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APPLICATION OF ISOTOPIC CORRELATION
SAFEGUARD TECHNIQUES TO VERIFY THE PLUTONIUM
CONTENT OF HUMBOLDT BAY SPENT FUEL

CONTRACT NUMBER ACDA/WEC-216

PREPARED FOR
U.S. ARMS CONTROL AND DISARMAMENT AGENCY

PREPARED BY

PACIFIC NORTHWEST LABORATORIES
a division of
BATTELLE MEMORIAL INSTITUTE
3000 Stevens Drive
Richland, Washington 99352

MASTER

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by

D. E. Christensen

MARCH 20, 1972

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PACIFIC NORTHWEST LABORATORIES
a division of
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ABSTRACT

This report presents a specific example of applied isotopic correlation safeguard techniques. The techniques were applied to measured chemical reprocessing plant input and product data from the processing of Humboldt Bay Reactor spent fuels. The objective was to verify the measured Pu/U value of each input batch. Humboldt Bay is a boiling water reactor and the $(\text{Pu/U})/^{235}\text{D}$ values of 6 batches of fuel evidenced a void effect. A significant development was that the isotopic correlation techniques were applied successfully through the use of several isotopic relationships each applicable where another was limited.

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APPLICATION OF ISOTOPIC CORRELATION SAFEGUARD TECHNIQUES TO VERIFY THE PLUTONIUM CONTENT OF HUMBOLDT BAY SPENT FUEL

I. INTRODUCTION

Humboldt Bay reactor spent fuel data represent the most complete data base for applied safeguards via isotopic correlation safeguards techniques that has been received to date. Information has been made available from Pacific Gas and Electric (PG&E) whereby each fuel assembly was followed from the point of being charged in the Humboldt Bay reactor core to its discharge a few years later and up to the point of storage and dissolution of the assembly at Nuclear Fuel Services, Inc. (NFS). Calculated data which included predicted spent fuel composition per assembly were provided as part of the preprocessing data received. Following the collection of these data, the measured compositions of each dissolved fuel batch obtained during chemical reprocessing were provided by NFS making the collection of pertinent data the most complete.

This report demonstrates how isotopic correlation techniques can be used by presenting a specific example of applied isotopic correlations. Presented also are how the techniques were used and under what conditions the techniques were applied to Humboldt Bay spent fuels. A more general and complete presentation of the use of isotopic correlations is contained in Reference 1.

Four general aspects of isotopic correlations are treated in the report. First, an example is presented which shows how preprocessing data would be used by a safeguards agency in conjunction with isotopic correlations. Secondly, the influence of voids in boiling water

reactors on isotopic relationships useful in safeguards is treated. The third subject of this report is the presentation of results from the detailed development of isotopic correlations from boiling water reactor data. Finally, it is shown how isotopic correlation safeguards techniques are strengthened through the use of available preprocessing data. This is particularly true in a case where no previous experimental data exist as was the case with Humboldt Bay fuel.

The Humboldt Bay campaign represented a routine test case in that isotopic correlations were applied with success. However, the campaign also permitted an indepth analysis since linear relationships other than the plutonium to uranium ratio (Pu/U) versus the depletion of ^{235}U due to irradiation (^{235}D) were developed and used to augment the ratio (Pu/U)/ ^{235}D . The analysis of Humboldt Bay spent fuel data 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." PG&E and NFS have provided the data presented and their contributions to this report are gratefully acknowledged.

II. SUMMARY AND CONCLUSIONS

The data pertaining to Humboldt Bay spent fuels, both measured and preprocessing, have made a valuable contribution to the development of isotopic correlations. The data have been particularly useful to investigate the effect of voids in a boiling water reactor on isotopic relationships, since void effects were evident both from the Pu/U and isotopic data.

The successful application of isotopic correlations to Humboldt Bay was seen to depend on the use of more than one isotopic relationship. The $(\text{Pu}/\text{U})/^{235}\text{D}$ relationship was determined to be void dependent and thus not constant for single enrichment fuels where voids were changing. The $(\text{Pu}/\text{U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ relationship was determined to be void independent but was seen to vary when assemblies differing in exposure by more than 3000 MWD/MTU were mixed together in a dissolution batch. However, by determining which assemblies had resided in an environment of like void during irradiation, the $(\text{Pu}/\text{U})/^{235}\text{D}$ relationship was used to verify the measured Pu/U values of the batches containing these assemblies. Where the exposure of assemblies included in a batch varied less than 3000 MWD/MTU but differing voids were in effect, the $(\text{Pu}/\text{U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ relationship was used to verify the measured Pu/U values.

Other relationships not presented substantiated these results.⁽²⁾ A significant development was the successful application of the concurrent use of several relationships, each applicable where another may be limited. In this instance the use of a single relationship would not have been as successful. In an actual safeguards application using isotopic correlations under similar circumstances, independent measurement of a larger number of the input batches would have been needed if $(\text{Pu}/\text{U})/^{235}\text{D}$ were

the only relationship that could have been formed. Thus, it was seen that all isotopic data normally measured during reprocessing were vital to the success of isotopic correlations in the case of Humboldt Bay.

The success of isotopic correlations was also due in part to the availability of preprocessing data prior to the reprocessing. Isotopic correlation techniques were strengthened by the analysis of these preprocessing data. As a result, rather than performing safeguards functions in a relatively uninformed environment, the functions were identified prior to dissolution. Since most of the data pertaining to each individual fuel assembly during its lifetime was made available, the report is an example of maximum use of already available data to perform safeguard functions. Also, as a result of using the data in a safeguards capacity, the user is oriented in concept to the total fuel cycle rather than oriented to the processes of a single facility.

Since this was the first campaign involving Humboldt Bay fuels, all the preprocessing data were examined and found useful in indicating variations in the physical and nuclear processes which affect isotopic composition changes. In future campaigns, such a complete analysis as performed for the first campaign may not be needed. This will be the case if it is possible to verify that the conditions affecting future spent fuels are the same as the conditions affecting the fuel in this campaign. The information which was most useful in this application of preprocessing data was:

- a). Final total uranium for each assembly and the initial ^{235}U enrichment.
- b). Assemblies which make up each batch.
- c). Calculated exposures and isotopic data per assembly.
- c). Core loading diagrams, reactor power history and storage location at reprocessing plant.

III. APPLIED ISOTOPIC CORRELATIONS

A. Outline of Principles and Methods Used

The principle upon which the use of isotopic correlations is based is the physical law that the plutonium in irradiated fuel is inherently related to other quantities that also change during irradiation, namely the amount of uranium and the uranium and plutonium isotopic compositions. The application is accomplished by determining relationships from spent fuel data measured at a chemical reprocessing plant that relate the measured plutonium in the form of Pu/U values directly to other measured quantities. Particularly useful have been linear relationships as a function of increasing exposure. Use of the relationships has been termed isotopic correlations.

The objective in applying isotopic correlations to any set of spent fuel data is to verify the validity of measured Pu/U values so that the Pu/U ratio method can be used with greater confidence in verifying the plutonium input. This is done by using several levels of comparison of isotopic relationship values involving the Pu/U values as follows:

1. Compare relationship values from previously measured batches of spent fuel from a given reactor to those being measured. The previous data define what the relationship values are and the conditions under which the values remain the same. The current values should agree with the previously measured values when irradiation conditions and the design of the assemblies have been the same for both sets of fuel.
2. Compare relationship values for internal self-consistency or agreement among the batches representing fuel for which irradiation conditions and design were equivalent. The measured values should agree with the average value within two standard deviations.

3. Compare relationship values from Agency and operator measurements. The Agency measurement values should be the same as those derived from operator measurements, within the uncertainties involved. One basic reason for using isotopic correlations is to reduce the number of batches which the Agency measures in verifying the operator batch measurements.
4. Compare relationship values from measurements with those generated from preprocessing data. These values are calculated values using computerized physics burnup codes.

In the case of Humboldt Bay spent fuels there were no previously measured data from which relationship values could be determined. This example is typical of the situation during the years to come as each new reactor comes on line. In these situations, greater emphasis would be given levels of comparison 2, 3 and 4. For example, a few more input batches would be selected for Agency measurement and a more detailed analysis of the preprocessing data would be done.

An example of the safeguards use of preprocessing data, including the selection of input batches for Agency measurement, is presented in Appendix A. The preprocessing data for Humboldt Bay fuel were received in March and April, 1971, prior to the beginning of the processing of the fuels the last of April, 1971.

The selection of batches for Agency measurement was hypothetical in that no arrangements were made to obtain such samples. However, Nuclear Audit and Testing Company (NATCO) under contract to PG&E was acting as third party surveillance and measured all input batches. These data have been received and are used, for the purposes of this example, as though they were Agency measurements. Thus, prior to the processing of Humboldt Bay Fuels, emphasis was given levels 3 and 4 above, with the result that a working knowledge of

isotopic correlations as they pertain to Humboldt Bay was available and was ready to be put to use with the receipt of the measured data.

Once the measured data were available and isotopic correlations had been applied, the last steps used to verify the plutonium input for this example would be (a) to use the Pu/U ratio method,⁽³⁾ that is, to multiply verified Pu/U values by verified uranium (from calculated data) per batch and campaign to determine the plutonium independently and (b) to compare plutonium and uranium isotopic compositions and isotopic relationships from input and product totals for agreement.

B. Humboldt Bay Measured Data

PG&E sent to NFS a total of 270 spent fuel assemblies for reprocessing. The assemblies were irradiated in the Humboldt Bay Reactor, a boiling water reactor, from 1963 through 1970, and had accumulated exposures varying from an estimated 5800 to 16000 MWD/MTU. A total of 20.984 metric tons of slightly enriched uranium fuels were involved containing an estimated 90.4 kgs. of plutonium. The assemblies were dissolved in 23 dissolver batches which resulted in 20 3D-1 accountability tank batches from which representative samples were obtained for measurement by NFS. The initial ^{235}U enrichments and the makeup of each batch are given in Table I. The assemblies were of three different initial enrichments, 2.578 weight percent for type A assemblies, 2.310 weight percent for type B assemblies and 2.103 weight percent for type C assemblies. Type A assemblies were stainless steel clad and type B and C were Zircaloy-2 clad. Further details are given in Appendix A.

The measured data received from NFS included not only the total uranium, ^{235}U , total plutonium and $^{239+241}\text{Pu}$ gram weights but also the complete isotopic measurements of each input and product batch which are routinely

TAB E 1
HUMBOLDT BAY MEASURED DATA SUMMARY

Batch Makeup and Initial Enrichments															Isotopic Relationship Values				
NFS Measured Chemical Process Input																			
3D-1 Batch Number	Number and Type of Assemblies			235U Initial Enrichment	Uranium Isotopic Composition				Plutonium Isotopic Composition					Total Uranium	Total Plutonium	Pu/U		235D	
					234U	235U	236U	238U	238Pu	239Pu	240Pu	241Pu	242Pu			Pu/U	235D	Pu/U	235D
	A	B	C	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	kgs	Grams	Grams/Tonne	wt%	Gms/Tonne	wt%
29*	10	0	0	2.578	0.014	1.919	0.128	97.938	0.165	83.603	12.996	2.919	0.318	810	2232	2756	0.658	4181	
28	11	0	0	2.578	0.014	1.874	0.134	97.978	0.183	82.676	13.600	3.171	0.370	873	2462	2820	0.704	4006	
25	11	0	0	2.578	0.014	1.860	0.140	97.985	0.180	82.051	14.120	3.238	0.410	859	2512	2924	0.718	4073	
26	11	0	0	2.578	0.014	1.847	0.143	97.996	0.256	81.759	14.138	3.417	0.430	863	2704	3133	0.731	4286	
27	11	0	0	2.578	0.015	1.837	0.144	98.004	0.199	81.925	13.921	3.504	0.451	858	2821	3288	0.741	4437	
17	12	0	0	2.578	0.014	1.810	0.151	98.025	0.212	81.321	14.231	3.743	0.493	924	3152	3455	0.768	4498	
4	24	0	0	2.578	0.014	1.764	0.161	98.061	0.240	79.947	14.991	4.202	0.620	1854	7268	3920	0.814	4816	
16	12	0	0	2.578	0.013	1.751	0.164	98.072	0.239	79.928	14.976	4.215	0.641	937	3690	3938	0.826	4762	
14	12	0	0	2.578	0.013	1.709	0.171	98.106	0.279	78.813	15.661	4.526	0.720	919	3693	4018	0.869	4624	
15	12	0	0	2.578	0.014	1.706	0.171	98.109	0.351	78.316	15.928	4.632	0.772	942	3842	4079	0.872	4677	
13*	12	0	0	2.578	0.014	1.683	0.173	98.130	0.300	78.309	15.863	4.729	0.799	910	3855	4236	0.895	4733	
22*	0	12	0	2.310	0.011	1.250**	0.205	98.534	0.462	70.683	19.992	7.217	1.647	928	4821	5195	1.060	4901	
21	0	12	0	2.310	0.012	1.223	0.210	98.556	0.488	69.786	20.629	7.333	1.765	901	4762	5219	1.087	4801	
20*	0	12	0	2.310	0.011	1.167	0.215	98.607	0.525	69.043	20.680	7.892	1.861	884	4772	5398	1.143	4723	
5	11	1	0	2.557	0.014	1.706	0.167	98.113	0.298	78.293	15.947	4.672	0.789	931	3660	3931	0.851	4620	
12	23	1	0	2.567	0.015	1.655	0.175	98.156	0.317	77.573	16.308	4.937	0.865	1879	7956	4234	0.912	4643	
23*	4	6	0	2.402	0.012	1.441	0.186	98.362	0.397	73.517	18.524	6.229	1.333	874	4000	4577	0.961	4762	
2	13	8	3	2.433	0.013	1.423	0.192	98.873	0.442	72.968	18.480	6.682	1.428	1805	8657	4796	1.010	4749	
19	0	10	2	2.276	0.011	1.106	0.220	98.662	0.565	68.078	21.066	8.274	2.018	882	4918	5576	1.168	4774	
18*	0	9	3	2.258	0.010	1.087	0.223	98.681	0.575	67.544	21.191	8.557	2.134	903	5184	5741	1.171	4903	
All Batches	189	73	8	2.495	0.013	1.593	0.174	98.220	0.359	75.805	17.173	5.561	1.102	20736	86941	4193	0.902	4648	
24, 30, 31 Sumps	Flush Batches													15	38				
Hulls														4	2				
														41	25				
Totals						1.593								20796	87006	4184	0.902	4639	

* Batches identified from preprocessing evaluation for Agency measurement as an example of safeguards application of isotopic correlations.

** Was reported as 1.205 by NFS. Later reported as 1.250.

measured at this plant. The data for each input batch are also given in Table I where all isotopic values are shown as weight percents rather than gram amounts. (The ^{235}U weight percent of batch 22 was changed from a reported value of 1.205 to 1.250 since the uranium weight percents for batch 22 did not add up to 100 percent. This was probably due to a typing or transcription error. Adding isotopic weight percents is a simple check which uncovers such errors from time to time. The 1.250 value was later substantiated when the NATCO measured values were received.)

The measured input batch data are identified by the 3D-1 batch numbers rather than the dissolver batch numbers, see Appendix A, and the ordering of the data in Table I has been done according to the initial enrichment and final ^{235}U content per batch. That is, all batches representing type A materials only have been grouped together and ordered according to decreasing ^{235}U weight percent. This essentially orders the batches of a given initial ^{235}U enrichment according to increasing exposure. Isotopic relationships, such as the relationship between decreasing ^{235}U weight percent and increasing ^{236}U weight percent or between the increasing Pu/U values are thus indicated from the tabulated data. Following the 2.578 weight percent ^{235}U initially enriched fuels, the 2.310 weight percent ^{235}U initially enriched batches are listed after which the batches which included assemblies of differing initial enrichments are listed. The initial enrichment of the mixed batches shown were determined from proper weighting of the initial weights of the assemblies involved. The same approach in tabulating input batch data is followed throughout the report.

C. Isotopic Correlations Applied to Measured Data

Additional information given in Table I includes the identification of the batches selected for postulated Agency measurement. These batches were identified during the evaluation of the preprocessing data. Also, the Pu/U values, ^{235}D values and $(\text{Pu/U})/^{235}\text{D}$ values are shown. From past experience and theoretical considerations, it was known that $(\text{Pu/U})/^{235}\text{D}$ varied with initial enrichment and various other design parameters. Such was the case as indicated by the $(\text{Pu/U})/^{235}\text{D}$ values in Table I, where the values increase as a function of decreasing initial enrichment. The increase was not as large as normally experienced when enrichments only are changed due to the change in cladding from stainless steel to Zircaloy-2. This change compensated for some of the normal increase which accompanies decreasing initial enrichment. However, the variations observed in the $(\text{Pu/U})/^{235}\text{D}$ values for a given initial enrichment had not been encountered before. As can be seen from Table I for batches of an initial enrichment of 2.578 weight percent, the $(\text{Pu/U})/^{235}\text{D}$ values for batches 17, 25, 26, 27, 28 and 29 were markedly lower than the other values shown. However, rather than reject the values as being in error (the lowest being approximately 20 percent low), sufficient evidence had been made available to indicate that an indepth look at void effects in boiling water reactors should be done as part of the data consistency analysis (comparison level 2, above). For example, the preprocessing data had indicated that most of the assemblies which were included in these batches had resided in the outer regions of the core (see Appendix A) where it was logical to expect that the voids were different than in the center of the core.

1. Void Considerations

Theoretical arguments indicate that as voids in a boiling water reactor increase, $(\text{Pu/U})/^{235}\text{D}$ values increase. Thus, the variation from high to low voids as a function of core position could cause $(\text{Pu/U})/^{235}\text{D}$ values to decrease

as the tabulated results do in Table I. However, if $(\text{Pu}/\text{U})/^{235}\text{D}$ values change due to void variations, evidence of the void variations should be substantiated purely from the isotopic data as well. The consistency evaluations were therefore also used to substantiate that a void effect was present and evident in the isotopic data.

The void effect was evident from the isotopic data, particularly through the use of, but not limited to, ^{239}Pu , ^{240}Pu and ^{242}Pu isotopic data. The methods whereby the void effect was verified are outlined in Appendix B. Evidence was thus available from the isotopic data which substantiated the variations in the $(\text{Pu}/\text{U})/^{235}\text{D}$ values for initially enriched 2.578 weight percent fuel.

It also follows that if the void effect can be observed by appropriate isotopic parameters, the effect will cancel when the Pu/U ratio and an appropriate isotopic ratio are considered, one versus the other. Therefore, the measured isotopic data were examined to find a parameter which when plotted versus the Pu/U ratio would result in a linear function insensitive to voids. It was determined that the isotopic ratio $(^{239}\text{Pu} \cdot ^{242}\text{Pu})/(^{240}\text{Pu}^2)$ gave the desired parameter. These developments are also explained in greater detail in Appendix B. (A theoretical treatment of this effect is given in Reference 4.) The results provided a linear combination, $(\text{Pu}/\text{U})/(^{239}\text{Pu} \cdot ^{242}\text{Pu}/^{240}\text{Pu}^2)$, as an excellent test for the consistency of the Pu/U ratio (level of comparison #2) when compared to the ^{239}Pu , ^{240}Pu and ^{242}Pu isotopic data. Thus, the consistency analysis results indicated that linear functions were available from boiling water reactor data, even though the Pu/U versus ^{235}D may not be linear due to void variations. The need to investigate all forms of linearity from data of different reactor types in addition to Pu/U versus ^{235}D was therefore apparent.

Thus, the isotopic data confirmed the void effect and provided a linear function insensitive to void to test the consistency of the Pu/U data as well.

The $(\text{Pu}/\text{U})/^{235}\text{D}$ ratio augmented by the $(\text{Pu}/\text{U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ ratio showed that the Pu/U ratios were consistent and measured correctly based on the consistency of all the measured data. The analysis was then ready to proceed to the use of independent measurements and the final verification of the measured plutonium.

2. Postulated Agency Data

The measurement data for each batch by NATCO have been received and these data are used in place of the hypothetical Agency measurements for the batches indicated in Table I. All the NATCO measured data along with the $(\text{Pu}/\text{U})/^{235}\text{D}$ relationship values are given in Table II and those particular input batches identified for Agency measurement during the preprocessing evaluation are indicated. The data have been arranged in the same order as the NFS data in Table I for easy comparison.

The basis of comparison (level of comparison #3 above) between measured (NFS) data and the postulated Agency (NATCO) data was that the same relationship values must result, within measurement errors, from both measurements. The fact that certain relationship values are the same regardless of exposure level for all batches of fuel of a given initial enrichment eliminates the need to measure independently every batch. In this case, where void had an effect on the $(\text{Pu}/\text{U})/^{235}\text{D}$ relationship, another relationship which was insensitive to voids was used in concert with $(\text{Pu}/\text{U})/^{235}\text{D}$. The relationship used was $(\text{Pu}/\text{U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$. If there had been access to only the data from which to form $(\text{Pu}/\text{U})/^{235}\text{D}$, then hypothetically, there would have been a need to measure independently several more batches because these values for the 2.578 weight percent initial enrichment fuel were not constant. However, since $(\text{Pu}/\text{U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ values were constant, this relationship replaced the $(\text{Pu}/\text{U})/^{235}\text{D}$ relationship in checking the consistency between the two sets of

TABLE II
NATCO MEASURED CHEMICAL PROCESS INPUT

3C-1 Batch Number	URANIUM ISOTOPIC COMPOSITION				PLUTONIUM ISOTOPIC COMPOSITION					Total Uranium Kgs	Total Plutonium Grams	Pu/U Grams Tonne	235D wt%	Pu/U 235D Gms/tonne wt%
	239U	235U	236U	238U	238Pu	239Pu	240Pu	241Pu	242Pu					
	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%					
29*	0.014	1.916	0.128	97.943	0.165	83.681	12.942	2.897	0.315	819	2246	2742	0.662	4143
28	0.014	1.874	0.131	97.983	0.168	82.799	13.541	3.127	0.365	875	2453	2803	0.704	3982
25	0.014	1.862	0.136	97.988	0.196	82.163	14.022	3.210	0.409	850	2517	2961	0.716	4136
26	0.014	1.864	0.145	97.977	0.186	81.917	14.100	3.377	0.420	862	2683	3112	0.714	4359
27	0.015	1.836	0.142	98.008	0.190	81.834	14.030	3.492	0.454	855	2824	3303	0.742	4451
17	0.014	1.822	0.151	98.014	0.214	81.240	14.292	3.759	0.495	927	3227	3481	0.756	4605
4	0.014	1.768	0.162	98.057	0.246	80.054	14.941	4.142	0.618	1885	7306	3876	0.810	4785
16	0.013	1.754	0.163	98.070	0.250	79.950	14.982	4.200	0.618	940	3658	3891	0.824	4723
14	0.013	1.703	0.169	98.114	0.261	78.830	15.686	4.505	0.711	925	3715	4016	0.875	4590
15	0.014	1.707	0.170	98.110	0.294	78.389	15.920	4.623	0.774	945	3829	4052	0.871	4652
13*	0.014	1.692	0.172	98.122	0.286	78.353	15.894	4.688	0.779	914	3862	4225	0.886	4769
22*	0.010	1.250	0.202	98.538	0.467	70.686	19.961	7.234	1.652	927	4810	5189	1.060	4895
21	0.010	1.225	0.204	98.561	0.511	69.782	20.602	7.329	1.776	900	4713	5237	1.085	4826
20*	0.010	1.181	0.215	98.594	0.520	69.018	20.719	7.910	1.833	875	4757	5437	1.127	4815
5	0.014	1.718	0.166	98.102	0.296	78.152	16.089	4.681	0.792	936	3676	3927	0.839	4681
12	0.014	1.678	0.176	98.132	0.315	77.612**	16.315	4.899	0.859	1875	7964	4247	0.889	4778
23*	0.012	1.446	0.186	98.356	0.403	74.493	18.474	6.285	1.345	873	4010	4593	0.956	4805
2	0.012	1.432	0.193	98.363	0.459	73.085	18.432	6.609	1.417	1806	8676	4804	1.001	4799
19	0.011	1.119	0.221	98.649	0.569	68.030	21.117	8.260	2.024	884	4927	5574	1.157	4817
18*	0.009	1.096	0.221	98.624	0.588	67.397	21.188	8.615	2.212	901	5173	5741	1.162	4941
All Batches	0.013	1.600	0.173	98.214	0.357	75.829	17.168	5.544	1.102	20774	87026	4189	0.895	4681
24,30,31	Flush Batches									16	38			
Sumps										4	2			
Hulls										41	25			
Totals		1.600								20835	87091	4180	0.895	4670

* Batches identified from preprocessing evaluation for Agency measurement in example.
** Was reported as 77.162

measurements. These results are illustrated in the discussion that follows.

The values of both the $(\text{Pu/U})/^{235}\text{D}$ and the $(\text{Pu/U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ relationships are summarized in Table III for all input batches from NFS and NATCO measurements. The hypothetical Agency measurement batches are further compared by showing the percent difference between NFS and NATCO results. The agreement between both sets of results for the six batches verified that these six Pu/U values were measured correctly. Although NATCO values for the two relationships are shown for all batches, the hypothetical safeguards example involves only the six batches and only these six are discussed.

The verification of the Pu/U values for the input batches not selected for Agency measurement would be accomplished by using relationships whose values should be the same as for the batches directly verified by measurement. For example, the Pu/U value of batch 21 was verified since the $(\text{Pu/U})/^{235}\text{D}$ value for batch 21 was essentially the same as the Agency (hypothetical) measured values for batches 20 and 22 where all three batches had the same initial enrichment (2.310 weight percent).

In the case of the initial 2.578 weight percent fuel, the agreement of the NFS and NATCO $(\text{Pu/U})/^{235}\text{D}$ values was excellent for batch 13 and also for batch 29. However, due to the void effect, the values of $(\text{Pu/U})/^{235}\text{D}$ for batch 29 did not agree with batch 13 and the individual agreement of the two batches could not be extended to the other batches of initially enriched 2.578 weight percent fuel. Using the $(\text{Pu/U})/(^{239}\text{Pu}^{242}\text{Pu}/^{240}\text{Pu}^2)$ relationship values, the individual agreement for batches 13 and 29 was also good. The fact that the other nine NFS values also agreed with batches 13 and 29 verified these nine values from the isotopic correlation point of view. The fact that there was agreement between NFS and NATCO

TABLE III
SUMMARY OF RELATIONSHIP VALUES

3D-1 Input Batch Number	$\frac{\text{Pu/U}}{^{235}\text{D}}$			$\frac{\text{Pu/U}}{(^{239}\text{Pu} \cdot ^{242}\text{Pu}) / (^{240}\text{Pu}^2)}$		
	Grams/Tonne			Grams/Tonne		
	wt%			x10 ⁻⁴		
	NFS	NATCO	Difference	NFS	NATCO	Difference
29*	4181	4143	-0.9%	1.751	1.734	-0.5%
28	4006	3982		1.705	1.701	
25	4073	4136		1.733	1.732	
26	4286	4359		1.781	1.799	
27	4437	4451		1.724	1.750	
17	4498	4605		1.745	1.768	
4	4816	4785		1.777	1.749	
16	4762	4723		1.724	1.768	
14	4624	4590		1.737	1.763	
15	4677	4652		1.711	1.693	
13*	4733	4769	+0.8%	1.704	1.749	+2.2%
22*	4901	4895	-0.1%	1.784	1.770	-0.8%
21	4801	4826		1.803	1.793	
20*	4723	4815	+1.9%	1.797	1.845	+2.7%
5	4620	4681		1.618	1.642	
12	4643	4778		1.678	1.696	
23*	4762	4805	+0.9%	1.602	1.586	-1.0%
2	4749	4799		1.572	1.576	
19	4774	4817		1.801	1.805	
18*	4902	4941	+0.8%	1.788	1.729	-3.3%
All Fuel	4639	4670		1.477	1.474	

values for the nine batches not identified for Agency measurement is also seen, but these NATCO measurements would not have been available through the hypothetical Agency measurements. (The expected one standard deviation for $(\text{Pu/U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ is 2.56%. Generally, for differences greater than 2%, the cause can be traced directly to the ${}^{242}\text{Pu}$ weight percent which has an expected one standard deviation of 1.5% and the difference is not due to a significant difference in the Pu/U values.) The $(\text{Pu/U})/({}^{239}\text{Pu} \cdot {}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ relationship could also have been used with the initial 2.310 ${}^{235}\text{U}$ weight percent enrichment batches.

The last six batches listed in Table III represent mixtures of different fuels and present a much more difficult situation for the application of isotopic correlations. This is because of the large number of differences involved—initial enrichment, cladding, exposure and void fraction. Nevertheless, the $(\text{Pu/U})/{}^{235}\text{D}$ was successfully used in the example to verify the measured data for the batches not directly verified. An examination of the preprocessing data indicated that batches 5 and 12 should have $(\text{Pu/U})/{}^{235}\text{D}$ values close to that for batch 13 since the initial enrichments were nearly the same and the assemblies came from similar regions of the core. The measured $(\text{Pu/U})/{}^{235}\text{D}$ values for these three batches were within 2.8%. Thus, the values for batches 5 and 12 could be verified by the direct verification of batch 13. Batches 23 and 2 both had a weighted initial enrichment of approximately 2.4 and thus batch 2 values could be verified by comparison with the batch 23 values for $(\text{Pu/U})/{}^{235}\text{D}$. Batches 18 and 19 both had a weighted initial

enrichment of approximately 2.26 and batch 19 would be verified by comparison with batch 18.

The $(\text{Pu}/\text{U})/({}^{239}\text{Pu}/{}^{242}\text{Pu}/{}^{240}\text{Pu}^2)$ relationship was not as effective for these six batches due to the mixing property of plutonium isotopes. For example, the growth of ${}^{242}\text{Pu}$ is approximated very well by an exponential function. The ${}^{242}\text{Pu}$ rate of increase in Humboldt Bay fuel at exposures less than 7000 MWD/MTU is slow and on the very non-linear part of the exponential curve, while the rate of increase for higher exposure fuel is much faster and on the more linear part of the exponential curve. As a result, when an assembly of a higher exposure is mixed with a lower exposure assembly at exposures less than 7000 MWD/MTU and the difference is greater than 3000 MWD/MTU, the ${}^{242}\text{Pu}$ content of the batch is weighted in the higher exposure direction. The ${}^{242}\text{Pu}$ value would be off (and high) the original exponential curve. The relationship values are thereby reduced as seen from Table III. This effect is most clearly illustrated by the very low value of 1.47×10^4 determined for the relationship value for all fuel and listed at the bottom of Table III. This is the result of mixing fuel of different exposures from 5800 to 16000 MWD/MTU. However, the preprocessing data gave the exposure differences of the assemblies in each batch beforehand (see Appendix A) so that the mixing property could be taken into account. None of the single initial enrichment fuel batches were made up of assemblies that differed more than 2900 MWD/MTU in exposure. The $(\text{Pu}/\text{U})/{}^{235}\text{U}$ relationship is not significantly affected by the mixing property. Nevertheless, the mixing property can be used to advantage in safeguards as will be illustrated later.

The Pu/U values were thus successfully determined to have been correctly measured based on two levels of comparison. That is, the values were consistent with the isotopic data (level of comparison #2) and consistent with the six hypothetical Agency measured data (level of comparison #3) through the use

of two isotopic relationships. The preprocessing data provided the information as to the conditions under which the two relationships were constant and thus when to apply them. Several other relationships such as $(\text{Pu/U})/\Delta^{236}\text{U}$ ($\Delta^{236}\text{U}$ is the change in ^{236}U during irradiation) and $(\text{Pu/U})/({}^{240}\text{Pu}/{}^{239}\text{Pu})$,⁽²⁾ were also formed and these relationships substantiated the results given above. Space does not permit that these relationships be considered in detail.

3. Pu/U Ratio Method

The above completes the example of the use of isotopic correlations, except for a few points to be considered later. The next step as outlined in Section III-A is to use the Pu/U ratio method⁽³⁾ by multiplying the verified Pu/U ratio by the calculated total uranium to obtain the total plutonium content of Humboldt Bay fuels. Certain options are available as to how to proceed which will indicate the versatility of the methods involved. In the past, where the relationship $(\text{Pu/U})/{}^{235}\text{U}$ has been a constant for all batches of a given initial enrichment, the simplest approach has been to determine the weighted Pu/U value from the individual batch values. Then the total uranium for all batches involved was used to multiply the weighted Pu/U value where the total uranium used was the shipper's values for total uranium. Thereby the total plutonium amount was determined as a check on the measured values of total input. In the present case the same approach can be used, although it is not as straightforward since all batches do not have the same $(\text{Pu/U})/{}^{235}\text{U}$ values. An alternative approach is to multiply the Pu/U value for each batch by the uranium values for that batch again using the shipper's uranium values. The results from both approaches are nearly the same and are given in Table IV. The difference between the two values (87.988 and 87.798 kg) arises from the fact that the weighted Pu/U ratio used included the plutonium assigned to hulls, sumps and flushes (see Table I). The agreement with the

TABLE IV

INDEPENDENT DETERMINATION OF PLUTONIUM
PRODUCED IN HUMBOLDT BAY SPENT FUELS

<u>3D-1 Batch Number</u>	<u>Pu/U Measured</u>	<u>Total U_s Shippers Values</u>	<u>Pu From (Pu/U)*U_s</u>
29	2756	792.2	2183
28	2820	867.2	2445
25	2924	871.3	2548
26	3133	870.0	2726
27	3288	869.1	2858
17	3455	948.7	3278
4	3920	1895.1	7429
16	3938	948.1	3734
14	4018	945.0	3797
15	4079	945.7	3858
13	4236	945.2	4004
22	5195	901.1	4681
21	5219	901.5	4705
20	5398	898.8	4852
5	3931	942.6	3705
12	4234	1886.6	7988
23	4577	916.0	4193
2	4796	1846.8	8857
19	5576	897.0	5002
18	5741	896.2	5145
Total		20984.2	87988
Weighted Average	4184	20984.2	87798

measured plutonium, 87.006 kg, is to within one percent.

The success of the Pu/U ratio method depends on the shippers values for uranium. In this case, the shippers values agreed within 0.9 percent of the measured total by NFS. The agreement was not as good as had been experienced previously. An extensive comparison is given in reference 2.

An additional verification of the total plutonium produced in Humboldt Bay fuels was accomplished by comparing the weighted isotopic values and relationship values for the total uranium and plutonium for the campaign at input and product. The product data are summarized in Table V per product batch, five for uranium and four for plutonium product. The weighted isotopic composition for all the total uranium and total plutonium at input and product are given also, the input values being from Table I. For verification of both input and product totals, the weighted isotopic compositions for input and product totals should be the same within measurement variations. The isotopic compositions should be the same since there are no physical or nuclear processes at the chemical plant which can alter them. Even losses should not alter the values since most losses of uranium and plutonium during the separation and recovery processes occur randomly throughout a campaign rather than occurring all at once. Or if some losses are not random and occur more at certain times than at other times, losses larger than those presently encountered at NFS would need to occur to alter the isotopic compositions.

Had there been disproportioned losses or diversion by substitution of plutonium, the mixing property of plutonium would reveal this. That is, if losses occurred only when the low content plutonium was processed, the relationship $(\text{Pu/U})/({}^{239}\text{Pu}/{}^{240}\text{Pu}^2)$ would have been reduced for the product material relative to the input material. If lower exposure plutonium had been substituted for some of the higher exposure plutonium, the $(\text{Pu/U})/({}^{239}\text{Pu}/{}^{240}\text{Pu})$

TABLE V
SUMMARY OF MEASURED PRODUCT DATA AND COMPARISON OF ISOTOPIC COMPOSITION AND
RELATIONSHIP VALUES OF TOTAL PRODUCT AND INPUT

URANIUM PRODUCT COMPOSITIONS				TOTAL URANIUM	PLUTONIUM PRODUCT COMPOSITIONS					TOTAL PLUTONIUM	Pu/U $\frac{235}{238}$	Pu/U $\frac{239Pu+242Pu}{240Pu^2}$
^{234}U wt%	^{235}U wt%	^{236}U wt%	^{238}U wt%	kgs	^{238}Pu wt%	^{239}Pu wt%	^{240}Pu wt%	^{241}Pu wt%	^{242}Pu wt%	Grams		
0.012	1.628	0.174	98.186	4287	0.356	76.971	16.399	5.274	1.001	16967		
0.013	1.664	0.173	98.150	4312	0.289	78.346	15.875	4.701	0.789	23427		
0.012	1.452	0.191	98.345	4291	0.483	71.153	19.512	7.187	1.665	21439		
0.013	1.634	0.166	98.188	4261	0.309	77.219	16.518	5.008	0.946	24118		
0.012	1.633	0.166	98.189	3486								
					<u>PRODUCT TOTAL</u>							
0.012	1.601	0.174	98.212	20637	0.356	75.964	17.066	5.520	1.093	85951	4659	1.461
					<u>INPUT TOTALS</u>							
0.013	1.593	0.174	98.220	20796	0.359	75.805	17.173	5.561	1.102	87006	4639	1.477

$^{240}\text{Pu}^2$) value of the product would have been higher than 1.461. The $(\text{Pu/U})/^{235}\text{D}$ values would not have been affected in either case. Thus, the mixing property of plutonium serves as a safeguards indicator of preferential losses of plutonium or plutonium substitution because relationships involving plutonium isotopic values are sensitive to mixing.

By the above procedure, the measured plutonium content of Humboldt Bay spent fuels could be verified. As additional measurements are made by the product receiver, converter, etc., the isotopic composition values will serve to verify further the input and product measurements as well as to identify the plutonium as originating from Humboldt Bay spent fuels. The data and results tabulated in this report will serve as historical data and experience to indicate the relationships useful to isotopic correlation safeguards techniques during future dissolutions of Humboldt Bay assemblies. This source of experience will also be aided by data and results from other boiling water reactors as well as continued development using the above data. A knowledge of isotopic relationships and the conditions under which they should be applied are thus available at any future time when Humboldt Bay fuels are processed.

IV. USE OF REACTOR DATA STRENGTHENED ISOTOPIC CORRELATIONS

The examination of preprocessing data, the bulk of which was received from PG&E (reactor data), provided significant information important to the application of isotopic correlation safeguard techniques. Each assembly, identified by number, was followed from the point of being charged in the Humboldt Bay Reactor core by the use of core loading diagrams and power generation histograms. Correlation of core positions with dissolver batch content was thereby possible; core position proved to be directly related to the void effect observed from the measured data.

However, the observation of this correlation would not have been possible without two other sets of information. The first was the shipping records provided by PG&E. The fuel assemblies were ordered in shipping by PG&E according to plutonium content, the highest first and the lowest last. The shipping sequence naturally grouped the assemblies according to similar core positions and generally according to similar irradiation histories. The second item of additional information was the subsequent storage location record at NFS (provided by NFS) which made it possible to ascertain the probable batch makeup beforehand. It was seen that it was possible to retain the natural grouping of assemblies which had resulted in shipping. Retaining this order limited mixed initial enrichment batches and gave a larger exposure range from batch to batch. As a result, the dissolution sequence was fixed through the mutual agreement of NFS, PG&E and ACDA and permitted relating batch makeup to assembly core position and the void effect. Retaining the shipping order during dissolution was done at no extra cost to PG&E, NFS or ACDA because the order did not require shuffling of fuel assemblies in the storage area.

Although the above was not absolutely necessary in implementing isotopic correlations, relating the batch makeup to assembly core position was useful and strengthened the analysis. However, the concept was deemed as useful to a reactor operator as to safeguards and worth explaining. That is, the ordering of the fuel assemblies as explained above provided more correlatable information. In the case of the reactor operator, ordering the fuel sent to a reprocessing plant will also provide additional information to correlate to his calculation methods to design and operate the reactor.

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APPENDIX A

APPENDIX A

PREPROCESSING DATA AND EVALUATION

Prior to the dissolution of Humboldt Bay fuels at NFS, PG&E and NFS consented to provide ACDA with as much information pertaining to the fuel to be processed as possible. The information was received, analyzed and a summary of the results written in a paper entitled "Isotopic Relationship Safeguard Techniques Applied to Humboldt Bay Spent Fuels," BNWL-SA-4078.⁽⁵⁾ This appendix updates the tables given in that report and provides further details obtained through the analyses of the preprocessing data.

The purpose of this appendix, and the earlier report, is to present an example of how preprocessing data can be used by a safeguards agency in planning for and carrying out safeguards at a chemical reprocessing plant. It includes the use of these data to determine in advance what isotopic relationships are likely to be useful in the subsequent evaluation of measurement data. It illustrates the use of preprocessing data to identify the ordering of assemblies in making up input batches so as to maximize the effectiveness of isotopic correlations. The use of these data as the basis for determining which input batches would be independently measured by the Agency is also treated. This appendix does not, however, cover all aspects of the safeguards use of isotopic data at a chemical reprocessing plant; a more general and complete presentation is given in Reference 1.

The first four tables presented, Tables A-I, A-II, A-III, and A-IV are the tables that were included in the above report.⁽⁵⁾ They have been updated to reflect the actual dissolution of the fuel at NFS and an explanation of the information they contain as well as other tabulated data are presented in the discussions that follow.

The information received, which included the calculated final fuel content from computerized burnup codes, is outlined in Table A-I. The value of the preprocessing data, which in this case included calculated values for all uranium and plutonium isotopes, was that the data were used to investigate isotopic relationships for the material involved prior to the actual processing. In particular, the data were used to determine isotopic relationships of the material

TABLE A-I

Applied Isotopic Correlation Safeguards Techniques - -
Data Collected and Specific Use of Data Prior to Dissolution

Humboldt Bay Spent Fuel

FROM	DATA COLLECTED	USE OF DATA
PG&E	<ul style="list-style-type: none"> Statement of Material to be Delivered for Reprocessing-Tables for each shipment which listed assembly number, <u>initial</u> total U, ^{235}U, and ^{235}U enrichment; and <u>final</u> exposure, total U, ^{235}U, ^{236}U, ^{238}U, ^{235}U enrichment, total Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, fissile Pu and the percent of fissile Pu. Also, Individual Fuel Assembly Accountability Tables were provided which listed the same data. 	Values of isotopic relationships, for example, $(\text{Pu}/\text{U})/^{235}\text{D}$, were determined for each assembly - done simply with computer.
PG&E	<ul style="list-style-type: none"> Reactor power history from 1963 through 1970 and core loadings for cores 1-A through 6 which pertained to the assemblies to be processed. 	Charging and discharging dates and core positions were determined for each assembly. Assemblies having like irradiation histories were identified.
NFS	<ul style="list-style-type: none"> Assembly storage location at NFS prior to dissolution and probable order of assembly dissolution. 	Possible makeup of dissolution batches was identified and isotopic relationship values were determined for each batch using the above data. The initial and final total U, ^{235}U , etc., per batch, were determined from the calculated data by summing contributions from each assembly making up the batch.

PURPOSE OF DATA COLLECTION: To provide inspectorate with a working knowledge of the isotopic relationships of the material to be processed. The data essentially represent collection of fabrication and reactor operation information pertaining to the fuel.

RESULTS FROM USE OF THE DATA: Inspectorate prepared to evaluate how successful isotopic relationship safeguards will be during dissolution campaign.

TABLE A-II
Applied Isotopic Correlation Safeguards Techniques - -
Preprocessing Evaluation
Humboldt Bay Spent Fuel

QUESTIONS TO BE EVALUATED		EVALUATION			
• Were the assemblies of different designs and of different initial enrichments which cause isotopic relationships to change?	Yes:	Fuel Assembly Identification Prefix	Type of Assembly (Cladding)	Initial ²³⁵ U Enrichment	Number of Assemblies
		A	304 SS	2.578	189
		B	Zr-2	2.310	73
		C	Zr-2	2.103	8
The 18.4% decrease in initial enrichment coupled with the change from 304 SS to Zr-2 cladding resulted in an approximate 13% increase in (Pu/U)/ ²³⁵ D (see Table A-V) from calculated data.					
• Did other factors affect the isotopic relationships?	Yes:	The (Pu/U)/ ²³⁵ D values of the "A" assemblies varied 5% due to their different irradiation histories indicating a probable void effect. The "A" assemblies of like history had the same relationship values (see Table A-V)			
• Was there a dissolution order more advantageous to isotopic relationship safeguards techniques that did not require extra work on the part of NFS?	Yes:	The assemblies were sent to NFS in an order according to plutonium content which made it possible to limit the batches of mixed type "A" and type "B" and "C" assemblies. A revised order from that initially indicated by NFS was confirmed and calculated batch values were redetermined, where necessary.			
• How many dissolution batches were expected and which batches were expected to have the same relationship values and which batches were expected to have varying values?	There were expected to be 23* dissolution batches if 12 assemblies were dissolved per batch. Dissolver batches expected to have the same relationship values were <ol style="list-style-type: none">Batches 3, 4, 5, 7, 8, 9, 10, 11, 18, 19, 20, 21, 22 and 23 involving "A" type assemblies only. Batches 7 and 23 were expected to contain highest and lowest exposure material, respectively, of "A" assemblies.Batches 14, 15, and 16 involving "B" type assemblies only. These three batches were expected to have approximately 7% higher relationship values than batches of "A" assemblies. Batches where the relationship values were expected to vary include batches 1 and 2, a mixture of "A", "B" and "C" assemblies, batches 6 and 17, a mixture of "A" and "B" assemblies and batches 12 and 13, a mixture of "B" and "C" assemblies. Batch 16* was expected to have the highest relationship value.				

RESULT: Inspectorate aware of calculated isotopic relationship value variations and acquainted with the characteristics of the material to be processed.

* In report BNWL-SA-4078 the expected number of dissolution batches was incorrectly given as 22 and batch 12, rather than 16, was incorrectly identified as the batch expected to have the highest (Pu/U)/ ^{235}D value.

TABLE A-III
Applied Isotopic Correlation Safeguards Techniques - -
Duties of Inspectorate and Inspector During Dissolution
Humboldt Bay Spent Fuel

INSPECTORATE

- Communicate results of preprocessing evaluation to inspector.*
- If expected dissolution order changes, redetermine batch isotopic relationship values from calculated data.
- Independently measure duplicate samples of batches 7, 12, 14, 16**, 17 and 23.
- Determine detailed isotopic relationship values from chemical plant measured data in order to
 - a. Verify measured Pu input (Table VII).
 - b. Identify anomalous results, if any using isotopic consistency analyses procedures.
 - c. Confirm preprocessing evaluation results.*
 (The procedures will have greater influence the closer in time to the actual measurements that they are applied. Procedure application concurrent with measurement would be the most effective.)

INSPECTOR

- Understand results of preprocessing evaluation.
- Verify and keep accurate records of assembly makeup per batch. Keep inspectorate aware of any changes in expected dissolution order and the number of assemblies per batch.
- Make arrangements to obtain duplicate samples of all batches.
- Transmit all data including product data, to the inspectorate as it is measured.
- Form certain isotopic relationship values from chemical plant measured data (input) as soon after measurement as possible. Judge whether or not measured values appear consistent with preprocessing results. Also, compare total U values per batch and per campaign, both measured and calculated. Use Pu/U verification relationships to judge measured Pu/U values.

- * The relationship values determined from measured data are expected to be different (biased) by a few percent from the values determined from calculated data due to an expected bias between measured and calculated data, except for the total U. Also, based on past experience with other reactor fuels and due to the assembly makeup of the dissolution batches, the variation of relationship values of batches involving "A" type assemblies is expected to be less than the 5% indicated in Table II.
- ** Sample 16 has replaced Sample 1 from the original numbers given in Reference 5.

TABLE A-IV
Applied Isotopic Correlation Safeguards Techniques - -
Verification of Plutonium Input
Humboldt Bay Spent Fuel

WHAT TO DO

- Compare isotopic relationship values determined from chemical plant measurements and from remeasurements to verify measured Pu/U values.
- Multiply Pu/U ratio values by corresponding calculated uranium (as reported by reactor operator) per batch and per campaign to determine plutonium produced in fuel. *
- Compare weighted isotopic compositions of the total uranium and plutonium of input and product for the campaign.
- Compare weighted isotopic compositions** of total uranium and plutonium of product as measured by the chemical plant and by the product receiver.

* Reference 1 includes a technique for confirming total uranium content based on verified chemical plant isotopic data and verified fabrication total uranium.

** Corrected for ^{238}Pu and ^{241}Pu Decay.

DESIRED RESULT

- Relationship values as determined from Agency measurements and chemical plant measurements should agree as follows:

<u>Agency Measurements</u>	<u>Chemical Plant Measurements***</u>
13, 29	-- 4, 13, 14, 15, 16, 17, 25, 26, 27, 28, and 29
20, 22	-- 20, 21, and 22
18, 23	-- 18, 23 (respectively)
- Plutonium as reported by chemical plant operator and plutonium as determined from Pu/U ratio method should agree for verification.
- The weighted isotopic compositions of the total uranium and plutonium of input and product should be identical since there is no way to change the isotopic compositions at the chemical plant except:
 - a. By falsification of either input or product isotopic compositions
 - b. By diverting an amount of uranium or plutonium greater than can be accounted for in waste streams
- Again, the weighted isotopics of total uranium and plutonium should be identical since there are no physical processes at this point in the fuel cycle to change them.

*** Batches 2, 12 and 19 which are mixtures of different type fuels, are expected to have (Pu/U)/ ^{235}D relationship values intermediate to those measured by the Agency.

involved in order to evaluate whether or not isotopic correlation safeguard techniques could be used successfully during the subsequent dissolution campaign. This is the approach which would be desirable for each new reactor dissolving its first spent fuel.

Isotopic relationships were evident from the preprocessing data. One particular relationship mentioned in Table A-II was the Pu/U ratio over the change due to irradiation in the ^{235}U enrichment, ^{235}U depletion or ^{235}D . Various parameters, some of which are mentioned in Table A-III, caused the relationship between Pu/U and ^{235}D to change. The calculated values of $(\text{Pu}/\text{U})/^{235}\text{D}$ are summarized in Table A-V and the one standard deviations of the values for the number of assemblies given are shown. For a given initial enrichment, the values $(\text{Pu}/\text{U})/^{235}\text{D}$ were essentially constant for assemblies having the same charge and discharge dates. The values did not change as a function of exposure although each discharge represented a range in exposures. For example, the assemblies in the first discharge ranged from 6800 to 9800 MWD/MTU in exposure. The $(\text{Pu}/\text{U})/^{235}\text{D}$ values grouped according to charge-discharge dates varied from 4548 to 4326, a variation of five percent, for the 2.578 ^{235}U weight percent initially enriched fuel assemblies. This variation was assumed to be due to variations in voids during reactor operation rather than that the calculated values decreased with increased exposure. Therefore at this point, all dissolution batches which included type A fuel assemblies only were expected to have generally the same relationship values. This expectation was based on past experience with Yankee Rowe and Dresden⁽⁵⁾ chemical dissolution data where $(\text{Pu}/\text{U})/^{235}\text{D}$ had been constant for single enrichment fuels.

In addition to differences in the initial enrichments of A, B and C type assemblies listed in Table A-II, other physical differences between type A and types B and C assemblies existed. There was a difference in cladding, as noted, and cladding thickness. The Zircaloy-2 (Zr-2) cladding was thicker than the stainless steel (SS). It was known that the lower enrichments of B and C assemblies would increase the $(\text{Pu}/\text{U})/^{235}\text{D}$ values while the change from SS to Zr-2 cladding would decrease $(\text{Pu}/\text{U})/^{235}\text{D}$ and that the two changes tend to compensate each other. Along with these changes, the fuel pellet diameters of B and C type assemblies were slightly less and the total rod diameters were slightly larger than type A assemblies to accommodate the thicker Zr-2 cladding.

TABLE A-V
Summary of Calculated (Pu/U)/²³⁵D Values

Initial ²³⁵ U Enrichment of 2.578 Weight Percent			
Number of Assemblies	Dates and Cores		(Pu/U)/ ²³⁵ D
	Charge	Discharge	
52	7/31/63 - Core 1-A	9/20/65 - Core 1-B	4548 ± 10
69	7/31/63 - Core 1-A	11/18/66 - Core 2-A	4414 ± 16
19	7/31/63 - Core 1-A	3/29/67 - Core 3-A	4373 ± 15
13	7/31/63 - Core 1-A	10/4/67 - Core 3-B	4326 ± 15
4	7/31/63 - Core 1-A	9/20/65 - Core 1-B	
	12/20/66 - Core 3-A	3/29/67 - Core 3-A	4485 ± 10
14	7/31/63 - Core 1-A	11/18/66 - Core 2-A	
	4/5/67 - Core 3-B	10/4/67 - Core 3-B	4375 ± 9
1	7/31/63 - Core 1-A	9/20/65 - Core 1-B	
	4/5/67 - Core 3-B	10/4/67 - Core 3-B	4477
6	9/13/64 - Core 1-B	10/4/67 - Core 3-B	4449 ± 14
8	9/13/64 - Core 1-B	11/18/66 - Core 2-A	
	4/5/67 - Core 3-B	10/4/67 - Core 3-B	4497 ± 11
2	12/1/65 - Core 2-A	6/14/69 - Core 4-B	4348
1	12/20/66 - Core 3-A	6/14/69 - Core 4-B	4417
Total 189			
Initial ²³⁵ U Enrichment of 2.310 Weight Percent			
31	12/1/65 - Core 2-A	6/14/69 - Core 4-B	4811 ± 21
6	12/20/66 - Core 3-A	6/14/69 - Core 4-B	4937 ± 2
29	12/20/66 - Core 3-A	4/19/70 - Core 5	4754 ± 6
7	12/1/65 - Core 2-A	4/19/70 - Core 5	4680 ± 8
Total 73			
Initial ²³⁵ U Enrichment of 2.103 Weight Percent			
8	10/28/67 - Core 4-A	4/19/70 - Core 5	5028 ± 4
Total 8			

The preprocessing data indicated that the overall effect would be an increase in $(\text{Pu}/\text{U})/^{235}\text{D}$ values for B and C type fuel as listed in Table A-V. The values of $(\text{Pu}/\text{U})/^{235}\text{D}$ for B type assemblies varied from 4680 to 4937, a variation of five percent, and this was assumed to be due to different voids as a function of time during reactor operation.

Other evaluations performed using the preprocessing data are also indicated in Table A-II. The evaluation was carried to the point of postulating the makeup of each dissolver batch based on the storage location of the assemblies at NFS and the probable dissolution order as indicated by NFS. This was done to note possible variations of relationships on a dissolver batch basis, identify the batches which would represent maximum and minimum ^{235}U content for batches of single enrichment fuels, and ascertain whether or not the dissolution order indicated by NFS was the most advantageous to isotopic correlation safeguards techniques. As a result, a revised dissolution order was confirmed between NFS, PG&E and ACDA which tended to limit the number of mixed initial enrichment fuel batches. Also, dissolver batches of a single initial enrichment which represented maximum and minimum ^{235}U content, batches 7 and 23 for 2.578 weight percent fuel and 14 and 16 for 2.301 weight percent fuel, were selected as being particularly useful for measurement by the Agency. Dissolver batches 12 and 16 which were of mixed initial enrichments were selected for Agency measurement also. The selection was based on their weighted initial enrichments in that they were different from both the 2.578 and the 2.301 fuels but similar to the weighted initial enrichments of other mixed batches.

Thus, by use of the preprocessing data, it was determined that isotopic correlation safeguard techniques were applicable to Humboldt Bay spent fuels. Had there actually been an inspectorate at this point in time, the inspectorate would have been in a position to define the safeguarding functions to be performed during the dissolution of the fuel as they apply to isotopic correlations. The preparations to accomplish safeguard functions were certainly more complete by use of the preprocessing data than if an inspectorate were to assign safeguard functions without use of the information.

Assuming the role of a safeguards agency, the hypothetical strategy decided upon before dissolution is given in Table A-III. Essentially the inspector was to keep the inspectorate informed of changes in expected dissolution sequenc-

ing and obtain a duplicate sample of input and product batches. By selecting a few samples for Agency measurement, all operator measured Pu/U values could be verified. Since there was no inspector and no samples were obtained for independent measurement, the above was strictly hypothetical.

As it turned out, there were changes in the actual dissolution sequence as compared to the postulated sequence (see Table A-VI). The batch (type A assemblies) postulated for processing just before the mini flush was actually processed much earlier. Also, a type B fuel assembly which had been assumed would be dissolved in batch 1, was dissolved in batch 5 (see Table I). In addition, it had been assumed that all dissolver batches would be measured separately in the 3D-1 accountability tank. During the actual dissolution, dissolver batches 1 and 2, 3 and 4, and 6 and 7 were mixed in the 3D-1 tank. Since batch 7, the highest exposure type A material, was mixed with batch 6 which contained a type B assembly, dissolver batch 8 replaced batch 7 as the highest exposure (or lowest ^{235}U content) type A material batch selected for Agency measurement. Otherwise the ordering and dissolution were as expected.

Note from Table A-VI that the 3D-1 batch numbers were generally different from the postulated and actual dissolver batch numbers. In translating dissolver batch numbers to 3D-1 batch numbers, dissolver batch 8 becomes 3D-1 batch 13, for example. In terms of 3D-1 batch numbers, batches 13, 18, 20, 22, 23, and 29 had been identified for Agency measurement based on the preprocessing data and the subsequent dissolution numbering at NFS. These batches are starred in Table A-VI. The 3D-1 batch numbers are utilized in the remainder of this appendix including Table A-IV in discussing individual batches. This table is the only table not arranged in the order discussed in the main report for the reason of translating postulated numbers to actual 3D-1 batch numbers.

Shown in Table A-VII are the number of assemblies per 3D-1 batch arranged to indicate the core and core shutdown date from which the assemblies were discharged, core position of the assemblies and the weighted exposures of each batch. Generally speaking, each assembly was identified as having resided in the core center region (C), intermediate region (I) or outer region (O), as defined in Figure A-1, if it resided at least two thirds of its total time in the reactor in that region of the core. The highest exposure batches of type A assemblies, 3D-1 batch 13 for example, included assemblies from the center and

TABLE A-VI
Processing Sequence and Postulated
Dissolver and 3D-1 Batch Numbers

Postulated Dissolver Batch Number	Actual Dissolver Batch Number	3D-1 Batch Number	Date of Dissolution
1, 2	1, 2	2	4/27/71
3, 4	3, 4	4	5/11/71
5	5	5	5/15/71
6, 7*	6, 7	12	6/7/71
8	8	13**	6/12/71
9	9	14	6/13/71
10	10	15	6/15/71
11	11	16	6/16/71
18	12	17	6/17/71
12*	13	18**	6/19/71
13	14	19	6/21/71
14*	15	20**	6/24/71
15	16	21	6/25/71
16*	17	22**	6/26/71
17*	18	23**	6/27/71
Mini Flush		24	6/27/71
19	19	25	6/28/71
20	20	26	6/29/71
21	21	27	6/30/71
22	22	28	7/1/71
23*	23	29**	7/3/71
		30	7/3/71
		31	7/4/71

* Identified prior to reprocessing for Agency measurement.

** Identified for Agency measurement on basis of actual reprocessing batching.

TABLE A-VII

Summary of Core Positions
and Exposures of Assemblies by Batch

3D-1 Input Batch Number	Core From Which Discharged and Core Discharge Date							Core Position†			Exposure	Exposure Range of Assemblies in Batch MWD/MTU
	Core 1-B 9/20/65	Core 2-A 11/18/66	Core 3-A 3/29/67	Core 3-B 10/4/67	Core 4-A 10/5/68	Core 4-B 6/14/69	Core 5 4/17/70	C	I	O	MWD/MTU	
29				10						10	6954	1065
28	1	1	1	5		3				11	7265	2870 (3/11)
25	1			10						11	7671	816
26		1		10						11	7834	1163
27	4	4	2	1					1	10	8139	506
17	4	3	1	4				1	3	8	8548	636
4	12	8	4					11	7	6	9127	2725 (2/24)
16	1	1						3	6	3	9228	1311
14		9	2						9	3	10001	661
15	2	8	2					2	8	2	9975	2046 (3/12)
13	5	7						1	11		9873	1194
22						12		3	9		13088	981
21						12			12		13528	794
20						4	8		12		14093	1970
5	2	7	2				1	1	9	2	9707	5805 (1/12)
12	3	11	9				1	3	19	2	10350	6255 (4/24)
23	2			2		8		1	9	2	11496	4773 (4/12)
2	4	9					11	8	15	1	12091	6574 (11/24)
19						1	11	1	9	2	14570	2358 (1/12)
18							12	3	8	1	15189	1445

†Position of assemblies in the reactor core is indicated by the following notation:

C - Resided predominately in the center region of the core.

I - Resided predominately in the intermediate region of the core.

O - Resided predominately in the outer region of the core, see Figure A-1.

LOCATION GIVEN WAS THE POSITION OF THE ASSEMBLY PRIOR TO DISCHARGE

FIGURE A-1. HUMBOLDT BAY CORE REGIONS DEFINED FOR THE PURPOSES OF THIS REPORT

intermediate regions only and the lowest exposure batch 3D-1, batch 29, included assemblies only from the outer region of the core. The core locations of the assemblies included in these two batches are shown in Figure A-1 also. Batches 25 through 29 of type A fuel contained assemblies which were essentially from the outer regions of the core while all other batches, except batch 17, predominately contained assemblies from the center and intermediate regions of the core. The batches thus represented distinct regions of the core where the percent voids during operation may have been different.

Also given in Table A-VII is information concerning the range of exposures of the assemblies making up each batch. For example, the exposures of the assemblies included in batch 29 varied from 6400 to 7465 MWD/MTU or a range of 1065 MWD/MTU. Where the exposure range was more than 2000 MWD/MTU, the number of assemblies whose exposures were outside this range of 2000 MWD/MTU is indicated along with the total number of assemblies in the batch. For example, three assemblies of 11 in batch 28 had exposures of 8637, 8489, 8314 MWD/MTU which exceeded by more than 2000 MWD/MTU the lowest exposed assembly of 5767 MWD/MTU. This information is important in the case of plutonium isotopic values since the higher exposure assemblies have formed ^{242}Pu , for example, at a higher rate. The mixing of a high and low exposure assembly will cause the ^{242}Pu value to appear in error because it will not be on the burnup path represented by the individual assembly values.

The calculated isotopic compositions for each input batch based on the assemblies actually dissolved in each batch and the calculated isotopic content of each assembly are shown in Table A-VIII. The table is an updated version of earlier tables generated from the preprocessing data during the preprocessing evaluation. The grouping was done according to initial ^{235}U enrichment and the ordering within a group was done according to final ^{235}U weight percent. (Batches 14 and 15 are exceptions since the ^{235}U values were essentially identical and the ordering was based on the ^{239}Pu weight percents.) The purpose of the table was to establish the ordering of each batch for later comparison to the ordering from measured data rather than to compare calculated and measured isotopic values. Values for total uranium from the calculated data are also shown in Table A-VIII.

TABLE A-VIII
PG&E Calculated Input Summary

3D-1 Batch Number	²³⁵ U Initial Enrichment	Uranium Isotopic Composition			Plutonium Isotopic Composition				Total Uranium	Total Plutonium	Pu/U GMS Tonne	²³⁵ D wt%	(Pu/U)/ ²³⁵ D GMS/tonne wt%
		²³⁵ U	²³⁶ U	²³⁸ U	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu					
	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	Kgs	GMS			
29*	2.578	1.880	0.132	97.988	80.608	14.593	4.352	0.447	792.2	2449	3091	0.698	4427
28	2.578	1.854	0.137	98.009	79.772	15.120	4.599	0.508	867.2	2757	3179	0.724	4391
25	2.578	1.820	0.144	98.036	78.900	15.647	4.895	0.558	871.3	2910	3340	0.758	4408
26	2.578	1.807	0.146	98.046	78.497	15.883	5.032	0.589	870.0	2958	3400	0.771	4412
27	2.578	1.785	0.152	98.064	77.747	16.290	5.321	0.642	869.1	3077	3541	0.793	4464
17	2.578	1.751	0.158	98.091	76.854	16.844	5.589	0.713	948.7	3470	3657	0.827	4424
4	2.578	1.709	0.167	98.124	75.547	17.562	6.067	0.824	1895.1	7371	3890	0.869	4475
16	2.578	1.705	0.167	98.129	75.156	17.731	6.252	0.861	948.1	3749	3954	0.873	4527
14	2.578	1.643	0.179	98.178	73.702	18.663	6.635	1.000	945.0	3899	4126	0.935	4415
15	2.578	1.644	0.179	98.177	73.564	18.703	6.711	1.022	945.7	3922	4147	0.934	4440
13*	2.578	1.637	0.181	98.182	73.288	18.841	6.826	1.045	945.2	3974	4203	0.941	4465
22*	2.310	1.221	0.205	98.574	66.285	23.450	8.404	1.861	901.1	4759	5281	1.089	4848
21	2.310	1.193	0.210	98.597	65.472	23.943	8.599	1.985	901.5	4846	5376	1.117	4815
20*	2.310	1.158	0.216	98.626	64.586	24.488	8.817	2.109	898.8	4958	5517	1.152	4787
5	2.557	1.651	0.173	98.176	73.909	18.596	6.482	1.012	942.6	3821	4054	0.906	4475
12	2.567	1.611	0.183	98.206	72.729	19.227	6.925	1.119	1886.6	8019	4251	0.956	4444
23*	2.402	1.403	0.189	98.408	69.228	21.642	7.599	1.531	916.0	4311	4706	0.999	4713
2	2.433	1.389	0.197	98.414	68.077	22.222	7.996	1.704	1846.8	8921	4830	1.044	4628
19	2.276	1.098	0.220	98.682	63.510	25.155	9.067	2.268	897.0	5060	5641	1.178	4789
18*	2.258	1.049	0.225	98.726	62.364	25.770	9.413	2.453	896.2	5216	5820	1.209	4814
All Fuel	2.495	1.552	0.179	98.269	71.442	20.155	7.081	1.322	20984.2	90447	4310	0.943	4571

* Batches identified for Agency Measurement during preprocessing evaluation.

Thus the preprocessing data gave evidence of isotopic relationships, provided the initial ^{235}U enrichments and total uranium, permitted ordering of batches according to ^{235}U content, and provided the basis for the selection of batches for Agency measurement. On a batch basis, the uranium measured compared to the uranium tabulated in Table A-VIII can vary due to variations of the heels that remain in dissolver tanks when the dissolved material is pumped to the 3D-1 accountability tank.

APPENDIX B

APPENDIX B

DEVELOPMENT OF ISOTOPIC RELATIONSHIPS WHICH ARE INSENSITIVE TO VOID EFFECTS

As mentioned in the discussion of the Humboldt Bay measured data, the values of the relationship $(\text{Pu/U})/^{235}\text{D}$ varied approximately 20% which was not necessarily a function of exposure but more probably due to a void effect. In what follows, supporting evidence of the void effect is presented from purely isotopic data and then the development of a relationship involving Pu/U values but insensitive to voids is discussed. In addition to Humboldt Bay measured data, other boiling water reactor data, both calculated and measured, are included to aid in showing the void effect and in determining a void insensitive relationship. The calculated data⁽⁷⁾ used were of Browns Ferry chosen because they were calculated as a function of increasing void. The basic purpose of these data was that they gave the functional form of the relationships investigated. The measured data⁽⁶⁾ were from the chemical reprocessing of earlier Dresden 1 fuels which data had not evidenced a strong void effect.⁽²⁾ The purpose of these data was to observe the functional form of various relationships from measured boiling water reactor data not strongly influenced by varying voids. Both non-linear and linear relationships as a function of increasing exposure are considered by plotting the data.

To illustrate the use of calculated data and Dresden 1 data along with Humboldt Bay data, the calculated results of $(\text{Pu/U})/^{235}\text{D}$ as a function

of increasing voids for Browns Ferry are shown in Figure B-1. Although the initial enrichment of Browns Ferry fuel, 2.19 weight percent ^{235}U , as well as other physical design parameters were not the same as Humboldt Bay fuels, the curves shown nevertheless indicated the functional form of the Pu/U versus ^{235}D relationship and the changes that occurred due to changing the void. That is, the relationship was reasonably linear for a given void and $(\text{Pu/U})/^{235}\text{D}$ increased as the void percent increased. It is seen that if voids decreased as a function of core position, that is, lower in the outer regions of the core, $(\text{Pu/U})/^{235}\text{D}$ would also decrease. Keeping in mind the functional form, the measured data from Humboldt Bay and Dresden 1 are shown in Figure B-2. It is seen that the Dresden 1 data describe linear functions reasonably well, both for the 1.5 and the 2.5 weight percent ^{235}U initially enriched fuels (the initial enrichments are indicated on the figures). The conclusion from this result was that all of the reprocessed Dresden 1 fuel of a given initial enrichment had been irradiated in environments where void histories had been essentially the same.

In the case of Humboldt Bay, however, it is seen that the lower exposure data points (particularly four points) deviate from a linear function depicted by the remainder of the data. (Remember that the decrease in $(\text{Pu/U})/^{235}\text{D}$ due to decreasing the initial enrichment from 2.578 to 2.310 weight percent was somewhat compensated for by a change from stainless steel to Zircaloy-2 cladding.) The conclusion reached was that the lower lying data points were consistent with the fuels having been irradiated in environments where the void percents had been lower than for the fuels represented by the remaining data points.

FIGURE B-1. Pu/U RATIO VERSUS ^{235}U DEPLETION FROM
CALCULATIONS OF A BOILING WATER REACTOR

FIGURE B-2. Pu/U RATIO VERSUS ^{235}U DEPLETION FOR HUMBOLDT BAY AND DRESDEN 1 DATA

The above methods of presentation are used also to show evidence of the void effect purely from the measured isotopic data not involving either the Pu/U ratio or the ^{235}U . The void effect was evident from isotopic data by plotting the combination of the $^{242}\text{Pu}/^{240}\text{Pu}$ ratio values versus the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio values. The calculated data, Figure B-3, indicated that the void effect was predominantly evident using such ratios by the fact that the burnup curves representing each void were well separated. The functional form of these curves was non-linear.

The measured data of these same ratios for Humboldt Bay and Dresden 1 are shown in Figure B-4 where the curves have been drawn in by inspection based on the shape predicted by the calculated data, Figure B-3. The Dresden 1 data for each of the two initial enrichments lie on single, non-linear curves and show no effects due to void variations, confirming the conclusion drawn from Figure B-2 that these fuels had essentially the same void histories. In comparison, the lower exposure data points of 2.578 weight percent Humboldt Bay fuel described a different non-linear curve than did the remaining data points of the 2.578 weight percent fuel indicating the void effect. Thus, the void effect observed in the $(\text{Pu}/\text{U})/^{235}\text{D}$ values was verified by the ^{239}Pu , ^{240}Pu and ^{242}Pu isotopic data.

As has been seen, plotting the measured data was a useful method whereby the void effect was observed. Plotting also proved useful in determining a relationship that was linear with exposure and insensitive to variations in voids as follows. The calculated data gave evidence

FIGURE B-3. $^{242}\text{Pu}/^{240}\text{Pu}$ RATIO VERSUS $^{240}\text{Pu}/^{239}\text{Pu}$ RATIO FROM CALCULATIONS OF A BOILING WATER REACTOR

FIGURE B-4. $^{242}\text{Pu}/^{240}\text{Pu}$ RATIO VERSUS $^{240}\text{Pu}/^{239}\text{Pu}$ RATIO
FOR HUMBOLDT BAY AND DRESDEN 1 DATA

that the Pu/U ratio versus the $(^{239}\text{Pu} \cdot ^{242}\text{Pu}) / ^{240}\text{Pu}^2$ ratio resulted in describing a function nearly linear and somewhat insensitive to voids due to the fact that the burnup curves representing different void percents were close together, see Figure B-5. The measured data are shown in Figure B-6 and a linear burnup curve described the Dresden 1 data very well. They were not influenced by voids. A linear burnup curve nearly identical to the Dresden 1 burnup curve was seen to also describe the Humboldt Bay data of an initial enrichment of 2.578 weight percent. A second linear burnup path was described by the 2.310 weight percent fuel and was seen to be close to the burnup path of the 2.578 weight percent fuel. The data points which represented the mixed initial enrichment batches, particularly the batches where the exposures of the different assemblies in the batch were different by 5000 MWD/MTU, were somewhat lower than all other data points. The reason for these data points being low was discussed in the body of the report and was referred to as the mixing property of plutonium isotopes.

Thus the isotopic data confirmed the void effect and provided as well a function which was linear over the exposure range examined and which was not dependent on void variations. However, the function was dependent on the mixing of assemblies of different exposures in a batch. As long as the exposure differences were not large (less than 3000 MWD/MTU) the linear function was not affected. It was also seen that, in addition, the relationship was somewhat insensitive to initial enrichments.

FIGURE B-5. Pu/U RATIO VERSUS $\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{(^{240}\text{Pu})^2}$ RATIO
FROM CALCULATIONS OF A BOILING WATER REACTOR

FIGURE B-6. Pu/U RATIO VERSUS $\frac{^{239}\text{Pu} \cdot ^{242}\text{Pu}}{^{240}\text{Pu}^2}$ RATIO
FOR HUMBOLDT BAY AND DRESDEN 1 DATA