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CAPABILITY OF A TYPICAL
MATERIAL BALANCE ACCOUNTING SYSTEM
FOR A CHEMICAL PROCESSING PLANT

R. A. Schneider and D. P. Granquist

May 1970

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CAPABILITY OF A TYPICAL MATERIAL BALANCE
ACCOUNTING SYSTEM FOR A CHEMICAL PROCESSING PLANT

By

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ABSTRACT

This paper, BNWL-SA-2728 was prepared for and delivered at the IAEA Panel on Safeguards Systems Analysis of Nuclear Fuel Cycles, at Vienna, Austria on August 25 through 29, 1969. It discusses the development of two basic models--a capability model and a performance model--for a chemical processing facility. The capability model represents the best performance (the smallest error) that the measurement system is theoretically capable of achieving in an operating environment. The performance model represents what might more generally be expected in practice from a high quality measurement system.

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CAPABILITY OF A TYPICAL MATERIAL BALANCE ACCOUNTING SYSTEM FOR A CHEMICAL PROCESSING PLANT

R. A. Schneider and D. P. Granquist

INTRODUCTION

Several years ago the authors⁽¹⁾ considered the problem of safeguarding a chemical processing plant using surveillance, containment, and material balance accounting as the main elements of safeguards. At that time the effectiveness of material balance accounting was considered in the context of the overall measurement uncertainty which might be expected when certain proposed or "expected to be used" methods were employed for the measurement of nuclear materials during the chemical processing of power reactor fuels. Since that time, more experience has been gained as to the effectiveness and choice of accountability methods for chemical processing. As a consequence, it appeared appropriate to consider in a more exact manner the effectiveness of material balance accounting in detecting the loss of nuclear materials during chemical processing.

A loss of material during a chemical processing plant campaign can be indicated by the material balance accounting index, Material Unaccounted For (MUF). For an across-the-plant material balance:

$$\text{MUF} = (I_B + \text{Inp}) - (I_E + \text{Pr} + W)$$

where:

I_B = Beginning Inventory

Inp = Input or Receipts to the plant

I_E = Ending Inventory

Pr = Product Shipments

W = Measured Waste Discards.

From the equation, it is apparent that a positive MUF indicates an unmeasured loss of material across the chemical processing operation while a negative MUF indicates the recovery of material presumably lost from previous campaigns. However, in actual practice, a measurement error is associated with each of the terms appearing in the equation. The problem, then, is one of determining in a statistical sense if an observed MUF is significantly different from zero after including these measurement errors in the calculated material balance. Consequently, in this report the effectiveness of material accounting is considered in the context of the imprecision or variance (σ^2) of the index, MUF. For safeguards purposes, the effectiveness of the material balance system can be considered as the probability with which the system will signal an alarm when a given amount of material has been lost (or diverted). If the variance of MUF is small, then the material accounting system can be a sensitive indicator of loss. It is evident that the converse is also true. Thus, if an observed positive MUF differs from zero by an amount significantly larger than would be expected from measurement uncertainties, then the system would signal the need for an investigation. The actual cause of an excessive MUF could result from many causes. These reasons include: chance alone, unmeasured process losses, unsuspected process holdup, and diversion. Hence, diversion is only one of the many causes of apparent material shortages. Only through investigation and the development of specific evidence could diversion actually be proven.

Two approaches have generally been used to estimate the variance of MUF. The first one is by a study of historical data. The other commonly used technique is to synthesize the overall measurement uncertainty of the material balance from the individual components of error. In this study, the method of synthesis is used, since historical data are often confounded with true unmeasured losses.

This report does not consider the problems resulting from accounting errors or from the falsification of records or measurements to mask diversion.

SUMMARY AND CONCLUSIONS

A material balance model was constructed for a commercial chemical processing facility. A Purex-type flow-sheet was assumed for a plant operating on a clean-plant-to-clean-plant basis. It was further assumed that a campaign consists of 40 tonnes of irradiated low enriched uranium containing 10,000 g/tonne of plutonium. A process waste loss of 1% of input and a between-campaign inventory of 0.5% of input were assumed.

Estimates of the random and systematic errors associated with sampling, analysis, volume and weight measurements were made for all individual batch measurements and shipper-receiver measurements. These estimates of error were combined by conventional techniques of error propagation to yield estimates of the variance of MUF. Two basic models were developed--one a capability model and one a performance model. The capability model represents the best performance (the smallest error) that the measurement system is theoretically capable of achieving in an operating environment. The performance model represents what might more generally be expected in practice from a high quality measurement system.

The effectiveness of material accounting in detecting a loss of material during a chemical processing campaign is shown in Table 1. The variance (σ^2) of MUF is shown in terms of the relative standard deviation (σ) as a percent of plant input, e.g., 40 tonnes uranium and 400 kg of plutonium. The performance indices are σ_{MUF} , Pu = 0.31% and 0.62% while σ_{MUF} , U = 0.25% and 0.52% respectively for the capability and performance models. The probability of the system signaling an alarm

TABLE 1. Probability of Detecting a Loss of Material
by Material Balance Accounting at a Chemical
Processing Plant

	% Probability of Observed MUF Exceeding Alarm Level ^(a)	Loss or Diversion, % of Input	
		Capability Model ^(b)	Performance Model ^(c)
<u>Plutonium</u>	50	0.51	1.02
	80	0.77	1.54
	90	0.91	1.81
<u>Uranium</u>	50	0.41	0.86
	80	0.62	1.30
	90	0.73	1.53

- (a) One-sided test for MUF at Type I error of 5% in such a manner that the critical region = $1.645 \sigma_{MUF}$.
- (b) The capability model represents the best performance (the smallest error) that the measurement system is capable of achieving in an operating environment.
The plutonium $\sigma_{MUF} = 0.31\%$ of Input
uranium $\sigma_{MUF} = 0.25\%$ of Input
- (c) The performance model represents what might more generally be expected in practice from a high quality measurement system.
The plutonium $\sigma_{MUF} = 0.62\%$
uranium $\sigma_{MUF} = 0.52\%$

is shown for a Type I error frequency of 5% for various levels of loss or diversion. The capability model would signal the need for an investigation 80% of the time if 0.77% of the plutonium or, if 0.62% of the uranium in a campaign were diverted. Similar values for the performance model are 1.54% of the plutonium or 1.30% of the uranium.

The limiting factor in reducing the variance of MUF is the systematic error associated with the analysis, sampling, and volume measurement of inputs and products. The contributions of inventory and waste measurements are small in the example because of the relatively small quantities assumed for the model. When waste losses become of the order of 2 to 3% of input, waste measurement errors become an important contribution to the variance of MUF. About 90% of the variance of MUF in the examples considered here result from systematic error. Because of the dominant role of systematic error, successive relative MUFs, when induced by measurement error alone and not other causes, tend to be relatively constant around some non-zero value.

Excellent independent checks are potentially available to the chemical plant material accounting system for uranium input and uranium product. Fabricator values for the total uranium content of a low-enriched uranium reactor core, after correcting for reactor burnup, are estimated to have a relative standard deviation of 0.16%. Similarly, product receiver measurements for UNH product are estimated to have a relative standard deviation of 0.10% for the receipt of 40 tonnes of uranium as UNH. Independent checks of similar reliability are not currently available for plutonium input. Predicted values for plutonium from power reactors are estimated to be good to a relative standard deviation of 3 to 5% for a typical core. Product receiver measurements for plutonium nitrate product solutions are potentially as reliable as those made at the

chemical plant. However, when the nitrate is stored for long periods of time before remeasurement at fuel fabrication, these checks are not available to the system.

An additional check on plutonium input is available to the system by use of the plutonium to uranium ratio accountability method for measuring total plutonium input. The ratio method is free of certain analytical, sampling and volume errors. Since it basically uses fabrication values for total uranium receipts to determine total plutonium (by ratio), it provides a partially independent check to the plants system for its direct measurement of total plutonium input.

DISCUSSION

PROCESS AND ACCOUNTABILITY SYSTEM DESCRIPTION

Figures 1 and 2 show a bare outline of a typical Purex plant operating at 1 tonne of uranium per day. Furthermore, it is assumed that a single reactor core of 40 tonnes U forms a single campaign between plant cleanouts. An exposure of about 25,000 MWd/tonne and a plutonium content of 10,000 g/tonne U are also assumed. The ionic concentrations and solution volumes are shown to illustrate the size, number, and frequency of the batch measurements which make up the material balance. An asterisk marks the areas (accountability points) that are needed to form a material balance. The number of batches and typical quantities which constitute the material balance for a campaign are shown in Table 2. The balance shown in Table 2 is typical of a "so-called" across-the-plant balance in which the sum of inputs and beginning inventory is balanced against the sum of ending inventory, product shipments, and waste discards. Positive values for MUF, when judged to be significantly greater than what would be expected from measurement uncertainties, are usually interpreted as an indication of unmeasured processing losses or unmeasured holdup of material in the plant.

* Denotes Primary Accountability Station

FIGURE 1. Outline of Purex Flowsheet - Part 1

* Denotes Primary Accountability Station

FIGURE 2. Outline of Purex Flowsheet - Part 2

TABLE 2. Material Balance Model for a Chemical Processing Plant Campaign

Material Balance Component	Batch Size		Batches Per Campaign	Totals	
	kg U	kg Pu		kg U	kg Pu
<u>Input</u>					
1. Dissolver Solution	1000	10	40	40,000	400
2. Cladding Waste (Hulls and Fines)	0.5	0.0050	40	20	0.20
<u>Product</u>					
UNH	3958		10	39,580	
Pu(NO ₃) ₄		22	18		396
<u>Waste</u>					
1. Cladding Waste	(Same as above)			20	0.20
2. Salt Waste	25	0.25	16	400	4
3. Low-Level Waste	0.04	-	100	4	-
<u>Inventory</u>					
In-Process Inventory	20	0.20	10 ^(a) (Vessels)	200	2

(a) In-Process Inventory is assumed to be distributed among 10 process vessels.

In terms of the variance of MUF, all the components shown in Table 2 except cladding waste measurements contribute to the variance of MUF. When cladding waste is treated in the accounting system as plant input and then subtracted as a plant waste, it contributes neither magnitude nor variance to the across-the-plant balance. In this case, errors in cladding waste measurements go into shipper-receiver difference. The alternative is to use the shipper value for total uranium and correct for burnup in the reactor. In this case, any unmeasured losses in head-end (presumably only cladding losses) would be included in the plant MUF. In practice, both methods are used concurrently. Usually one method is chosen as the official accountability method and the other is used to test for the possibility of unmeasured head-end losses.

There are additional measurements which should be made occasionally for accountability purposes that are not shown. Over a long period of time such components as filters, vent lines, headers, upper regions of scrubbers, stripping columns and towers, can collect small amounts of uranium and plutonium. Periodically, efforts should be spent in measuring this form of holdup. Monitoring of any "cold" exit streams which do not pass through the low level waste tank also should be done periodically to ensure that material is not being lost via leaks in such equipment as steam and water jackets and cooling coils. Process control information of internal flows such as recycle acid and solvent allow a better understanding of the process, although theoretically they are not required to form the overall balance. There are plants however where recycle acid containing appreciable quantities of uranium and plutonium is used for fuel dissolution and thus must be included in the material balance.

TABLE 3. Chemical Processing Plant Accountability Measurement Methods

Material Balance Component	Process Vessel	Measurement Methods		
		Analytical	Volume or Weight	Sampling
<u>Input</u>				
1. Dissolver Solution-Pu and U	Accountability Tank	Isotopic Dilution	Dip Tube-Manometer and Specific Gravity	Circulating Sampler
2. Hulls and Fines Pu and U	Mechanical Cell	U-Fluorimetric and Pu/U	Piece Count Weight/Piece	Mechanical
<u>Product</u>				
Pu(NO ₃) ₄	Pu Product Receiver	Volumetric Titration	Weight-Specific Gravity	Circulating Sampler
UNH UO ₂ (NO ₃) ₂ •6H ₂ O	U Product Receiver	Volumetric Titration Gravimetric	Weight-Specific Gravity Dip-Tube Manometer	Circulating Sampler
<u>Waste</u>				
1. Salt Waste				
Pu	Acid Waste Tank	Alpha Counting	Dip-Tube Manometer or Recorder	Circulating Sampler
U	Acid Waste Tank	U-Fluorimetric	Dip-Tube Manometer or Recorder	Circulating Sampler
2. Low Level Waste - U	Retention Basin	U-Fluorimetric	Fixed Level Per Batch or Flow	Proportional Sampler
<u>Inventory</u>				
Pu and U	10-Process Vessels	Product, Feed, and Waste Methods	Dip-Tube Manometer or Recorder	Circulating Sampler

MEASUREMENT ERRORS

Typical measurement methods used in current and proposed commercial chemical processing plants are listed in Table 3. Detailed discussions of the analytical methods, sampling equipment, and volume measurement equipment are given in References 2, 5, and 8. With the exception of leached hulls (jackets), all measurements are made on solutions using the volume-concentration method. Samples of the process solutions are obtained from the various accountability tanks through built-in sampling lines. The uranium or plutonium concentrations of the sample solutions are measured in the plant laboratory. The total amount of uranium or plutonium in an accountability batch is obtained from the product of solution volume and chemical concentration. Each complete measurement--sampling, analysis, and volume determination--represents a discrete quantity or batch of material. The material balance for a campaign is formed by summing up all the individual batch quantities for such factors as inputs, products, and waste and striking the material balance shown previously.

Estimates of the systematic and random error associated with the individual measurements are shown in Table 4. The estimates are based on the current literature, historical experience, and the personal experience of the authors and their colleagues. Where possible, the estimates shown for measurement capability are based upon the best performance that has been reported or observed. Estimates of what might generally be termed good or high quality performance in a production environment are based, where possible, upon quality control data and historical experience at U.S. sites. Because of the wide range of values reported for a given method and the effects of particular applications on a given method, some judgment and averaging were required. Some sampling errors are listed as less than values because of the difficulty in distinguishing between analytical and sampling error.

TABLE 4. Estimates of Random and Systematic Error for Chemical Processing Plant Accountability Measurements

Material Balance Component			Relative Percent Standard Deviation of a Single Measurement					
			Analytical		Volume		Sampling	
			Random	Systematic	Random	Systematic	Random	Systematic
<u>Input</u>								
Pu	Capability (C)		0.40	0.15	0.10	0.15	<0.10	<0.10
	Performance (P)		1.00	0.25	0.30	0.30	<0.30	<0.20
U	(C)		0.40	0.10	0.10	0.15	<0.10	<0.10
	(P)		1.00	0.20	0.30	0.30	<0.30	<0.20
<u>Product</u>								
Pu	(C)		0.25	0.15	0.05	0.05	<0.20	<0.10
	(P)		0.70	0.30	0.10	0.10	<0.50	<0.20
U	(C)		0.10	0.05	0.10	0.05	<0.10	<0.05
	(P)		0.25	0.10	0.20	0.20	<0.20	<0.10
<u>Waste</u>								
<u>Salt Waste</u>								
Pu	(C)		10	5	1	1	3	3
	(P)		20	10	2	3	6	6
U	(C)		10	5	1	1	3	3
	(P)		20	10	2	3	6	6
<u>Low Level Waste</u>								
U			20	15	10	10	5	5
<u>In-Process Inventory Difference</u>								
Pu and U			5	--	5	--	5	--
(10 Process Vessels Per Inventory)								

Input Measurements

In the material balance model shown in Table 2, the only input errors which contribute to the across-the-plant MUF are those associated with dissolver solution measurements. Cladding waste measurement errors, as was noted earlier, contribute to the shipper-receiver difference variance for input.

Analytical Errors

Total uranium and total plutonium concentrations of dissolver solution are normally measured by the isotopic dilution method using a thermal ionization mass spectrometer. A measured aliquot of the sample is "spiked" or mixed with known amounts of ^{233}U and ^{242}Pu . After isotopic exchange has been obtained by chemical treatment, uranium and plutonium are separated from each other and extraneous constituents. The atom ratios of $^{238}\text{U}/^{233}\text{U}$ and $^{239}\text{Pu}/^{242}\text{Pu}$ are measured on a mass spectrometer. From those ratios, and other known volumes, concentrations, and measured isotopic compositions of the unspiked material, uranium and plutonium concentrations are computed. The method is capable of a precision of 0.20 to 0.40 relative percent standard deviation under carefully controlled conditions. More typical of good operating performance are relative standard deviations of the order of 1.0 to 1.5%. The method is highly specific and not as subject to interference by other constituents as are many chemical methods.

Under carefully controlled conditions, systematic errors can be kept in the range of 0.10 to 0.20 relative percent standard deviation. Systematic errors arise mainly from the fact that spike solutions are "standardized" or "calibrated" by the same method against a pure reference standard of uranium or plutonium. Since only a finite number of calibration runs are made, the value assigned to the spike solution has an associated error of 0.10 to 0.30% relative standard deviation.

Systematic errors can also arise from shifts in the "mass effect." Measured atom ratios are normally corrected by about 0.25 to 0.50 relative percent per mass unit to obtain "true" ratios. Bias correction factors are usually determined experimentally. These factors do change with time as a result of changes in source conditions and detector response.

Volume

Measurement of the volume of input dissolver solution is normally done with a dip-tube manometer system. That system measures the weight of solution above the bottom dip-tube (near the bottom of the tank). Conversion to volume units is made by dividing the weight by the specific gravity of the solution. Specific gravity is measured either at the plant laboratory or by a dip-tube manometer system which measures the weight of solution between two dip-tubes of exactly known separation. At key accountability points, a large (150-in.) water manometer is usually used. Readings can be reproducible to within ± 0.05 to 0.10 relative percent when special cares are taken. The precision of a single volume measurement which is normally a transfer volume involving two readings is estimated to have a "best" reproducibility of 0.10 relative percent standard deviation. For the performance model $\pm 0.30\%$ is considered good practice.

The limiting factor in reducing volume measurement error is the systematic error associated with the calibration curve or calibration table. To relate manometer readings to weight of solution, the system must be calibrated by adding known volumes (usually weight) of water to the empty tank to establish the relationship between manometer reading and solution weight. The tanks themselves are usually not perfect right cylinders. They also contain such items as coils, spargers, and agitators which have volume. Thus, it is not unusual for a tank to have as many as three or four calibration curves to

cover the entire usable height of the vessel. Under carefully controlled and nearly ideal conditions, calibration curves have been produced with reported accuracies of 0.10 relative percent standard deviation. In practice, a calibration error of $\pm 0.30\%$ or better⁽¹¹⁾ is considered good.

Sampling

Dissolver solution is nearly a true solution of uranyl nitrate containing only traces⁽⁹⁾ of solids (0.2 to 0.5 vol%). The main sources of sampling error postulated for dissolver solution are: (1) concentration or dilution of the sample by the air lift system used to circulate the process solution from the tank to sample port, (2) incomplete mixing of the solution in the tank prior to sampling, (3) failure to maintain clean sampling lines, (4) unrepresentative samples because of the presence of solids, and contaminated sample containers. To date, most sampling studies of dissolver solutions have shown that sampling error is difficult to distinguish from analytical error. As a result, estimates of sampling error are shown as less than values. In general, the possibility of sampling error is usually tested by comparing the sample specific gravity to the specific gravity measured in the tank itself after correcting for temperature differences. These checks usually agree within 0.10 to 0.20%. Normally, several samples are taken for each dissolver solution accountability batch.

The problem of undissolved solids which may carry or contain plutonium and uranium is part of the input shipper-receiver problem rather than a contributor to plant MUF. Presumably, such solids would not be extracted or be measured in plant waste solutions. Thus, as long as they don't enter the across-the-plant balance by being measured in input, waste, or product, they have no effect on MUF. There are definitely traces

of undissolved solids in dissolver solutions of high-exposure fuels. These are believed to consist mainly of noble metal fission products and solid solutions of certain fission products formed at high temperatures. The exact extent to which such solids may carry or contain uranium or plutonium has not been fully established. The best estimate of the effect of such solids on plant accountability (shipper-receiver differences) is that, at the worst, it is in the order of a few tenths of a percent.

Product Measurements

Plutonium

Plutonium product is usually sampled in the plutonium product receiver or product storage tanks. These tanks are critically safe tanks which use air sparging or liquid circulation to mix the nitrate solution. Samples are obtained from built-in sampling equipment which circulate the samples through a sample port. Usually these tanks hold about 100 to 150 liters of solution. The accountability procedure consists of determining the plutonium concentration of the solution in the tank by sampling and laboratory analyses. The solution in the tank is then transferred to 10-liter shipping bottles which are weighed before and after filling. A laboratory specific gravity determination is made on the tank samples to convert weight units to volume. The total plutonium in a final product batch then consists of the product of the total volume transferred and the mean of laboratory analyses for plutonium concentrations.

Analytical Error. Volumetric oxidation-reduction titration methods are commonly used for the analysis of plutonium nitrate product solutions. Reported precisions range from 0.20 to 1.00 relative percent standard deviation. For the capability model, a precision of 0.25% was regarded as a "best"

performance estimate for a production laboratory and a precision of $\pm 0.70\%$ for good quality performance. Systematic errors in volumetric methods are usually in the order of 0.05 to 0.15 relative percent standard deviation. Because of the large magnitude of between-method differences and between-laboratory differences (0.64 to 1.6%) that have been observed,^(3,7) it is felt that 0.15% systematic error represents the current "best" state-of-the-art. For the performance model, 0.30 relative percent standard deviation was selected as the limit of systematic error which could be associated with good performance. There are a number of reasons for the apparent systematic errors associated with analyses of production-grade plutonium nitrate. First is the fact that the oxidation-reduction methods which are used tend to measure (titrate) some of the impurities as plutonium. These impurities (except for uranium) have valence change to mass ratios 5 to 10 times greater than plutonium. Thus, even seemingly low levels of certain impurities (200 to 1000 ppm) can produce a significant bias. Usually, some after-the-fact correction is made for such impurities. When these corrections are made from composite samples, a bias correction factor is applied to all results. In this case, the systematic error is the error associated with the bias correction factor itself. Other extenuating circumstances which tend to increase the random and systematic error associated with such analyses are the intense alpha activity and the health hazard of plutonium itself. The intense alpha activity tends to produce concentration changes and undesirable by-products. Concentrated plutonium solutions require special handling and working facilities which make the analytical task more difficult.

Volume Measurement. The volume of plutonium product solutions is usually based upon weight divided by specific gravity. These measurements are capable of being performed with high

accuracy. Measurement capability is estimated to be 0.05 relative percent standard deviation for both random and systematic error. For good performance, it is expected that volume measurements could be controlled to within 0.10% relative standard deviation.

Sampling of Plutonium Product Solutions. Historically, the sampling of plutonium nitrate product solutions has been frequently subject to question. The large (1 to 2%) shipper-receiver differences that have been observed over the years has prompted many investigations and studies.^(6,7) However, in many of these investigations, it was not possible to isolate a significant sampling error from other errors. As a result, sampling errors for the plutonium nitrate product are listed as "less than" values. However, in view of the still unresolved shipper-receiver differences, the estimates given in Table 4 may be too optimistic (error levels are too small).

The causes of sampling errors which were discussed previously for dissolver solution are also true for plutonium product solutions. In many cases, solids have been observed. Also, the critically safe storage tanks are not amenable to good mixing as evidenced by the sampling errors which have been observed. In addition, definite differences have been observed between tank contents and sample contents. However, for the most part, these errors have been explained in such a manner that proper precautions would largely eliminate or minimize such errors.

Uranium Product Measurements

The uranium product is measured from fairly large storage tanks. Some of the newer plants use weigh tanks to improve final product accountability. Product accountability consists of sampling through built-in lines and obtaining laboratory analyses for uranium concentration or uranium weight/weight

of solution and specific gravity. When a weigh tank is used, the total uranium in a product batch is the product of the solution weight transferred out and the laboratory result for weight U/wt of solution. When uranium concentration is used, then the laboratory specific gravity is used to convert tank weight to volume or laboratory concentration measurements to weight units. Conventional dip-tube manometer systems are also used to measure the volume of uranium product solutions. The actual accountability transfer involves the transfer of UNH from the storage tank to a tank car or tank truck or trailer. When scales are available for loading the tank car or truck, an additional weight check is available for the product transfer.

Measurements. In general, the measurements associated with uranium product accountability represent nearly the "best" state-of-the-art. The values given in Table 4 tend to approach the inherent limits of the measurement system rather than represent errors which arise from the particular materials being measured or limitations imposed by the nature of the processing equipment. Good quality performance approaches what might be termed the capability of the system in an operating environment. The measurements themselves are well-documented in references ^(2,15) and do not require further discussion.

Liquid Waste Accountability

The main waste exit of a chemical separations plant is via the waste neutralization tank from which neutralized waste is sent to underground storage or to waste solidification. Alternatively, the highly acid wastes may be stored in stainless steel tanks without neutralization. In any event, the total amount of uranium and plutonium sent to underground storage is determined by the volume-concentration method.

Samples of the waste are taken in acid waste accountability tanks. From the laboratory analysis and the volume of solution transferred, the total uranium and plutonium sent to storage is computed.

Small amounts of uranium are discarded via the low-level waste system which usually discharges to a swamp or lagoon. Low-level waste consists mainly of steam condensate, and certain evaporator overheads. Flows are of the order of 42,000 liters per day. Accountability and health hazard control are usually maintained by testing each batch before discharge. Usually such wastes are collected in large retention basins or tanks so as to allow some time for corrective measures in case a high result is obtained.

Salt Waste Measurements

At a loss rate of about 1% of input, waste measurement errors are not an important contributor to the variance of MUF. The main concern, at this loss level, is that the waste loss is not grossly (>10%) over or understated. As a result, the same level of effort is not expended on waste measurements as on inputs and outputs.

Salt waste, which is the main waste stream, is a highly radioactive acid solution containing some 2 to 5 vol/% solids.⁽¹⁰⁾ Over-concentration of the waste leads to viscous slurries which are difficult to sample or analyze.

The analytical methods most commonly used for the analysis of salt waste are the TTA extraction-alpha counting method for plutonium, and the hexone extraction-fluorimetric method for uranium. These methods are discussed in detail in References 2 and 5.

The TTA extraction-alpha counting method has a limiting precision of about 10% relative standard deviation when used for high exposure plutonium waste analyses. For plutonium

produced in thermal reactors at an exposure of 20,000 to 25,000 MWd/tonne U, ^{238}Pu is the major alpha activity. It is, however, only a minor weight constituent. As a result, conversion of alpha activity to weight plutonium requires special specific activity measurements which themselves contribute to the random and systematic errors. In addition, the presence of beta and gamma emitters in the sample solution can further complicate the analysis. Unless special precautions are used, trace levels of fission products and transuranic elements will extract and carry through to the counting disk, giving rise to erratic and biased alpha activity measurements. Good performance under the many complicating circumstances associated with salt waste analysis is estimated to be 20% relative standard deviation for random error and 10% for systematic error.

The hexone extraction-fluorimetric method for uranium also has two basic sources of error. First are those which are inherent in the method itself. Second are those which arise from the particular properties of the sample.

On pure standards, random and systematic errors of the order of 5% are considered good. In application to salt waste solutions which contain many constituents which can cause serious error, the method is limited to a random error level of about 10% and a systematic error level of about 5%. For good performance, error levels below 20% for random error and 10% for systematic error are considered good.

Volume. Normally high precision volume measurement devices are not used in waste accountability tanks. A 1% error level is about the "best" that could be achieved with normal oil manometers or chart recorders. A 3% error level is considered good performance.

Sampling. Because of the presence of solids and the "salt-like" nature of concentrated acid waste, sampling errors of 3 and 6% relative standard deviation are postulated for the capability and performance model.

Low Level Waste Measurements

Low-level waste is monitored more to guard against the possibility of uranium leaving the system without detection than to establish the exact loss level. Normally, the volume of such streams are based on engineering estimates or liquid flows. Sampling is either by dipping or by the use of a proportional sampler in the inlet pipe. Normally, uranium sampling is done by the fluorimetric method after the sample has been concentrated to give sufficient sensitivity.

Inventory Measurements

For the model plant, it is assumed that the bulk of the in-process inventory which is left between campaigns is distributed among some 10 process vessels. It is assumed that a mixture of methods would be used, e.g., product, waste, and input methods. It is further assumed that the contents of each vessel can be measured with an average random error of 5% relative standard deviation for each component of error (5% sampling, 5% volume, 5% analytical). Since only the inventory difference is important in the variance of MUF, it is assumed that systematic errors, would for the most part, be self-canceling. Also, the relatively small amount of inventory postulated does not warrant more rigorous treatment.

ERROR PROPAGATION

In the context of this report, measurement error is considered to consist of two main components--random and systematic error. Random error refers to those errors which affect the reproducibility or precision of a measurement method without affecting the expected or mean value estimated by the method.

Systematic error refers to those errors which cause the expected value to differ from the true value. For the material balance model previously considered, random error is assumed to average out with consecutive batch measurements while systematic error is assumed to persist throughout a campaign.

Two basic sources of error are considered. First is the error inherent to the method itself such as what might be observed in the measurement of pure standards. Second are the errors which result from particular properties of the material being measured. Errors from this source are reflected in precision observed when process materials are measured, changes in such precision with time, and between-method differences. Errors arising from "process effects" can also be estimated from theoretical cause and effect relationships, e.g., a certain impurity level causes a certain response in a method.

For the systematic errors shown in Table 4, it is assumed that quality control data are used to control systematic error. Furthermore, it is assumed that any bias correction factors which are used are applied to the entire campaign. It is important to note that the systematic errors shown in Table 4 are the residual errors or the errors remaining after known biases have been taken into account. Thus, they represent the error in the bias correction factors, rather than the biases themselves.

The random and systematic components of error in the measurement of a single batch; e.g., total plutonium or uranium in an input, product, or waste batch, are given in Table 5. Relative errors for sampling, analysis and volume measurement were combined by the "sum of squares" method as follows: (see Reference 13).

TABLE 5. Combined Error Estimates for Individual Batch Measurements for a Chemical Processing Plant

Material Balance Component	Relative Percent Standard Deviation of a Single Batch Measurement			
	Random Error		Systematic Error	
	Capability	Performance	Capability	Performance
<u>Input</u>				
Pu	0.31	0.80	0.23	0.44
U	0.31	0.80	0.21	0.41
<u>Product</u>				
Pu	0.23	0.62	0.19	0.37
U	0.14	0.30	0.09	0.24
<u>Waste</u>				
1. <u>Salt Waste</u>				
Pu	10	21	6	12
U	10	21	6	12
2. <u>Low Level Waste</u>				
U		23		19
<u>In-Process Inventory Difference</u>				
Pu and U				4

$$\sigma_{\text{Rel. Random}} = \sqrt{\frac{\sigma_{\text{ar}}^2}{n} + \sigma_{\text{vr}}^2 + \frac{\sigma_{\text{sr}}^2}{n}}$$

where

$\sigma_{\text{Rel. Random}}$ = combined relative random error for sampling, analysis, and volume measurement for a single accountability batch.

σ_{ar} = analytical relative random error.

σ_{vr} = volume measurement relative random error.

σ_{sr} = sampling relative random error.

n = number of replicate measurements applied to a single batch.

Systematic errors were combined as follows:

$$\sigma_{\text{Rel. Systematic}} = \sqrt{\sigma_{\text{as}}^2 + \sigma_{\text{vs}}^2 + \sigma_{\text{ss}}^2}$$

where

σ_{as} , σ_{vs} , σ_{ss} are the relative systematic errors of analysis, volume measurement, and sampling that are associated with the measurement of a single batch quantity.

For example, the relative errors associated with a plutonium input batch were combined as below assuming two samples, and two plutonium assays per batch: (i.e., one assay per sample).

$$\begin{aligned}\sigma_{\text{Pu random}} &= \sqrt{\frac{(0.40)^2}{2} + \frac{(0.10)^2}{1} + \frac{(0.10)^2}{2}} \\ &= \sqrt{0.095}\end{aligned}$$

$$\sigma_{\text{Pu random}} = 0.31\% \text{ (Capability model)}$$

$$\sigma_{\text{Pu systematic}} = \sqrt{(0.15)^2 + (0.15)^2 + (0.10)^2}$$

$$\sigma_{\text{Pu systematic}} = 0.23\% \text{ (Capability model)}$$

From the number of batches and quantities per batch, the propagated errors for total inputs, total outputs, and total waste in a campaign were determined. For purposes of exposition, the combined and individual components of error are shown in relative units in Table 6. The calculations for the combined error for total plutonium input are shown below:

$$\sigma_{\text{Pu, Input}}^2 = n[(\text{kg/batch})(\sigma_r)]^2 + n^2[(\text{kg/batch})(\sigma_s)]^2$$

(Capability model)

$$\sigma_{\text{Pu, Input}}^2 = 40[(10)(0.0031)]^2 + (40)^2[(10)(0.0023)]^2$$

$$\sigma_{\text{Pu, Input}}^2 = 0.03844 \text{ kg}^2 + 0.8464 \text{ kg}^2$$

$$\sigma_{\text{Pu, Input}}^2 = 0.8848 \text{ kg}^2 \text{ (Capability model)}$$

Inventory errors were propagated on the basis of 10 process vessels containing 0.20 kg of Pu and 20 kg of U, each measured with 8.66% relative standard deviation for combined random error. Taking into account both the beginning and the ending inventory, results in a 4% relative (relative to the total amount in inventory) error for the inventory difference as shown in Table 5, e.g., $\frac{8.66\sqrt{2}}{\sqrt{10}} \approx 4$. Relative to input, the error in measuring the inventory is 0.02 relative percent standard deviation.

The variance of MUF for the performance and capability models was calculated by adding the variances of the individual components of the material balance as follows:

$$\sigma_{\text{MUF}}^2 = \sigma_{\text{Input}}^2 + \sigma_{\text{Output}}^2 + \sigma_{\text{Waste}}^2 + \sigma_{\Delta\text{Inventory}}^2$$

TABLE 6. Combined Error Estimates for Material Balance Components of a Single Campaign at a Chemical Processing Plant

Material Balance Component	Batch Size, kg	Number of Batches	Total Material in Campaign		Relative Standard Deviation as a Percent of Plant Input ^(a)					
					Combined Error		Random		Systematic	
					Capability	Performance	(C)	(P) ^(b)	(C)	(P)
<u>Total Input</u>										
Pu	10	40	400		0.24	0.46	0.050	0.127	0.23	0.44
U	1000	40		40,000	0.22	0.43	0.050	0.127	0.21	0.41
<u>Total Product</u>										
Pu	22	18	396		0.19	0.39	0.054	0.146	0.19	0.37
U	3958	10		39,580	0.10	0.26	0.044	0.094	0.089	0.24
<u>Total Waste</u>										
1. <u>Salt Waste</u>										
Pu	0.25	16	4		0.065	0.13	0.025	0.053	0.060	0.12
U	25	16		400	0.065	0.13	0.025	0.053	0.060	0.12
2. <u>Low Level Waste</u>										
U	0.04	100		4		0.002		0.0002		0.002
<u>In-Process Inventory Difference</u>										
Pu	0.20	10	2			0.02				
U	2	10		20		0.02				

(a) Relative Standard Deviation of entire component, e.g., total input, total product, etc.

(b) (C) = Capability (P) = Performance

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The contributions of the individual components to the overall variance in MUF are shown in Table 7. For the plutonium material balance, about 60% of the variance of MUF is contributed by input measurements. For the uranium balance, about 70% of the overall variance is caused by input measurement error. In both cases, the major component of error is systematic error.

TABLE 7. Contribution of Individual Material Balance Component to the Overall Variance of MUF
Chemical Processing Plant

Plutonium Balance Component	% Contribution to Total MUF		% of Random and Systematic Error			
	Capability Model	Performance Model	Capability Model		Performance Model	
			Random	Systematic	Random	Systematic
Input	59	56	5	95	8	92
Product	37	40	7	93	13	87
Waste	4	4	15	85	16	84
Inventory Difference	~0.4	~0.4			100	
	% of Input					
$\sigma_{\text{MUF, Pu}}$	0.31	0.62	6	94	10	90
Uranium Balance						
Component						
Input	77	69	6	94	9	91
Product	16	25	20	80	13	87
Waste	6	6	15	85	16	84
Inventory Difference	~0.6	~0.1	--	--	100	--
	% of Input					
$\sigma_{\text{MUF, U}}$	0.25	0.52	9	91	10	90

SHIPPER-RECEIVER DIFFERENCES

The expected precisions of shipper-receiver differences are given in Table 8 in terms of expected capability of shipper and receiver measurements. Because of the limited data⁽¹²⁾ on reactor versus chemical plant on plutonium input, a range of values is given.

TABLE 8. Expected Precision of Difference Between Chemical Plant and Shipper and Receiver-Capability Model

<u>Material</u>	<u>Amount, kg</u>	<u>Shipper-Receiver Point</u>	<u>Relative Standard Deviation of Difference, %</u>
U	40,000	Input Area	0.30
U	39,580	Product Area	0.14
Pu	400	Input Area	2.0-4.0
Pu	396	Product Area	0.27

The shipper-receiver difference is treated as a random normal variable with a mean of zero and a standard deviation of σ . The values shown for relative standard deviation are based on the combined errors shown in Table 6 for the chemical plant and estimated capabilities of shippers to the chemical plant and receivers of product materials. For the product measurements, it is assumed that the product receiver has the same measurement capability as does the chemical processing plant. For the uranium input difference, it is assumed that the fabricators total uranium is good to 0.10% relative standard deviation and that 3.6% of the total uranium is consumed in the reactor. It is further assumed that the burnup can be estimated to 5% relative standard deviation. The range shown for reactor versus chemical plant measurements on plutonium input is what might be expected on successive cores from the same reactor.

PLUTONIUM TO URANIUM RATIO METHOD

An alternative method of determining total plutonium input for a campaign is by the plutonium to uranium ratio method.⁽¹⁴⁾ This method is only applicable when the total uranium input from fabrication is known to a high degree of certainty. In the ratio method, the weighted plutonium to uranium ratio obtained from all dissolver solution input batches is applied to the shipper's values for total uranium (fabrication values corrected for burnup.)

The weighted Pu/U ratio is computed as follows:

$$\text{The weighted ratio Pu/U} = \frac{\sum_{i=1}^n V_i U_i \frac{\text{Pu}_i}{U_i}}{\sum_{i=1}^n V_i U_i}$$

where

V_i = the total volume of the i th input batch

U_i = uranium concentration of the i th input batch

Pu_i = plutonium concentration of the i th input batch

The variance of the weighted ratio is approximately equal to the sum of the plutonium and uranium analytical variance for the campaign, e.g.,

$$\text{The weighted ratio } \sigma_{\text{Pu/U}}^2 \approx \sigma_{\text{Pu analytical}}^2 + \sigma_{\text{U analytical}}^2$$

This is true, since (1) volume systematic errors are self-canceling, (2) sampling systematic error tends to cancel, and (3) volume measurement random errors are perfectly correlated. When the "mixed spike" isotopic dilution method is used, analytical errors are somewhat reduced since dilution and pipetting errors are self-canceling. The estimated precision of the weighted ratio obtained for the capability model is:

$$\text{The weighted ratio } \sigma_{\text{Pu/U}} \approx 0.16 \text{ relative percent (capability)}$$

Total plutonium input by the ratio method has the following estimated variance:

$$\sigma_{\text{Pu, ratio method}}^2 \approx \sigma_{\text{Pu/U}}^2 + \sigma_{\text{U (reactor-fabrication)}}^2$$

$$\sigma_{\text{Pu, ratio method}}^2 \approx (0.16)^2 + (0.21)^2$$

σ_{Pu} , ratio method $\approx$ 0.26 relative percent for standard deviation of total plutonium input in a campaign.
(capability)

It should be noted that the above values are for the capability of the measurement system. Corresponding performance values are on the order of σ_{Pu} , ratio method = 0.40 relative percent.

When the ratio method is used as the official accountability method, the errors in cladding waste measurements enter into the variance of MUF. Normally, cladding waste losses are sufficiently small that even fairly large measurement errors (15 to 20%) do not contribute significantly to the variance of MUF.

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REFERENCES

1. D. P. Granquist and R. A. Schneider. Application of Safeguards to Nuclear Fuel Processing Plants, BNWL-301. Battelle-Northwest, Richland, Washington, 1966.
2. Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle, edited by R. J. Jones, TID-7029. Division of Technical Information Extension, Oak Ridge, Tennessee, 1963.
3. M. W. Lerner. Evaluation of the Methods in TID-7029, NBL-231. New Brunswick Laboratory, AEC, New Brunswick, New Jersey, 1966.
4. C. G. Hough. Statistical Analysis-Accuracy of Volume Measurements in a Large Process Vessel, HW-62177. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, October, 1959.
5. Chemical Processing Department Analytical Technical Manual, edited by R. A. Schneider and K. M. Harmon, HW-53368. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, 1961.
6. D. W. Hoba and R. A. Schneider. "Redox SS Materials Balance Test," AEC Contractors and SS Materials Management Meeting, Germantown, Maryland, TID-7581. Division of Technical Information Extension, Oak Ridge, Tennessee, May 1959.
7. J. T. Byrne, R. L. Delnay, et al. Measurements of Plutonium Nitrate Shipping Solutions, RFT-436. Rocky Flats Division, The Dow Chemical Company, Golden, Colorado, 1964.
8. R. P. Wischow, J. R. Clark, and I. B. Roll. Safeguards Procedures Manual for the NFS Reprocessing Plant, Volumes 1, 2, 3. Nuclear Fuels Services, Inc., 1967.
9. R. A. Schneider, W. Y. Matsumoto, M. W. Goheen, and A. C. Leaf. Unpublished Data. Battelle-Northwest, Richland, Washington. (Internal Reports 1966-1967: Analysis of Shippingport and Yankee Fuel Samples)
10. L. A. Bray. Unpublished Data. Battelle-Northwest, Richland, Washington, 1969. (Personal Communication: Composition of Concentrated Purex Acid Waste)
11. L. F. Wirfs. Unpublished Data. Battelle-Northwest, Richland, Washington, 1969. (Personal Communication: Historical Calibration Data for Large Volume Process Vessels)

12. G. H. Daly. "USAEC Surveillance Activities in an Irradiated Fuel Reprocessing Plant," IAEA Panel on Safeguards Technical Practices for Irradiated Fuel Plants, Vienna, Austria, April, 1968.
13. K. B. Stewart and J. L. Jaech. "The Role of Statistics in Safeguards," IAEA Panel on Safeguards Systems Analysis of Nuclear Fuel Cycles, Vienna, Austria, August, 1969.
14. R. A. Schneider. Measurement of the Nuclear Materials Content of Non-Production Fuel, HW-60533. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, 1959.
15. C. J. Rodden, Editor. Analysis of Essential Nuclear Reactor Materials, USAEC, Superintendent of Documents, Washington, D.C.
16. Nuclear Materials Controls in Irradiated Fuel Reprocessing, by personnel of the Hanford Atomic Energy Works and the Idaho Falls Chemical Processing Plant of the AEC, TID-12100. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, 1961.
17. Fuels Reprocessing Branch. Reprocessing of Dresden Fuel, SRO-123. Savannah River Operations Office, Aiken, South Carolina, 1968.
18. Fuels Reprocessing Branch. Reprocessing of Yankee Fuel, SRO-124. Savannah River Operations Office, Aiken, South Carolina, 1968.

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