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SUMMARY OF EXPERIENCE WITH THE USE OF
ISOTOPIC CORRELATION SAFEGUARD TECHNIQUES

CONTRACT NUMBER ACDA/WEC-216

PREPARED FOR
U.S. ARMS CONTROL AND DISARMAMENT AGENCY

PREPARED BY

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by

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ABSTRACT

Isotopic correlation safeguard techniques which have been developed to verify Pu/U ratio values are derived from burnup relationships in spent fuel data measured at the input to a chemical reprocessing plant. Relationships have been observed from all the chemical reprocessing plant isotopic data received to date at BNW and thus isotopic correlations have been applied successfully to these data. Because the physical laws governing the irradiation of nuclear fuels are immutable, isotopic relationships are a common characteristic of all spent fuels from any given reactor. It remains to be seen what the characteristics of some of the relationships will be for reactor types not yet studied and for newer fuel designs in reactors already investigated. There is, however, little doubt that relationships will be evident for all reactor fuels which will be appropriate for verifying the Pu/U ratio at the reprocessing plant input.

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I. INTRODUCTION

Isotopic correlation safeguard techniques in concert with the Pu/U ratio method have been developed to provide a means to accomplish the following:

- A. Provide independent verification of plutonium content of spent nuclear fuels measured at the input to a chemical reprocessing plant.
- B. Reduce the number of remeasurements required for verification, lowering safeguards inspection efforts.
- C. More effectively utilize data relating to individual fuel assemblies currently available throughout the nuclear fuel cycle.

The standard procedure developed to date to apply to measured spent fuel data has been to apply isotopic correlation techniques to measured Pu/U ratio values in order to verify that they were correctly measured. After this is applied, the Pu/U ratio method is applied to validated Pu/U values to determine independently the plutonium content of the fuel.

As more and more data from the fabrication, irradiation and reprocessing of nuclear fuel assemblies become available, the more the dimensions of isotopic correlation safeguard techniques have unfolded. To date, data from pressurized light water reactors (PWR's), boiling light water reactors (BWR's) and a pressurized heavy water reactor (PHWR) have been received and analyzed. All the measured data had one common characteristic, namely, that isotopic relationships were evident from each data group. The relationships served as a tool to verify that the plutonium content of each lot of spent fuel was measured correctly. The experience gained throughout the analyses of each individual data set also indicated the value of analyzing the data from each fuel lot collectively. It was felt that such a collective data

analysis performed when sufficient data were available from several reactor types would facilitate the further empirical development of useful isotopic relationships.

The purpose of this report is to present results from a current study of the analysis of collective data. The report also serves as a summary of all data received to date, referencing published reports on data and results as well as presenting certain chemical plant measurement data not previously reported. The work was performed by Battelle-Northwest for the Arms Control and Disarmament Agency (ACDA) under contract number ACDA/WEC-199 entitled "Applications of Minor Isotope Safeguards Techniques (MIST) to the Nuclear Fuel Cycle-Nuclear Materials Safeguarding."

II. DATA SUMMARY

The purpose of this section is to summarize the data on nuclear reactor fuel received to date at Battelle-Northwest Laboratories for the development of isotopic correlations. The data consist of information generated by the fabricator, the reactor operator and the chemical reprocessing plant. Ten different reactors are represented. There are three PWR's (YANKEE ROWE, TRINO and SENA); five BWR's (DRESDEN 1, HUMBOLDT BAY, VAK, BIG ROCK POINT and KRB); one PHWR (CANDU) and one experimental reactor (CdN). The PWR initial enrichments ranged from 2.9 to 4.9 wt% ^{235}U while the BWR enrichments ranged from 1.5 to 2.6 wt%. (Some higher enrichment BWR data currently available have not yet been analyzed.) The data on the 10 reactors included 20 cores or fuel lots. Complete measured data for six fuel lots have not yet been received, three of which represent nonreprocessed fuel so that chemical plant data are not available. These data are by no means a complete set of all that might be available or useful for developing isotopic

correlations. Moreover, only a very few are the result of activities pre-planned for the purpose of enhancing their utility. Most are simply a result of reviewing historical records. The most complete sets of data are those which include independent remeasurement of chemical plant input and product data.

Two sets of data are reported for the first time in the Appendix A and B of this report. The status and identification of all data is shown in Table I along with the report reference where detailed isotopic correlation information may be found.

It can be seen that data useful for the development of isotopic correlations concern individual fuel element history from fabrication through recovery. Moreover, the more complete the chain of data, the more confidence can be ascribed to any particular part. ". . . Accounting for the initial isotopic ratios and contents, irradiation histories, and final compositions of individual fuel assemblies, coupled with accurate calculations of the burnup and production of fuel in assemblies during irradiation in a reactor, can provide precise experimentally tested prediction techniques for use in systematic, optimal design of nuclear reactor fuels and other components. It is difficult to imagine that reactor engineers and physicists would not closely follow the results of the use of such safeguards techniques for many years to come."⁽¹³⁾

The most complete information received has been concerning HUMBOLDT BAY fuels and more recently concerning DRESDEN 1 fuel lot IV. It will be noted from Table I that some data analyses such as YANKEE ROWE cores I through IV depended almost entirely upon the measured chemical plant data. Additional information was documented in this case after the analysis.^(14,15)

TABLE I

SUMMARY OF DATA RECEIVED DURING DEVELOPMENT OF ISOTOPIC CORRELATIONS

REACTOR IDENTIFICATION			FABRICATION DATA			REACTOR DATA				CHEMICAL PLANT DATA				REMEASURED DATA				ANALYZED (REFERENCE)	
										INPUT		PRODUCT		INPUT		PRODUCT			
TYPE	NAME	FUEL LOT	Total U (Kgs)	Initial Enrichment	Assembly Number	Total U & Pu	Isotopic Data	Exposure	Irradiation History	Plant	Total U & Pu	Isotopic Data	Total U & Pu	Isotopic Data	Total U & Pu	Isotopic Data	Total U & Pu	Isotopic Data	
PWR	YANKEE ROWE	Core I		3.4	X	X				NFS	X	X	X	P					1, 2, 3, 4, 5
		Core II & III		3.4	X	X				NFS	X	X	X	P					1, 2, 3, 4, 5
		Core IV		4.1	X	X				NFS	X	X	X	P					1, 2, 3, 4, 5
		Core V & VI	X	2.9, 4.1, 4.9	X	X	P	X	P	NFS	X	X	X	X	X	X	X	X	3, 4, 5, 6, 7
		Core VII	X	3.5, 4.9	X	X	P	X		NFS	X	X	X	X	X	X	X	X	This Report
		Core VIII	X	4.9	X	X	X	X		NFS	A	A							
PWR	TRINO	II	X	2.9, 3.1						E	X	X							8
		III	X	2.9, 3.3	X	X				E	X	X			X				9, 10
		IV	X	3.1, 3.9	X	X	P	X		E	A	A							
PWR	SENA	I	A	A	A	A	A			A	A								
BWR	DRESDEN 1	I		1.5	X	X				NFS	X	X	X	P					11, 2
		II		1.5	X	X				NFS	X	X	X	P					11, 2
		III		2.5	X	X				NFS	X	X	X	P					11, 2
		IV	X	1.5, 1.8, 2.3	X	X	X	X	X	NFS	X	X	X	P	X	X			This Report
BWR	HUMBOLDT BAY	I	X	1.5, 1.8, 2.3	X	X	X	X	X	NFS	X	X	X	X	X	X	P	P	12
BWR	VAK	II	X	2.3, 2.5		X				E	X	X				X			9, 10
BWR	BIG ROCK POINT	I		2.8, 3.2, 3.7	X	P		X		NFS	X	P							
		II	A	3.0, 3.7, 4.5	X	A		X		NFS	A	A							
BWR	KRB		X	2.2	X	X	P	X		E	A	A							
PHWR	CANDU	I	X	NAT U.						E	A	A							9, 10
Exp	CdN	I	X	4.5		X				E	X	X				X			9, 10

X = Data Received
 NFS = Nuclear Fuel Services, Inc.
 E = Eurochemic
 P = Partial
 A = Anticipated

Some other analyses were substantially aided by fabrication and reactor data. It has been true during the development of isotopic correlations that the more complete the data received, the more conclusive have been the data analyses results. The ability to project these results from a first analysis of data from a given reactor to a second lot of spent fuel for the same reactor has also been related to the total amount of information received. That is, for YANKEE ROWE spent fuels, the results from the analysis of cores V and VI were more conclusive and were projected with more confidence to cores VII and VIII than were the results from cores I through IV which were projected to cores V and VI. This was due to the information made available that indicated that design, irradiation histories, etc., of the assemblies in the cores VII and VIII were similar to those of earlier fuels in core VI. Thus, it can be seen that extrapolating past results to new measurements is done with greatest confidence when the assembly histories are well known. As more experience is gained with isotopic correlations, the total information indicated in Table I may not be necessary for each spent fuel lot. However, enough information is required to determine what changes, if any, in design, irradiation history, etc., have occurred which may change previous isotopic correlation results.

The fuel lots listed in Table I do not account for all the fuels that have been reprocessed either at NFS or at Eurochemic. However, successful application of isotopic correlation techniques to all fuels to verify that batch Pu/U ratio values were correctly measured would be expected. The basis for this conclusion is illustrated in the sections that follow.

The success of the Pu/U ratio method⁽¹⁶⁾ to be subsequently applied to validated Pu/U values requires the reactor's statement of final total uranium (that is, fabricated total uranium corrected for burnup). In order

to arrive at a conclusive statement of how successful this method can be, reactor and measured total uranium values for sixteen reprocessing campaigns are listed in Table II.⁽¹⁷⁾ The fuels were processed at NFS in the period from 1966 through July 1970. Again, the values do not represent all fuels reprocessed at NFS. However, it is seen that over a period of approximately four years, the calculated uranium totals per campaign have agreed with measured totals to better than 0.5% for twelve of the sixteen campaigns. Only for four campaigns, 6, 16, 17 and 23, has the shipper-receiver difference approached 1.0% (four campaigns by including the campaign for HUMBOLDT BAY, Campaign 23). Thus, under a process situation such as at NFS, calculated values for total uranium indicate that the Pu/U ratio method can succeed generally to within 0.5% in providing an independent verification of the measured plutonium content of spent fuels per campaign once the measured Pu/U ratio values are verified by isotopic correlations. (The differences between measured plutonium and plutonium by the Pu/U ratio method are equivalent to those between measured and calculated uranium in Table II.) Values available to show a similar comparison of measured values at Eurochemic are found in Reference 18.

A factor in the independent verification of measured plutonium content of spent fuels has been a comparison between the isotopic compositions of total uranium and plutonium at input to the isotopic compositions of total uranium and plutonium at product (across-the-plant isotopics). Input and product values represent two separate sets of measurements of the same quantities, since there are no physical or nuclear processes at a reprocessing plant which can alter the relative isotopic compositions of the totals. Eleven data sets have been available where comparisons between input and

TABLE II⁽¹⁷⁾

NFS CHEMICAL PROCESSING HISTORICAL SHIPPER-
RECEIVER DIFFERENCES - SINGLE CAMPAIGNS

<u>NFS Campaign Number</u>	<u>Shipper Total U Kg</u>	<u>NFS Total U Kg</u>	<u>S-R % D</u>
1	19706	19716	-0.051
2	28661	28814	-0.53
3	46765	46681	+0.18
4	49994	50017	-0.046
5	50054	49759	+0.59
6	26367	26618	-0.94
7	26013	26116	-0.40
8	42471	42396	+0.18
9	39070	38837	+0.50
10	55352	55330	+0.04
12	48665	48882	-0.45
13	19542	19571	-0.15
14	30416	30295	+0.40
15	21488	21544	-0.26
16	15742	15557	+1.19
17	9364	9280	<u>+0.90</u>

$$\sigma_0 = 0.56\%$$

$$\text{Accum } \bar{X} = +0.05\%$$

$$\text{Ave } |X| = 0.43\%$$

product could be made. These comparisons are listed in Table III and in three cases all the uranium and plutonium isotopes were available for comparison. In the other eight cases only the ^{235}U and $^{239+241}\text{Pu}$ compositions were available for comparison. These comparisons provide an indication of the differences between input and product isotopic compositions that can be expected in the future. The values shown have not been corrected for either waste losses or beginning and ending inventories. However, if losses and inventories are small, those effects are small and the data represent the agreement based on measurement variations only. In the case of the major isotopes, ^{235}U , ^{238}U , ^{239}Pu and $^{239+241}\text{Pu}$, the agreement is seen to be within 0.2%.

III. ISOTOPIC RELATIONSHIP SUMMARY

The experience gained in the past based on the analysis of individual spent fuel data sets has indicated a need to analyze the data from each fuel lot listed in Table I collectively. It was recognized that a collective isotopic relationship analyses presented certain advantages which were not readily available from the individual analyses of each fuel lot. The advantages are as follows:

- a). Individual relationships as they result from data of different reactors as well as from different reactor types are easily compared. The comparisons are facilitated if the collective data are plotted. Linear relationships which have been determined to be the most useful in safeguards as well as nonlinear relationships are especially amenable to further study from plotted data.
- b). The range of applicability of individual relationships as pertaining to different reactors and reactor types are evident from the

TABLE III

NFS CHEMICAL PROCESSING DATA FOR THE WEIGHTED ISOTOPIC COMPOSITIONS OF INPUT AND PRODUCT

NFS CAMPAIGN NUMBER	URANIUM COMPOSITIONS				PLUTONIUM COMPOSITIONS						$\frac{\text{Pu}}{\text{U}}$
	^{234}U Wt %	^{235}U wt %	^{236}U wt %	^{238}U wt %	^{238}Pu wt %	^{239}Pu wt %	^{240}Pu wt %	^{241}Pu wt %	^{242}Pu wt %	$^{239+241}\text{Pu}$ wt %	
1 Input		0.931								99.0	
Product		0.934								98.9	
2 Input		0.937								99.0	
Product		0.938								99.0	
3 Input		0.821								91.16	
Product		0.819								91.21	
4 Input		1.006								79.938	3819
Product		1.006								79.932	3677
5 Input		2.509								86.470	5729
Product		2.510								86.392	5634
6 Input		0.712								87.12	
Product		0.714								87.13	
7 Input		0.733								87.79	
Product		0.734								87.72	
13 Input	0.016	2.708	0.398	96.879	0.820	75.087	14.154	8.585	1.356	83.672	8995
Product	0.016	2.704	0.399	96.881	0.797	75.190	14.148	8.515	1.340	83.714	8907
15 Input		0.704								73.937	4856
Product		0.704								74.047	4790
17 Input	0.015	2.724	0.480	96.780	1.067	72.397	14.913	9.942	1.681	82.339	10273
Product	0.015	2.722	0.479	96.784	1.076	72.301	14.972	9.954	1.696	82.255	9960
23 Input	0.015	1.593	0.174	98.220	0.359	75.805	17.173	5.561	1.102	81.366	4184
Product	0.012	1.601	0.174	98.212	0.356	75.964	17.066	5.520	1.093	81.484	4165

comparisons of a) above. The applicability of a given isotopic ratio, for example, is based on whether or not the slope values of the relationship from individual batches can be represented by a constant. (The slope values have been termed isotopic ratios.)

- c). Further development of useful isotopic relationships is made possible by the comprehensive overview which is obtained by using the collective data.

These advantages are illustrated in the discussions that follow by considering four isotopic relationships used to verify that the Pu/U ratio values were correctly measured. Presenting comparisons of the data by plotting is a qualitative approach which yields quantitative information, i.e., reminiscent of an old axiom "a picture is worth a thousand words." This approach is used in preference to tabulating isotopic ratio values since tabulated results are sometimes misleading unless they are explained with the aid of needed graphical displays of the data. The mean values and their one standard deviations of several isotopic ratios for individual data sets are shown on the figures to aid the quantitative interpretation of the plotted data. Curves representing the burnup paths that are evident from the data are also shown and have been drawn in by inspection. The initial data or zero burnup data points are included in the plots for the purpose of presenting an overall view of the burnup paths being traced by the data. These values are zero for the plutonium isotopes and Table I shows the reported initial ^{235}U values. The ^{236}U values are assumed on the basis of values known to be typical for uranium fabricated during the particular time span except for YANKEE ROWE, core VI.⁽¹⁹⁾ Care has been taken to assure that the shape of the burnup paths in regions where no data exist are consistent with theory.⁽²⁰⁾

A. Relationships Involving the Pu/U Ratio

The ratio Pu/U itself is a relationship in that the Pu/U values increase as the exposure increases. The ^{235}U content decreases as a function of exposure or, equivalently, the depletion of ^{235}U (^{235}D) increases so that it too is a relationship. Considering Pu/U values as a function of ^{235}U or ^{235}D has provided a linear relationship for PWR spent fuels which is particularly useful and convenient in safeguards. The relationship was useful because the isotopic ratio, $(\text{Pu}/\text{U})/^{235}\text{D}$, of individual batches were constant within measurement variations for fuels of a given initial enrichment. This was the case, even though each of the components, Pu/U and ^{235}D , varied with exposure over an appreciable range. The relationship was convenient because the Pu, U and ^{235}U values, from which the isotopic ratio was formed, were available through the accountability procedures in use. The real strength of isotopic correlations is derived from the existence of a number of relationships involving Pu/U values and several isotopes^(9,12) and the $(\text{Pu}/\text{U})/^{235}\text{D}$ ratio and other isotopic relationships are presented after which the conditions under which each should be applied are summarized.

Linear relationships have proven to be very useful in isotopic correlations and the linearity of a relationship can be easily observed by plotting. For example, the Pu/U ratio is shown plotted as a function of the ^{235}U weight percent of the irradiated fuel in Figure 1 for the collective data. (All data are shown except the CdN data.) Several symbols were used for plotting the data. A single symbol was used to indicate the batch data from a given fuel lot even though several different initial ^{235}U enrichment fuel batches were included in the fuel lot, as in the cases of HUMBOLDT BAY and DRESDEN 1 fuel lot IV. Where the batch data from two fuel lots were well separated, for example, DRESDEN 1 fuel lots I and II, one symbol has

FIGURE 1. Pu/U RATIO VERSUS ^{235}U WEIGHT PERCENT FOR THE COLLECTIVE DATA

been used for both. The lines shown are used to indicate the various linear burnup paths that the data evidenced and were drawn in by inspection. The same symbols used for each fuel lot in Figure 1 and the use of curves drawn from inspection are employed when other plotted data are presented in the remainder of the report. The choice of ^{235}U weight percent rather than ^{235}D was made because this format presents the data from the various reactors spread out for more convenient identification. Pu/U versus ^{235}D presents the data in a crowded format where it was difficult to identify individual spent fuel lots and compare the burnup paths being traced by the data points. Also, it will be recognized that the slope of the curves in either format are represented by the ratio $(\text{Pu/U})/^{235}\text{D}$, the only difference being that the slope is negative in Figure 1 and positive in the Pu/U versus ^{235}D format.

In either format, the important characteristic to compare and note is the linearity of the burnup paths described by the data points since the slope value of a linear function is a constant. It is seen that each data set evidences a relationship between the Pu/U ratio and ^{235}U . In particular, for a given initial enrichment, the batch data from CANDU, DRESDEN 1 fuel lots I, II and III, YANKEE ROWE cores I, II, III, IV, V, VI and VII reasonably describe linear burnup paths. The burnup paths have been indicated by drawing in a straight line from the initial ^{235}U values through the batch data points by inspection. In this case, where a straight line was reasonable, the absolute value of the slope, $(\text{Pu/U})/^{235}\text{D}$, formed from the individual batch values representing single initial enrichment fuels approached being constant within measurement variations. The mean $(\text{Pu/U})/^{235}\text{D}$ values for the various groups of single initial enrichment

batches are shown in Figure 1. (Due to the amount of data, it was not possible to show values for all the lower initial enrichment fuels for HUMBOLDT BAY, see Reference 12, and some of the values for DRESDEN 1 fuel lot IV, see Appendix B.) Where there were five or more batches representing single initial enrichment fuels, a one standard deviation based on the variance of the mean values are also shown. The standard deviations are an indication of the distribution of the individual batch $(Pu/U)/^{235}D$ values. It will be seen that the one standard deviations for the fuel lots mentioned above range from 0.7% to 2.7% which corresponds closely to measurement uncertainties. It is also seen that the values of the $(Pu/U)/^{235}D$ ratio varied inversely with the initial enrichment.

The variations of $(Pu/U)/^{235}D$ with initial enrichment for YANKEE ROWE spent fuel (stainless steel clad) have been resolved by the development of a second linear function which related the $(Pu/U)/^{235}D$ values to the various initial enrichments, see References 3, 5 and 7. The relationship is given in Figure 1. The particular numerical constants in this relationship do not apply to other reactors as can be verified by inserting initial enrichment values of other reactors into the function, nor do they apply to the YANKEE ROWE batches of initially enriched 2.9 and 3.5 weight percent fuel which were from Zircaloy-2 clad assemblies. However, the concept is valid and indicates a method whereby initial enrichment variations may be related for other reactors. The concept may be applicable to other variables affecting isotopic relationships also.

Further comparison in the case of HUMBOLDT BAY and DRESDEN 1 fuel lot IV of initial enrichments of 2.578 and 1.474 wt% respectively, indicates that single straight lines through the initial enrichments and the

batch data points would not adequately represent the data of these fuels. HUMBOLDT BAY and DRESDEN 1 fuel lot IV data points indicate a departure from a single linear function. This is consistent with the effects of void variations on isotopic relationships in a BWR.⁽¹²⁾ The resulting standard deviations are also larger which supports this observation. It appears that the data points from these fuels having a given initial enrichment value are evidencing two (or more) burnup paths which result from different average neutron spectrum environments possibly caused by variation of voids that the assemblies resided in during irradiation. The other alternative is that the data points are low and represent incorrectly measured Pu/U or ^{235}U values.

There is a need to observe the Pu/U values in a different linear relationship insensitive to void variations in order to resolve whether the values are low due to error or are consistent with void variations. If all of the Pu/U values are consistent, a constant will represent the slope value of the second relationship. If the values for the same batches are again low for this isotopic ratio (and the relationship is independent of any other possible cause of variations) the Pu/U values can be concluded to be low. However, before presenting a relationship which appears to be independent of void variations in a BWR, additional relationships which also show variations due to void variations are presented.

Thus, with respect to linearity resulting in a single constant slope value, $(\text{Pu/U})/^{235}\text{D}$ is seen to be applicable to all single initial enrichment PWR and PHWR data received and to BWR data under the condition that the BWR fuels processed have resided in environments of similar void history during irradiation. A detailed examination of this phenomenon

pertaining to HUMBOLDT BAY is presented in Reference 12 while DRESDEN 1 fuel lot IV data are considered in this report.

The above discussion of the $(\text{Pu}/\text{U})/^{235}\text{D}$ ratio illustrates the advantages of intercomparing the collective data by plotting the Pu/U ratio versus measured isotopic values. The goal is to have available several relationships from linear or nearly linear functions. Some should have ranges of application that are both similar as well as different to the range of application of $(\text{Pu}/\text{U})/^{235}\text{D}$. For example, a relationship not affected by void variations in a BWR to show that the HUMBOLDT BAY and DRESDEN 1 fuel lot IV data are not the result of low Pu/U values is useful. However, such a relationship may not be linear for PWR data and not apply to a PWR. Through the use of a set of mutually supporting relationships, all variable conditions affecting isotopic ratios can be treated with at least one or more relationships applying to a certain condition such as void variations in a BWR while other relationships may not.

Two other functions that were developed in the past are shown in Figures 2 and 3. The Pu/U ratio values versus the ^{236}U weight percents are shown in Figure 2 and the isotopic ratio of safeguards value is $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$ (the $\Delta^{236}\text{U}$ is read as the change in ^{236}U). The mean values and one standard deviations of the ratio for single initial enrichment fuel batches which numbered five or more are also shown. Due to the crowded nature of the data points representing DRESDEN 1 fuel lot IV, HUMBOLDT BAY and YANKEE ROWE core I, II and III, several lines were not drawn in. Also $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$ values for single initial enrichment fuel batches numbering three or less were not shown. (The same approach is used with several of the figures yet to be presented.)

FIGURE 2. Pu/U RATIO VERSUS ^{236}U WEIGHT PERCENT FOR THE COLLECTIVE DATA

FIGURE 3. Pu/U RATIO VERSUS $^{240}\text{Pu}/^{239}\text{Pu}$ RATIO FOR THE COLLECTIVE DATA

It is seen from Figure 2 that the relationship between Pu/U and ^{236}U weight percent was evidenced by all the data. Also, the relationship was reasonably linear for all PWR, PHWR and BWR fuels not affected by void variations. This conclusion is supported by the observation that the standard deviations for these fuels range from 0.8% to 2.9%. The void effect is evident for HUMBOLDT BAY and DRESDEN 1 fuel lot IV data points and the respective standard deviations (5.0% and 4.6%) are also larger. The ratio $(\text{Pu/U})/\Delta^{236}\text{U}$ is an excellent back up to $(\text{Pu/U})/^{235}\text{D}$ because it relates the plutonium produced in a fuel to the ^{236}U content. It is also seen that this ratio is applied under the same conditions as $(\text{Pu/U})/^{235}\text{D}$.

The Pu/U ratio values versus the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio values are shown plotted in Figure 3. In this instance, the burnup functions are not exactly linear if the initial point (0,0) is included with the data points of a given fuel lot. However, in the range of the data points, the burnup curves are linear as shown and the isotopic ratio of the form $(\text{Pu/U}-a)/(^{240}\text{Pu}/^{239}\text{Pu})$ approaches being a constant under the same conditions outlined for $(\text{Pu/U})/^{235}\text{D}$. In addition, the quantity "a" which is the y-axis intercept of the straight line portion of the curves when extrapolated to the y-axis, is positive for the BWR and CANDU data and negative for the PWR data indicating characteristic trends for different reactor types. Nevertheless, neglecting the intercept value and using the ratio in the form of $(\text{Pu/U})/(^{240}\text{Pu}/^{239}\text{Pu})$ has been of value^(9,10) and the mean values of the ratio in this form are also shown in Figure 3.

The standard deviations given in Figure 3 are not indicative of the distribution of the slope values as pertain to the curves shown but are indicative of the distribution of the slope values of a straight line through (0,0).

Where the standard deviations are less than 3%, a straight line through (0,0) was a good approximation for the curves shown. Where the standard deviations are larger than 3%, a straight line through (0,0) was not a good approximation (in the case of PWR fuels) or void variations caused the larger deviations (in the case of BWR fuels). Note that two burnup paths, or low data points, are evident from Figure 3 for HUMBOLDT BAY and DRESDEN 1 fuel lot IV indicating that the relationship is void sensitive.

It is thus seen from Figure 3 that the relationship between Pu/U and $^{240}\text{Pu}/^{239}\text{Pu}$ was evidenced by all the data. This relationship is an excellent addition to the two relationships given previously because it relates the plutonium produced in a fuel to the ^{239}Pu and ^{240}Pu isotopic values.

The last isotopic ratio involving the Pu/U ratio to be considered in this paper is $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$. This ratio was developed during the analysis of HUMBOLDT BAY fuels⁽¹²⁾ and was shown to be insensitive to void variations in a BWR. The relationship for all the collective data is shown in Figure 4 where the Pu/U ratio values are plotted as a function of the ${}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2$ values. The mean values and one standard deviations of the isotope ratio for individual batches where there were five or more batches representing single initial enrichment fuels are also shown. Based on observations of the data presented in Figure 4, that is, observing that:

- a). The data points evidence very few (or closely spaced) burnup paths;
- b). The mean values of the ratio for BWR fuels are reasonably the same and
- c). The differences between mean values for BWR and PWR or CANDU fuels are not as large as differences for other relationships presented,

certain conclusions were evident. They are as follows:

- 1). The relationship is not strongly dependent on different initial enrichments, fuel designs and void variations in the case of the BWR fuels

FIGURE 4. Pu/U RATIO VERSUS $\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{^{240}\text{Pu}^2}$ FOR THE COLLECTIVE DATA

considered. The standard deviations support this conclusion and are seen to be 2.1% or less for three of the five BWR fuel lots. Possible reasons for the other two standard deviations being over 3% (3.3% and 3.4%) are presented in Appendix B.

- 2). The dependence on initial ^{235}U enrichment is retained for CANDU fuels and PWR fuels, although the dependence is not large for PWR fuels except for the fuel from YANKEE ROWE core I.
- 3). The data points appear to be describing a single burnup path that is somewhat common to all the data, except CANDU. The burnup path is seen to be linear for BWR data at lower exposures while tending to be non-linear for PWR data at higher exposures.

The ratio is dependent upon what has been termed the mixing property of plutonium isotopes.⁽¹²⁾ Also, since additional isotopic values are included in the ratio, it is more subject to the random error in measurement, particularly in the measurement of ^{242}Pu . The results of the mixing effect and measurement random variations are evident from the data points that lie separated from the bulk of the data points (see Appendix B for further discussion of mixing).

The value of the $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ ratio is that it can be applied under the condition that the spent fuels were irradiated in different void environments in a BWR. In the case that variations seen for the previous ratios presented for certain HUMBOLDT BAY and DRESDEN 1 fuel lot IV batches were the result of void variations, the $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ values for these batches would be the same as the values of the remaining batches. In the case that the Pu/U values were low, the $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$

values would also be low. It was seen that the ratio values for HUMBOLDT BAY⁽¹²⁾ and DRESDEN 1 fuel lot IV (see Appendix B) were reasonably constant for all batches which represented 2.578 and 1.474 initially enriched fuels. This result indicated that variations in the previous isotopic ratios considered were due to void effects.

The $(\text{Pu/U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ ratio is thus important for a BWR since it applies under conditions where the first three ratios do not. A summary of these conditions is listed in Table IV. Also, an additional isotope, ${}^{242}\text{Pu}$, not included in the other ratios, was shown to be related to the plutonium produced in a fuel. Again, it can be said that the relationship between Pu/U and ${}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2$ was evident from all the data shown in Figure 4.

It is therefore apparent from all the analyses completed to date that the various data sets have one common characteristic, namely, consistent isotopic relationships were evident from each set of measured data. It is expected that this characteristic is common to all spent fuels from any given reactor due to the physical laws governing the irradiation of nuclear fuels. It remains to be seen what some of the relationships will be for certain reactor types and for newer fuel designs in reactors previously investigated. There is, however, little doubt that appropriate relationships will be evident for all reactor fuels. It also follows that there will be relationships which relate Pu/U values to isotopic values thus providing a means to verify that the Pu/U values were correctly measured. It is expected also that to accomplish the above goals, the measured chemical reprocessing plants isotopic data as well as uranium and plutonium totals at input and product will be needed.

TABLE IV
SUMMARY OF CONDITIONS WHICH WHEN VARIED CHANGE THE VALUES OF
THE ISOTOPIC RATIOS LISTED

ISOTOPIC RELATIONSHIP	VARIABLE PARAMETERS												
	PWR				BWR					PHWR			
	Initial ²³⁵ U Enrichment	Cladding	Assembly Dimensions	Mixing Effect	Initial ²³⁵ U Enrichment	Cladding	Assembly Dimensions	Void History	Mixing Effect	Initial ²³⁵ U Enrichment	Cladding	Assembly Dimensions	Mixing Effect
$\frac{\text{Pu/U}}{^{235}\text{U}}$	X	X	X		X	X	X	X		X	X	X	
$\frac{\text{Pu/U}}{\Delta ^{236}\text{U}}$	X	X	X		X	X	X	X		X	X	X	
$\frac{\text{Pu/U}}{^{240}\text{Pu}/^{239}\text{Pu}}$	X	X	X	S	X	X	X	X	S	X	X	X	S
$\frac{\text{Pu/U}}{^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2}$	X	X	X	X	S	X	X		X	X	X	X	X

X = Relationship changes.

S = Slight but not a strong effect.

B. Relationships not Involving the Pu/U Ratio

All of the relationships discussed thus far have involved both the Pu/U ratio and various isotopic values. An additional basic part of the investigation of isotopic correlations has been the development of linear as well as nonlinear relationships which involve uranium and plutonium isotopes only. The purpose of these investigations has been to show that the isotopic values have been correctly measured. Then, in the event an anomalous value of any one of the four relationships considered in the last section is observed, it will be known which parameter is at fault, the Pu/U ratio or an isotopic value. Also, it should be possible to substantiate variations in any one of the four Pu/U isotopic ratios caused by an initial enrichment change, varying voids, etc. This part of isotopic correlations has been termed "consistency cross checks" and also includes checking the measured data to verify that they are consistent with previous history (that is, any previously measured chemical plant data, fabrication data or reactor data) and remeasured data. The previous history and remeasured data are mentioned to delineate consistency cross checks fully, however, these checks are considered in more detail in other papers. (21,12)

As an example of a reasonably linear function involving isotopic values only, the ^{236}U weight percent is shown as a function of ^{235}U weight percent for the collective data in Figure 5. The isotopic ratio of safeguards value is $\Delta^{236}\text{U}/^{235}\text{D}$ (this is the absolute value of the negative slope shown in Figure 5). The mean values and one standard deviations for the $\Delta^{236}\text{U}/^{235}\text{D}$ ratio are also shown. The relationship is initial enrichment

FIGURE 5. ^{236}U WEIGHT PERCENT VERSUS ^{235}U WEIGHT PERCENT FOR THE COLLECTIVE DATA

dependent and dependent on void variations in a BWR. The void dependence is not strong as evidenced by the fact that the standard deviations for HUMBOLDT BAY and DRESDEN 1 fuel lot IV are within 2%. The dependence was most readily observed by enlarging the plotting scale for HUMBOLDT BAY data plots. The $\Delta^{236}\text{U}/^{235}\text{U}$ ratio provides a means to verify that ^{236}U and ^{235}U were measured correctly under the same conditions listed for $(\text{Pu}/\text{U})/^{235}\text{U}$ in Table IV. Again, it will be noted that the relationship between ^{236}U and ^{235}U was evidenced by all the data.

It is important to realize that for every linear burnup function shown thus far, several nonlinear functions also are available for safeguards use. This includes those of isotopic values alone and those involving the Pu/U. Some nonlinear functions are as useful as linear functions to indicate what happens when enrichments, voids, etc., change. For void variations for example, nonlinear functions involving isotopic data are the only functions available to readily substantiate the variations seen in the isotopic ratios involving Pu/U values.

For the purpose of illustrating a nonlinear function, the $^{236}\text{U}/^{235}\text{U}$ ratio versus the $^{236}\text{U}/^{238}\text{U}$ ratio data are shown in Figure 6. As can be seen the data again evidenced distinct burnup paths although nonlinear. The contribution to safeguards is that such a plot can be used to check data consistency. In this case, the burnup paths appear to be independent of void variations and change only due to changes in initial enrichments. The ^{235}U initial enrichment is indicated for each burnup path. Another example involving isotopic data only is shown in Figure 7 where the $^{242}\text{Pu}/^{240}\text{Pu}$ ratio data are plotted versus the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio data.

FIGURE 6. $^{236}\text{U}/^{235}\text{U}$ RATIO VERSUS $^{236}\text{U}/^{238}\text{U}$ RATIO FOR THE COLLECTIVE DATA

FIGURE 7. $^{242}\text{Pu}/^{240}\text{Pu}$ RATIO VERSUS $^{240}\text{Pu}/^{239}\text{Pu}$ RATIO FOR THE COLLECTIVE DATA

In addition to the variations in initial enrichment, these isotopic data show the void effect (that is, the same data points which described two burnup paths in Figures 1, 2 and 3 for HUMBOLDT BAY and DRESDEN 1 fuel lot IV describe two burnup paths in Figure 7). The isotopic data, in this case ^{239}Pu , ^{240}Pu and ^{242}Pu , thus substantiate the variations seen in the Pu/U values earlier. Each burnup function that can be observed via plotting by an appropriate choice of format makes a contribution to the overall usefulness of isotopic correlations by showing data consistency. One relationship may show the dependence of the data on void variations, another may not (as indicated in Figure 6 and 7). The goal is to develop numerous plots some of which evidence a single variation and some which are independent of the particular characteristics of the fuel.

C. Reactor Characterization

The relationships shown and discussed in previous sections serve to identify measured data as originating from a given reactor type as well as a given reactor. The numerical values of the isotopic ratios presented in Figures 1 through 5 for a reactor have generally been unique relative to values for the other reactors considered. Thus, the ratios characterize a reactor. As an example of characterizing different reactor types from isotopic data, batch data from a PWR and a BWR reactor at approximately 15000 MWd/MTU are compared in Table V. The most marked differences between the isotopics for each reactor type, neglecting the ^{235}U and ^{236}U values which are dependent on initial enrichments, appear in the plutonium isotopic chain, particularly for ^{240}Pu and ^{242}Pu . The differences are due to the different neutron spectra associated with each reactor type. As long

TABLE V
ISOTOPIC COMPOSITIONS OF A PWR AND A BWR AT 15,000 MWd/MTU EXPOSURE

Exposure, MWd/MTU	Reactor Type	Initial Enrichment	Uranium Isotopic Composition				Plutonium Isotopic Composition					$\frac{\text{Pu}}{\text{U}}$
			^{234}U	^{235}U	^{236}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	
		wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %	wt %	$\frac{\text{Grams}}{\text{Tonne}}$
15,800	PWR	4.101	0.019	2.675	0.325	96.981	0.577	77.961	13.257	7.220	0.985	7762
15,200	BWR	2.258	0.010	1.087	0.223	98.681	0.575	67.544	21.191	8.557	2.134	5741

as different reactor types are characterized by such differences, the plutonium isotopes will identify reactor types by the relative abundance of each plutonium isotope.

IV. FUTURE APPLICATION

The collective data analysis using plotting as a tool has been successful in providing the desired advantages, indicated at the beginning of section III, of easily comparing the individual data sets and noting the range of applicability of several isotopic relationships. The third advantage mentioned, that of further developing useful isotopic relationships using the collective data has also been realized, even though the analysis is still incomplete. It now appears that as a result of the additional insights derived from using the growing body of information on isotopic correlations, collective data analysis is the prime tool to extract the maximum amount of information useful for safeguard application from the various individual data sets.

An interesting prospect has developed from the collective data analysis which has resulted from the examination of data plots such as the ones shown in Figures 1 through 7. The question has been asked: Is there a burnup function, linear or nonlinear, relating isotopic values to Pu/U values and/or to other isotopic values that is common to all measured data received to date such that all plotted data points would describe the same burnup path and thus be independent of initial enrichments, void variations, cladding differences, etc.? The number of possible functions to investigate in search for an answer is formidable. For example, beginning with the relationship of

one isotope versus another, several possibilities of plotting the data are as follows:

- a. A ratio of isotopes versus a single isotope,
- b. A ratio of isotopes versus other isotopic ratios,
- c. The sum of two isotopes versus a single isotope or ratio,
- d. The above variations where one or the other parameter is raised to some power, n ,

and so on. It is difficult to visualize the total number of plotting formats that can be devised in developing common burnup functions. However, that such functions may be available has been indicated by plotting formats such as shown in Figure 4.

The value of common burnup functions to safeguards is understood when it is realized that if a common function were available, then no matter what fuel from any of the reactors listed in Table I were processed, the resulting measured data would agree with the established function. The safeguarding agency would always be equipped with foreknowledge of how the isotopic and Pu/U measurements should agree with each other and would verify the measurements when agreement was observed.

Functions which represent the other extreme, that is, functions which provide relationships that are distinctly different and depend on reactor type, void variations, initial enrichments, etc., are equally useful since they provide identification of the fuel and the particular characteristic conditions present during irradiation of the fuel. For example, the

plotting format shown in Figure 6 can provide general verification of the initial enrichment independent of void effects by virtue of where the data points lie on the figure. Then, the plotting format shown in Figure 7 can be used to show that in addition to enrichment effects, void effects were either present or not present. By developing those plotting formats that show enrichment variations independent of void variations, for example, and another format that shows void variations independent of enrichment variations, etc., isotopic correlations would serve as strong tools to verify measurements, initial enrichments and irradiation conditions. Therefore, the collective data analysis has been directed toward the development of common functions as well as distinct functions in order to develop isotopic correlation safeguard techniques further.

The use of the collective data has also indicated the need to develop a computerized information data file to utilize and compare fabrication, reactor and chemical plant data readily and effectively. Although not specifically mentioned previously during the discussions pertaining to Figures 1 through 7, a real part of verifying that any one individual set of data measurements is correct (YANKEE ROWE core VII or DRESDEN 1 fuel lot IV, for example) is that an individual data set appears consistent compared to all other data sets, and therefore believable. All previously measured data, whether from the same reactor or not, thus aid the verification of new measurements. It is of value then to quickly access and manipulate data previously measured, analyzed and stored to aid the verification of new measurements via an

information data file. Such an approach also offers a means of having a comprehensive overview of all data measurement results and their implications that a safeguarding agency requires for decision making. In view of the volume of data involved, such a means can only be realized through the use of a computerized data file.

Some considerations of such a file are as follows: The chemical reprocessing plant measurements are vital in that they presently are the most accurate measurements of spent fuel content and thus would be the focal point of the data file. To supplement these data and assist in reaching valid conclusions, all information available about a particular batch of fuel would be included on the data file. Measurements on spent fuel assemblies other than by the chemical reprocessing plant may become available in the future, such as gamma scanning measurements, and these data would also constitute a part of the data file. Another feature of the information data file is the requirement of long term storage and recovery since the time from availability of fabrication data until chemical plant data extends to 10 or more years. The specific data to be considered for the information file are discussed in detail in other papers⁽²¹⁾ and were partially summarized in Table I of this report.

A graphical view of one information file concept which has been given the acronym NUMIST is shown in Figure 8. The essential parts of the concept are the inspector, data analysis center and the information data file. The information data file includes all historical data relevant to isotopic correlations including previously reported and collected data from fuel fabrication and the reactor. The inspector could provide any chemical plant data not routinely reported in addition to other data directly to the analysis center via teletype in real time. The function of the analysis center

FIGURE 8. DATA INFORMATION FILE (NUMIST) CONCEPT

includes forming isotopic correlation relationships from the following sources:

- Chemical plant data
 - Previous campaign data
 - Fabrication data
 - Preprocessing data (calculated data)
 - Gamma scan data
- } Information data file

Through the comparisons thus available, it will be possible for the analysis center to decide how many remeasurements are necessary and which batches should be remeasured. This recommendation will then be relayed to the inspector using an answer file via teletype. Once the remeasurements have been completed and the results received at the analysis center, the final comparisons would be made including the comparison of total uranium as measured and as calculated. The decision as to whether or not plutonium content as measured by the chemical plant is correct would then follow through application of the Pu/U ratio method which verifies the plutonium content and the uncertainty. The information data file is thus seen to be an effective vehicle for employing isotopic correlation safeguard techniques to their fullest effectiveness.

V. CONCLUSIONS

The success of isotopic correlation safeguard techniques to verify Pu/U ratio values derives from the existence of burnup relationships which were evident in plots of all the data sets analyzed to date. Thus, the basis for successful application has been observed and is expected to be observed in future data representing different reactors as well as different reactor types. Examples of linear and nonlinear relationships have been given, each applicable under various conditions. That the use of several relationships is a distinct advantage has been shown as well. In fact, the power of iso-

topic correlations is most effectively employed when it is possible to take advantage of all the historical information pertaining to a given batch of spent fuel which is available throughout the nuclear fuel cycle.

It was seen through a comparison of measured total uranium and calculated total uranium per campaign, that the application of the Pu/U ratio method to verified Pu/U values using calculated values of total uranium can successfully provide an accurate independent measure of total plutonium. Another useful verification tool for additional substantiation of plutonium content was seen to be a comparison of isotopic composition of input and product total uranium and plutonium.

Based on the collective data analyses results to date, further empirical and theoretical development of isotopic relationships is needed. The immediate goals include the following:

- a). Complete the six data sets mentioned in Section II as well as obtain data sets from other reactor types and include them in the collective data analysis.
- b). Develop further both common and distinct burnup relationships as indicated in Section IV.
- c). Make data available to the Agency on a broad basis as indicated in comments on a data information file.

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APPENDIX A

APPENDIX A

Chemical Reprocessing Input Data for YANKEE ROWE Core VII

YANKEE ROWE core VII fuel lot consisted of 36 spent fuel assemblies of which 32 were stainless steel clad and identical in design to the assemblies processed previously from core VI⁽⁵⁾. Of the 32 assemblies, 4 were charged to the reactor on 11/9/65, were exposed during cores V, VI and VII and reached an exposure level of 30,000 MWD/MTU ⁽²²⁾. The remaining 28 assemblies were charged on 11/7/66, were exposed during cores VI and VII and ranged in exposures from 19,000 to 27,000 MWD/MTU. Four of the 36 assemblies were zircaloy clad and were also different in other design respects compared to previously processed assemblies from YANKEE ROWE. They were charged on 5/1/68, were exposed during core VII and reached an exposure level of 12,000 MWD/MTU. All assemblies were discharged from core VII on 8/2/69.

Using previous isotopic correlation analysis results of YANKEE ROWE core I through VI and the preprocessing data for core VII, it was possible to predict empirically the value of several isotopic ratios to be expected from the chemical plant measured data for this core. The expected values for $(\text{Pu/U})/^{235}\text{D}$ were given in Reference 10, volume II, (Page 36) and those values are again shown in Table A-I of this report along with measured values. The individual Batch measured value of the ratios considered in the body of this report are given in Table A-II. The mean $(\text{Pu/U})/^{235}\text{D}$ values of Table A-I were determined from the individual measured values given in Table A-II. The actual chemical processing input data are given in Table A-III. The batch data in Tables A-II and III have been grouped according to initial enrichment values and ordered according to decreasing ^{235}U weight percent within an initial enrichment group.

TABLE A-1
SUMMARY OF EXPECTED AND MEASURED VALUES FOR THE RELATIONSHIP $(Pu/U)/^{235}_D$
FOR YANKEE ROWE CORE VII

Fuel Type	$^{235}_U$ wt %	Expected Number of Chem Plant Input Batches	EXPECTED EMPIRICAL VALUES			MEASURED VALUES			
			Mean $\frac{Pu/U}{^{235}_D}$	2σ Range for Individual Batches - $\frac{Pu/U}{^{235}_D}$	2σ Range for Mean - $\frac{Pu/U}{^{235}_D}$	Number of Actual Batches and $^{235}_U$ wt %	Mean $\frac{Pu/U}{^{235}_D}$	2σ Range for Individual Batches - $\frac{Pu/U}{^{235}_D}$	2σ Range for Mean - $\frac{Pu/U}{^{235}_D}$
Stainless Clad (32)	4.935	8 - 14	4591	4861 - 5121	4921 - 5061	12 4.935 *	4951 (4966)	4809 - 5093 (4866 - 5066)	4910 - 4992 (4936 - 4996)
Zircaloy Clad (2)	3.533	0 - 1	5247	5090 - 5404		1 3.59 **	5150		
Zircaloy Clad (2)	3.470	0 - 1	5275	5117 - 5433		1 3.50 **	5158		
TOTAL	4.791	10 - 15	4999		4913 - 5085	14	4970		

* From Preprocessing Data

** Assumed from Correlation Analysis

TABLE A-II
SUMMARY OF ISOTOPIC RELATIONSHIPS FOR YANKEE ROWE CORE VII

Batch Number	VALUES OF ISOTOPIC RELATIONSHIPS									
	Pu/U gms/T	²³⁵ D	$\frac{Pu/U}{^{235}D}$	$\Delta^{236}U$	$\frac{Pu/U}{\Delta^{236}U}$ ($\times 10^{-4}$)	$\frac{^{240}Pu}{^{239}Pu}$	$\frac{Pu/U}{^{240}Pu/^{239}Pu}$ ($\times 10^{-4}$)	$\frac{^{239}Pu \cdot ^{242}Pu}{^{240}Pu^2}$	$\frac{Pu/U}{^{239}Pu \cdot ^{242}Pu / ^{240}Pu^2}$ ($\times 10^{-4}$)	$\frac{\Delta^{236}}{^{235}D}$
	1	2	3	4	5	6	7	8	9	10
12	9619	1.950	4934	0.402	2.39	0.1878	5.121	0.5110	1.882	0.206
11	9842	2.020	4873	0.415	2.37	0.1935	5.086	0.5165	1.906	0.205
9	10119	2.022	5006	0.423	2.39	0.1949	5.193	0.5215	1.940	0.209
10	9964	2.026	4919	0.423	2.36	0.1944	5.125	0.5237	1.903	0.209
1	9751	2.039	4783	0.414	2.36	0.1931	5.049	0.5136	1.898	0.203
8	10553	2.138	4937	0.442	2.39	0.2038	5.178	0.5465	1.931	0.207
2	10787	2.163	4988	0.449	2.40	0.2088	5.165	0.5562	1.939	0.208
4	11128	2.210	5037	0.460	2.42	0.2120	5.248	0.5609	1.984	0.208
3	11261	2.246	5015	0.463	2.43	0.2141	5.259	0.5653	1.992	0.206
6	11521	2.299	5012	0.479	2.41	0.2001	5.235	0.5873	1.962	0.208
5	11717	2.360	4966	0.490	2.39	0.2257	5.191	0.5961	1.966	0.208
7	12046	2.438	4942	0.500	2.41	0.2282	5.270	0.6153	1.958	0.205
			4951 $\pm$ 1.4%*		2.39 $\pm$ 0.8%*		5.177 $\pm$ 1.4%*		1.938 $\pm$ 1.8%*	0.207 $\pm$ 0.9%*
13	6597	1.281	5150	0.263	2.50	0.1923	3.431	0.3671	1.797	0.205
14	6608	1.251	5158	0.258	2.56	0.1929	3.425	0.3553	1.860	0.206
All Batches	10273	2.067	4970	0.429	2.42	0.2060	4.987	0.5471	1.878	0.208

*Mean value and the one standard deviation.

Initial ²³⁶U values: For $^{235}U_0 = 4.935$, $^{236}U_0 = 0.0564$ (Measured (19))
 For $^{235}U_0 = 3.59$, $^{236}U_0 = 0.023$
 For $^{235}U_0 = 3.50$, $^{236}U_0 = 0.022$ } Assumed
 For $^{235}U_0 = 4.791$, $^{236}U_0 = 0.051$

TABLE A-III
YANKEE ROWE CORE VII INPUT DATA SUMMARY

Batch Number	235 _U * Init. Enrich	NF ₂ MEASURED CHEMICAL PROCESS INPUT										Total U Kgs	Total Pu Grams
		Uranium Isotopic Composition***				Plutonium Isotopic Composition***							
		234 _U	235 _U	236 _U	238 _U	238 _{Pu}	239 _{Pu}	240 _{Pu}	241 _{Pu}	242 _{Pu}			
12	4.935	0.016	2.985	0.458	96.541	0.888	74.549	11.022	9.094	1.346	695.9	6694	
11	4.935	0.016	2.915	0.471	96.598	0.916	73.949	14.309	9.397	1.430	703.4	6923	
9	4.935	0.016	2.913	0.479	96.593	0.945	73.740	14.369	9.436	1.460	687.2	6954	
10	4.935	0.016	2.909	0.479	96.597	0.938	73.757	14.434	9.505	1.460	698.3	6958	
1	4.935	0.016	2.896	0.470	96.618	0.905	73.979	14.286	9.413	1.417	694.5	6772	
8	4.935	0.016	2.797	0.498	96.690	1.063	72.505	14.777	10.008	1.646	714.3	7538	
2	4.935	0.016	2.772	0.505	96.708	1.101	71.938	15.023	10.193	1.745	693.8	7484	
4	4.935	0.015	2.725	0.516	96.744	1.142	71.504	15.162	10.389	1.803	691.3	7693	
3	4.935	0.015	2.689	0.519	96.777	1.170	71.240	15.253	10.491	1.846	701.9	7904	
6	4.935	0.014	2.636	0.535	96.815	1.288	70.487	15.512	10.708	2.005	689.7	7946	
5	4.935	0.014	2.575	0.546	96.865	1.371	69.794	15.755	10.960	2.120	673.2	7888	
7	4.935	0.012	2.497	0.556	96.935	1.477	69.385	15.833	11.112	2.223	693.0	8348	
13	3.527	0.018	2.309	0.286	97.387	0.456	76.254	14.663	7.592	1.035	457.5	3018	
14	3.470	0.018	2.219	0.280	97.482	0.431	76.301	14.719	7.540	1.009	486.4	3214	
ALL BATCHES	4.791	0.015	2.724	0.480	96.780	1.067	72.397	14.913	9.942	1.681	9280.4	95334	
15**		0.018	2.267	0.289	97.426	0.496	76.010	14.880	7.838	0.779	0.2	1	
16		0.018	2.572	0.434	96.796	0.830	74.128	14.691	8.944	1.407	0.0	1	
TOTAL											9280.6	95336	

* From Preprocessing Data

** The Isotopic Values from Flush Batches 15 and 16 are not used.

*** Wt%

From the expected and measured results given in Table A-I, it can be seen that the expected $(\text{Pu/U})/^{235}\text{D}$ values were observed during the processing of the fuel. Two values are given for the 12 measured batches of 4.935 fuel. The first value represents the mean value for all 12 batches while the second value (in parenthesis) represents the mean of 11 batches and excludes the value for batch #1, see Table A-II. The $(\text{Pu/U})/^{235}\text{D}$ value for batch #1 was low and outside the 2σ range for individual batches. Three plausible explanations presented themselves:

1. The Pu/U value really was low,
2. The ^{235}U value was low resulting in a high ^{235}D value, or
3. The batch had become contaminated with lower initial ^{235}U content uranium such that the weighted initial enrichment of batch #1 was actually too high causing ^{235}D using 4.935 to be high.

It was concluded by an examination of all isotopic data for batch #1 that the ^{235}U weight percent was probably low (1 to 2% low) which may have been contributed to by contamination. This conclusion was reached by noting that all isotopic values for batch #1, both ^{236}U and all plutonium isotopic values, and the Pu/U values, see Table A-II and A-III, indicated that the ^{235}U value should have been comparable to (and possibly higher than) the value of batch #11. Thus, all isotopics except ^{235}U indicated that batch #1 was at an exposure level similar to batch #11 (so did the preprocessing data). Also, none of the other relationships values for batch #1 involving Pu/U, Table A-II, were outside the 2σ range while the $\Delta^{236}\text{U}/^{235}\text{D}$ relationship was low (again due to low ^{235}U). All data plots involving ^{235}U in addition depict the data point of batch #1 as being 1 to 2 percent low. Remeasured isotopic values for batch #1 gave the same result (that is, low ^{235}U weight

percent while all other isotopics were again similar to batch #11) indicating possible contamination of the ^{235}U . Thus, the Pu/U value for batch #1 was considered to be correctly measured.

In the cases of batches 13 and 14, which included the Zircaloy clad assemblies, weighted initial enrichments different from those from the preprocessing data were used with the measured data. This was not done to imply that the preprocessing data were incorrect. This was due to the fact that dissolver heels from the 4.935 fuel were mixed with the 3.533 material raising the weighted initial enrichment of batch 13. The same could be said of batch 13 heels and batch 14. The initial ^{235}U values of 3.59 and 3.50, respectively, indicated from data plots Figures 5 and 6 of the main report were thus used. The adjusted measured $(\text{Pu}/\text{U})/^{235}\text{D}$ values were within the predicted 2σ range for these two batches as seen from Table A-I.

The measured weighted $(\text{Pu}/\text{U})/^{235}\text{D}$ for the total fuel of core VII was seen to be in excellent agreement with the expected empirical value from Table A-I. The weighted Pu/U value for core VII, 10273 grams/tonne (from Table A-II), was thus considered to be validated. Using the Pu/U ratio method where a uranium calculated total for core VII of 9364 Kgs was available from the preprocessing data, the independent determination of the plutonium content of core VII fuels was 96196 grams. This value is 0.9% higher than the measured value, 95336 grams (due to the 0.9% difference between measured 9280.6 Kgs and calculated uranium values).

The product data for core VII are given in Table A-IV along with the isotopic values and a summary of isotopic ratios for the product totals. From a comparison of isotopic values for input, Table A-III, and product, Table A-IV, (or Table II in the body of the report), it is seen that agreement was to within 0.2% for the major isotopes. In the case of isotopic

TABLE A - IV

YANKEE ROWE Core VII Product Data and
Isotopic Relationship Summary

Measured Data

Shipment Number	Uranium Isotopic Compositions and Uranium Total					Shipment Number	Plutonium Isotopic Compositions and Plutonium Total					
	^{234}U wt %	^{235}U wt %	^{236}U wt %	^{238}U wt %	Total U Kgs		^{238}Pu wt %	^{239}Pu wt %	^{240}Pu wt %	^{241}Pu wt %	^{242}Pu wt %	Total Pu Grams
1	0.014	2.679	0.521	96.786	4160.9	1	1.066	72.118	15.012	10.086	1.708	12197
2	0.017	2.757	0.444	96.783	2677.2	2	1.066	72.121	15.014	10.087	1.707	10388
3	0.016	2.752	0.444	96.786	2346.0	3	1.245	70.629	15.555	10.606	1.969	11606
						4	1.338	70.092	15.652	10.837	2.078	16375
						5	0.983	73.368	14.504	9.614	1.523	22323
						6	0.864	74.217	14.519	9.016	1.384	18587
						PRODUCT TOTALS						
	0.015	2.722	0.479	96.784	9184.1		1.076	72.301	14.972	9.954	1.696	91476

Isotopic Relationship Summary

Pu/U	^{235}D	Pu/U ^{235}D	$\Delta^{236}\text{U}$	$\frac{\text{Pu/U}}{\Delta^{236}\text{U}}$ ($\times 10^{-4}$)	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$	$\frac{\text{Pu/U}}{^{240}\text{Pu}/^{239}\text{Pu}}$ ($\times 10^{-4}$)	$\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{^{240}\text{Pu}^2}$	$\frac{\text{Pu/U}}{\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{^{240}\text{Pu}^2}}$ ($\times 10^{-4}$)	$\frac{\Delta^{236}\text{U}}{^{235}\text{D}}$
9960	2.069	4814	0.428	2.33	0.2071	4.810	0.5470	1.821	0.207

ratios involving the Pu/U (Tables A-II and A-IV), the product values are different due to the fact that different losses result for uranium and plutonium during the chemical reprocessing. However, the values involving only isotopic values agree very well.

As a result of the isotopic correlation analysis, application of the Pu/U ratio method, and comparison of across the plant isotopics, the measured plutonium content of YANKEE ROWE core VII spent fuel is considered validated.

APPENDIX B

APPENDIX B

Chemical Reprocessing Input Data for DRESDEN 1 Fuel Lot IV

During the fifth refueling of DRESDEN 1, 96 fuel assemblies were removed from the core and were later shipped to NFS for reprocessing. Accompanying these spent fuel assemblies were 106 assemblies which had been removed from the core during the fourth refueling. The total 202 assemblies represented different initial ^{235}U enrichments and different design types⁽²³⁾ as follows:

Type	Weighted Initial Enrichment ^{235}U wt %	Number of Assemblies	Range of Exposures MWD/MTU	Cladding Material
I	1.474	165	6038 - 16966	Zircaloy
III _b	1.831	29	5924 - 14896	Zircaloy
III _f	2.253	8	7311 - 11693	Zircaloy
TOTAL		202		

The measured chemical reprocessing plant data are presented in Table B-I. There were 18 dissolution batches, 12 batches of initially enriched 1.474 fuel, 1 batch of initially enriched 1.831 fuel and 5 batches representing mixtures of the initially enriched 2.253, 1.831 and 1.474 fuels. The batches are listed by decreasing initial ^{235}U enrichment and in the case of the 12 batches of 1.474 fuel, the batches are ordered according to the final ^{235}U weight percent. The data presented in Table I have been included in the collective data analysis and have been shown in Figures 1 through 7 of the body of the report.

It was indicated in the body of the report that the plotted data, both the Pu/U data and isotopic data, evidenced a void effect. A summary of the isotopic ratio values discussed in the report are given in Table B-II. From

TABLE B-I
DRESDEN 1 FUEL LOT IV DATA SUMMARY

Batch Number	²³⁵ _U Init. Enrich.	NFS MEASURED CHEMICAL PROCESS INPUT										Total _U kgs	Total _{Pu} grams
		Uranium Isotopic Composition*				Plutonium Isotopic Composition*							
		²³⁴ _U	²³⁵ _U	²³⁶ _U	²³⁸ _U	²³⁸ _{Pu}	²³⁹ _{Pu}	²⁴⁰ _{Pu}	²⁴¹ _{Pu}	²⁴² _{Pu}			
22	2.180	0.010	1.221	0.174	99.595	0.362	72.636	19.344	6.241	1.417	784	3497	
6	1.831	0.006	0.784	0.195	99.015	0.632	64.805	23.563	8.341	2.659	793	4320	
1	1.767	0.005	0.709	0.194	99.093	0.670	63.654	24.306	8.538	2.832	1195	6621	
5	1.617	0.006	0.775	0.156	99.054	0.517	67.925	22.478	7.084	1.996	1237	5607	
21	1.596	0.006	0.755	0.152	99.087	0.506	67.461	22.692	7.151	2.190	1302	6295	
2	1.586	0.004	0.696	0.162	99.138	0.587	65.797	23.615	7.619	2.383	1241	6116	
8	1.474	0.005	0.751	0.130	99.114	0.413	70.288	21.787	6.018	1.494	1283	5093	
9	1.474	0.003	0.721	0.136	99.140	0.462	68.770	22.394	6.546	1.828	1285	5490	
12	1.474	0.005	0.718	0.137	99.140	0.486	68.773	22.536	6.437	1.769	1102	4700	
7	1.474	0.005	0.695	0.142	99.159	0.532	67.498	22.934	6.969	2.067	1246	5698	
3	1.474	0.005	0.681	0.141	99.173	0.517	67.700	22.992	6.867	1.923	872	4009	
15	1.474	0.007	0.644	0.148	99.202	0.563	65.396	23.456	7.530	2.555	1302	6745	
16	1.474	0.007	0.637	0.149	99.208	0.546	65.882	23.487	7.566	2.520	1350	6990	
18	1.474	0.007	0.636	0.150	99.207	0.553	65.618	23.652	7.523	2.654	1304	6773	
13	1.474	0.003	0.632	0.149	99.215	0.568	65.765	23.577	7.589	2.501	1309	6743	
10	1.474	0.004	0.624	0.151	99.221	0.586	65.378	23.821	7.643	2.572	1290	6646	
11	1.474	0.004	0.616	0.152	99.223	0.593	64.699	24.131	7.819	2.759	1306	6796	
20	1.474	0.007	0.611	0.155	99.227	0.616	64.858	23.938	7.772	2.817	1309	6988	
All Batches	1.550	0.005	0.704	0.153	99.138	0.547	66.572	23.185	7.361	2.335	21510	105027	
Corrected Subtotal**											21510	104504	
Flush Batches											6	2	
Sumps, Hulls											28.5	136	
TOTAL											21544	104642	

* wt % ** Analytical Bias Correction Factor for Plutonium = 0.995

TABLE B-II
SUMMARY OF ISOTOPIC RELATIONSHIPS VALUES FOR DRESDEN 1 FUEL LOT IV

VALUES OF ISOTOPIC RELATIONSHIPS										
Batch Number	Pu/U gms/T	^{235}D	$\frac{\text{Pu/U}}{^{235}\text{D}}$	$\Delta^{236}\text{U}$	$\frac{\text{Pu/U}}{\Delta^{236}\text{U}}$ ($\times 10^{-4}$)	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$	$\frac{\text{Pu/U}}{^{240}\text{Pu}/^{239}\text{Pu}}$ ($\times 10^{-4}$)	$\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{^{240}\text{Pu}^2}$	$\frac{\frac{\text{Pu/U}}{^{239}\text{Pu}} \cdot \frac{^{242}\text{Pu}}{^{240}\text{Pu}^2}}{(\times 10^{-4})}$	$\frac{\Delta^{236}}{^{235}\text{D}}$
	1	2	3	4	5	6	7	8	9	10
22	4460	0.959	4651	0.165	2.70	0.2663	1.675	0.2751	1.621	0.172
6	5448	1.047	5179	0.187	2.90	0.3636	1.498	0.3106	1.754	0.179
1	5541	1.058	5237	0.187	2.96	0.3818	1.451	0.3051	1.816	0.177
5	4533	0.842	5383	0.150	3.03	0.3309	1.370	0.2683	1.689	0.178
21	4835	0.841	5749	0.146	3.31	0.3364	1.437	0.2869	1.685	0.174
2	4928	0.890	5537	0.156	3.16	0.3589	1.373	0.2812	1.752	0.175
8	3970	0.723	5490	0.125	3.19	0.3100	1.281	0.2212	1.795	0.173
9	4272	0.763	5674	0.131	3.27	0.3256	1.312	0.2507	1.704	0.172
12	4265	0.756	5641	0.132	3.24	0.3277	1.301	0.2395	1.781	0.175
7	4573	0.779	5870	0.137	3.35	0.3398	1.346	0.2653	1.724	0.176
3	4597	0.793	5797	0.136	3.39	0.3396	1.354	0.2463	1.866	0.172
15	5180	0.830	6241	0.143	3.63	0.3559	1.455	0.3060	1.693	0.172
16	5178	0.837	6186	0.144	3.61	0.3565	1.452	0.3010	1.720	0.172
18	5117	0.838	6107	0.145	3.54	0.3604	1.420	0.3113	1.644	0.173
13	5151	0.842	6118	0.144	3.59	0.3585	1.437	0.2959	1.741	0.171
10	5152	0.850	6061	0.146	3.54	0.3644	1.414	0.2963	1.739	0.172
11	5204	0.858	6056	0.147	3.55	0.3730	1.395	0.3065	1.698	0.171
20	5338	0.863	6186	0.150	3.57	0.3691	1.446	0.3188	1.674	0.174
			$5952 \pm 4.2\%$		$3.46 \pm 4.6\%$		$1.384 \pm 4.6\%$		$1.732 \pm 3.4\%$	$0.173 \pm 0.9\%$
All Batches	4858	0.846	5742	0.147	3.31	0.3483	1.395	0.2892	1.680	0.174

*Mean values and one standard deviations.

Initial ^{236}U values: For $^{235}\text{U}_0 = 2.180$, $^{236}\text{U}_0 = 0.0088$
 For $^{235}\text{U}_0 = 1.831$, $^{236}\text{U}_0 = 0.0070$
 For $^{235}\text{U}_0 = 1.767$, $^{236}\text{U}_0 = 0.0067$
 For $^{235}\text{U}_0 = 1.617$, $^{236}\text{U}_0 = 0.0064$
 For $^{235}\text{U}_0 = 1.596$, $^{236}\text{U}_0 = 0.0059$
 For $^{235}\text{U}_0 = 1.586$, $^{236}\text{U}_0 = 0.0059$
 For $^{235}\text{U}_0 = 1.474$, $^{236}\text{U}_0 = 0.0054$

Table B-II, it is seen that the $(\text{Pu}/\text{U})/^{235}\text{D}$, $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$ and $(\text{Pu}/\text{U})/(^{240}\text{Pu}/^{239}\text{Pu})$ ratio values varied with changes in the ^{235}U enrichment for batches 1, 2, 5, 6, 21 and 22 as expected. The $(\text{Pu}/\text{U})/^{235}\text{D}$ and $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$ values increased and the $(\text{Pu}/\text{U})/(^{240}\text{Pu}/^{239}\text{Pu})$ values decreased as the initial ^{235}U enrichment decreased from 2.180 to 1.586 weight percent. The ratio $(\text{Pu}/\text{U})/(^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2)$ appeared to be more affected by measurement variations in the ^{242}Pu values than by initial enrichment changes and varied about an average value of 1.72×10^4 . The mixing effect of plutonium isotopes may be the cause for some of the variations seen that were lower than 1.72×10^4 . However, the variation above 1.80×10^4 was reasonably explained as the result of measurement variations in ^{242}Pu . The mixing effect would not be as evident in this case as with HUMBOLDT BAY fuels because all the data are on the more linear parts of the burnup paths even though some of the exposures are in the 6000 MWd/MTU range. That is, the lower enriched fuels arrive at linear parts of the plutonium isotopic burnup paths sooner than higher enriched fuels. Thus mixing assemblies whose exposures differed more than 3000 MWd/MTU does not have as strong an effect as in the case of HUMBOLDT BAY fuels. The last ratio given in Table B-II, $\Delta^{236}\text{U}/^{235}\text{D}$, appeared to be reasonably constant for batches 1, 2, 5, 6, 21 and 22 and indicated that the ^{235}U and ^{236}U measured values were measured correctly. The assumed initial ^{236}U weight percents used to determine $\Delta^{236}\text{U}$ are listed at the bottom of Table B-II.

DRESDEN 1 fuel of an initial enrichment and design similar to the initial enriched 1.474 weight percent fuels has been processed in previous campaigns. The isotopic ratio values that resulted are as follows:

Fuel Lot	$\frac{\text{Pu/U}}{^{235}\text{D}}$	$\frac{\text{Pu/U}}{\Delta^{236}\text{U}}$	$\frac{\text{Pu/U}}{^{240}\text{Pu}/^{239}\text{Pu}}$	$\frac{\text{Pu/U}}{^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2}$	$\frac{\Delta^{236}\text{U}}{^{235}\text{D}}$
I	6234 $\pm$ 1.4%	3.52 $\pm$ 1.6%	1.563 $\pm$ 1.1%	1.739 $\pm$ 1.4%	0.177 $\pm$ 1.0%
II	6070 $\pm$ 1.6%	3.49 $\pm$ 2.9%	1.451 $\pm$ 1.9%	1.734 $\pm$ 2.1%	0.173 $\pm$ 1.3%

It is seen that the mean values given for $(\text{Pu/U})/^{235}\text{D}$, $(\text{Pu/U})/\Delta^{236}\text{U}$, $(\text{Pu/U})/(^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2)$ and $\Delta^{236}\text{U}/^{235}\text{D}$ are reasonably the same for fuel lots I and II. The one standard deviations of the mean values are given and the mean values agree to within two standard deviations. The $(\text{Pu/U})/(^{240}\text{Pu}/^{239}\text{Pu})$ values represent the slopes of two straight lines (see discussion in body of report) and should not be the same. The ratio values from fuel lots I and II thus indicate what the values should be for assemblies of fuel lot IV of similar enrichment, design and irradiation history.

It is seen by comparing the above values with the batch values for 1.474 fuel in Table B-II that the $(\text{Pu/U})/^{235}\text{D}$, $(\text{Pu/U})/\Delta^{236}\text{U}$ and $(\text{Pu/U})/(^{240}\text{Pu}/^{239}\text{Pu})$ values of batches 10, 11, 13, 15, 16, 18 and 20 agreed with the respective ratio values for fuel lots I and II. It is also seen that five batches, batches 3, 7, 8, 9 and 12, have lower values for these ratios. This variation was correlated directly to core positions of the assemblies involved and was determined to result from probable void variations as a function of core position in the DRESDEN 1 reactor. The correlation to core positions is explained in the next several paragraphs and is summarized in Table B-III.

As indicated earlier, a total of 165 type I assemblies were processed. Of these assemblies, 89 were charged to the core during 1960, 67 of which resided in zone 3 or the outer region of the core. The core regions are defined in Figure B-1. Only 3 of the 89 assemblies were charged to zone 0, or the center of the core, and the remaining 19 were charged either to zone 1

TABLE B-III

SUMMARY OF CORE POSITIONS AND EXPOSURES OF ASSEMBLIES BY DISSOLUTION BATCH

Batch Number	Core Position of Assemblies				Type and Number of Assemblies Per Batch				Exposure Distribution Summary Mwd/MTU	Average Weighted Exposure Mwd/MTU
	Zone				I	III _b	III _f	Total		
	3	2	1	0						
22	0		3	5	0	2	6	8	3 at 7300 to 7800; 5 at 10500 to 11700.	9810
6	0	1	1	6	0	8	0	8	1 at 8000; 7 at 13400 to 14700.	13148
1	1		2	9	2	10	0	12	1 at 8400; 11 at 12300 to 14600.	13388
5	6	3		3	7	5	0	12	1 at 5900; 7 at 8200 to 10'00; 4 at 11030 to 14300.	10419
21	3	4	1	4	10	0	2	12	5 at 8600 to 9500; 7 at 11'00 to 13600.	10788
2	7			5	8	4	0	12	6 at 9500; 1 at 11600; 5 at 14000.	11550
8	10	2			12	0	0	12	All 7400 to 10000.	8492
9	9	3			12	0	0	12	All 7600 to 10200.	9077
12	8	2			10	0	0	10	All 7800 to 10400.	9088
7	8	3		1	12	0	0	12	1 at 6000; 1 at 8400; 9 at 9200 to 10200; 1 at 17000.	9680
3	7	1			8	0	0	8	All 9460 to 10200.	9792
15	3	3	2	4	12	0	0	12	4 at 9100 to 9400; 8 at 10300 to 13300.	11252
16	1	5	5	1	12	0	0	12	All 9000 to 12900.	11041
18	2	5	3	2	12	0	0	12	7 at 9400 to 10100; 5 at 11600 to 13800.	10704
13	2	2	4	4	12	0	0	12	3 at 8800 to 9600; 9 at 10600 to 12900.	11423
10	5	2		5	12	0	0	12	3 at 8900 to 9700; 9 at 11400 to 13300.	11712
11	2		2	8	12	0	0	12	2 at 7700; 10 at 12000 to 13700.	11954
20	3	3	2	4	12	0	0	12	1 at 7600; 1 at 9876; 10 at 11300 to 13500.	11881

* See Figure B-1 for definition of zones.

51 52 53 54 55 56 57 58 59 60 61 62 63 64 65 66 67 68 69 70 71 72 73 74

FIGURE B-1. DRESDEN 1 CORE ZONES DEFINED FOR THE PURPOSES OF THIS REPORT

or zone 2. The 89 assemblies resided in the core until 1967 or 1968 and, although a number of them resided in more than one position, they were generally not moved to a position in a different zone. Thus, 67, or a third of all the assemblies represented a core region where the void history during operation was probably lower than the void history in other zones of the core.

The remaining 76 type I assemblies were charged during 1963 and, except for 2 assemblies, they were charged to zones 0, 1 and 2. Sixty-eight of the 76 assemblies were discharged in 1967 and the remaining 8 were discharged in 1968. Of the type III_b and III_f assemblies, 28 were charged during 1964 and 9 were charged during 1965 to zones 0 and 1 of the core. Two assemblies were discharged in 1967 and 35 in 1968.

The core history information thus indicated that for the first time a large number of zone 3 assemblies from DRESDEN 1 were reprocessed. Earlier discharges had generally included assemblies irradiated in zones 0, 1 and 2 of the core. The order of processing during the campaign was random, that is, the assemblies in a batch represented different assembly types as well as assemblies of different irradiation histories. The probability that any one batch would contain all zone 3 assemblies was therefore small. The number of assemblies from zone 3 dissolved per batch is given in Table B-III along with a summary of the core positions and assembly types of all the assemblies involved in this campaign.

It is seen from Table B-III that the assemblies included in batches 3, 7, 8, 9 and 12 were predominately zone 3 assemblies. Zone 3 assemblies were also included in all other 1.474 fuel batches but the number was always less than half the total number of assemblies in each batch. In addition to

the direct correlation to core position, the plotted isotopic data which represented batches 3, 7, 8, 9 and 12 appeared to be on different burnup curves (see Figure 7 of the main report) which were lower than the burnup curve for the remainder of the 1.474 fuel batches from fuel lot IV and for the previous 1.474 batches from fuel lots I and II. This phenomenon can result from void variations, that is, the assemblies resided in regions of lower void. Thus, the isotopic data substantiated the variations noted for the $(\text{Pu}/\text{U})/^{235}\text{D}$, $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$ and $(\text{Pu}/\text{U})/(^{240}\text{Pu}/^{239}\text{Pu})$ ratio values for 1.474 fuels.

The $(\text{Pu}/\text{U})/(^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2)$ ratio for the 12 batches of 1.474 fuels was seen to have a mean value of 1.732×10^4 and a standard deviation of 3.4%. The mean value agreed with the mean values of previous fuel lots I and II. Comparing the standard deviations of other data sets determined for $(\text{Pu}/\text{U})/^{235}\text{D}$, $(\text{Pu}/\text{U})/\Delta^{236}\text{U}$, and $\Delta^{236}\text{U}/^{235}\text{D}$ (see Figures 1, 2 and 5 of the body of the report), a 3% standard deviation appeared to be an upper bound for the case where batch data points were describing a linear function. The standard deviation in this instance was larger but was considered to be within reason. The cause of the larger deviation was probably due to the random variations in ^{242}Pu measurement rather than a mixing effect. Three considerations supported this conclusion, 1) the data were on a linear portion of the burnup curve (see Figure 7 of the main report); 2) batches which contained assemblies of exposure difference greater than 3000 MWd/MTU were not always seen to be the batches having low relationship values (see Tables B-II and III); and 3) remeasured ^{242}Pu values that were provided concurrent with the NFS ^{242}Pu values differed

on the average by 3.4%, indicating a larger random variation in ^{242}Pu than the expected 1.5 to 2.0%.

This was the second time a standard deviation of more than 3% was encountered when applying the $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ ratio to BWR fuel. The first occurred for DRESDEN 1 fuel lot III where the fuel was of a different initial enrichment (2.50 weight percent ^{235}U) and of a different design (stainless steel cladding). The mean value for the ratio was 1.750×10^4 and the standard deviation was 3.3%. However, the cause in this case was more likely due to the mixing effect instead of random variations in ^{242}Pu because the initial enrichment and the burnup of the fuel were similar to HUMBOLDT BAY fuels. In addition, the ^{242}Pu measurements of fuel lots I and II which were processed during the same campaign as lot III fuels did not evidence larger random variations.

Based on the observations that the isotopic data for batches 3, 7, 8, 9 and 12 had evidenced a void effect and that the ratio $(\text{Pu}/\text{U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ was reasonably consistent, the Pu/U ratio values of batches 3, 7, 8, 9 and 12 were considered to be verified. The last ratio shown in Table B-II, $\Delta^{236}\text{U}/^{235}\text{U}$, for the 12 batches of 1.474 fuel was also in good agreement with the mean values from fuel lots I and II. The mean value was $1.73 \times 10^4 \pm 0.9\%$ and the good agreement indicated that the ^{236}U and ^{235}U values for the 12 batches were correct. The values of the five isotopic ratios discussed in the body of the report were thus seen to be in agreement with previous DRESDEN 1 spent fuel data. The variations seen in three of the ratios were determined to be consistent with void variations present in a BWR. As a result, all the measured batch Pu/U values were considered to have been verified.

To verify the total plutonium measured for fuel lot IV, a weighted Pu/U value of 4857 grams/tonne was used in applying the Pu/U ratio method. This Pu/U value included flush batch, sump and hull measurements given in Table B-I. (The Pu/U value given in Table B-II, 4858 grams/tonne, represented the corrected subtotal values from Table B-I.) The calculated total uranium from preprocessing records was given as 21.488 tonnes. Multiplying the weighted Pu/U ratio by the calculated total uranium gave the result of 104,368 grams of plutonium as having been produced in DRESDEN 1 fuel lot IV fuels. The agreement with the measured total plutonium of 104,642 grams was to 0.26%.

The product data available permitted the comparison of ^{235}U and $^{239} + ^{241}\text{Pu}$ weight percents from total input and total product. The product data are given in Table B-IV along with the comparison of input and product totals. There were 6 uranium product batches and 5 plutonium product batches. From Table B-IV, it can be seen that the ^{235}U and $^{239} + ^{241}\text{Pu}$ weight percents of input and product totals agree to 0.15%. The total plutonium of DRESDEN 1 fuel lot IV measured at input was thus considered validated.

TABLE B-IV

DRESDEN 1 FUEL LOT IV PRODUCT DATA AND COMPARISON TO INPUT DATA

Shipment Number	^{235}U wt %	Total U kgs	Shipment Number	$^{239} + ^{241}\text{Pu}$ wt %	Total Pu grams
1	0.726	4057	1	73.768	20475
2	0.725	4216	2	74.913	18855
3	0.663	4281	3	73.633	26590
4	0.703	2874	4	73.705	29088
5	0.706	2780	5	75.773	6146
6	0.702	3077			
		TOTAL PRODUCT ISOTOPIC VALUES			
	0.704			74.047	
		TOTAL INPUT ISOTOPIC VALUES			
	0.704			73.933	