus $2s_\varepsilon$ (i.e., the upper bound in Figure 11) must not exceed 200g. Based on the equations used to derive Figure

11 the measured mass must be below 77g in order to meet this criterion for drums containing glass waste. (Again the uncertainty will be somewhat greater when isotopic ratios are considered.)

SUMMARY AND DISCUSSION

The glass data appear to indicate greater bias but less precision error than did the combustible data. The bias for combustible drums was estimated at -16% with an approximate 95% confidence interval covering from -28% to -4%. For glass waste drums the bias was -24% with a 95% confidence interval from -30% to -18%. The degree of overlap in the confidence intervals indicates that the difference in the bias for the two waste types is not statistically significant, but the broad confidence bands indicate that it is not necessarily safe to assume the bias values are in fact the same. The estimated difference between the glass and combustible drums is -8% with 95% confidence interval bounds of -21% and +5%, so there is the potential for a sizeable negative or moderate positive difference in the bias values.

Figure 12 compares the glass and combustible data to each other as well as to the graphite data previously analyzed. (Data for the glass content codes have been shifted slightly to the right in the plot to avoid obscuring the combustible data.) The graph shows that the least bias (-6%) occurred in the graphite measurements. The lower 95% confidence bound on the graphite bias was -8.6% (Harker, et al., 1995), which overlaps the interval reported above for combustibles, but not for glass (suggesting a statistically significant difference for the comparison with glass, but not for combustibles). More striking differences are evident in the amount of variability among the measurements for the three different waste codes. Graphite showed by far the least variability (i.e., the best precision). The glass waste showed considerably better precision than the combustible at low Pu quantities (60g or less) but approximately the same precision above that level. The more rapid increase in the variability for the glass data is reflected in the fact that the regression fit to the increasing trend in the glass standard deviation data was a quadratic equation while that for combustibles was only linear.

The combined results in Figure 12 clearly show that uncertainty patterns can vary considerably among waste types. Thus, for accurate assessment of a system's measurement uncertainty, it is essential to perform separate analyses for different categories of waste.

Figure 12. Combined waste form uncertainty analysis results
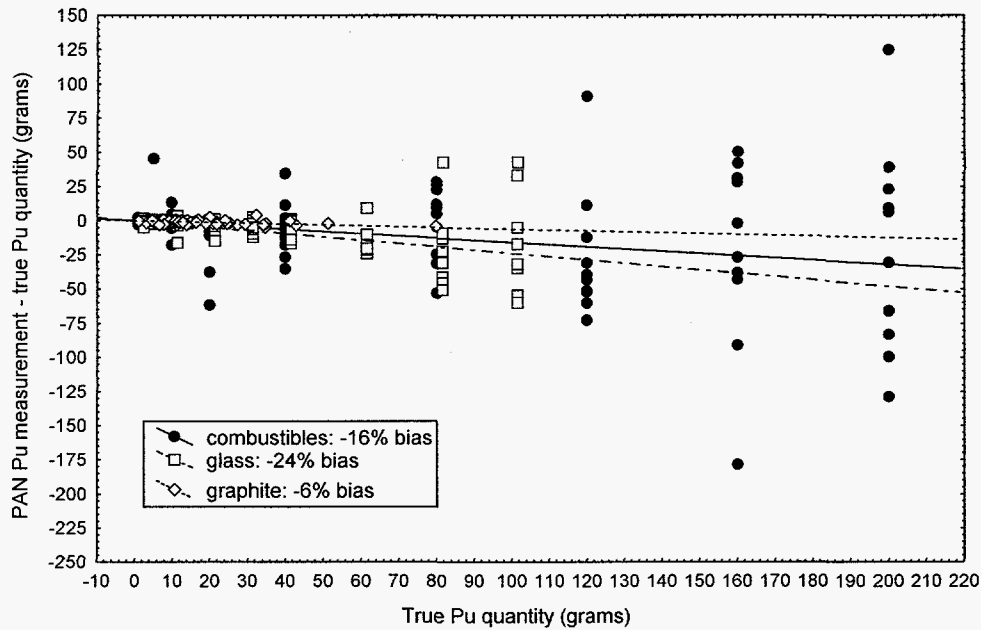
Figure 12. Combined waste form uncertainty analysis results

ACKNOWLEDGMENT

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