The Need for Experimental Data for LEMUF Calculations

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The calculation of the limit of error of MUF (LEMUF) must be based entirely on measured quantities of special nuclear material (SNM) that appear in the material balance. Measurement of these quantities must be technically valid and based on good experimental data. This paper illustrates, through a sample calculation of the propagated limits of error across the entire material balance of a typical fuel facility, how the systematic and random components of measurement errors contribute to the LEMUF. The example gives a typical list of the materials that must be measured, a set of individual measurement errors that must be estimated and incorporated into the propagated error, and the results of the sample calculations.

Designation of material balance areas (MBA) is essential to a materials accounting system for safeguards and make it possible to draw a measured material balance. Figure 1 shows a storage area MBA where transfers in and out of the area may be made on the basis of item identification in case of fuel pins or assemblies or by measured tag values in the containers that are sealed. In this MBA no materials unaccounted for (MUF) is possible unless there has been an accounting error or a diversion has taken place. A physical inventory of fuel pins or assemblies consists of a simple item count. Sealed identifiable containers enter and leave the MBA on the measured tag values. The value of the limit of error on the contents of a storage MBA is a propagated error of the measured values on the tags. Again no MUF is possible in a storage area MBA unless an accounting error has been made or a real loss has occurred.

Figure 2 is a diagram of a process MBA. In the process MBA the measured values of transfer in are not identical with the measured transfers out, some losses occur and in general, the values of MUF will vary around zero and LEMUF will differ from zero. In the process MBA flow in and flow out is measured and a complete measured material balance is possible only by a thorough cleanout of the process area or by an accurate measurement of the material remaining in the process equipment.

Clearly, in the process MBA item accounting is not valid and the measured material balance depends upon quantity accounting. Quantity accounting provides for a sampling and measuring plan for feed, product and discards flowing across the MBA boundaries. Also essential to quantity accounting is a calibration program for measurements used to determine the flow and the inventory of SNM. Finally, quantity accounting must provide for a determination of limits of error, both systematic and random, based on experimental data.

An illustrative calculation applied to a hypothetical plutonium uranium oxide fuel conversion and fabrication facility indicates the kinds of measurement data that are required to perform an analysis and evaluation of LEMUF. In this example it was assumed that the facility would have a thruput of 1/2 tonne of fuel per day and that the nominal accounting period would be once every 10 days. The results are based on purely hypothetical data but it is felt that the quantities and the sampling frequencies that were assumed are realistic for a commercial facility of this type. The assumed data and results do not apply to any particular plant that is now in operation nor do the assumptions reflect AEC policy.

Figure 3 gives the expected flows and the proposed sampling frequency. Figure 4 gives the quantities of material that would be found in a total ending inventory after a cleanout. In this example it was assumed that the total ending inventory contained a quantity of Pu that was equivalent to four months of receipts. To illustrate the importance of a cleanout of in-process material, Figure 5 gives the distribution of the estimated quantities of plutonium hold-up in the process equipment.

The single measurement errors that have been assumed for the assay of the various materials in the facility are given in Figure 6. Preferably these measurement errors should be based on experimental data obtained by the facility or on the best applicable information available to the facility. The contributions of weighing, sampling and analysis to the individual measurement errors for each material provide the basis for calculating the components of LEMUF. Measurement error data has been published by Wirfs (1) for low enriched U facilities but no similar summary exists in the literature for mixed oxide facilities. However, some of the errors assumed to be valid in Figure 6 were based on the Wirfs' data on the assumption that a commercial mixed oxide facility for thermal reactor

fuel fabrication would resemble, in some aspects, a low enriched UO_2 facility. For example, mixed oxide bulk powders should be subject to about the same relative weighing errors as low enriched UO_2 bulk powders. Measurement errors applicable to $\text{Pu}(\text{NO}_3)_4$ receipt have also been published by Wirfs (2).

Values for Pu assay based on coulometric titration of sintered pellets appear in recently published information for the Integrated Safeguards Experiment (3) and in a recently published study by Los Alamos for FFTR fuel (4,5). Measurement uncertainties for spent fuel processing plants can be found in a report by Schneider and Granquist (6).

Figure 7 shows the working equations that were used to calculate the limits of error associated with the measurement of each material type and the equation used to combine these quantities to determine the LEMUF. Figure 8 presents the square of the propagated uncertainties for each material type after elimination of some covariance items (items that appear in both the beginning and ending inventory and do not contribute to the LEMUF). The results of this calculation are given on the bottom of Figure 8 showing that LEMUF for this hypothetical case is 2.97 kg of Pu for an assumed accounting period of 10 days.

Both Figure 7 and Figure 8 indicate which measurement errors make large contributions to LEMUF. Improved measurement techniques that would reduce the systematic error would have a strong effect on the value of LEMUF. One of the most important improvements that can be foreseen will be the introduction of nondestructive assay techniques for plutonium. Assays based on calorimetry, passive neutron counting and passive gamma ray counting will be especially useful in a mixed oxide facility and offer the possibility of reducing the systematic error of determination of total Pu in items to a very low level. The primary gain would be through reduction of the sampling error.

The determination of LEMUF is only valid if the results are based on measurements whose values have been evaluated by careful experiments and tests. These data should provide not only the precision of the technique but must also provide an evaluation of the systematic and random errors for bulk

measurement, sampling, chemical analysis and isotopic analysis (where applicable) for all significant measurements of SNM. To distinguish between real losses and measurement errors, requires reliable measurement data to evaluate limits of error and a program for reducing the LEMUF values.

ITEM ACCOUNTABILITY STORAGE MBA

FIGURE 1

PROCESS MBA

$$MUF \equiv BI - EI + R - S - NOL$$

FIGURE 2

Hypothetical 1/2 te (U+Pu)/Day Mixed Oxide Fabrication Facility

Material	Average Production (P) per 10 days					Chemical Assays	
	Pu Weight kgs	Bulk Weight kgs	Weighing Unit kgs	No. of units		Frequency	No.
Pu Nitrate Receipts	125.0	947 [†]	12.8	74		1/20 kgs Pu	6
Pu Nitrate Blend	125.0	1488 [†]	125	12		1/20 kgs Pu	6
U-Pu Nitrate Blend	141.3	68592 [†]	2384	29		1/1 blend	29
Mixed Powder	137.5	6278	8	785		2/day	20
Milled Powder	137.1	6260	8	782		2/day	20
Green Pellets	134.3	6132	5	1226		2/day	20
Sintered Pellets	133.0	6045	5	1209		3/day	30
Ground Pellets	127.6	5800	5	1160		5/day	50
Rods	126.2	5736	4	1434		*	49*
Rod Bundles	124.9	5677	4	1419		*	49*
Liquid Waste	.004	10 ^{8†}	3.3 (10 ⁵)	300		1/1 dump	300
Solid Waste	.096	-	-	19		**	**
Dissolved Scrap	16.3	7913 [†]	2384	3		1/1 unit	3

[†] Weight of solution

* Pu content based on chemical analysis of Ground Pellets

** Solid waste measured by NDT

Inventory Distribution After Cleanout

<u>Material</u>	<u>Pu weight kgs</u>	<u>No. of units</u>	<u>I_E/P</u>	<u>No. of chem. assays</u>
Pu Nitrate Receipts	75	44	.59	3
Pu Nitrate Blend	75	7	.58	3
U-Pu Nitrate Blend	225	46	1.59	46
Mixed Powder	105	599	.76	15
Milled Powder	225	1284	1.64	33
Green Pellets	75	685	.55	11
Sintered Pellets	85	773	.64	19
Ground Pellets	225	2045	1.76	88
Rods	150	1704	1.19	58
Rod Bundles	150	1704	1.20	59
Dissolved Scrap	<u>110</u>	22	7.33	22
	1500			

FIGURE 4

**DISTRIBUTION OF IN-PROCESS MATERIAL AND
FORM AT TIME OF CLEANOUT**

<u>LOCATION</u>	<u>Pu WEIGHT KGS</u>	<u>CLEANOUT FORM</u>	<u>NO. OF UNITS</u>
WET PROCESSES IN CO-PRECIPITATION	30	MIXED POWDER	171
SINTERING FURNACE	10	SINTERED PELLETS	91
PELLET GRINDER	5	SCRAP SOLUTION	1
SCRAP RECOVERY	30	SCRAP SOLUTION	6
TOTAL IN-PROCESS	75 KG.		

FIGURE 5

Single Measurement Uncertainties (%)
95% Confidence Level

	<u>bulk*</u>		<u>sampling</u>		<u>analysis</u>		<u>Total</u>		
	<u>sys</u>	<u>ran</u>	<u>sys</u>	<u>ran</u>	<u>sys</u>	<u>ran</u>	<u>sys</u>	<u>ran</u>	<u>combined</u>
Pu(NO ₃) ₄ Receipts	.1	.1	.25	.50	.3	.5	.40	.71	.81
Pu(NO ₃) ₄ Blends	.1	.1	.25	.50	.3	.5	.40	.71	.81
U-Pu Blends	.1	.1	.25	.50	.6	1.2	.66	1.30	1.46
Mixed Powder Powder	.1	.14	.25	.50	.6	1.2	.66	1.31	1.46
Milled Powder	.1	.14	.25	.50	.6	1.2	.66	1.31	1.46
Pellets	.1	.1	.05	.10	.6	1.2	.61	1.21	1.35
Liquid Waste	2.0	7.0	15.	30.	5.	12.	15.9	33.1	36.7
Solid Waste	---	---	---	---	5.**	15.**	5.	15.	15.8
Scrap Sol'n.	.1	.1	.30	.60	.6	1.2	.68	1.34	1.51

* weight or volume

** Pu content measured directly by NDT (approximate errors based on 5 gms Pu and 200 gms U in 5 gal. cans).

FIGURE 6

WORKING EQUATIONS

$$Q_j = \bar{c}_j \sum_{N_W} w_j$$

$$LE^2(Q_j) = Q_j^2 \left[LE^2(W \text{ REL. SYS.}) + \frac{1}{N_W} LE^2(W \text{ REL. RAN.}) \right]$$

$$+ Q_j^2 \left[LE^2(S \text{ REL. SYS.}) + \frac{1}{N_S} LE^2(S \text{ REL. RAN.}) \right]$$

$$+ Q_j^2 \left[LE^2(C \text{ REL. SYS.}) + \frac{1}{N_C} LE^2(C \text{ REL. RAN.}) \right]$$

$$LE^2(I_B) = \sum_N LE^2(Q_j)$$

$$LE^2(MUF) = LE^2(I_B) + LE^2(R) + LE^2(S)$$

$$+ LE^2(D) + LE^2(I_E)$$

FIGURE 7

Square of Measurement Errors ($\text{kgs}^2 \text{ Pu}/10 \text{ days}$)

Hypothetical Co-Precipitation Mixed Oxide Facility ($1/2 \text{ Te U-Pu/day}$)

Excluding Covariance Items

Material	I_B		Receipts or Removals		I_E	
	Sys	Ran	Sys	Ran	Sys	Ran
Pu Nitrate Receipts	.0898	.0922	.0418	.0643	.0000	.0000
Pu Nitrate Blends	.0645	.0668	-	-	.0645	.0668
U-Pu Nitrate Blends	.8773	.1189	-	-	.8773	.1189
Mixed Powder	.4104	.1146	-	-	.4104	.1146
Milled Powder	.8118	.1587	-	-	.8118	.1587
Green Pellets	.1700	.0662	-	-	.1700	.0662
Sintered Pellets	.2240	.0513	-	-	.2240	.0513
Ground Pellets	.6065	.0472	-	-	.6065	.0472
Rods	.0001	.0003	-	-	.5932	.0471
Rod Bundles	.0000	.0000	.0000	.0000	.0000	.0000
Scrap Solution	.1109	.0436	-	-	.1109	.0436
Liquid Waste	.0000	.0000	.0000	.0000	.0000	.0000
Solid Waste	.0000	.0000	.0003	.0001	.0000	.0000
	3.3653	.7598	.0421	.0644	3.8686	.7144

$$\text{LEMUF} = [(3.3653 + .7598) + (.0421 + .0644) + (3.8686 + .7144)]^{1/2}$$

$$= 2.97 \text{ kgs Pu}/10 \text{ days}$$

FIGURE 8

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