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SUBJECT: Problems in Accountability Measurements Associated
with the Interim Chemical Processing Program

TO: F. L. Culler

FROM: E. D. Arnold and A. T. Gresky

Abstract

Available knowledge of precision limits in S.S. accountability measurements and/or calculations by reactor and chemical processing groups is surveyed and summarized. Experience in comparisons of reactor (production and research) calculations versus chemical plant accountability measurements is also reported. A general tentative conclusion is that available precisions ($\pm$ 0.54 to $\pm$ 0.78%) in chemical plant measurements (bulk and analytical) for fissionable material accountability is superior to the variable precision ($\pm$ 1.0 to $\pm$ 11.0%) possible by calculations (nuclear and/or engineering) of power reactor systems; however, with operation and empirical experience (e.g. after two or three core loadings), it is believed that calculations for given reactors can attain acceptable precisions, e.g. less than $\pm$ 1.0%

It may be proposed that fuel payments be made as follows: 90% of fuel value based on reactor calculations, an additional 5% based on dissolver analyses, and final settlement based on chemical plant material balance (product plus loss analyses).

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Problems in Accountability Measurements Associated
with the Interim Chemical Processing Program

(This memo has been prepared as requested by F. L. Culler,
April 28, 1959, in a letter to A. T. Gresky, re: Outline for
Accountability Work to Use in Power Reactor Negotiations. The
cooperation and assistance of L. T. Corbin, W. H. Lewis,
W. T. McDuffee, G. S. Sadowski, J. A. Harvey, and L. Dresner
of ORNL, and J. Vanderryn of the AEC-ORO are gratefully acknow-
ledged.)

Introduction and Summary

Accountability of source and special materials is recognized as a
major problem affecting transfer of irradiated fuels from private reactor
operations to the chemical reprocessing plants and/or the AEC. Owing to
the high value of fissionable materials, it is of primary concern that
the chemical operations attain high precision and accuracy in bulk and
analytical measurements of fuel solutions (because small percentage
errors in fissionable material content can represent relatively large
monetary losses or gains).

The purpose of this brief survey of accountability problems is to
summarize available existing knowledge of precision limits in the chemical
plant operations (bulk and analytical measurements) and in the reactors
(S.S. material/content in fuel elements before and after the reactor
cycle), i.e. to assist in determinations of policies and contractual
agreements between reactor operators and the AEC. Since the only existing
knowledge of accountability problems has involved experience in AEC
production reactor fuel cycles (relatively much less complex than expected
for the power reactor cycles), it may be assumed that the precision data
reported here is somewhat conservative. However, it is expected that
future statistical and developmental experience in both the irradiation
and processing of power reactor fuels can approach or even exceed the
precisions indicated.

The following brief conclusions tentatively summarize the more
pertinent aspects of the survey:

Fissionable material content in fuel elements containing U-235 has been
measured by gamma scanning with accuracies of $\pm$ 0.2 to $\pm$ 2%. However,
similar data are not available for elements containing U-233 or Pu-239.

Mass assay of uranium isotopic ratios is accurate to $\pm$ 0.001 to $\pm$ 0.1%.
Assay of plutonium isotopic ratios is accurate to $\pm$ 0.5 to $\pm$ 1.0%.

The various physical constants employed in reactor irradiation calculations are generally accurate to about $\pm$ 1.0%. However, in high temperature systems where the ratio of resonance-to-total neutron density is high, the contribution of error by unknown resonance cross-sections (of fissionable and fertile isotopes, etc.) may be quite high. Resonance integrals are accurate to only $\pm$ 20 to $\pm$ 50%, which might lead to errors of about $\pm$ 1.0 to $\pm$ 10.0%, and could be even higher if an error existed in the evaluation of the spectral index. (In two cases, reactor (MTR) burn-up estimates have deviated from chemical plant measurements by -4.2 to -8.77%.)

Measurement of reactor coolant temperatures are quite accurate, e.g. $\pm$ 0.5°F; however, measurements of high coolant flowrates may be accurate to only $\pm$ 2 to $\pm$ 10%, the error generally increasing with the rate.

Bulk measurement of fuel solutions in the chemical plants are accurate to $\pm$ 0.4 to $\pm$ 0.65%.

Analytical accuracy for fissionable and fertile elements in the measurement of highly radioactive feed solutions ranges from $\pm$ 2 to $\pm$ 4%; however, uranium and plutonium measurements in the total plant product and waste solutions are considerably more accurate, e.g. $\pm$ 0.2 to $\pm$ 0.64% (analysis of thorium products is accurate to $\pm$ 2.0%.)

In several ORNL pilot plant programs, reactor estimates versus chemical plant measurements of uranium and/or plutonium have shown deviations of -1.95% to -6.67%.

On the basis of present practical knowledge, it is believed that the chemical plant accuracy for analysis of fissionable materials (product + waste determination) of $\pm$ 0.54 to $\pm$ 0.78% affords a basis for accountability which is definitely superior to that of reactor estimates with accuracies of $\pm$ 1.0 to $\pm$ 11%. Continued analytical and engineering development in both the fuel processing plants and reactor operations may well be expected to improve the above precisions markedly. As operational statistics increase, production reactor experience suggests that future close agreements between power reactor estimates and chemical plant measurement can be expected.

A bibliography of reports pertinent to the accountability problem is appended. These reports are recommended for more detailed descriptions of precision data as well as several other important phases of accountability (as summarized in the following brief survey).

I. Determination of Accuracy and Precision of Methods Currently Used in ORNL Pilot Plant Programs

(Fuel materials handled: conventional production-type irradiated elements, i.e. aluminum-jacketed uranium and thorium metal slugs. Accountability measurements have been made on the basis of (A) analysis of dissolved

fuel solution fed into the processing plant, and (B) analysis of product solutions and waste losses leaving the plant.

(A) Analysis of Feed Solutions

(Differs from B in that the major portions of measured solutions are highly radioactive. Therefore, order of difficulty and/or inaccuracy is higher for (1) bulk measurement of volumes and densities in the plant, and (2) analysis of samples in the laboratory.)

(1) Bulk Measurement

(Measured by solution liquid-level and density data as determined with remote pneumatic-type instruments.)

Tests with equipment properly designed and operated with respect to instrumentation, calibration, and solution mixing have indicated accuracies, or limits of error, of about $\pm 0.65\%$.

(2) Sample Analyses

(Precision values, as per L. T. Corbin, are limits of error of one determination based on 95% probability level, with no bias existing.)

a) Uranium: Spectrophotometric ammonium thiocyanate method, $\pm 4\%$. (The coulometric method, now under development and believed to be available soon, should allow a precision of $\pm 1\%$.)

b) Thorium: Spectrophotometric-thoron method, $\pm 2\%$.

c) Plutonium: Thenoyltrifluoroacetone extraction method, $\pm 2.5\%$. (Requires that specific activity of the plutonium be known.)

d) Uranium-233: Hexone extraction method, $\pm 4\%$.

In four pilot plant uranium campaigns the combined limits of error (bulk measurement and analysis) were measured for uranium at $2.5-3.7\%$, and for plutonium at $1.6-2.7\%$.

(B) Analysis of Products and Waste Losses

(This case is considered on the basis of pilot plant experience, to represent the most valid and/or accurate analysis for accountability.)

(1) Bulk Measurement

The "cold" product and waste solutions, normally accounting for greater than 99% of original material fed to the process, can be measured in directly maintained equipment by determinations of solution weight and density data. Tests indicate limit of error is $\pm 0.4\%$.

The radioactive waste solutions (e.g. aqueous waste from initial "hot" cycle), normally accounting for less than 1% of original material fed to the process, must be measured by determinations of solution liquid level and density data (as is case A) which permit limits of error of $\pm$ 0.65%.

(2) Sample Analyses

Product Solutions

Uranium: Automatic potentiometric ferric sulfate method, $\pm$ 0.2%.

Thorium: Spectrophotometric-thoron method, $\pm$ 2%.

Plutonium: Potentiometric ceric sulfate method, $\pm$ 0.5%.

Uranium-233: Automatic potentiometric ferric sulfate method, $\pm$ 0.2%.

Hot Waste Solutions

Uranium: Fluorometric method, $\pm$ 10% (for low uranium concentrations, e.g. < 0.01 g/liter).

Thorium: Spectrophotometric-thoron method, $\pm$ 2%.

Plutonium: Thenoyltrifluoroacetone extraction method, $\pm$ 15% (for low plutonium concentrations, e.g. < 0.00005 g/liter).

Uranium-233: Hexone extraction method, $\pm$ 10% (for low uranium concentrations, e.g. < 0.00005 g/liter).

(Table 1 summarizes the above limits of error for cases A and B, and suggests an estimate for the combined probable error, i.e. including both the bulk measurement and analytical errors.)

High accuracies in mass assay are possible, as follows: uranium isotopic ratios (natural to 5% enriched), $\pm$ 0.01 to $\pm$ 0.001% if working against standards, or $\pm$ 0.1 to $\pm$ 0.01% if by direct measurement; plutonium isotopic ratios, $\pm$ 0.5 to $\pm$ 1.0%.

II. Chemical Plant Accountability Determinations Versus Reactor Estimates in ORNL Pilot Plant Campaigns

Table 2 summarizes comparisons, for four major pilot plant campaigns, of (1) shipper's estimates versus (2) chemical plant feed measurements, and (3) chemical plant loss and product measurements, i.e. in the determinations of uranium, uranium-233, and plutonium. Limits of error in the two methods of chemical plant measurement were as follows:

Table 1. Summary of Limits of Error in Bulk Measurements
and Sample Analysis

	Method	Fuel	Fraction of Feed	Bulk Measurement Error ± %	Analytical Error ± %	Combined Error ± %	Combined Errors (Overall) ± %
A)	Feed						
	Measurement	U	1.0	0.65	4.0*	4.05*	4.05*
		Th	1.0	0.65	2.0	2.11	2.11
		Pu	1.0	0.65	2.5	2.58	2.58
		U-233	1.0	0.65	4.0*	4.05*	4.05*
B)	Product and Loss	Product:					
	Measurement	U	>0.99	0.4	0.2	0.45	0.54
		Th	>0.99	0.4	2.0	2.04	2.04
		Pu	>0.99	0.4	0.5	0.64	0.78
		U-233	>0.99	0.4	0.2	0.45	0.54
		Loss:					
		U	<0.01	0.65	10.0	10.0	
		Th	<0.01	0.65	2.0	2.11	
		Pu	<0.01	0.65	15.0	15.0	
		U-233	<0.01	0.65	10.0	10.0	

* Values may be reduced by future coulometric method to 1.0%.

Table 2. <u>Irradiated Fuel Recovery</u>

Program	Shippers Estimate			Feed Measurements			Recovery Product and Losses		
	U,kg	U-233, g	Pu, g	U, kg	U-233, g	Pu, g	U, kg	U-233, g	Pu, g
Clementine Fuel	-	-	15,005.5±?	-	-	14,716+430	-	-	14,713±103
Thorex	-	92,143±?	-	-	92,509±5551	-	-	87,299±1053	-
SCRUP-2	5620.4±?	-	2163.4±?	5386±200	-	2028±57	5329±38	-	2019±13
Pu-Al	-	-	1153.7±?	-	-	1120±145	-	-	1111±6

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Program	Feed	Product and Loss
Clementine	± 2.7% (Pu)	± 0.7% (Pu)
Thorex	± 6% (U-233)	± 1% (U-233)
Scrup-2	± 3.7% (U)	± 0.7% (U)
	± 2.8% (Pu)	± 0.6% (Pu)
Pu-Al	± 12.9% (Pu)	± 0.5% (Pu)

The data illustrates the very marked advantage of accuracy available in the case of product-plus-loss determinations.

The agreement of shipper's estimates with the two chemical plant measurements were as follows:

Program	Shipper vs Feed*	Shipper vs Product and Loss*
Clementine	- 1.9 ± 2.9% (Pu)	- 1.95 ± 0.7% (Pu)
Thorex	+ 0.4 ± 6.0% (U-233)	- 5.25 ± 1.2% (U-233)
Scrup-2	- 4.2 ± 3.6% (U)	- 5.18 ± 0.7% (U)
	- 6.2 ± 2.6% (Pu)	- 6.67 ± 0.6% (Pu)
Pu-Al	- 2.9 ± 12.9% (Pu)	- 3.71 ± 0.5% (Pu)

The large deviations in the Scrup-2 and Pu-Al programs were believed due, in part, to the fact that the elements had been sheared and stored at the reactor site (arriving at ORNL covered with fragile oxide layers which flaked off during storage and dissolver loading operations).

The large deviations in the Thorex case are believed due, in part, to several poorly defined nuclear parameters involved in the Th,n irradiation process (as well as several unknowns believed to exist in the estimations of specific pile-loading characteristics).

III. Reactor Estimates Versus Chemical Plant Measurements at Other Sites

Table 3 shows comparisons of some reactor estimates and ICPP measurements for total uranium and U-235. As an example, it will be noted that good agreement (about 1.0%) was possible in the LITR case, but was somewhat poorer (~3%) in the MTR case. (Burnup calculations in the latter case differed by 9%, as discussed in section IV.)

Experience at the production sites seems to support a general contention that long-term operational knowledge of given reactors, in conjunction with appropriate analytical data from the chemical plants, can be used to establish consistently good agreement between reactor estimates and chemical plant measurements. Such evidence suggests the probability that operators of given power reactors, after experience with a few cores, can establish very accurate calculation procedures, i.e. after resolving empirical questions concerning specific nuclear and burn-out characteristics (which can be experimentally determined with the help of chemical and isotopic data on their recovered fuels).

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* Note that ± values refer to accuracy of the chemical plant measurements.

Table 3.   MTR and LITR Estimates vs ICPP Measurements

Reactor	Fuel	Reactor Estimate (grams)	Chem. Plant Meas. (grams)	Average Deviation* (%)
MTR (1956)	Uranium	21,450 ($\pm$ 2.5%)	20,875 ($\pm$ 1.1%)	+ 2.75
	U-235	18,745 ($\pm$ 3.3%)	18,177 ($\pm$ 1.1%)	+ 3.12
LITR	Uranium	13,370 ($\pm$ 3.2%)	13,287 ($\pm$ 4.5%)	+ 0.62
	U-235	11,323 ($\pm$ 3.8%)	11,194 ($\pm$ 4.5%)	+ 1.15
MTR (1954)	Uranium	51,080 (-)	50,275 (-)	+ 1.60
	U-235	45,661 (-)	44,729 (-)	+ 2.08
MTR (1958)	Uranium	40,213 (-)	39,662 (-)	+ 1.39
	U-235	. 30,194 (-)	29,761 (-)	+ 1.45
AECL**	Uranium	1,282 (-)	958 (-)	+ 33.8**
	U-235	1,068 (-)	754 (-)	+ 41.6**

* Reactor versus chemical plant.

**Major source of discrepancy is probably due to large errors in metallurgical
analysis of original booster rods shipped from Y-12 to AECL.

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Table 3a. AECL-Hanford Fuel Measurements

Shipment Number	AECL Reactor Calculations		HANFORD Measured Values		Variance (%)	
	Pounds U	Grams Pu	Pounds U	Grams Pu	U	Pu
1	1,807.0	548.6	1,770	456	- 2.1	- 20.3
2	3,730.7	869.6	3,677.0	821.3	- 1.5	- 5.9
3	5,285.1	897.3	5,292.1	852.2	+ 0.1	- 5.3
4	9,120.8	2,587.8	8,949	2,377	- 2.0	- 8.9
5	10,026.2	3,840.40	9,896	3,560.54	- 1.3	- 7.8
6	8,513.6	2,428.60	7,656	2,090	- 11.2	- 16.2
7	9,955.2	3,069.3	10,000.2	3,164.9	+ 0.45	+ 3.0
8	9,902.8	3,292.9	9,858.00	3,100.00	- 0.45	- 6.2
9	4,253.4	1,264.3	4,377.00	1,234.80	+ 2.8	- 2.4
Totals	62,594.8	18,798.8	61,475	17,656.7		
Variance			-1,120	-1,142	(-1.8%)	(-6.5%)

IV.   Accuracies and/or Uncertainties of Reactor Measurements

The accuracy of reactor measurements for accountability will be affected by a multiplicity of errors inherent in (A) values of basic physics parameters and (B) engineering measurements at the reactor. It is believed that the latter measurements are limited by the greater unknowns and will vary considerably in severity for specific power reactor cases.

(A)   Accuracy of Basic Physics Parameters

Small errors inherent in the values of nuclear parameters such as available energy per fission, isotopic cross-sections, neutrons per fission, neutrons per absorption, non-fission capture-to-fission ratios impress some limits on the accuracy of given reactor calculations of burnup, etc. However, as indicated in Tables 4, 5, 6, and 7, most of the pertinent values are accurate to about ± 1.0 to $\pm 2.0\%$; so, in themselves, these possible errors would not seem to place major limits on reactor calculations.

Though multigroup calculations for specific power reactor designs can closely evaluate a spectral index (resonance neutron density/total neutron density), it is believed that several large unknowns involving neutrons of intermediate-energy and resonance cross-sections (± 20 to $\pm 50\%$ errors) can contribute major errors in calculations, particularly in fuels containing plutonium. Table 8 indicates the order of changes in pile-neutron cross-sections in high temperature operation, e.g. where the spectral index will approach a significant value.

The Effective Cross-Section for a Reactor Spectrum

The effective cross-section for a thermal reactor can be given by either of the following relationships:

$$\hat{\sigma}_1 = \sigma_o(1)\left[g(1) + rS(1)\right] \text{ or}$$

$$\hat{\sigma}_1 = \sigma_o(1)g(1) + r\left(\sqrt{\frac{4T}{\pi T_o}}\ I'_o(1) - 1.01\ \sigma_o(1)g(1)\right)$$

where $\hat{\sigma}_1$ = effective 2200 m/sec cross-section for actual reactor specimens

$g(1)$ = non 1/v correction factor

r = spectral index or resonance neutron density/total neutron density

S = resonance integral correction factor for 1/E tail of neutron spectrum above 5 KT

$\sigma_o(1)$ = 2200 m/sec cross-section

$I_o(1)$ = effective resonance integral

S can be obtained from the above expressions, i.e.

Table 4. Values of Cross-Sections and Resonance Integral
for Important Fissile, Fertile or Heavy Isotope

U.S. Accepted Values (BNL-325)

Nuclide	σ	Reson. Integ., I_o	Comments
Th-232	a 7.33 ± 0.12	(70^1)	(Strong resonance absorber)
Pa-233	a 68 ± 6.5	700	(Strong resonance absorber)
U-233	f 527 ± 4.0 a 581 ± 7.0	805 922	(Moderate resonance absorber) (Moderate resonance absorber)
U-234	a 97 ± 5.0	700	(Strong resonance absorber)
U-235	f 582 ± 6.0 a 694 ± 8.0	271 370	(Moderate resonance absorber)
U-236	a 7 ± 2.0 eff 16 - 48	350	(Very strong resonance absorber) (Depending on pile spectrum)
U-238	a 2.71 ± 0.02	(289^1)	(Strong resonance absorber)
Pu-239	f 746 ± 8.0 a 1026 ± 13.0	319* 478*	(Strong resonance absorber and non-1/v)
Pu-240	a 250 ± 40.0	8700	(Very strong resonance absorber; non-1/v; resonance absorption depends on size of fuel element and scattering x-sections per absorbing atom)
Pu-241	f 1025 ± 10.0 a 1400 ± 80.0	1358 1869	(Strong resonance absorber)
Pu-242	a 30 ± 2.0	1015	(Strong resonance absorber)

[1] Infinite dilution resonance integral - effective resonance integral is much less than this; depends on size and spacing of fuel elements.

* Does not include 0.30 ev resonance.

NOTE: The terms strong or moderate resonance absorber refer to the comparison of relative absorption rates of resonance neutrons and thermal neutrons for the specified isotope.

Table 5. World Values of Fissile Cross-Section

Country	U-233		U-235		Pu-239	
	Fiss.	Abs.	Fiss.	Abs.	Fiss.	Abs.
U.S.	533±10	585±10	584±10	689±7	748±15	1025±15
U.K.	520±12	577±10	596±14	725±13	717±16	1006±25
USSR	525±15	590±20	570±15	695±20	715±30	1035±20
France	-	-	584±20	-	770±20	-
Canada	524±4	-	578±6	-	754±9	-
Norway	521±20	-	587±16	-	-	-
India	-	-	555±15	-	-	-
World weighted averages	525±3	582±7	579±4	697±6	747±6	1025±13
World consistent set	527±4	581±7	582±6	694±8	746±8	1026±13

Table 6. U.S. Values of η, ν, and α

(Values are for 2200 m/sec neutrons only)

Fissionable Isotope	η $\frac{\text{neuts.}}{\text{Absorp.}}$	ν $\frac{\text{neuts.}}{\text{Fiss.}}$	α $\frac{\text{capt. x-section}}{\text{Fiss. x-section}}$
U-233	2.28 ± 0.02	2.51 ± 0.03	0.102 ± 0.005
U-235	2.06 ± 0.03	2.43 ± 0.04	0.18 ± 0.01
Pu-239	2.10 ± 0.02	2.90 ± 0.04	0.38 ± 0.02

Table 7. Available Energy Per Fission*

Mev/fission	Source
192 ± 5.5	A. M. Weinberg, ORNL
192	Benedict, MIT
195	Glasstone-Edlund, ORNL
192	Arnold, ORNL
198	(Tingey) IDO

* Neutrinos not considered.

$$S = \frac{\sqrt{\frac{4T}{\pi T_o}} \; I'_o(1)}{\sigma_o(1)} - 1.01 \; g(1)$$

For a reactor having a spectral index of 0.08 and operating at a neutron temperature of 600°K, the effective 2200 m/sec cross-section for U-236 can be calculated as

$$
\begin{aligned}
\hat{\sigma}_{26} &= 7 \times 1 + 0.08 \; (1.6147 \times 350 - 1.01 \times 7 \times 1) \\
&= 7 + 0.08 \; (565.1 - 7.1) \\
&= 7 + 0.08 \times 558 = 7 + 44.6 = 51.6 \text{ barns}
\end{aligned}
$$

Thus the effective U-236 cross-section would be 7.3 times the 2200 m/sec value.

The cross-sections shown in Table 8 were calculated by E. D. Arnold for hypothetical plutonium and U-233 recycle reactors having a spectral index r of 0.09 and a neutron temperature of 600°K. The right hand column lists the accepted U.S. 2200 m/sec values and the percent difference between the "pile" cross-section and the 2200 m/sec value. These effective pile cross-sections indicate the effects of increased temperature and resonance neutron density. Some typical values of spectral index for known reactors are: NRX (0.07), Calder Hall (0.075), MTR (0.083). A value of 0.09 was used for the listed cases as typical for a H_2O moderated, close packed, high temperature power reactor.

The value of the effective resonance integral cannot be obtained directly from thin foil measurements. A thin foil measurement will give what is called an infinitely dilute resonance integral. However, the effective resonance integral must be reduced below the infinitely dilute resonance value by a factor relating self shielding and Doppler broadening effects. For example the effective resonance integral for U-238 for a 0.6 in. diameter uranium rod might be 12-15 barns as compared to an infinitely dilute value of 281 barns. Also, the effective resonance integral for something like Pu-240 $(I_o = 8700b \; (\infty))$ will vary with its concentration by the relationship

$$I'_o(1) = \frac{I_o(1)}{1 + \frac{I_o(1) \; \frac{N_i}{N_G}}{\sigma_s}}$$

where N_i/N_G = concentration of isotope i in fertile material G

σ_s = energy independent scattering cross-section for fertile material matrix

As an example for the case of 1% Pu-240 in $U^{238}O_2$ with σ_s = 20 barns

$$I'_o(40) = \frac{8700}{1 + \frac{8700 \times 0.01}{20}} = \frac{8700}{5.35} = 1626 \text{ barns}$$

Table 8. Effective Cross-Sections for Recycle Reactors

Isotope	Effective 2200 m/sec x-Section for Pile Neutrons $T = 600^{O}K, \ r = 0.09*$	Accepted 2200 m/sec (Pure Thermal) x-Section $T = 293^{O}K, \quad r = 0$	% Difference
U-233 σ_f	618	527	+ 17.3
σ_c	67	54	+ 24
σ_a	685	581	+ 18
U-234 σ_a	188	97	+ 94
U-235 σ_f	539	582	- 7.5
σ_c	111	112	- 0.9
σ_a	650	694	- 6.4
U-236 σ_a	56	7	+ 700
Pu-239 σ_f	1192	746	+ 60
σ_c	659	280	+ 135
σ_a	1851	1026	+ 80.4
Pu-240 σ_a	839	250	+ 236
Pu-241 σ_f	1369	1025	+ 33.5
σ_c	526	375	+ 44
σ_a	1895	1400	+ 35.3
Pu-242 σ_a	129	30	+ 330

* r = spectral index = resonance neutron density/total neutron density.

Thus the effective resonance integral for Pu-240 will vary over a wide range during reactor operation from a starting value of 8700 barns to 1600 barns or less for the case of plutonium recycle.

Estimated Error in Effective Pile Cross-Section

Using the expression for effective pile cross-section of

$$\hat{\sigma} = \sigma_o g + r\left(\sqrt{\frac{4T}{\pi T_o}}\ I'_o - 1.01\ \sigma_o g\right),$$ it can be shown that the square

of the error in $\hat{\sigma}$ $(\Delta\hat{\sigma})^2$ can be obtained from an expression of the form

$$(\Delta q)^2 = \left(\frac{\partial F}{\partial x}\right)^2 (\Delta x)^2 + \left(\frac{\partial F}{\partial y}\right)^2 (\Delta y)^2 + \left(\frac{\partial F}{\partial z}\right)^2 (\Delta z)^2$$

where cross-product effects are neglected, and where $q = F(x,y,z)$ and Δx, Δy, Δz are errors in x, y, and z, respectively. Using the previous expression for $\hat{\sigma}$, the error in $\hat{\sigma}$ becomes

$$(\Delta\hat{\sigma})^2 = (g - 1.01\ rg)^2 (\Delta\sigma_o)^2 + (\sigma_o - 1.01\ r\ \sigma_o)^2 (\Delta g)^2$$

$$+ \left(\sqrt{\frac{4T}{\pi T_o}} - 1.01\ \sigma_o g\right)^2 (\Delta r)^2 + \left(\sqrt{\frac{4T}{\pi T_o}}\ r\right)^2 (\Delta I_o)^2$$

As an example, consider the following hypothetical case:

$$\sqrt{\frac{4T}{\pi T_o}} = 1.60$$

$\sigma_o = 400b$	$\Delta\sigma_o = \pm 10b$
$g = 1.05$	$\Delta g = \pm 0.02$
$r = 0.10$	$\Delta r = \pm 0.05$
$I'_o = 800b$	$\Delta I'_o = \pm 200b$

then

$$(\Delta\hat{\sigma})^2 = (1.05 - 0.106)^2 (10)^2 + (400 - 40.4)^2 (0.02)^2$$

$$+ (1280 - 424)^2 (0.05)^2 + (0.16)^2 (200)^2$$

$$= (0.944)^2 (100) + (359.6)^2 (0.0004) + (856)^2 (0.0025)$$

$$+ 0.0256(40,000)$$

$$= \quad 89.1 \qquad + 51.7 \qquad + 1831.8 \qquad + 1024$$

$$\text{Due to } \Delta\sigma_o \quad \text{Due to } \Delta g \quad \text{Due to } \Delta r \quad \text{Due to } \Delta I'_o$$

$$(\Delta\hat{\sigma})^2 = 2996.6$$

$$\Delta\hat{\sigma} = \pm 54.8$$

The direct calculation of σ gives

$$\hat{\sigma} = 505.6$$

Thus the expected value σ with its deviation would be

$$\hat{\sigma} = 505.6 \pm 54.8b$$

i.e. an error of $\pm 10.8\%$.

B. Engineering Measurements at the Reactor

The measurement of reactor coolant temperatures is quite accurate; however the measurement of coolant flow-rates (for obtaining energy balances) will be subject to large limits of error, i.e. in the order of ± 5 to $\pm 10\%$ (the error increases with high flow-rates). Experience in the MTR operation is discussed below:

Accuracies of Heat Measurement in MTR

The only direct measurement used at the MTR is gross flow of primary coolant and temperature differential.

The error in the differential temperature measurement is believed to be $\pm 0.5^{0}F$.

The error in the gross flow at 40 MW operation (at a flow of about 22,000 gpm) is $+ 1700$ to $- 1600$ gpm, or approximately $\pm 8\%$.

The total burnup computation error by this method is $\pm 9\%$. An example is given in Table 9.

A large part of the discrepancy between the MTR measurement of U-235 burnup and the chemical plant analysis is probably due to the use of 198 Mev/fission instead of the more preferred value of 192 Mev. (See Table 7.)

V. Determination of Fissionable and Fertile Materials in Fuel Elements by Reactor Operator

Argonne has studied two methods for determining fissionable material content in fuel elements prepared by co-extrusion of an aluminum and uranium oxide core and an aluminum cladding. (It is important, for purposes of calculating reactivity of reactors, that the total uranium and its distribution in each element be accurately measured.)

One method involved the transmission of collimated 84 Kev gamma rays (from thulium-170) through the fuel element to scan and measure transmitted radiation in energized CdS or NaI crystal detectors. Accuracies of $\pm 2\%$ were reported.

Table 9. Fuel Content Following Irradiation in MTR

Starting Material		MTR Calc.		ICPP Meas.	
SS (Net)	U-235	SS (Net)	U-235	SS (Net)	U-235
26,429±300	24,656±300	21,450±538	18,745±611[1] (Calc. burnup of 5,911±529)	20,875±221	18,177±193 (Meas. burnup of 6,479±356)*

(Variation of averages = - 8.77%)

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* $E_o U_o - E_1 U_1$ = measured burnup obtained by total U content and U-235 assay before and after irradiation.

[1] Includes uncertainties in fuel fabricator's values.

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The other method, tested on elements of nominal 20% enrichment, employed NaI crystal detection of the 184 Kev gamma rays from the U-235 to scan the elements. Accuracies of $\pm$ 1% were reported. It was felt that no limitations prevent even greater accuracy where scanning time is not an important consideration.

Savannah River has employed a method similar to the latter for scanning of billets and report accuracies of about $\pm$ 0.2%.

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APPENDIX A

Analytical Methods Used in Pilot-Plant Programs for Accountability Work

(Supplied by L. T. Corbin)

The following is a list of analytical methods used in the pilot plant for accountability work and precision of each method.

I.   Uranium, Spectrophotometric Ammonium Thiocyanate Method - $\pm$ 4%

II.   Uranium, Fluorometric Method - $\pm$ 10%

III. Uranium-233, Hexone-Extraction Method
      Dissolver solution $\pm$ 4%
      Waste solution $\pm$ 10%

IV.   Plutonium-Thenoyltrifluoroacetone Extraction Method
      Dissolver solution $\pm$ 2.5%
      Waste solution $\pm$ 15%

V.   Thorium-Spectrophotometric-Thoron Method - $\pm$ 2%

These precision figures are limit of error of one determination based on 95% probability level with no bias existing.

The above figures are on radioactive samples. In the near future we hope to be determining uranium in dissolver solutions by the Coulometric Method with a precision of $\pm$ 1%.

Product methods are as follows:

I.   Uranium - Automatic Potentiometric Ferric Sulfate Method - $\pm$ 0.2%

II.   Plutonium - Potentiometric Ceric Sulfate Method - $\pm$ 0.5%

(A copy of each of the above methods will be made available for the discussions with AEC personnel.)

All mass assay work is done at Y-12.

I.   Uranium - natural - 5 percent enriched $\pm$ 0.01 - 0.001% of the ratios working against standards or 0.1 - 0.01% if by direct measurements.

II.   Plutonium - 0.5 - 1.0% between ratios.

For the thermal emission method of assay, the L. E. for a single determination of the isotopic analysis of 80-100% range is $\pm$ 0.25%; as compared to the L. E. of $\pm$ 0.08% for a galvanometer determination of the same material in the form of a gas, that is, uranium hexafluoride. A breakdown on the thermal assay is as follows:

Range	% L. E.$_{(0.5)}$ (Individual)
80 - 100	0.25%
10 - 80	1.0%
1 - 10	5.%
0.1 - 1.0	14.0%
<.1	22.%

On the mean of different loadings of the same sample the numbers are improved by about % L.E/1.4. The internal precision on a single instrument is usually much better than this but these values should encompass all conditions.

APPENDIX B

Measurements Evaluation Program
Reactor Calculations and Reactor Source Data Requirements

(Letter, R. J. Dickeman, HAPO
to J. T. Christy, AEC, HOO.)

Necessary Reactor Data

The differential equations representing plutonium buildup and various decay schemes existing within an operating reactor can be readily derived. The various reactor parameters such as, resonance escape probability, fast effect, and age, that appear in the solutions of these equations, can be calculated to within ± 1% by present analytical methods for Hanford type reactors. These analytical methods have not as yet been checked for other type reactors but the expected accuracy should be comparable to that for the Hanford type reactors with the one possible exception, the age calculation. However, the prediction accuracy is fairly insensitive to the age value. These reactor parameters will be of little concern to the reactor operators; however, the operators will be directly concerned with the independent variable in the solutions, i.e., the number of fissions that have taken place within the irradiated fuel. Therefore, it will be necessary that the reactor operators maintain an accurate system for determining the number of fissions that have occurred in the irradiated fuel. The most commonly utilized method for determining the total number of fissions that have taken place is to measure the amount of energy released in form of heat to the reactor system.

The exposure of metal in individual fuel channels can be readily determined by measuring the coolant flow and the input and output coolant temperatures for the individual channels. The heat transferred to auxiliary components, such as the shield and control rods, is added to the metal exposures and the contribution to each channel is determined from the power factor weighting. It will be necessary that the reactor operators maintain an accurate record of the heat output of each channel and heat transferred to the other reactor components.

Monitoring of each channel will not be possible for reactors in which the heat energy is transferred directly to a circulating moderator. The total number of fissions can then be determined from the total heat transferred to the moderator and other components. Since the fission events do not occur uniformally over the reactor core, and the plutonium buildup and uranium burnout expressions are not linear with exposure, a good deal of bias will be introduced in the predictions for moderator cooled reactors. To predict the bias in the calculations due to this effect would involve an accurate knowledge of the neutron flux distriubtion. These distributions can be roughly estimated, but would certainly introduce an additional source of error in the calculations.

Estimated Accuracy of Reactor Calculations

It is Hanford's opinion that the determination, based on reactor calculations and precise exposure data, of the SSNM content of the first material discharged from a power reactor will be accurate to about $\pm$ 10% for plutonium and $\pm$ 4% for uranium-235. However, it is expected that the above uncertainties will become somewhat smaller after experience with a particular reactor is obtained. It is expected that calculated values will eventually be accurate to about 3% for plutonium and about 1% for uranium-235. Also, the predictions for first discharge material will become more accurate as experience with long irradiated, > 5,000 MWD/Ton, materials is gained.

It has been the experience at Hanford that the calculated values agree on a long term average with the chemical measurements within 1-2% for plutonium and about 1% of uranium-235. However, individual batch values may differ by as much as 5-10%. The uncertainties associated with assigning exposure values to specific batches of metal limit the accuracy of the calculations rather than the accuracy of the basic equations.

Uncertainties in calculating the isotopic content, % Pu-240 and Pu-241, of the total amount of plutonium will be large compared to the uncertainties in predicting the total quantity of plutonium. The isotopic content of plutonium is highly sensitive to the neutron temperature and the self shielding of Pu-240 on the effective cross section of Pu-240; whereas the total plutonium quantity is not as sensitive to these effects. However, studies are presently being made to develop analytical methods for accurately predicting the isotopic content of plutonium vs exposure. Certainly a good deal of progress will be made in the near future since the performance of the power reactors themselves depend on the buildup of the various isotopes.

APPENDIX C

Proposed Basis for Payment

(Letter, C. E. Center, UCC, to
S. R. Sapirie, Oct. 2, 1958.)

It might be desirable and necessary to use the several bases (i.e., reactor calculations, hot feed analysis, product-plus-loss determination) proposed for payment for each fuel in the following manner:

(a) Provisional payment of 90% of total fuel value, based on "certified" reactor records, presuming an approved recording and calculation procedure for reactor performance.

(b) Negotiated settlement of 5% of the fuel value based on dissolver (or other designated measuring points) analyses if appropriate.

(c) Final settlement based on material balance or product-plus-loss determination.

(d) Final payment for fuels that will be stored for an extended period of time, such as three years or longer, could be made on the basis of reactor calculations.

(e) Final payment for fuels that will be dissolved and blended with other reactors could be made on dissolver analyses.

At Oak Ridge, where we will be processing a variety of fuels in relatively small batches, it is probable that we will never develop our measurements and our over-all accountability to a high confidence level. Each reactor core and blanket will differ, at least for a few years, from the previous one because of the developmental nature of power reactors; the reprocessing procedures will all be new, so new that the first few years must be regarded as a development period.

For these reasons we suggest that care be used in the designation of accuracy, precision, and confidence levels in the preparation of processing contracts. Perhaps these might be renegotiated periodically as experience is accumulated.
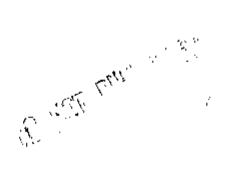
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