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FEB 20 2001

ENGINEERING DATA TRANSMITTAL

Page 1 of 1
1. EDT 605745

2 To (Receiving Organization) Engineering Files	3 From (Originating Organization) PFP Analytical Laboratory	4 Related EDT NO N/A
5 Proj./Prog./Dept./Div.: PFP/Residues	6. Design Authority/Design Agent/Cog. Engr.: DM Fazzari	7 Purchase Order No N/A
8. Originator Remarks: This EDT is to release supporting documentation to Engineering Files for retention and retrieval.		9 Equip /Component No.: N/A
11. Receiver Remarks:		10 System/Bldg /Facility: 234-5Z
11A. Design Baseline Document? <input type="checkbox"/> Yes <input checked="" type="checkbox"/> No		12. Major Assm. Dwg. No.: N/A
		13. Permit/Permit Application No.: N/A
		14. Required Response Date: N/A

15. DATA TRANSMITTED					(F)	(G)	(H)	(I)
(A) Item No.	(B) Document/Drawing No.	(C) Sheet No.	(D) Rev. No.	(E) Title or Description of Data Transmitted	Approval Designator	Reason for Transmittal	Originator Disposition	Receiver Disposition
i	HNF-6944	N/A	0	TMU FOR PFP SGSAS	Q	2	1	

16. KEY

Approval Designator (F)	Reason for Transmittal (G)	Disposition (H) & (I)
E, S, Q, DORNIA (See WHC-CM-3-5, Sec. 12.7)	1. Approval 2. Release 3. Information	4. Review 5. Post-Review 6. Dist. (Receipt Acknow. Required)
		1. Approved 2. Approved w/ comment 3. Disapproved w/ comment
		4. Reviewed no comment 5. Reviewed w/ comment 6. Receipt acknowledged

17. SIGNATURE/DISTRIBUTION
(See Approval Designator for required signatures)

(G) Reason	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN	(G) Reason	(H) Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN
		Design Authority				2	1	SPO: JL Maupin	<i>JL Maupin</i>	2/15/01	T4-06
		Design Agent				2	1	TL Welsh	<i>TL Welsh</i>	2-9-01	T4-40
2	1	Cog. Eng. DM Fazzari	<i>DM Fazzari</i>	2-14-01	T5-06	2	1	GA Westsik	<i>GA Westsik</i>	2-14-01	T5-53
2	1	Cog. Mgr. EW Curfman	<i>EW Curfman</i>	2-9-01	T5-05						
2	1	OR Groth	<i>OR Groth</i>	02-07-01	T4-15	2	1	SPO: P.J CRANE	<i>P.J CRANE</i>	2/15/01	T4-06
		Safety									

18. Signature of EDT Originator <i>DM Snyder</i> DM Snyder Date: 2-9-01	19. Authorized Representative for Receiving Organization <i>EW Curfman</i> EW Curfman Date: 2-9-01	20. Design Authority Cognizant Manager <i>EW Curfman</i> EW Curfman Date: 2-9-01	21. DOE APPROVAL (if required) Ctrl No. _____ <input type="checkbox"/> Approved <input type="checkbox"/> Approved w/ comments <input type="checkbox"/> Disapproved w/ comments
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HNF-6944
Revision 0

Total Measurement Uncertainty For the Plutonium Finishing Plant Segmented Gamma Scanner Assay System

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Project Hanford Management Contractor for the
U.S. Department of Energy under Contract DE-AC06-96RL13200

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Document Type: RPT

Division: PFP

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Date Published
January 2001

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Official / ADC:

William F Russell, Jr.
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Date: 2/15/01

William F Russell, Jr.
Release Approval Date 2/15/01

FEB 20 2001		
DATE:	HANFORD	ID: 24
STA: 5	RELEASE	
Release Stamp		

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Printed in the United States of America

Total Pages: 18

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Total Measurement Uncertainty
For the Plutonium Finishing Plant
Segmented Gamma Scanner Assay System

Table of Contents

1	INTRODUCTION.....	3
2	SYSTEM OPERATION	3
3	TOTAL MEASUREMENT UNCERTAINTY FOR THE SGSAS SYSTEMS.....	4
3.1	Primary Sources of Uncertainty.....	4
3.2	Counting Statistics Uncertainties	5
3.3	Self Absorption Uncertainties	5
3.4	Matrix Uncertainties.....	7
3.5	Non uniform Source Distribution Uncertainties.....	8
3.6	End Effects.....	9
3.7	Summary Uncertainty Estimates for the SGS System	9
3.8	Isotopic Uncertainties.....	11
3.9	Corrections Based on Safeguards Data Evaluations	12
3.9.1	Example Case	14
3.9.2	Application of the TMU correction factor	14
4	REFERENCES.....	15

1 Introduction

This report presents the results of an evaluation of the Total Measurement Uncertainty (TMU) for the Canberra manufactured Segmented Gamma Scanner Assay System (SGSAS) as employed at the Hanford Plutonium Finishing Plant (PFP). In this document, TMU embodies the combined uncertainties due to all of the individual random and systematic sources of measurement uncertainty. It includes uncertainties arising from corrections and factors applied to the analysis of transuranic waste to compensate for inhomogeneities and interferences from the waste matrix and radioactive components. These include uncertainty components for any assumptions contained in the calibration of the system or computation of the data. Uncertainties are propagated at 1 sigma. The final total measurement uncertainty value is reported at the 95% confidence level.

The SGSAS is a gamma assay system that is used to assay plutonium and uranium waste. The SGSAS system can be used in a stand-alone mode to perform the NDA characterization of a container, particularly for low-medium density container matrices. The SGS system provides a full gamma characterization of the container content.

This document is an edited version of the Rocky Flats TMU Report for the Can Scan Segment Gamma Scanners which are in use for the plutonium residues projects at the Rocky Flats plant'. The can scan segmented gamma scanners at Rocky Flats are the same design as the PFP SGSAS system and use the same software (with the exception of the plutonium isotopes software). Therefore all performance characteristics are expected to be similar. Modifications in this document reflect minor differences in the system configuration, container packaging, calibration technique, etc. These results are supported by the QAO counts², safeguards test data, calibration data, etc. for the PFP SGSAS system. Other parts of the TMU analysis utilize various modeling techniques such as Monte Carlo Neutron Photon (MCNP) and In Situ Object Counting Software (ISOCS).

2 System Operation

The SGSAS system is capable of assaying the gamma content of a variety of sizes of cans ranging in diameter up to 12 in. At the PFP facility the actual container size which contains plutonium bearing materials is currently a billet can with a maximum diameter of 5.5 in and a height of 7.0 in. The SGSAS performs the measurement in a number of 0.5 in vertical segments. The number of segments for the billet can is 15. The germanium detector utilizes a lead shield and collimator to limit the field of view of the high purity germanium detector and define the vertical segments. The collimator dimensions used for the Canberra SGSAS systems are 0.5 in vertical opening x 6 in depth. The width of the collimator is wide enough that the detector has an unattenuated view of the container radially. During the assay, the container is rotated on a turntable to minimize the potential source non uniformity in the measurement.

A shielded transmission source is located directly opposite the container from the detector/collimator package and is synchronized vertically with the detector/collimator package. The transmission source is used to measure the absorption of the gamma radiation in the container matrix. By assaying the container in small vertical segments, the SGS system can correct for matrix variations in the container. The assay systems use a Se-75 source that provides transmission lines of 136,265, and 400 keV, where the 400 keV line is the key transmission line for the 414 keV plutonium line and the 265 keV line is used for the 129 keV plutonium line. The 136 keV line is currently not being used since it is fully attenuated by absorbers on the transmission source.

The assay system has an efficiency calibration which covers the energy range from 50 keV to 1500 keV and directly measures several gamma emitting nuclides including Pu-239 and Am-241. The system will also quantify U-233, U-235, Np-237 and U-238. Additional nuclides can be added to the library if they are found in the residues, although it is not expected that there will be other nuclides in the residues waste stream.

The system will also perform a plutonium isotopics measurement using the Multi Group Analysis³ (MGA) isotopics software. The MGA software will provide the relative weight percents for the Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, Np-237, and U-235 isotopes. Under normal operation the measured Pu-239 assay value will be combined with the relative weight percents to calculate the plutonium mass for each of the plutonium isotopes. A separate high resolution, low energy, germanium detector is setup and optimized for the isotopics measurements.

The SGSAS system is calibrated using a number of Eu-152 and Am-241 point sources, distributed in the billet can geometry. The calibration was performed using the standard Canberra Gamma Waste Assay Software⁴ (GWAS) efficiency calibration technique. This provides an efficiency calibration which covers the full energy range from 50 keV – 1500 keV. Additional detail on the SGSAS calibration can be found in the calibration document⁵.

3 Total Measurement Uncertainty For the SGSAS Systems

3.1 Primary Sources of Uncertainty

The primary components of the total measurement uncertainty in the SGSAS gamma-ray assay include the following sources:

- Calibration source uncertainties
- Counting statistics
- Matrix absorption
- Source self absorption uncertainties (lumps)
- Source non uniformity
- End Effects

Although there are other potential sources of measurement uncertainty, they are typically quite small in comparison to the ones listed above.

The results from the validation testing of the SGSAS system for assays over a range from 9 to 142 g shows that the typical overall uncertainty associated with counting statistics and calibration uncertainties is typically less than 5% based on a review of the QAOs run on the SGSAS. This is due primarily to the uncertainty associated with the fabrication of the calibration sources, which typically have a maximum calibration uncertainty of 3%. A smaller portion of the uncertainty is associated with the calibration counting statistics and fit of the calibration data to the calibration curve. This uncertainty is automatically calculated and propagated in the GWAS software so that measurement uncertainties will reflect the calibration uncertainty. Algorithms for propagation of the calibration source uncertainties are contained in the Model S431 Genie-PC Gamma Waste Assay Software Technical Reference Manual⁶ Section 6.4.

3.2 Counting Statistics Uncertainties

Counting statistics uncertainties are very small when significant quantities of material are measured but become significant as the radioactive source strength decreases. Since the system is only being qualified for the 2 highest QAO ranges at the present time, the statistical uncertainty is not expected to be a major source of uncertainty (typically in the range of 3% or less based on the precision results from the QAOs). Because this uncertainty term is propagated in the GWAS software, the TMU analysis will pick up the measured counting statistics uncertainty values so that this term is valid for any measurement range. Data taken during the initial measurement configuration with standards shows a relatively constant standard deviation of about 3% which tracks with the counting statistics uncertainty for the lowest gram levels, but should have dropped to approximately 1.5% or less at the high gram levels based on the counting statistics.

The counting statistics tend to be the primary effect in the precision of the measurements. QAO counts taken with the SGSAS demonstrate a precision of approximately 3% for a 9 g standard of weapons grade Pu, using a standard 50 sec/segment assay time. The precision is almost totally related to counting statistics uncertainties.

Algorithms for propagation of the counting statistics uncertainties are contained in the Genie-PC algorithms manual section B.4.2. (Also see section B.9.1), and the Model S431C Genie-PC Gamma Waste Assay System Technical Reference Manual Section 6.

3.3 Self Absorption Uncertainties

Self absorption uncertainties depend both on the quantity of plutonium in a "lump", lump density, and the waste material type. Certain waste streams such as residues may be likely to produce lumps of plutonium where significant self absorption can occur. Using equations from page 163

of Passive Nondestructive Assay of Nuclear Materials'' a worst case underestimate for an SGS assay of a 1 gram spherical lump of pure plutonium metal using the Pu-239 gamma-ray peak at 413 keV at **50%** assuming no additional corrections are applied. For PuO₂ the underestimate drops to about **25%**. If a flatter shape is assumed as might be expected and the material was plated onto a crucible or other matrix form, it can be calculated that changing the geometry to a less spherical shape would reduce the self absorption underestimation to on the order of **5%** to **10%**. Going through the same exercise for a larger single 10 g spherical lump, the underestimation would be approximately **75%**, again assuming no differential peak correction. Reconsidering this as a PuO₂ rather than a metal changes this to a **50%** underestimation and considering the material in a more plated form would change the underestimation to about **10%** - **25%**. Furthermore the probability of a single 10 g lump is much less probable than a number of smaller lumps summing to 10 g. Finally for containers with high gram loadings (over 100 g) there is a probability of many lumps of plutonium with varying sizes. Since the data review procedure looks for localized concentrations of activity the maximum total gram value for any single position must be considered to be significantly smaller than the total gram value. In summary there is a wide range of potential uncertainties due to the presence of lumps of plutonium in the container, which increases as the total quantity of the plutonium increases in the container.

Although the effect of lumps or self absorption is always a negative bias uncertainty, the SGSAS system utilizes a differential peak correction in the calculation of the results for the Radioassay Data Sheet. Differential peak correction is described in the software requirement specification for the TMU software'. This applies a correction for the Pu result based on the increased absorption of the 129 keV line over the 414 keV line. The differential peak correction will tend to minimize the effects caused by self absorption and in some cases may actually overestimate the result. Therefore for the purposes of the TMU uncertainty, the self absorption effect will be considered a random uncertainty rather than a bias uncertainty.

Since it is not possible to directly quantify the extent of any self absorption in the cans being assayed, the following are assumptions that will be used to determine the self absorption effect in the TMU analysis. Results shown below are reported as percentages at a 1 sigma uncertainty for the assay value.

- For Pu assays < 1 gram: 0
- For gram loadings between 1g < Pu < 10g: **2.5%**
- For gram loadings between **10g** < Pu < 100g: **5%**
- For gram loadings greater than 100g: **7.5%**

For the ash matrix the differential peak correction is not used due to the density of the matrix. However the material is screened and therefore not expected to have significant lumps of plutonium.

3.4 Matrix Uncertainties

Uncertainties due to matrix absorption are small for uniform matrices and source distributions. The assay system corrects for this absorption by calculating the matrix density using the transmission correction technique. This technique measures the absorption of the gamma radiation for the matrix by beaming an external source through the container with a gamma energy close to the energy of the primary assay peak. This directly accounts for both the density and the Z effects of the matrix. Therefore the effects of the elemental composition of the matrix is directly accounted for in the correction technique. Based on the segregation of materials by material type, it is not likely that there will be significant variations in the overall density or Z of the materials in a single can. The most likely matrix effect will be the presence of voids in the material.

To evaluate the measurement uncertainty associated with a heterogeneous matrix distribution Canberra has modeled the response of a measurement segment. Several test cases are described below:

For the following group of tests the matrix was uniform except for a 10 cc sphere (dia. of approximately 1 in). This size was chosen, because it represents about the largest size non uniformity which could be present without significantly modifying the transmission measurement. Therefore it should represent the worst case uncertainty. The non uniformity was positioned off axis so that it would not be in the transmission beam during the full sample rotation. Once the non uniformity is large enough the transmission correction will adequately handle the average matrix effect and the overall uncertainty will actually be smaller.

The source distribution was considered to be uniform to keep source distribution uncertainties separated from matrix distribution uncertainties. The following are results for the various test cases run where the results shown are the ratio of measured result to the correct result, and the sphere is either placed at the center of the container or at the outside edge:

	Center	Outside edge
I) Container was primarily cellulose at 0.5 g/cm ³ .		
1) Inhomog. was a void sphere:	0.98	1.00
II) Container was primarily ash at 1.0 g/cm ³ .		
1) Inhomog. was a void sphere:	0.96	1.01

Since the SGSAS assays the can in small vertical segments, each of which receives a transmission correction, the vertical effects of waste matrix inhomogeneity are insignificant. This minimizes the potential uncertainty associated with stratified matrices of differing densities.

The only significant matrix inhomogeneity effect seems to be a matrix material which is significantly different in density from the average matrix and which is centered radially in the container to the extent that the transmission source is always being effected by the inhomogeneity.

Since each segment receives a separate transmission correction measurement, these examples are considered to be relatively extreme instances and that matrix heterogeneity uncertainty is reduced when averaged over the typical 15 segments. Therefore from the experiment it will be assumed that +/- 4% represents a 2 sigma uncertainty. For the purposes of the TMU calculation, all uncertainties are initially combined using the 1 sigma value or in this case a matrix correction uncertainty of 2 %.

3.5 Non Uniform Source Distribution Uncertainties

The "Billet" can to be used for the residues at PFP is the 5.5 in dia. billet can. This is smaller than the 7 in dia. of the 8808 container used for the TMU analysis at Rocky Flats. The source non uniformity is dependant on the container diameter. Since the containers are similar in size the source non uniformity for the billet can will be very similar to the source non uniformity for the 8808 container. Because the billet can diameter is slightly smaller than the 8808 can the non uniformity equation can be considered somewhat conservative when using the analysis from the Rocky Flats TMU.

In order to establish an estimate of the uncertainty for the source non uniformity uncertainty a series of measurements were modeled using Canberra's ISOCS software. The assumption in the modeling is that a source in the center of the can represents the worst case underestimate measurement and a source at the outside of container represents a worst case overestimate measurement. Based on a probable distribution of sources in a container, these two measurements represent a 3 sigma limit for the source non uniformity. Since the calibration is based on a uniform distribution of activity throughout the container, the worst case underestimate and overestimates are taken as a ratio to the uniform distribution of activity. This was calculated for densities ranging from 0.2 g/cc to 2.5 g/cc. From these measurements, the range of variation in the signal were plotted as a function of matrix density. In addition, the corresponding maximum and minimum signals were determined as a function of density for a gamma energy of 400 keV. In equation form, these maximum and minimum values are given below and are used as a basis for the uncertainty source non uniformity.

$$\begin{aligned} \text{ERR}_{\text{Max}} &= (0.035 * \text{AverageDens}^2) + 0.18 * \text{AverageDens} + 1.01 \\ \text{ERR}_{\text{Min}} &= (-0.03 * \text{AverageDens}^2) - 0.16 * \text{AverageDens} + 0.99 \end{aligned}$$

The estimated uncertainty (1σ) due to non uniform source distribution is then determined as

$$\text{ERR} = (\text{ERR}_{\text{Max}} - \text{ERR}_{\text{Min}}) / 6$$

Although not run for the SGSAS geometry, Canberra has run large numbers of Simulations^{7,8} for random source distributions that support the assumptions above

3.6 End Effects

A special case of the source non uniformity uncertainty is known as end effects. It will be considered a separate uncertainty term as discussed below.

Although the collimator used on the SGSAS system minimizes the volume of the container that is measured in each segment there is some overlap between segments. In general a point source of material can actually be somewhat detected over approximately 3 adjacent vertical segments. Since the assay systems are not set up to assay below the bottom of the outer container, there is an additional uncertainty that may become significant when most of the activity is located at the bottom of the container. In this case the material is only measured in 1 or 2 segments whereas when the material is located at a higher position in the container it is measured in 3 segments. For containers where the plutonium is reasonably distributed throughout the container this uncertainty is quite small. However if all of the plutonium is located at the bottom of the container, this uncertainty can be as large as 20 – 35%⁹.

In order to minimize this effect on the system, a **0.75** in high spacer has been placed on the system so that there is an under scan of the container. In addition administrative procedures define a minimum fill requirement for the containers. Therefore end effects will not be a measurable effect on this system.

End effects at the top of the container are not considered a credible effect since the container is not usually filled completely to the top and the possibility that most of the activity would be at the top of the matrix is very small.

3.7 Summary of Uncertainty Estimates for the SGS System

All of the uncertainties which are used to propagate the TMU uncertainty are either random or have utilized a correction term which makes the uncertainty in the measured value now a random uncertainty. Therefore all of the uncertainties can be summed in quadrature to calculate the final TMU uncertainty value.

Table 2 is shown below as an example of how the TMU would be expected to vary as a function of three different container densities. The values used are based on values calculated based on the above analysis for a waste container containing **5 g** of plutonium. The uncertainties are propagated as the square root of the sum of the squares. Uncertainties associated with source non uniformity dominate the measurement uncertainty. All of the individual uncertainties are shown as 1 sigma limits. The TMU in the bottom row of the table is corrected to 1.96 sigma limits. Results are reported as percentages of the assay value. The **5 g** value was chosen to demonstrate the contribution of a self absorption correction uncertainty while still being small enough to demonstrate counting statistics uncertainties. The different matrices were chosen to actually represent different typical container densities. The densities represent the range that is considered

acceptable for use with an SGS system. The range of matrix densities shown below is between 0.5 and 1.5 g/cc. However based on a review of a number of assay reports the typical ash densities tend to run between 1.3 – 1.5 g/cc. Therefore the 1.5 g/cc value will tend to represent the expected uncertainties for the ash matrix.

The focus of Table 2 is to show that the primary uncertainty term under most conditions is the source positioning uncertainty term. Table 3 demonstrates that at **high** gram loadings the self absorption uncertainty also becomes a significant term.

Table 2

Table 2. SGS Uncertainties for approximately 5g Pu loading @ 1sigma

Matrix Density	(1.5 g/cc)	(1.0 g/cc)	(0.5g/cc)
Counting Statistics (Cstat)	5%	5%	4%
Self Absorption Uncertainties (Lerr)	2.5%	2.5%	2.5%
Matrix uncertainties (Merr)	2%	2%	2%
Source non uniformity uncertainties (Serr)	12%	7%	2.5%
Calibration Uncertainty (CALerr)	3%	3%	3%
Total RMS Uncertainty @ 1 sigma	13.7%	9.7%	5.6%
Total Measurement Uncertainty @ 95% Confidence	27%	19%	11%

Table 3

Table 3. SGS Uncertainties for i 11 150g P loading @ 1sigma

Matrix Density	(1.5 g/cc)	(1.0 g/cc)	(0.5g/cc)
Counting Statistics (Cstat)	3%	3%	2%
Self Absorption Uncertainties (Lerr)	7.5%	7.5%	7.5%
Matrix uncertainties (Merr)	2%	2%	2%
Source non uniformity uncertainties (Serr)	12%	7%	2.5%
Calibration Uncertainty (CALerr)	3%	3%	3%
Total RMS Uncertainty @ 1 sigma	14.5%	11.2%	8.7%
Total Measurement Uncertainty @ 95% confidence	29%	22%	17%

The algorithm used to calculate TMU at the 95 % confidence level was:

$$\text{TMU} = 1.96 * 100 * \text{Sqrt} ((\text{Merr})' + (\text{Comb})^2 + (\text{Serr})' + (\text{Lerr})^2)$$

Where:

Merr is the uncertainty due to matrix absorption.

Comb is the combination of the propagated counting and calibration statistics from the assay software. $= \text{Sqrt}(\text{Cstat}^2 + \text{CALerr}^2)$

Lerr is the uncertainty due to source self absorption effects and is calculated as:
Less than 1 g: 0
1.0 < g < 10: 2.5%
10 < g < 100: 5%
100 < g: 7.5%

Serr is the uncertainty due to nonuniform source distributions. It is calculated according to the discussion in Section 3.5.

For the ash matrix, standards measurements were performed at PFP utilizing containers of actual ash material that had been assayed in a calorimeter to compare with the assay results from the SGS system. A review of this data in section 3.9 demonstrates that the TMU results shown above are considered to be conservative for the ash matrix.

3.8 Isotopic Uncertainties

The only uncertainties that are not included in the above TMU calculations are the potential uncertainties associated with the plutonium isotopic ratios. The PFPTMU software requires the use of either measured or declared Plutonium isotopics for the total calculation of values that must be reported to WIPP.

For the SGSAS systems, the software MGA will be used to directly measure the plutonium isotopics. This software calculates the relative weight percents for each of the plutonium isotopes as well as Am-241 and U-235 and Np-237 if detected. The uncertainty that is calculated for the isotopics encompasses all of the uncertainties associated with the measurement and therefore is a total measurement uncertainty. For normal assays where the MGA result has been evaluated and considered acceptable the plutonium isotopics will be combined with the self absorption corrected Pu-239 assay result to supply the assay values for all of the plutonium isotopes as well as U-235 and Np-237. Under most conditions the measured Am-241 result from the SGSAS assay will be used over the MGA calculated Am-241 result.

If the **MGA** software can not perform a good measurement then a set of declared isotopics are used. This is determined based on the uncertainty for the Pu-238 and Pu-240 results. The maximum acceptable uncertainties for these isotopes are:

Isotope	1 sigma Uncertainty
Pu-238	200%
Pu-240	40%

If either uncertainty is above these values, then a set of default isotopics will be used for calculating the isotopics. The default isotopics are listed in the data analysis procedure.

The americium result is based on the measured result from the **SGSAS** assay. This uses the 662 keV line to calculate the result. If Np-237 or U-235 are measured by **MGA** these ratios will be using the isotopic uncertainties even if the default isotopics are used. The TMU report will indicate whether measured or default isotopics were used.

The isotopics uncertainties are summed in quadrature with the TMU uncertainty to produce the final TMU result for each of the plutonium isotopes.

3.9 Corrections Based on Safeguards Data Evaluations

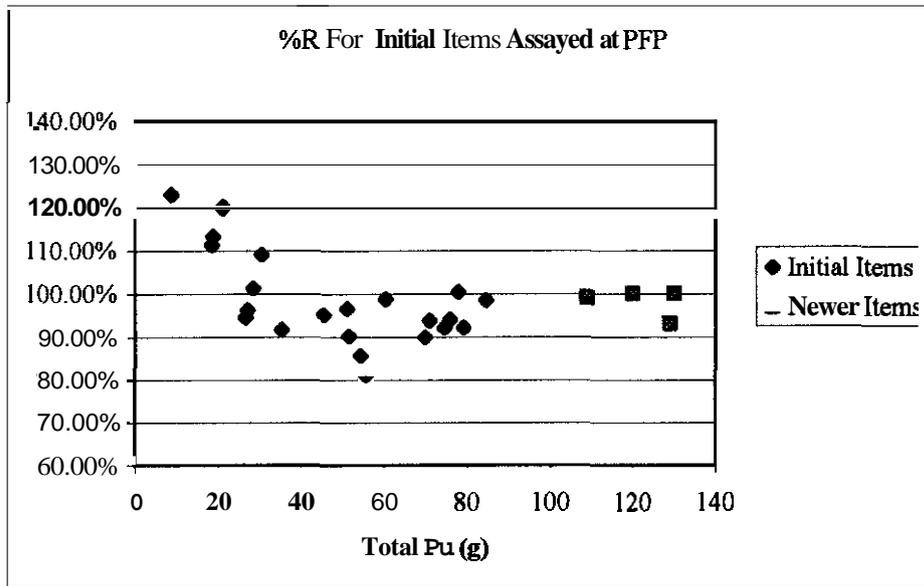
A safeguards review of the validation for the system has identified a bias on the results, based on the measurement of standards. The review compares calorimetry results to the **SGSAS** assay results. To minimize the potential biases in the safeguards inventory, the safeguards program has established a bias correction equation which is applied to the gamma assay results at the present time. The bias problems appear to have been initially due to some system precision and linearity problems, as well as problems with creating the optimum plutonium calibration when using a number of mixed gamma point sources.

4 Evaluation of the TMU for the Rocky Flats Ash Matrix

The above TMU analysis is designed to cover a variety of potential waste matrices. Characteristics of the ash matrix tend to minimize some of the TMU uncertainties described above so that the actual TMU should be somewhat less than the physics based analysis would predict. Some of these characteristics include:

- The ash is sieved to eliminate any significant lumps of material. This will tend to reduce the overall self absorption uncertainty.
- Material is mixed with a surrogate prior to filling the billet cans. This tends to ensure that an already relatively uniform source and matrix distribution will be very uniform.

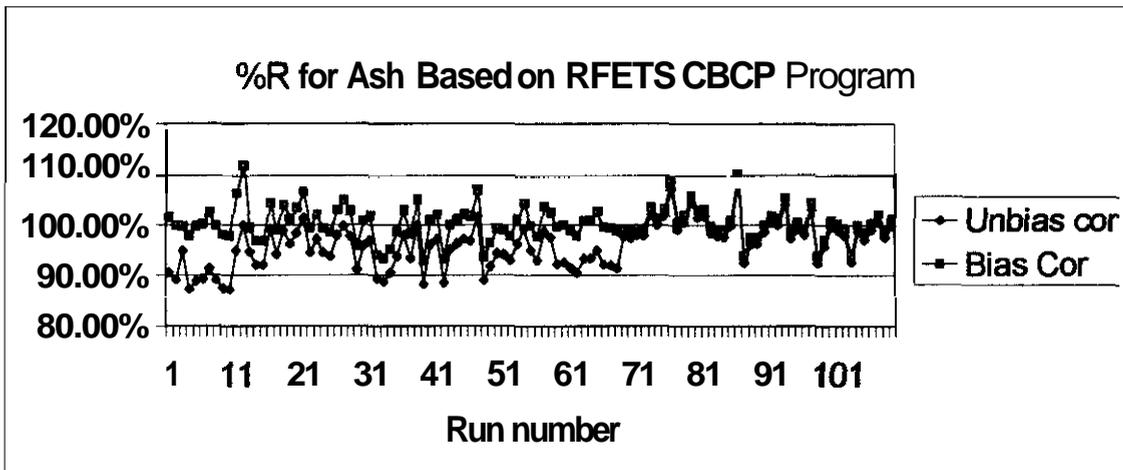
Calorimetry results for a number of the cans which were initially assayed have been compared to the bias corrected assay result. A plot of the % recovery for this data (based on the assumption that the calorimetry data has a small uncertainty) is shown below.



A few assays below 10 g were eliminated from this data since the uncertainty based on the calorimetry in this gram range is typically as large or larger than the gamma assay data. This data shows that the %R based on this data goes from 80% to 120% or approximately 20% TMU rather than the predicted 27 – 29% TMU from the calculations above. The largest uncertainties are in the initial items that had more significant precision and linearity problems. The newer items appear to have even smaller uncertainties.

This is further confirmed by data from the Rocky Flats Continuous Bias Correction Program (CBCP). The CBCP data was provided to Canberra by email as a part of the TMU evaluation at Rocky Flats. This data shows the comparison between Rocky Flats calorimetry and the Rocky Flats SGS systems for a series of randomly selected ash containers which were routinely checked to monitor the Rocky Flats bias correction program.

The Rocky Flats data is from a very similar process, and utilized very similar instrumentation (Canberra can scan systems running the GWAS software) for the gamma assay. Air bath calorimeters were used at Rocky Flats as opposed to water bath calorimeters at PFP. Although not an exhaustive comparison, the Rocky Flats data shows a similar range of recoveries to the PFP data and therefore would tend to support the assumption that the TMU value is conservative. Results from this data shown below:



Since the ash currently being assayed at PFP is the Rocky Flats ash (IDC 420P) this data should also reflect the expected TMU for the PFP SGSAS system.

Based on these two sets of data applying a correction factor of 0.7 to the calculated TMU value probably represents the actual system TMU for the ash matrix. Additional data and a more rigorous evaluation will be used to develop a final correction factor for the ash matrix.

4.1 Example Case

At the present time the one material type which is being assayed at PFP appears to have a much smaller TMU (approximately 10%) based on the experimental data, than the physics based TMU (approximately 20%) would predict.

Since the material type being assayed is an ash, the matrix tends to be relatively uniform and the source material should be reasonably distributed throughout the matrix. The physics based TMU calculations would be expected to be conservative since a term for source non-homogeneity uncertainty is included. Therefore once a complete review of the comparison data is completed a correction factor of some value less than 1.0 will be applied to the TMU uncertainty calculation.

4.2 Application of the TMU correction factor

The TMU correction factor will modify the TMU equation from section 3.7 as follows:

$$TMU = 1.96 * 100 * CF * \text{Sqrt} ((Merr)^2 + (Comb)^2 + (Serr)^2 + (Lerr)^2)$$

Where

CF is the correction factor described in the above sections.

5 References

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