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THE SAFEGUARDS VALUE OF ISOTOPIC CORRELATIONS

MASTER

By

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I. INTRODUCTION

The concept of using isotopic correlations as a means of safeguards verification and confirmation has evolved over a number of years. Historical applications at the Hanford Project have included their use in cross-checking analytical measurements made on spent fuel samples and in checking calculated values for spent fuel compositions and exposures. In recent years, further development of the concept has been sponsored by the Atomic Energy Commission's Office of Safeguards and Materials Management and by the U. S. Arms Control and Disarmament Agency (ACDA). Research sponsored by ACDA on isotopic correlations is included under the programmatic title of Minor Isotopes Safeguards Techniques which is denoted by the acronym MIST. The MIST program embodies the use of isotopic data for safeguards purposes at all parts of the fuel cycle. By contrast, the isotopic correlation techniques discussed in this report are concerned primarily with that part of the fuel cycle which extends from the end of fuel fabrication through the input stage of chemical processing.

A. Technical Basis

The potential safeguards value of isotopic correlations arises from the inherent relationships between the formation of Pu and the growth and depletion of the U and Pu isotopes during the irradiation process. As a result of the underlying interdependence provided by these relationships, the isotopic composition data tend to be internally self-consistent and at the end of irradiation, the isotopic compositions of U and Pu give testimony to the Pu content and confirm previous safeguards information about the fuel.

The key point for the application of these principles is at the input accountability tank at the chemical processing plant. Here, in many current processes, the unaltered ratios of the important nuclides are present in the dissolver solution. Current practice for input accountability in reprocessing spent power reactor fuel is to measure the total U, total Pu, and the corresponding U and Pu isotopic compositions for each batch. From data previously obtained on slightly enriched U fuels, the following empirical generalizations are apparent.

1. The integrating effect of dissolving whole rods and bundles provides an unusually high degree of consistency in the burnup relationships when viewing data from chemical plant input batches of the same spent fuel.
2. Spent fuel data from the same reactor, having the same design and initial enrichment, portray sets of burnup relationships which trace smooth curves over a wide range of exposure. In addition, some of the relationships between Pu content and the isotopic compositions are linear and have a consistency equivalent to the measurability of the relationship.

From that evidence, several potentially valuable safeguards applications were recognized by workers in both Europe⁽³⁾ and the U. S.^(2,9). These are discussed subsequently in sub-topics B, C, and D.

B. Verification of the Pu Content of Spent Fuels

It was recognized that the correlation relationships may be used to verify the Pu content of spent fuels in two main ways. The first approach, the historical method, is based simply on accumulating the data (Pu/U ratios

and U and Pu isotopic compositions on dissolved batches of spent fuels) for a particular reactor and fuel design, developing the correlations and then applying the correlations to future similar spent fuels. It was also recognized that a chemical plant operator, a central safeguards agency, and a reactor operator could use the historical method as a cross check on input measurements of batches of similar fuel.

A second method of potential application is to use the correlation relationships as a means of verifying those input batches which are measured only by the plant. That is to say the agency or the verification agent establishes relationships between the variables by using data from a few batches measured independently. The plant data on other batches, should, within the limits of statistical error, then bear the same relationships. In practice, both methods could be used simultaneously in that the verification agency could always make some redundant measurements but could vary the intensity of independent measurement with the nature and extent of the previously established data base. Again, it was recognized that the facility operator could also use the isotopic correlations to test the consistency of measurements made on a series of input batches.

C. Confirmation of Previous Safeguards Information

The continuous nature of the burnup paths and the feature that they are characteristic of spent fuel from a given reactor type can be used to confirm previous safeguards information. Typical applications would include confirmation of the initial enrichment, the reactor type, and the exposure. When extensive historical data are available, the confirmation may be extended to include reported changes in fuel design and reactor operating conditions.

D. Improving the Theoretical Base

The data, with these evident, inherent regularities, also offer the physicist a place where the reactor prediction equations can be checked both as to the kind of relationship observed and as to the actual amounts which are observed. Indeed the data may be useful for improving the estimates of constants (parameters) which are involved in the equations. For example the fact that $k_5 = (Pu/U)/(^{235}U \text{ depletion})$ values for a given design and enrichment are invariant on a batch basis over the range of integrated exposures in a core may be useful for understanding the relative importance in application of the different parts of the theoretical relationships.

E. Scope of Paper

This report is a follow on to a number of previous reports. Two papers^(2,6) on this subject were presented at last years meeting of the Institute. The intent of this paper is to 1) summarize the work completed to date in this area and 2) to use this base to illustrate the potential safeguards value of isotopic correlations to the various parties concerned.

II. SUMMARY

During the past year a broader experimental and theoretical base has been established for safeguards applications of isotopic correlation techniques. As a consequence, it is now possible to illustrate more clearly potential applications of the concept and also to define more exactly its quantitative aspects and limitations.

In the main body of the report potential safeguards uses of the isotopic correlation technique are discussed in some detail. Safeguards value is illustrated for three separate users and four separate objectives. Potential

users include:

1. A central safeguards agency;
2. A reactor operator or his agents, and
3. A chemical processing plant operator.

The four safeguards objectives discussed in the main body of the report are:

1. verification of the plutonium content of spent fuel,
2. confirmation of previous safeguards information,
3. cross-checking of analytical measurements made on dissolver solution,
4. and improvement of the theoretical base for reactor calculations.

These subjects are discussed separately in the subsequent sections of this report. Each application is illustrated with experimental data. Also, the statistical and theoretical basis for the applications are also included, when appropriate.

III. ILLUSTRATIONS OF POTENTIAL APPLICATIONS

A. Description of the Data

Much of the data which are used in the examples of potential applications came from Yankee Cores V and VI. "A total of 74 Yankee fuel assemblies, 2 discharged with Core IV, 36 from Core V and 36 from Core VI were reprocessed by NFS during the period May through July, 1969. Forty of the assemblies were initially 4.1 weight percent ^{235}U enriched uranium fuel (hereafter referred to as 4.1 fuel assemblies), 32 were initially 4.94 weight percent ^{235}U enriched uranium fuel (4.94 fuel assemblies) and 2 were initially 2.9 weight percent ^{235}U enriched uranium fuel (2.9 fuel assemblies). The 2.9 fuel assemblies differed from the other assemblies in that they were zircaloy clad instead of stainless steel clad. The 2.9 fuel assemblies replaced two-4.1 fuel assemblies in core V, and although the displaced 4.1 assemblies were discharged with Core IV, they were Core V material.

In past dissolution campaigns of spent reactor fuels, fuel assemblies from a given core have not been dissolved in any predetermined order. A unique feature of the dissolution of Yankee Cores V and VI was that the order of processing of the fuel was determined before the actual campaign began. Only assemblies of the same initial enrichment were grouped together in making up a batch. The estimated exposure ranged from approximately 9700 to 27,000 MWD/MTU*, and each dissolution batch contained fuel assemblies having approximately the same total exposure."(5)

1. An Example of Isotopic Production Data

The data in Tables 1.1 and 1.2 come from NFS production data in the processing of Yankee Rowe, Cores V and VI material. (5)

* Megawatt Days/Metric Ton (tonne) Uranium

Table 1.1

Input and Product Uranium Accountability Measurements

INPUT ACCOUNTABILITY DATA (24)							
NORMAL PLANT MEASUREMENTS							
Sample Number	Enrichment*	U ISOTOPES*				U (g/l)	Total U (grams)
		234	235	236	238		
1	4.1	0.0214	3.1755	0.2209	96.5823	246.48	533370.
2	2.9	0.0197	1.8690	0.2188	97.8925	112.15	467802.
3	4.94	0.0131	3.1750	0.4250	96.3869	179.90	796337.
4	4.1	0.0184	2.5136	0.3590	97.1090	222.64	1081296.
5	4.94	0.0130	2.8373	0.4990	96.6508	182.50	787352.
6	4.1	0.0187	2.6748	0.3253	96.9811	236.78	1056390.
7	4.94	0.0130	2.9782	0.4682	96.5407	178.72	789877.
8	4.1	0.0189	2.6388	0.3313	97.0109	237.37	1054889.
9	4.94	0.0126	2.9943	0.4599	96.5333	188.23	793613.
10	4.1	0.0187	2.6359	0.3282	97.0171	233.66	1065294.
11	4.94	0.0124	2.9734	0.4652	96.5490	167.82	788115.
12	4.1	0.0179	2.4754	0.3601	97.1466	237.57	1059897.
13	4.94	0.0124	2.7889	0.5043	96.6945	178.08	789749.
14	4.1	0.0187	2.6523	0.3296	96.9994	236.86	1055114.
15	4.94	0.0124	2.8324	0.4968	96.6584	190.08	797490.
16	4.1	0.0163	2.0467	0.4454	97.4916	225.25	1053376.
17	4.94	0.0118	3.0057	0.4598	96.5227	181.53	792471.
18	4.1	0.0175	2.2545	0.3993	97.3287	225.00	1045784.
19	4.94	0.0122	2.9907	0.4596	96.5376	175.30	796311.
20	4.1	0.0201	2.7958	0.2999	96.8843	244.81	1047457.
21	4.94	0.0124	2.8203	0.4946	96.6727	183.23	799544.
22	4.1	0.0180	2.4709	0.3589	97.1522	175.32	531435.
24	4.94	0.0131	3.0868	0.4430	96.4571	164.15	536060.
26	-	0.0130	3.0595	0.4437	96.4840		1388.
						Total	19530411
URANIUM PRODUCT DATA (5)							
1		0.0165	2.7377	0.3619	96.8839		4237000.
2		0.0168	2.8262	0.3908	96.7617		4113000.
3		0.0170	2.6590	0.4120	96.913		4284000.
4		0.0150	2.6790	0.4180	96.888		3838000.
5		0.0150	2.6790	0.4180	96.888		2779000.
						Total	19251000.

*wt %

Table 1.2

Input and Product Plutonium Accountability Measurements

NORMAL PLANT MEASUREMENT								
Sample Number	Enrichment*	Pu Isotopes*					Pu (g/g)	Total Pu (grams)
		238	239	240	241	242		
1	4.1	0.2890	85.0504	10.2940	4.0105	0.3561	1.222	2650.
2	2.9	0.4132	77.0680	14.8107	6.6196	1.0886	0.668	2787.
3	4.94	0.7263	76.9673	13.1553	8.0899	1.0612	1.551	6842.
4	4.1	0.6701	75.6664	14.2846	8.1138	1.2651	1.928	9338.
5	4.94	1.0372	72.9158	14.6825	9.7586	1.6059	1.905	8220.
6	4.1	0.5770	77.9612	13.2569	7.2199	0.9850	1.838	8199.
7	4.94	0.9674	74.5809	14.0261	9.0534	1.3722	1.756	7762.
8	4.1	0.6162	77.5050	13.4530	7.3824	1.0434	1.895	8420.
9	4.94	0.8561	75.1180	13.8339	8.8894	1.3024	1.827	7697.
10	4.1	0.5419	77.5738	13.4712	7.3680	1.0451	1.846	8448.
11	4.94	0.8865	74.7847	13.9596	9.0369	1.3323	1.650	7752.
12	4.1	0.6909	75.2167	14.5248	8.2264	1.3412	2.076	9261.
13	4.94	1.0769	72.5647	14.8030	9.8849	1.6705	1.954	8672.
14	4.1	0.5385	77.7337	13.4348	7.2783	1.0148	1.873	8446.
15	4.94	1.0239	72.9417	14.6952	9.7332	1.6060	1.986	8341.
16	4.1	1.2277	69.3089	16.3367	10.8526	2.2741	2.509	11712.
17	4.94	0.8486	75.0776	13.8645	8.9127	1.2966	1.758	7672.
18	4.1	0.9210	72.6915	15.4591	9.1778	1.7506	2.204	10242.
19	4.94	0.8592	75.0471	13.8537	8.9315	1.3086	1.709	1161.
20	4.1	0.6838	79.3513	12.5320	6.6510	0.7819	1.739	7441.
21	4.94	1.0391	72.8453	14.7559	9.7289	1.6308	1.929	8419.
22	4.1	0.6940	75.1394	14.5352	8.2871	1.3442	1.555	4710.
24	4.94	0.7886	75.9508	13.5383	8.4845	1.2378	1.493	4875.
26	-	0.7888	75.8790	13.619	8.5181	1.195	-	13.
							Total	175686
PLUTONIUM PRODUCT DATA (g)								
1		0.685	76.804	13.656	7.737	1.118		18678
2		0.705	76.209	13.775	8.121	1.190		21435
3		0.728	76.257	13.734	8.103	1.177		26533
4		0.775	75.216	14.193	8.490	1.326		24418
5		0.949	73.273	14.195	9.360	1.624		24605
6		0.931	73.267	14.902	9.273	1.626		19317
7		0.769	75.802	13.862	8.301	1.266		19955
8		0.838	74.664	14.279	8.825	1.394		19536
							Total	17477

*wt%

2. R_5 and R_6

Table 2 is derived from Table 1 and gives the values of R_5 and R_6 where

$$R_5 = (Pu/U) / ({}^{235}U_0 - {}^{235}U_f) = - (Pu/U) / \Delta {}^{235}U$$

$$R_6 = (Pu/U) / ({}^{236}U_f - {}^{236}U_0) = (Pu/U) / \Delta {}^{236}U$$

${}^{235}U_f$, and ${}^{236}U_f$ are the final weight percent values

and

${}^{235}U_0$, and ${}^{236}U_0$ are the initial weight percent values.

Then $-\Delta {}^{235}U = {}^{235}U_0 - {}^{235}U_f$ is the ${}^{235}U$ depletion and $\Delta {}^{236}U$ is the increase in the weight percent of ${}^{236}U$.

Table 2

 R_5 and R_6 Values From Yankee Rowe Cores V and VI

Batch	Enrichment	R_5	R_6
2	2.9	5778	29522
1	4.1	5371	27463
4	4.1	5444	27072
6	4.1	5446	27204
8	4.1	5463	27401
10	4.1	5417	27516
12	4.1	5378	27297
14	4.1	5377	27381
16	4.1	5413	27427
18	4.1	5306	27258
20	4.1	5448	27334
22	4.1	5441	27792
3	4.94	4868	23284
5	4.94	4965	23567
7	4.94	5009	23840
9	4.94	4985	24013
11	4.94	5002	24037
13	4.94	5105	24494
15	4.94	4963	23728
17	4.94	5005	23975
19	4.94	5004	24167
21	4.94	4968	24008
24	4.94	4907	23499

Some statistics pertinent to the 4.1 enrichment and 4.94 enrichment data are given in Table 3.

Table 3
Statistical Summary of R_5 and R_6 Results

	R_5		R_6	
Enrichment	4.1	4.94	4.1	4.94
Average	5419	4980	27377	23874
Standard Dev.	49.1	60.5	186.6	339.1
S.D. of Ave.	14.8	18.2	56.2	102.2
% S.D.	0.91%	1.21%	0.68%	1.42%
% S.D. of Ave.	0.25%	0.37%	0.21%	0.43%
95% CL on Ave.	± 33.0	± 73.5	± 125.2	± 227.7
95% CL in %	$\pm 0.56\%$	$\pm 0.82\%$	$\pm 0.47\%$	$\pm 0.96\%$

S.D. means standard deviation

CL means confidence limits

These results indicate the stability of the R_5 and R_6 ratios. For example with 4.1 enrichment R_5 values roughly 95% of the individual values will fall within $2(0.91)\% = 1.8\%$ of the mean.

3. Graphical Presentation of the Data Illustrating the Observed Relationships

Graphs 1-7 are used to illustrate some of the relationships which have been observed from a separations facility's accountability measurements.

The data are based on Yankee Core V and VI results as determined by the NFS separations facility.

<u>Graph</u>	<u>Dependent Variable</u>	<u>Independent Variable</u>
1	Pu/U	^{235}U
2	Pu/U	^{236}U
3	^{235}U	^{236}U
4	Pu/U	$^{240}Pu/^{239}Pu$
5	$^{240}Pu/^{239}Pu$	^{235}U
6	$^{240}Pu/^{239}Pu$	^{236}U
7	$^{240}Pu/^{239}Pu$	$^{236}U/^{235}U$

GRAPH 1

YANKEE DATA-CORES V AND VI

Pu/U VERSUS $^{235}\text{U}_f$

GRAPH 2

YANKEE DATA-CORES V AND VI

Pu/U VERSUS $^{236}\text{U}_f$

GRAPH 3

YANKEE DATA-CORES V AND VI

 $^{235}\text{U}_f$ VERSUS $^{236}\text{U}_f$

GRAPH 4

YANKEE DATA-CORES V AND VI

Pu/U VERSUS $^{240}\text{Pu} / ^{239}\text{Pu}$

GRAPH 5

YANKEE DATA-CORES V AND VI

 $^{240}\text{Pu} / ^{239}\text{Pu}$ VERSUS $^{235}\text{U}_f$

GRAPH 6

YANKEE DATA-CORES V AND VI

 $^{240}\text{Pu} / ^{239}\text{Pu}$ VERSUS $^{236}\text{U}_f$

GRAPH 7

YANKEE DATA - CORES V AND VI

 $^{240}\text{Pu} / ^{239}\text{Pu}$ VERSUS $^{236}\text{U}_f / ^{235}\text{U}_f$

B. Fulfillment of Objectives

1. Verification of the Plutonium Content of Spent Fuels

The isotopic correlations may be used to verify the plutonium concentration (Pu/U ratio) of spent fuels in several ways and by several organizations.

As was noted earlier, the technical basis for verification stems from the inherent consistency of the burnup relationships for spent, low enriched uranium fuels. This phenomenon is such that batches of chemical plant dissolver solution derived from similar spent fuel (same reactor, fuel design, initial composition, etc.) have the same relationships. That is burnup paths are smooth curves or linear. Several correlations involving the Pu/U ratio are fairly constant over a wide range of exposure. These include the terms:

$$\frac{\text{Pu/U}}{\Delta^{235}\text{U}}, \quad \frac{\text{Pu}}{\Delta^{236}\text{U}}, \quad \frac{\text{Pu/U} + \text{K}}{^{240}\text{Pu}/^{239}\text{Pu}}$$

The time frame for such applications may be equivalent to only a few dissolution batches or it may extend over a series of reactor lots dissolved over a period of years. These principles are illustrated for PWR fuel from the same core and same initial enrichment (Yankee Rowe Core V) in Table 4. The upper part of the table shows the raw data and the lower part shows the consistency of the key correlations for the Pu/U ratio.

Consistency Test

Verification of the Pu/U ratio by the correlation concept may be carried out with various degrees of rigor. The first test is simply to test form the relationships from the raw data and to test for consistency. The raw data for end-of-life shown in Table 4 and initial values (initial $^{235}\text{U} = 4.1$ wt. %) are used to form the correlations shown in the lower part of the Table.

Table 4

Example of Pu/U and Isotopic
Consistency Yankee Rowe Core V Data

Raw Data

Input Batch #	Total Pu g/tu	<u>Uranium Isotopics</u>		<u>Plutonium Isotopics</u>			
		^{235}U W/O	^{236}U W/O	^{239}Pu W/O	^{240}Pu W/O	^{241}Pu W/O	^{242}Pu W/O
1	4968	3.176	0.2209	85.05	10.29	4.01	0.36
20	7104	2.796	0.2997	79.35	12.53	6.65	0.78
10	7930	2.675	0.3253	77.57	13.47	7.37	1.04
22	8863	2.471	0.3589	75.14	14.54	8.29	1.34
18	9794	2.254	0.3993	72.69	15.46	9.18	1.75
16	11119	2.046	0.4454	69.31	16.34	10.85	2.50

Pu/U - Isotopic Correlations

Input Batch #	Total Pu g/tu	$\frac{\text{Pu/U}}{^{235}\text{U}}$	% Dev.	$\frac{\text{Pu/U}}{^{236}\text{U}}$	% Dev.	$\frac{\text{Pu/U} + \text{K}}{^{240}\text{Pu}/^{239}\text{Pu}}$	% Dev.
1	4968	5371	-0.52	27463	-0.01	54078	-0.32
20	7104	5448	+0.91	27334	-0.48	55026	+1.43
10	7930	5417	+0.33	27516	+0.19	54820	+1.05
22	8863	5441	+0.78	27792	+1.19	54054	-0.36
18	9794	5306	-1.72	27258	-0.75	53565	-1.27
16	11119	5413	+0.26	27427	-0.14	53970	-0.52
		$\bar{X} = 5399$		$\bar{X} = 27465$		$\bar{X} = 54252$	
		$\sigma = 0.98\%$		$\sigma = 0.67\%$		$\sigma = 1.03\%$	

Any batch statistically different from the majority is then marked for investigation, e.g. remeasurement, check supporting data, etc.

Comparison to Independent Measurement

A more rigorous test is to test both for consistency and bias. In this case, the verification group would independently measure a few batches and compare the relationships obtained by independent measurement with those measured by the plant.

Comparison to Historical Data

Once sufficient data has been accumulated on similar fuel from the same reactor, the reported values for the Pu/U ratio and isotopic compositions can be compared to historical values through the correlations. Correlations formed on current data are compared to correlations formed from past data. In this case the mean values ($\bar{X}$'s) shown in the bottom of Table 4 could be used to test future measurements of the same material e.g. more similar fuel from Yankee Core V.

When the material from the same reactor is irradiated under the same conditions and the difference between cores lies only in the initial enrichment, similar checks can be made. Once the change in the relationships with initial enrichment is established, the relationships for any future material which differs only in initial enrichment can be predicted. The effectiveness of this approach is shown in Table 5. In this table, an equation derived from experimental data is used to predict the relationships between Pu/U and $\Delta^{235}\text{U}$. The comparisons made in the table illustrate how such a technique might work once such an equation were known. As the data shows the check is good to one percent or better.

Table 5

Comparison of Plutonium Results by
Empirical ^{235}U Depletion Method to Measured Values

<u>NFS</u> <u>Campaign</u>	$^{235}\text{U}_0$ <u>Wt. %</u>	<u>D</u>	<u>Grams</u> <u>Pu/Tonne U</u>	<u>Empirical</u> <u>Method</u> <u>Grams</u> <u>Pu/Tonne U</u>	<u>Measured</u> <u>Grams</u> <u>Pu/Tonne U</u>	<u>%</u> <u>Difference</u>
5-1	3.4042	0.8028	5723	4595	4552	+0.94
5-2	3.4058	1.0237	5722	5858	5922	-1.08
5-3	4.0278	1.4414	5425	7819	7812	+0.09
13	4.4315	1.7239	5232	9019	8996	+0.26
All Campaigns To Date - Weighted Average				6651	6650	+0.02

Grams Pu/Tonne
D Obtained By Equation $\frac{\text{g Pu/TU}}{\text{D}} = 7351 - 478.2 (^{235}\text{U}_0)$

In practice the verification technique can involve all three variations - consistency test, comparison to independent measurement, and comparison to historical values.

Conversion to Total Pu Input

Since the isotopic correlations, per se, relate only to ratios, some other means is needed to convert ratios to total quantities. The usual procedure is to apply the Pu/U ratio method where the weighted Pu/U ratio is applied to total U stated by the reactor (fabrication value converted for burnup). The chemical plant, of course, would simply check its ratio measurements by the technique and continue to use its primary accountability measurements of the total quantities involved.

Potential Users and Approaches

Potential users include a central safeguards authority, the chemical plant, and a reactor operator or his agent. The chemical plant can use the concept to test both Pu/U measurements as well as isotopic measurements. This subject and the numerical analysis techniques used for such checks are discussed later.

The reactor operator has a particular advantage in the historical approach since he has the most complete knowledge about his fuel and irradiation conditions. With all the supporting data in his possession, he can best judge those cases where it can be expected that the chemical plant measurement system should report the same relationships that were observed previously. Also, once some base experimental points are established, he can predict from theory how much he expects changes in conditions to effect the base number. For example, the term,

$$R_5 = - \frac{\text{Pu/U}}{\Delta^{235}\text{U}}$$

changes with the reactor conversion ratio which in turn is a function of the

initial enrichment, fuel design, cladding, fuel-to-moderator ratio, etc. If these conditions change, then he is in a unique position to predict what the expected value of the term should be for the new conditions.

A central safeguards agency is in the best position to carry out rigorous application of the concept as a verification technique. The central agency has the advantage of having all the independent pieces of information from all parts of the fuel cycle. It has fabrication data on the fuel, reactor data on the fuel to be processed, the chemical plant data after the fuel is processed, and product receiver data on shipments from the chemical plant. Since the isotopic compositions (e.g. wt. % ^{235}U) are not changed during chemical processing. The agency can check for isotopic biases by simply monitoring the across-the-plant isotopic wt. % and checking shipper-receiver isotopic data. In addition, the central agency can accumulate historical data for all reactor fuels and from all chemical plants. This subject of using the technique as a fuel cycle cross-check was discussed in more detail in last years paper⁽⁶⁾.

2. Confirmation of Previous Safeguards Information

The isotopic correlations as well as the isotopic measurements made on spent fuel dissolver solutions may also be used as a confirmation technique. In general, the information to be confirmed for safeguards purposes include the data features which identify the particular reactor or reactor class, the stated values for end-of-life nuclear materials content and initial composition of the fuel. For purposes of illustration, an example comparison is given in Table 6 . The example represents a composite of four reactors of four different types. The relative differences are typical. However, the mean values shown could not represent a real reactor.

The technique may also be used to confirm starting compositions. This feature was shown earlier in Graphs 1-2.

Table 6

Example Comparison of Preprocessing
Information With Measured Chemical Plant Data⁽¹⁾

<u>Reactor Fuel</u>	<u>Identifying Characteristic</u>	<u>Observed Value</u>	<u>Reactor Statement</u>
Composite Reactor	$\frac{\text{Pu/U}}{\Delta^{235}\text{U}}$	4475	4635
	$\frac{\text{Pu/U}}{\Delta^{236}\text{U}}$	33611	34300
	$\frac{\text{Pu/U}}{^{240}\text{Pu}/^{239}\text{Pu}}$	18780	19000
	Wt. % ^{235}U	1.601	1.611
	Wt. % ^{236}U	0.211	0.213
	Wt. % ^{239}Pu	69.25	69.15
	Wt. % ^{240}Pu	22.86	20.68
	Average Exposure MWD/Tonne	11000	11600
	Pu/U(gms/tonne)	3948	4204

(1) Mean values for data taken from four reactor of four different classes to illustrate typical agreement of data.

3. The Use of Isotopic Correlations to Cross-Check Results

There are several reasons for checking the internal consistency of the U and Pu isotopic correlation results. Among these are a) the need to pinpoint results in which the measurement error is excessive and b) the use of the correlations to assist in detecting any falsification of results. The procedures to accomplish these two objectives often involve the same statistical techniques.

a. Excessive Measurement Error

Graphs 1-7 indicate that the production isotopic values and the Pu/U results offer a variety of relationships which can be used as safeguards tools. The fact that the same variable can occur in a variety of functional relationships between the isotopic values can be used to aid in the isolation of an aberrant value.

If the relationship $y = \alpha + \beta x$ is fitted by least-squares, the residuals $r = y - \hat{y}$, where $\hat{y}$ is the predicted y value, can be used to test for aberrant values. The normalized residual form $(y - \hat{y})/\hat{\sigma}_r$ is especially useful in this context; r , the residual, and $r \times 100\%/\hat{\sigma}_r$, the percent residual, are useful in assessing the practical significance of a residual.

As graphs 1-7 indicate, there is generally little doubt about whether a particular fit is statistically significant. The values $\hat{\sigma}$, the residual standard deviation, $100\% (\hat{\sigma}/\bar{y})$ the coefficient of variation in percent and $\hat{\rho}$, the estimated correlation coefficient are useful in assessing the quality of the least-squares fit. In some cases it is of interest to test whether $\hat{\alpha}$, the least-squares estimate of α , is significantly different from 0. Here $(\hat{\alpha} - 0)/\hat{\sigma}_{\hat{\alpha}}$ is distributed as Student's t if $\alpha = 0$, and this statistic is of interest on occasion.

In the situation where $y = Pu/U$ and $x = {}^{235}U_f$, the solution of the equation for x when $y = Pu/U = 0$, will allow an estimate of ${}^{235}U_0$, the pre-irradiation enrichment, to be made. Here $\hat{{}^{235}U_0} = -\hat{\alpha}/\hat{\beta}$. Evaluating $\hat{\sigma}({}^{235}U_0)$ by propagation of errors techniques allows one to form a t value to test whether the stated initial enrichment values are credible. The assumption of linearity to the extrapolated region is crucial here. This procedure has, in practice, provided good estimates of the stated initial enrichments.

The patterns of the residuals are also useful in assessing whether the statistical model is adequate. For instance if the batches are ordered according to exposure a pattern of signs such as ++-----++ of the residuals would indicate that the model is inadequate. The quantitative values of the residuals, as well as the signs, can also be used.

b. Detecting Falsely Stated Values

The statistical regularity of the R_5 and R_6 ratios for a given reactor and enrichment has been indicated and demonstrated. Thus when R_5 and R_6 values occur which are below those which can be explained by random variation, the Pu/U and the Pu values may be suspected to be understated. If the Pu value but not the U value is understated the statistics based on the R_5 and R_6 values should determine this understatement. In order to be able to understate the Pu value of a batch and still maintain a correct R_5 value the U value may be purposely understated. The other isotopic correlations involving the Pu weight percents, however, create bounds on the extent that the Pu values can be altered. Dissembling in the sense of misstating certain isotopic values requires that the pattern be continued through the whole mosaic of Pu and U values and the isotopic weight percent values. Even if this falsification were successfully accomplished the

product data from the separations facility can be used to check on these isotopic weight percent numbers.

c. An Illustration of Cross-Checking

Tables 7.1 and 7.7 give some pertinent statistical results for individual batch measurements from 4.1 enrichment data from Yankee Rowe, Cores V and VI.

Table 7.1
Pu/U Versus $^{235}\text{U}_f$
4.1 Enrichment

<u>Batch</u>	<u>$^{235}\text{U}_f$</u>	<u>Pu/U</u>	<u>$\hat{\text{Pu/U}}$</u>	<u>Residual</u>	<u>Normalized Residual</u>	<u>Percent Residual</u>
1	3.1755	4968.4	5056.2	- 87.8	-1.55	-1.7%
4	2.5136	8635.9	8588.9	47.1	0.60	0.5
6	2.6748	7761.3	7728.5	32.8	0.42	0.4
8	2.6388	7981.9	7920.7	61.2	0.79	0.8
10	2.6359	7930.2	7936.1	- 59.4	-0.08	-0.7
12	2.4750	8737.6	8792.3	- 55.1	-0.71	-0.6
14	2.6523	7929.7	7848.6	81.1	1.04	1.0
16	2.0467	11118.5	11080.8	37.7	0.61	0.3
18	2.2545	9793.6	9971.7	-178.1	-2.45	-1.8
20	2.7958	7103.9	7082.7	21.1	0.28	0.3
22	2.4709	8862.8	8816.8	46.0	0.59	0.5

Table 7.2

Pu/U Versus $^{236}\text{U}_f$ 4.1 Enrichment

<u>Batch</u>	<u>$^{236}\text{U}_f$</u>	<u>Pu/U</u>	<u>$\hat{\text{Pu/U}}$</u>	<u>Residual</u>	<u>Normalized Residual</u>	<u>Percent Residual</u>
1	0.2209	4968.4	4957.5	11.0	0.26	0.2%
4	0.3490	8635.9	8731.5	- 95.6	1.63	-1.1
6	0.3253	7761.3	7810.5	- 49.2	- 0.84	-0.6
8	0.3313	7981.9	7974.5	7.4	0.13	0.1
10	0.3252	7930.2	7889.8	40.4	0.69	0.5
12	0.3661	8737.6	8761.5	- 23.9	- 0.41	-0.3
14	0.3296	7929.7	7928.0	1.6	0.03	0.0
16	0.4454	11118.5	11092.6	25.9	0.55	0.2
18	0.3993	9793.7	9832.8	- 39.2	- 0.71	-0.4
20	0.2999	7103.9	7116.4	- 12.5	- 0.22	-0.2
22	0.3589	8862.8	8728.8	134.1	2.29	1.5

Table 7.3

 $^{235}\text{U}_f$ Versus $^{236}\text{U}_f$

4.1 Enrichment

<u>Batch</u>	<u>$^{235}\text{U}_f$</u>	<u>$^{235}\text{U}_f$</u>	<u>$\hat{^{235}\text{U}_f}$</u>	<u>Residual</u>	<u>Normalized Residual</u>	<u>Percent Residual</u>
1	0.2209	3.1755	3.1927	-0.0172	- 1.47	-0.5%
4	0.3590	2.5136	2.4871	0.0265	1.61	1.1
6	0.3253	2.6748	2.6593	0.0155	0.94	0.6
8	0.3313	2.6388	2.6286	0.0102	0.62	0.4
10	0.3282	2.6359	2.6444	-0.0085	- 0.52	-0.3
12	0.3601	2.4754	2.4815	-0.0060	- 0.37	-0.2
14	0.3296	2.6523	2.6373	0.0150	0.91	0.6
16	0.4454	2.0467	2.0456	0.0011	0.08	0.1
18	0.3993	2.2545	2.7812	-0.0267	- 1.71	-1.2
20	0.2999	2.7958	2.7890	0.0068	0.42	0.2
22	0.3589	2.4709	2.4876	-0.0167	- 1.01	-0.7

Table 7.4Pu/U Versus $^{240}\text{Pu}/^{239}\text{Pu}$ 4.1 Enrichment

<u>Batch</u>	<u>$^{240}\text{Pu}/^{239}\text{Pu}$</u>	<u>Pu/U</u>	<u>$\hat{\text{Pu/U}}$</u>	<u>Residual</u>	<u>Normalized Residual</u>	<u>Percent Residual</u>
1	.12103	4968.4	5121.6	-153.2	-2.09	-3.0
4	.18878	8635.9	8654.1	- 18.2	-1.84	-0.2
6	.17005	7761.3	7677.1	84.3	0.86	1.1
8	.17358	7981.9	7861.2	120.7	1.22	1.5
10	.17366	7930.2	7865.4	64.8	0.66	0.8
12	.19311	8737.6	8879.5	-141.8	-1.44	-1.6
14	.17283	7929.7	7822.3	107.3	1.09	1.4
16	.23571	11118.5	11100.8	17.7	0.22	0.2
18	.21267	9793.6	9899.4	-105.8	-1.14	-1.1
20	.15793	7103.9	7045.4	58.4	0.61	0.8
22	.19344	8862.8	8897.1	- 34.2	-0.35	-0.4

Table 7.5

$$\frac{^{240}\text{Pu}}{^{239}\text{Pu}} \text{ Versus } ^{235}\text{U}_f$$

4.1 Enrichment

Batch	$^{235}\text{U}_f$	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$	Residual	Normalized Residual	Percent Residual
1	3.1755	.12103	.11988	0.00115	1.28	1.0%
4	2.5136	.18878	.18752	0.00126	1.02	0.7
6	2.6748	.17005	.17105	-0.00100	-0.85	-0.6
8	2.6388	.17355	.17473	-0.00115	-0.93	-0.7
10	2.6359	.17366	.17502	-0.00136	-1.11	-0.8
12	2.4754	.19311	.19143	0.00168	1.36	0.9
14	2.6523	.17283	.17335	-0.00052	-0.42	-0.3
16	2.0467	.23571	.23524	0.00047	0.48	0.2
18	2.2545	.21267	.21400	-0.00133	-1.16	-0.6
20	2.7958	.15793	.15868	-0.00075	-0.63	-0.5
22	2.4709	.19344	.19189	0.00156	1.27	0.8

Table 7.6

 $^{240}\text{Pu}/^{239}\text{Pu}$ Versus $^{236}\text{U}_f$ 4.1 Enrichment

Batch	$^{236}\text{U}_f$	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{240}\text{Pu}/^{239}\text{Pu}$	Residual	Normalized Residual	Percent Residual
1	0.2209	0.12103	0.11814	0.00290	1.89	2.5%
4	0.3590	0.18878	0.19023	-0.00145	-0.67	0.8
6	0.3253	0.17005	0.17264	-0.00259	-1.20	-1.5
8	0.3313	0.17358	0.17577	-0.00219	-1.01	-1.2
10	0.3282	0.17366	0.17415	-0.00050	-0.23	-0.3
12	0.3601	0.19311	0.19081	0.00230	1.06	1.2
14	0.3296	0.17283	0.17488	-0.00205	-0.95	-1.2
16	0.4454	0.23571	0.23534	0.00037	0.22	0.2
18	0.3993	0.21267	0.21127	0.00140	0.68	0.7
20	0.2999	0.15793	0.15938	-0.00145	-0.69	-0.9
22	0.3589	0.19344	0.19018	0.00326	1.51	1.7

Table 7.7

$^{240}\text{Pu}/^{239}\text{Pu}$ Versus $^{236}\text{U}/^{235}\text{U}$

4.1 Enrichment

Batch	$^{236}\text{U}/^{235}\text{U}$	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{240}\text{Pu}/^{239}\text{Pu}$	Residual	Normalized Residual	Percent Residual
1	0.06956	0.12103	0.12953	-0.00850	-2.29	-6.0
4	0.14282	0.18878	0.18607	0.00271	0.60	1.5
6	0.12162	0.17005	0.16971	0.00034	0.07	0.3
8	0.12555	0.17358	0.17274	0.00083	0.18	0.5
10	0.12451	0.17366	0.17194	0.00172	0.38	1.0
12	0.14547	0.19311	0.18812	0.00499	1.09	2.7
14	0.12427	0.17283	0.17175	0.00108	0.24	0.6
16	0.21762	0.23571	0.24380	-0.00809	-0.25	-3.3
18	0.17711	0.21267	0.21254	0.00013	0.03	0.1
20	0.10727	0.15793	0.15863	-0.00070	-1.59	-0.4
22	0.14525	0.19344	0.18795	0.00550	1.21	2.9

In addition the following results were obtained.

Table 8
Statistical Results Describing
the Least-Squares Fits

<u>Fit</u>	<u>y</u>	<u>x</u>	<u>$\hat{\rho}$</u>	<u>% CV*</u>	<u>$\hat{\alpha}$</u>	<u>$\hat{\beta}$</u>
1	Pu/U	$^{235}\text{U}_f$	-0.999	1.0%	22004	-5337
2	Pu/U	$^{236}\text{U}_f$	0.999	0.8	-1079	27328
3	$^{235}\text{U}_f$	$^{236}\text{U}_f$	-0.998	0.7	4.3213	-5.1093
4	Pu/U	$^{240}\text{Pu}/^{239}\text{Pu}$	0.999	1.3	-1189	52140
5	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{235}\text{U}_f$	-0.999	0.7	0.44440	-0.10219
6	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{236}\text{U}_f$	0.997	1.3	0.002817	0.05220
7	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{236}\text{U}_f/^{235}\text{U}_f$	0.988	2.6	0.07584	0.77182

*% CV means the coefficient of variation in percent.

These results indicate that all of these fits as judged by their relative precisions, with the possible exception of Fit 7, offer considerable promise in predicting results. In the case of Fits 2, 4, 6, and 7 it is of interest to see whether the estimated y intercepts are significantly different from the origin. Here the t values, $t = \hat{\sigma}/\hat{\sigma}_{\hat{\alpha}}$, are as follows,

fit	2	4	6	7
t	-9.0	-5.8	0.6	13.5

Thus only in Fit 6 would there be any doubt that the fitted line does not go through the origin and/or that the linear extrapolation is not valid.

In Fit 1, $^{235}\text{U}_0$, the initial enrichment is estimated as $^{235}\text{U}_0 = -\hat{\alpha}/\hat{\beta} = 4.12$ with 95% confidence limits of 4.12 ± 0.32 . The stated initial enrichment of 4.10 here is almost identical to the estimated initial enrichment and as a consequence is well within the confidence limits.

With random variation in the errors under a normal distribution the normalized residuals should generally fluctuate between -2 and +2. The exact interpretation is somewhat difficult, however, due to the fact that residuals from a least-squares fit are inherently correlated and to the fact that an outlier will have a large effect on the line itself and on the residual variance.⁽¹¹⁾ Nevertheless the normalized residuals provide a good basis for judging whether a particular result is atypical. The consequences of outliers should be judged in terms of their practical significance as well as their statistical significance.

Suppose, for example, that a normalized residual indicates that an excessive error has occurred for a given batch. The error may originate in either the independent or the dependent variable. The other relationships involving these two variables can be checked for this batch to see whether other statistically significant normalized residuals occur. A table such as the following one can be made up.

Fit	Batch	Normalized Residual	Indicated Possible Source of Error				
			Pu/U	$^{235}\text{U}_f$	$^{236}\text{U}_f$	^{240}Pu	^{239}Pu
1	18	-2.45	low	low			
2	22	2.29	high		low		
4	1	-2.01	low			high	low
7	1	-2.29		low	high	low	high

Batch 1 gives questionable values for the variable $^{240}\text{Pu}/^{239}\text{Pu}$ in relationships 4 and 7, but the pattern is not consistent. The general conclusion here is that while there are some individual normalized residuals which are somewhat out-of-line there is no consistent evidence against a particular measurement and considering the inherent measurement capability the measurement process is in control. After all there are only 4 questionable cases out of $7 \times 11 = 77$ possible cases for a rate of $(4/77) \times 100\% = 5\%$ which based on our use of ± 2 as criteria is about what would be expected.

4. Improvement of the Theoretical Base for Reactor Calculations

a. The Theoretical Basis for the Invariance of the R_5 Value

In a graph relating $y = Pu/U$ (grams/tonne) to $^{235}U_f$, the R_5 values are essentially estimates of the negative value of the slope of the line. This can be seen in Graph 1 since the slope is $(Pu/U-0)/(^{235}U_f-^{235}U_0) = -R_5$ where the numerator is the negative value of the ^{235}U depletion. The invariance of R_5 for a given reactor and enrichment over a wide range of exposure has been indicated and its importance in safeguards applications has been demonstrated. This property of invariance has been induced from compelling empirical evidence.

Efforts are now underway to establish a theoretical basis for the constancy of this slope of the Pu/U versus $^{235}U_f$ line for ^{235}U enriched light water reactors. When the complicated physics models which predict the constant slope are simplified the slope expression is of the form

$$\text{slope} = C \left[\frac{a-x}{b-x} \right]$$

where

C - is a slowly varying conversion factor (it varies by less than 2% over 25,000 MWD/MTU in Yankee Rowe),

a - is the neutron capture probability in ^{238}U at time t ,

b - is the fission probability in ^{235}U times the ratio of the neutron flux at $t = 0$ to the flux at another time t ,

and

x - is the fission probabilities of the Pu isotopes.

a and b are relatively slowly varying quantities and their variation tends to be in the same direction so they compensate each other to some extent. x varies strongly as a function of exposure, but since it appears in both the numerator and the denominator of the fraction the influence of its variation on the slope is muted. The sensitivity of the slope to variations in x is directly proportional to the difference between a and b.

The characteristics which have just been described combine to produce a slope which varies so slowly with exposure that on a batch basis essentially a straight line is produced between Pu/U and $^{235}U_f$. The linearity is so strong that the observed variations of the points about the fitted lines are commensurate with ordinary measurement errors in the Pu , U and $^{235}U_f$ values. Where duplicate measurements have been taken from the same batch it has been found that the between-duplicates, within-batch variation of R_5 is very similar to the variation of R_5 values from different batches. (10)

b. The Improvement of Reactor Codes

Table 9 is shown as an example of the quality of the Pu/U and isotopic weight percent data which are generated at the separations facility's head end accountability tank, where the tabular values are the observed correlation coefficients between the different batch Pu and isotopic results.

Table 9

Observed Correlation Coefficients Between Pu/U, U and Pu

Isotopic Values

	1	2	3	4	5	6	7	8	9
2	-.983								
3	-.999	.980							
4	.999	-.981	-.998						
5	.992	-.974	-.994	.992					
6	.921	-.852	-.924	.921	.918				
7	-.999	.979	.999	-.999	-.992	-.928			
8	.995	-.984	-.995	.994	.988	.889	-.995		
9	.998	-.978	-.995	.998	.988	.928	-.998	.991	
10	.981	-.956	-.985	.979	.979	.949	-.981	.964	.975

Legend:Variable

1	Pu/U
2	^{234}U
3	^{235}U
4	^{236}U
5	^{238}U
6	^{238}Pu
7	^{239}Pu
8	^{240}Pu
9	^{241}Pu
10	^{242}Pu

These results are from Yankee Rowe, Cores V and VI for 4.1 enrichment data. They are not intended to show that all these relationships are linear but rather to indicate that the inherent functional relationships on a batch basis clearly show through measurement and sampling errors which might tend to obscure them.

If reactor exposure monitors are used to calculate the exposures for the fuel assemblies which go into a batch, the reactor codes can be normalized to the observational data and this feedback of measured Pu/U, U and Pu isotopic weight percent data, with all their observed relationships, can eventually create codes in which the predicted weight percents and total Pu amounts are consistent with observational fact and in which the codes themselves are internally consistent.

Bibliography

1. D. E. Christensen and R. A. Schneider, "Consistency Evaluation of Accountability Data for a Chemical Processing Plant," BNWL-CC-2346 Rev. (Nov. 1969).
2. D. E. Christensen, R. A. Ewing, E. P. Gaines, Jr., R. Kraemer, R. A. Schneider, L. R. Stieff, and H. Winter, "A Summary of Results Obtained from the First MIST Experiment at Nuclear Fuels Services, West Valley, New York," TR-35 U.S. ACDA (May 1970).
3. H. H. Ph. Moeken and H. Bokelund, Eurochemic Technical Report, ETR-235 (1969).
4. H. H. Ph. Moeken and H. Bokelund, "Some Developments in Input Accountability at Eurochemic," IAEA/SM-133/77. IAEA Symposium on Progress in Safeguards Techniques, Karlsruhe, Germany (July 1970).
5. D. E. Christensen, R. P. Matson, R. A. Schneider, and W. C. Wolkenhauer, "The Safeguards Value of Chemical Plant Measurements Relating to Burnup - Yankee Cores V and VI," BNWL-1473 (June 1970).
6. R. A. Schneider, D. E. Christensen, and D. P. Granquist, "Uranium-235 Depletion as a Potential Independent Cross-Check of the Plutonium Content of Spent Power Reactor Fuels," BNWL-SA-3304, (March 1970), 11th Annual Meeting of Institute of Nuclear Materials Management, Gatlinburg, Tennessee (May 1970).
7. K. B. Stewart and R. A. Schneider, "Properties of the Pu Estimate Based on Weighted Pu/U Values," IAEA SM-133/55, IAEA Symposium on Technical Progress in Safeguards Techniques, Karlsruhe, Germany (July 1970).
8. W. Hafele and O. Nentwich, "Modern Safeguards of Reactors and Reprocessing Plants," IAEA SM-133/103, IAEA Symposium on Technical Progress in Safeguards Techniques, Karlsruhe, Germany (July 1970).
9. D. E. Christensen and R. A. Schneider, "Consistency Evaluation of Accountability Data for a Chemical Processing Plant," BNWL-CC-2346 Rev. (Nov. 1969).
10. D. E. Christensen, R. A. Schneider and K. B. Stewart, "Minor Isotopes Safeguards Techniques - Application of Isotopic Correlations to Spent Fuels," BNWL-SA-3749, May 15, 1971.
11. F. J. Anscombe, "Rejection of Outliers," Technometrics, Vol. 2; No. 2, May 1960, pp. 123-147.