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THE SAFEGUARDS VALUE
OF CHEMICAL PLANT MEASUREMENTS
RELATING TO BURNUP
-YANKEE CORES V AND VI -

June 1970

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THE SAFEGUARDS VALUE
OF
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-YANKEE CORES V AND VI-

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June 1970

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PREFACE

This report contains results of research sponsored by the US Arms Control and Disarmament Agency (ACDA) under Technical Services Agreement Y-49589 and the US Atomic Energy Commission (AEC) under USAEC Contract AT-(45-1)-1830.

ABSTRACT

Data evaluation methods relating to the area of burnup data from spent nuclear fuels have been applied to accountability data which result from the measurements of spent reactor cores at a chemical processing plant. It was determined that the methods are easily applied to accountability data and significant information of value to a safeguards program result from application of the evaluation methods.

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THE SAFEGUARDS VALUE OF CHEMICAL PLANT MEASUREMENTS RELATING TO BURNUP
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I. INTRODUCTION

In 1967, chemical plant input measurement data became available for the first time on spent power reactor cores^(1,2,3). Although not specifically required for accountability, U and Pu isotopic ratios along with total U and total Pu of dissolved fuel batches (~ 1 tonne U) were measured at the chemical plant. These measurements represented a first time in history for the direct and accurate measurement of both the U and Pu isotopic compositions of input dissolver solutions on a routine basis.

The relationships between the growth and depletion of the various nuclides of nuclear fuel undergoing irradiation have been used for a number of years as a basis for verification of analytical measurements on spent fuels. Well developed and applied techniques used as indices of the plutonium content of irradiated, low enriched uranium are (1) the plutonium alpha activity, (2) the americium-curium alpha activities and (3) the depletion of ^{235}U during irradiation.

With the increased emphasis on nuclear materials safeguards, it was appropriate to provide alternate and more accurate means of verifying the nuclear materials content of spent power reactor fuels. Thus, other related techniques describing the fuel burnup were examined. These techniques are based upon the mathematics describing the uranium and plutonium isotopic concentrations during irradiation. The techniques can be manipulated to yield parameters which testify as to the plutonium content in the fuel.

The key point in the application of such techniques in the overall fuel cycle is at the input accountability tank of the chemical processing plant where measurements are made on dissolved reactor cores. Safeguards verification at this point is extremely important. First, because this is the point at which plutonium can be extracted from the cycle in a useable form. Secondly, it is the first point at which material can be accurately measured after the irradiation process. Thus, chemical plant input measurements confirm (or reject) measurements and/or predictions of fuel content made at previous points in the fuel cycle (e.g., diffusion plant, fabrication plant or in-reactor) and represent the tie to the overall fuel cycle since re-conversion and fabrication of the recovered fuel are also tied to these measurements.

Useful knowledge has been gained from evaluation of burnup data⁽⁴⁾ which are similar to accountability data. The burnup data are the U and Pu isotopic ratios and concentrations of small (~5-20 gms U) spent fuel samples, e.g., 1/2 in. long fuel specimens cut from a fuel rod. One group of burnup data of particular interest is the data collected from Yankee core I fuels during the Yankee Core Program⁽⁵⁾ since they can be compared to accountability data that resulted from the dissolution of Yankee Core I.^(2,3) In the latter case, a single data point represents whole fuel assemblies or an integration of over a tonne of material, some five percent of the entire core. Comparisons of the small and the gross sample data are presented in Figure 1. The ^{241}Pu has been decay corrected to date of discharge for all data. Only representative burnup samples from different fuel assembly locations, as indicated by the different regions noted, are shown.

The criteria of consistency must be met in applying burnup-data evaluation techniques to the accountability data. Both data sets exhibit

FIGURE 1. The $^{241}\text{Pu}/^{239}\text{Pu}$ Isotopic Ratio Versus the $^{240}\text{Pu}/^{239}\text{Pu}$ Isotopic Ratio and the Pu/U Ratio Versus ^{235}U Depletion for Small Sample Burnup Data and Accountability Data from Yankee Core I.

consistency which is necessary for successful data evaluation. Consistency in such data is illustrated in Figure 1 by the ability of each data set to define a smooth curve that could be drawn through data sets by inspection. As expected, it can be seen that accountability data have greater consistency as evidenced by the decreased scatter of accountability data. Thus, data evaluation techniques which are based on consistency can be effectively applied to accountability data. Moreover, for this case, the accountability data meet the consistency criteria more successfully than do the burnup data.

Data evaluation procedures are techniques which can be used for safeguards purposes. The techniques were first applied to accountability data from Yankee Cores I through IV.⁽⁶⁾ Within the framework of the safeguards experiment, "Evaluation of Minor Isotopes Safeguards Techniques (MIST) in Reactor Fuel Processing,"⁽⁷⁾ evaluation procedures were further developed when applied to the initial U and final U and Pu isotopic ratios from analytical measurements on dissolution batches of Yankee Cores V and VI at Nuclear Fuel Services, Incorporated (NFS). In particular, an investigation was made to determine if the data yield the reactor characteristic(s) or a "fingerprint" of the core. The purpose of this report is to illustrate the techniques and the independent verification cross checks that are available for safeguarding purposes.

The experiment was sponsored by the Arms Control and Disarmament Agency (ACDA)⁽⁷⁾ with the support of the Atomic Energy Commission (AEC) and the International Atomic Energy Agency (IAEA). In addition to NFS, four other U.S. industrial firms cooperated in the program. Reactor data and fabrication data were supplied by Yankee Atomic Electric Company (YAEC) and Westinghouse Electric Company, respectively. Teledyne Isotopes, formerly Isotopes Incorporated (II) and Ledoux & Company, acting jointly as agents

for YAEC, provided results of their measurements on Yankee cores V and VI for comparison studies with measurements made by other laboratories. The experiment was further complemented by the National Bureau of Standards (NBS) who provided independent mass spectrometric measurements of all accountability samples. This work was particularly important in connection with measurements of the low yield isotopes. Independent measurements of all samples were made by New Brunswick Laboratories (NBL) at the request of the IAEA. Also involved in other aspects of the experiment were personnel from the Battelle Columbus Laboratory of Battelle Memorial Institute, from the German Atomic Energy Research Institute at Karlsruhe, Germany, and from the IAEA.

The purpose of this report is to demonstrate the value of chemical plant measurement data from Yankee cores V and VI from the following standpoints:

- As a means of verifying the plutonium input to a chemical processing plant.
- To look for a "fingerprint" which confirms safeguards information regarding the initial enrichment, exposure, reactor type, irradiation conditions, and fabrication data.
- As a basis for further developing evaluation techniques used for determining the internal consistency of chemical plant input measurements.
- As a basis for obtaining data useful for improving prediction of reactor parameters and for improving associated measurement techniques of nuclear material content by the reactor operator.

The current status of the data evaluation phase of the experiment and the extent to which experimental objectives were reached are discussed.

II. SUMMARY AND CONCLUSIONS

Safeguarding techniques based on consistency have been applied to accountability batch data of Yankee cores V and VI. To provide visual comprehension of the information available which is of value to a safeguards program, the results are summarized as levels of a pyramid in Figure 2. Nature has provided the fundamental laws that the techniques are based upon. These laws are described mathematically as the transmutation or burnup equations and constitute the foundation of all levels at which safeguarding information was obtained. Significant areas of information from the work completed to date are indicated as levels 1,2,3 and 4 of the pyramid in Figure 2. Each level is discussed below and considers that irradiated fuels arriving at a chemical processing plant have come from two possible reactor situations. These situations are noted as data cases I and II in Figure 2. That is, the accountability data of Yankee core V and VI are assumed to be the first dissolution data from the Yankee reactor and most of the work done assumed that premise to illustrate data case I. Previous accountability data from Yankee cores I thru IV were used to illustrate data case II.

A. Verification of Plutonium Input

The U and Pu isotopic compositions provide easily applied empirical relationships for verifying the Pu concentration of spent power reactor fuels. Of particular importance are the relationships between the Pu/U ratio, ^{235}U depletion, initial ^{235}U enrichment and Pu content, and between U and Pu isotopic ratios and Pu content. When these are combined with fabrication records for total U corrected for reactor burnup, independent cross checks can readily be made for total Pu input to a chemical plant. For application of the verification methods, the safeguards agency, or other users, need

DATA CASE I

NEW REACTOR-DISSOLUTION OF FIRST CORES-VERY LITTLE DATA HISTORY.

DISSOLVE FUEL ACCORDING TO ENRICHMENT, EXPOSURE, AND ASSEMBLY POSITION.

GO TO ALL LEVELS

1. FIRST GO TO LEVEL 3
2. DEVELOP LEVEL 2.
3. BEGIN TO DEVELOP LEVELS 1 AND 4

REMEASURE RANDOMLY SELECTED SAMPLES

DATA CASE II

DATA HISTORY FOR REACTOR ESTABLISHED

NO SPECIAL DISSOLUTION REQUIREMENTS

GO TO LEVELS 1 AND 3

REMEASURE RANDOMLY SELECTED SAMPLES

FIGURE 2. PYRAMID OF CONSISTENCY OF MEASURED ACCOUNTABILITY DATA

simply accumulate the proper data from the diffusion plant, fabrication plant, reactor operator and the chemical processing plant. The initial data base can be greatly enhanced by ordering fuel assembly dissolution according to initial enrichment, exposure and final core position, data case I, but this would not be necessary with subsequent cores, data case II.

B. Reactor Characteristics or "Fingerprint"

1. By Data Manipulation

A number of relationships may be used as a "fingerprint" to confirm stated information about the fuel i.e., reactor type, initial enrichment, etc. Each of the data parameters that are discussed in Section V thru VII are characteristic of the Yankee Reactor and therefore identify data that are consistent with the curves and tables shown as coming from measurements of irradiated Yankee fuel. The many data paths that can be constructed from various combinations of the data thus constitute a "fingerprint" of the core. Dissimilar cores exhibit separate data paths and therefore different fingerprints. Often, a small set of well chosen data plots are sufficient to fingerprint a core.

Among the numerous "fingerprint" plots that can be made, those that form a straight line are especially useful. For accountability data from cores V and VI, these are:

- o $^{236}\text{U}/^{238}\text{U}$ vs $^{235}\text{U}/^{238}\text{U}$ Figure 6
- o $^{240}\text{Pu}/^{239}\text{Pu}$ vs $(^{242}\text{Pu}/^{240}\text{Pu})/(^{236}\text{U}/^{238}\text{U})$ Figure 9
- o $^{241}\text{Pu}/^{239}\text{Pu}$ vs $^{242}\text{Pu}/^{241}\text{Pu}$ Tabulated Table III
- o $(^{242}\text{Pu}/^{241}\text{Pu})/(^{235}\text{U}/^{236}\text{U})$ vs $(^{241}\text{Pu}/^{240}\text{Pu})/(^{239}\text{Pu}/^{238}\text{U})$ Tabulated Table III
- o Pu/U vs ^{235}D (where $^{235}\text{D} = \text{initial } ^{235}\text{U wt\%} - \text{final } ^{235}\text{U wt\%}$) Figure 10

These plots are useful for two reasons. First, there is greater confidence in extrapolating the straight line of these plots when checking the reliability of data points well removed from the rest of the data. Secondly, all of the information about the plot is contained in a single number, the slope of the plot. Given the data point of the initial fuel, each sample has its representative slope which is constant, within measurement uncertainties, for all samples along the straight line. The results show that the slope parameters change with initial ^{235}U enrichment and therefore, presumably, also with other core design variables. Thus, the slopes of these plots constitute a concise, numerical and graphical fingerprint of a core for a given enrichment and a constant core design.

One set of parameters, $(^{242}\text{Pu}/^{241}\text{Pu})/(^{235}\text{U}/^{236}\text{U})$ as a function of $(^{241}\text{Pu}/^{240}\text{Pu})/(^{239}\text{Pu}/^{238}\text{U})$, appeared to be independent of initial enrichment and thus constitutes a part of the fingerprint that can be used without prior knowledge of enrichment for the Yankee reactor. Empirical relationships as a function of initial enrichment can be determined for the remaining slope parameters in the same manner as was done for $\frac{\text{Pu (grams)}/\text{U (tonne)}}{^{235}\text{D}}$ in Section VII. Empirical relationships involving other core design variables may be available upon further investigation, such that the fingerprint can be developed to include all possible core and fuel design changes.

Plots which are linear for ^{235}U enriched fuels (such as Yankee cores V and VI) may not be linear for other cores, especially those that are plutonium enriched. In this case, other linear plots may exist. At the present time more of the nuclear industry is using ^{235}U enriched fuel, so the plots suggested above should be quite useful.

2. Fingerprint by Cross Section Ratios

The isotopic concentrations of the uranium and plutonium isotopes are interrelated through transmutations equations. These equations have been cast in a form that includes the uranium and plutonium concentrations and the ratios of effective (flux averaged) cross sections. The isotopic concentrations are known experimentally from the ^{137}Cs and accountability data. A least squares fitting code was used to adjust the cross section ratios to obtain a best least squares fit of the transmutation equations to the experimental data. Good least squares fits to the data were obtained (see Figures 15-20). However, the resulting cross section ratios are inconsistent with expected patterns. This difficulty has been attributed to the simple analytical forms assumed for the cross section ratios in the current version of the least squares code. It was hoped that these cross section ratios would provide another approach for fingerprinting a core. Improvements in the code might still provide a means of attaining this worthwhile goal. This area also requires a continuing experimental and theoretical program involving data from different fuel designs and reactor types to fully delineate the concept.

3. Smudging of the Fingerprint

The fingerprint of a core can be altered (smudged) by changing core design and reactor operation variables. The effects of changing some of the more important design and operating variables upon the fingerprint are shown in Figures 24-30. Table XIII is an aid in the interpretation of Figure 25-29. For those cores where the requisite reactor information is available, a reliable reactor burnup code for studying the effects of design and operational changes on the fingerprint could be useful part of a materials safeguards program.

C. Internal Consistency Analysis

Separate from the concept of bias control and independent verification, is the concept of internal consistency analysis. Nuclear material safeguards for reprocessing plants are built on the foundation of accountability data. The reliability of these data is fundamental to the safeguards process. It is also vital to the closely related fingerprint of a core. The burnup relationships also provide an excellent basis for determining the internal consistency and thus the reliability of the chemical plant data. Internal consistency as a means of locating poorly measured data for subsequent remeasurement is extremely important in safeguards control situations. In addition, when the core is of the same initial enrichment, or processed according to initial enrichment, the consistency checks can be applied without having previous history.

The accountability data have been analyzed by a data evaluation technique previously used to establish the reliability of the closely related data from small, spent-fuel samples. The method consists of letting the collective data define a line or path that the individual data points can then be compared against. Exposure differences between the batches cause the data to be strung out along a line rather than grouped at a single point.

The data paths of cores V and VI were well defined, making the evaluation approach for determining the internal consistency of measured accountability data all the more effective. Individual deviations from a path were easily detected. This method successfully uncovered all of the incorrect measurements that were later turned up via the remeasurement process. The one exception occurred for an isolated sample (sample 2, initially 2.9 fuel) which lacks other samples of its own kind that it can be compared with.

Thus, when sufficient, high-quality data are available, complete remeasurement of the data is not required to establish the data's reliability. Testing the data for internal consistency can economically accomplish the same thing. Only samples labeled as "bad" by this evaluation technique need to be re-measured. If the product data consists of accountability batches from the same core with the same enrichment, the product data should fall on the same path as the data from the accountability tank. If, on the other hand, the product data is a mixture from different cores of different enrichments, as occurred in FT-62, this data must be remeasured to assure its reliability. Current practice is that the accountability data is remeasured by the product receiver.

Finally, it is often difficult to establish the reliability of a data point well removed from all others (e.g., sample #1), since there are no surrounding points to compare with it. Automatic remeasurement is a prudent procedure in this situation.

Analysis of paired measurements sets and comparison to independent sets of data measurements indicate that current plant measurement methods are sufficiently precise and accurate for application of the verification techniques. In general, excellent agreement was obtained between plant mass spectrometer measurements and those of NBS. The total uranium input stated by the reactor was in excellent agreement (0.2%) with plant measurements. This again confirms earlier belief that the total uranium in a spent fuel core can be known previous to chemical plant measurements (from fabrication measurements corrected for reactor burnup) to a high degree of certainty. Another interesting piece of information uncovered through the comparison is that approximately five percent of the fuel material estimated to have been in batch 2 was missing, according to the accountability measurements, from the fuel processing for that batch.

A comparison of the batch data with the final product data shows that 1.43% (279.4 kg) of the uranium and 2.40% (4.21 kg) of plutonium was lost during reprocessing. These discrepancies are considerably larger than can be accounted for by measurement uncertainties in the data and are attributed to reprocessing losses.

The concentration of the fission product ^{137}Cs in each fuel batch was measured. The ^{137}Cs measurements were combined with the accountability data for use in a least squares analysis program designed to provide a fingerprint based on ratios of effective cross sections. The average exposure of each batch was also determined from the ^{137}Cs measurements.

D. Improvement in Reactor Predictions

The accountability data were compared with the predictions of a burnup code. This code underestimates the end-of-life amount of ^{235}U for the fuel batches of cores V and VI (especially core VI). The total plutonium is overestimated for core V and underestimated for core VI (see Table VI). The exposure is overestimated in both cores as a direct result of ^{235}U being underestimated. Because ^{235}U was not estimated correctly, it is felt that the isotopic concentrations of plutonium, as calculated, are also in error. The ability to calculate the isotopic composition of both uranium and plutonium may add credence to the reactor operators estimates. To bring about better agreement, both the shape of the data curves and the numerical values of the slope parameters from accountability data should be compared to those from the calculation.

E. Need for Further Work

As yet, work on assessing the safeguards value and application of chemical plant input measurements relating to burnup, has pertained largely to one pressurized water reactor, namely Yankee Rowe with basically one fuel design.

To fully assess the value and also to obtain the best base for future applications, a consistent, continuing effort needs to be pursued both in broadening the data base and in extending the theoretical bounds to the relationships. The program also needs to be extended in terms of completeness of data base. Of particular importance are the data from the fabrication and irradiation stages of the fuel cycle.

The isotopic combinations developed in Section V form an easily identifiable fingerprint and are quite useful for establishing the internal consistency of the data. However, many of the linear relations found in Section V are expected to break down for fuels which are plutonium enriched instead of ^{235}U enriched. A general plan for all fuel types from all reactors is needed. The results of our study of the Yankee Cores V and VI accountability data lead us to propose the "bare bones" plan of Section XI for accomplishing this end. The plan monitors only the reprocessing plant. Other parts of the fuel cycle are not covered. Potentially useful interrelations between different parts of the cycle have been largely ignored.

III. DESCRIPTION OF EXPERIMENT

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

In past dissolution campaigns of spent reactor fuels, fuel assemblies from a given core have not been dissolved in any predetermined order. A unique feature of the dissolution of Yankee cores V and VI was that the order of processing of the fuel was determined before the actual campaign began. Table A-I of Appendix A lists the dissolution sequence of the fuel assemblies where the ordering takes into account the operation requirements of NFS, the initial enrichment of the fuel, the estimated total exposure of each assembly, and the final core position of each assembly. Only assemblies of the same initial enrichment were grouped together in making up a batch. The estimated exposure ranged from approximately 9700 to 27,000 MWD/MTU*, and each dissolution batch contained fuel assemblies having approximately the same total exposure. Each group of 2 or 4 assemblies of 4.1 fuel from core V were from symmetric locations in the core

*Megawatt Days/Metric Ton (tonne) Uranium

with reference to the positions of the 2.9 fuel assemblies as shown in Figure 3 (except batch No. 1 which contained the displaced 4.1 assemblies). Position symmetry of the 4.94 fuel assemblies from core VI, Figure 4, was not strictly maintained for each group of fuel because 3 instead of 4 assemblies were dissolved per batch.

The ordering of the fuel assemblies thus resulted in 11 dissolution batches of 4.94 fuel, 11 dissolution batches of 4.1 fuel and 1 dissolution batch of 2.9 fuel. From pre-irradiation data shown in Table A-1, the 74 assemblies represented initially 20,189.489 kgs of uranium, of which it was estimated that 19,571.020 kgs of uranium were still available to be processed. The estimated loss of uranium while under irradiation in the reactor was 3.04% and the amount of initial uranium converted to plutonium was estimated to be 0.87%. Totals on a batch per batch basis are also shown in Table A-I from the pre-irradiation data. (Batch sequence number 23 is replaced by number 24 and batch 23 does not exist).

The batch identity as discussed above was maintained during the cutting of the fuel assemblies for each batch, the subsequent dissolution of the cut fuel in the dissolution tank and during the time the dissolved fuel was in the input accountability tank where volume measurements were made and duplicate representative samples of the dissolved fuels were obtained and analysis. Cross contamination between batches of dissolved fuel (the dissolving sequence alternated between 4.1 fuel and 4.9 fuel) was kept at a minimum by rinsing the dissolver tank after transferring dissolved fuel to the accountability tank⁽⁷⁾. Each dissolver batch was transferred from the dissolver to the input accountability tank except for the jet heel

FIGURE 3 CORE LOCATION OF FUEL ASSEMBLIES DISCHARGED AS SPENT FUEL FROM CORE V

FIGURE 4 CORE LOCATION OF FUEL ASSEMBLIES DISCHARGED AS SPENT FUEL FROM CORE VI

in the dissolver, approximately 74 liters. The volume of dissolver batches ranged from about 3900 to 4300 liters. Following the transfer, 200 to 300 liters of rinse were added to the dissolver and the rinse solution was then transferred and added to the solution in the input accountability tank. After measurement and sampling, the batches were transferred to the solvent extraction system leaving only a jet heel of approximately 4 liters in the input accountability tank. Thus, each set of input batch analytical measurements should be representative of the fuel specified for that batch, smeared only by the tank heels of the dissolver and the input accountability tanks.

A summary of the samples taken throughout the processing of Yankee core V and VI is shown in Figure 5. Duplicate samples were obtained for 24 input batches, 23 batches of dissolved fuel assemblies and 1 batch of flush material, batch 26. Intermediate points between input and separated uranium and plutonium product were also sampled as shown (the data from these intermediate samples will not be discussed). Duplicate samples were also obtained from the 5 batches of uranium product and 8 batches of plutonium product that were the end result of the processing.

Analytical measurements on 2 duplicate samples from each input batch of dissolved fuel and each uranium and plutonium product batch were done in the analytical laboratory at NFS. These measurements are referred to hereafter as normal plant measurements and are listed on line A of Table I. Another unique feature of the dissolution of Yankee cores V and VI fuel was that additional measurements were made by NFS, lines B and C; by NBS, line D; by New Brunswick Laboratories (NBL), line E; and by II and Ledoux & Company acting jointly for YAEC, line F.

The input uranium and plutonium concentrations were measured by NFS using mixed spike isotopic dilution techniques in addition to normal plant methods to evaluate to mixed spike technique for the determination of solution content. Measurements were also done to determine ^{137}Cs to provide fission product data for each input batch. Data measurements by NBS and NBL were made to test the measurement accuracy and provide independent measurements of all the dissolver material. Measurements noted in lines A through E were originally scheduled in planning the experiment. However, the measurements by NBL were done at the request of the IAEA and will be incorporated only into the FT-62 final report. The data measurements by II were provided through a special agreement since these measurements were done for the Yankee Atomic Electric Company. The data to be discussed in this report are the data indicated on lines A through D and F of Table I.

The normal plant measurements are considered in this report to be the basic set of accountability data for the fuel from Yankee cores V and VI. It is these measurements that the remainder of the report is primarily concerned with. The other measurements are compared to this basic set of accountability data. All the data taken are summarized in Tables A-II, A-III, and A-IV in Appendix A. From the tables, it is noted that the isotopic compositions were reported as weight percents, uranium and plutonium concentrations are grams/liter and total grams, and the ^{137}Cs in disintegrations per minute per milliliter (dpm/ml). Approximate periods of time in days since fuel discharge to data of analysis for ^{241}Pu and ^{137}Cs are included in the tables in order to decay correct each to the data of discharge.

FIGURE 5 SAMPLING POINTS FOR FT-62

TABLE I

SUMMARY OF ANALYTICAL MEASUREMENTS DONE FOR DUPLICATE INPUT BATCH SOLUTION
AND URANIUM AND PLUTONIUM PRODUCT SOLUTION SAMPLES

	INPUT BATCH SOLUTION SAMPLES (24)			PRODUCT SOLUTION SAMPLES			
	URANIUM AND PLUTONIUM		^{137}Cs	URANIUM (5)		PLUTONIUM (8)	
	ISOTOPIC COMPOSITION	CONCENTRATION		ISOTOPIC COMPOSITION	CONCENTRATION	ISOTOPIC COMPOSITION	CONCENTRATION
A	NFS - MS ^a	NFS - MSDS ^b		NFS - MS ^a	NFS - MSSS ^c	NFS - MS ^a	NFS - MSSS ^c
B		NFS - MSMS ^d					
C			NFS - DC ^e				
D	NBS - MS ^a			NBS - MS ^a		NBS - MS ^a	
E*	NBL	NBL		NBL	NBL	NBL	NBL
F	II - MS ^a	II - MSMS ^d		II - MS ^a	II - G&V ^f	II - MS ^a	II - AT ^g

*Data not available

LEGEND:

- a. MS - Done using mass spectrometer.
- b. MSDS - Done using mass spectrometer isotopic dilution techniques - double spike.
- c. MSSS - Done using mass spectrometer isotopic dilution techniques - single spike.
- d. MSMS - Done using mass spectrometer isotopic dilution techniques - mixed spike.
- e. DC - ^{137}Cs determinations done by direct counting method using a Ge(Li) diode.
- f. G & V - Uranium determined both by gravimetric and volumetric methods by Ledoux & Company
- g. AT - Plutonium determined by amperometric titration at N.F.S. facilities.

IV. DATA EVALUATION

The property of internal consistency will be illustrated first to answer the question of measurement accuracy. By being consistent, it is meant that a data parameter traces a continuous distinct path as a function of exposure. Plotted data points are expected to show the burnup path and to lie close to a smooth curve that can be drawn by inspection through the data points. A data parameter can be part of the accountability data in the form it was measured or a quantity derived from the data. Generally speaking, the isotopic data expressed as weight percents are derived data parameters since the measured quantities are atom ratios. However, it is a simple matter to reform the measured atom ratios from the given weight percents and the uranium and plutonium totals. This was done by the use of the computer program ACTSUR, a program that not only forms the measured atom ratios, for example $^{235}\text{U}/^{238}\text{U}$, but manipulates the data to form other possible atom ratio combinations of ^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu . The inverse of any given ratio was not formed. In converting the data in weight percents to atom ratios, all the isotopes were first expressed as atoms per gram of uranium (atom/grams U). Of the 21 ratio parameters possible, 9 parameters, shown in Table II, form a convenient set to test measurement accuracy by plotting the data parameters as indicated in Table II.

A. Normal Plant Measurement Data

In the case of the uranium isotopic composition measurements made by normal plant methods, the $^{236}\text{U}/^{238}\text{U}$ data as a function of $^{235}\text{U}/^{238}\text{U}$ data are shown in Figure 6 for both the input batch samples and the uranium product samples. The initial or zero burnup points for each enrichment

TABLE II. Data Parameters Listed in Column A Have Been Plotted As A Function of Data Parameters in Columns B to Test Measurement Accuracy of Isotopic Compositions.

A		B			
1	$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{235}\text{U}}{^{238}\text{U}}$			
2	$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$		$\frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{236}\text{U}}{^{235}\text{U}}$	
3	$\frac{^{241}\text{Pu}}{^{240}\text{Pu}}$				
4	$\frac{^{242}\text{Pu}}{^{240}\text{Pu}}$				
5	$\frac{^{241}\text{Pu}}{^{239}\text{Pu}}$				$\frac{^{240}\text{Pu}}{^{239}\text{Pu}}$
6	$\frac{^{242}\text{Pu}}{^{239}\text{Pu}}$				
7	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}}$

Figure 6 Uranium Isotopic Composition Data from Input Batch Samples and Product Batch Samples for Yankee Cores V and VI.

are also shown and it is evident that the data are tracing continuous paths from zero exposure to exposures in the range of 25,000 to 27,000 MWD/MIU. (The initial ^{236}U values are not well known, except for the 4.94 fuel ⁽⁹⁾, and the values shown for 2.9 fuel and 4.1 fuel are estimated by extrapolation and examination from known ^{236}U values of other enrichments.) A straight line, as drawn by inspection, serves to represent the burnup paths where the slope, S, of the lines can be expressed as

$$S = \frac{\hat{\alpha}^{235}}{\hat{\alpha}^{235} + 1} - \frac{\hat{\sigma}_c^{236}}{\hat{\sigma}_a^{235}} \frac{N^{236}}{N^{235}} \quad (2)$$

(The ratio of capture to fission in ^{235}U is $\hat{\alpha}^{235}$). The consistency of the individual input batch data parameters is apparent from Figure 6 and by close examination of the graph, it is noted that each input data point lies within $\pm 1\%$ of the straight lines. None of the 4.1 and 4.94 input batch samples are in doubt as far as ^{236}U , ^{235}U , ^{238}U isotopic compositions are concerned. However, since there is only one 2.9 input batch, it is not possible to conclude whether the sample data are consistent or not. This would be possible by data evaluation methods only if other 2.9 data were present to show the burnup path.

The uranium product isotopic data parameters should also exhibit consistency. It is known that input batch identity is not maintained once the dissolved solutions are pumped from the accountability tank. Mixing of batches occur from this point on and means that at the start of the campaign, 2.9, 4.1 and 4.94 fuels were mixed and were present in the first uranium product batch. Subsequently, only 4.1 and 4.94 fuel were mixed on a scale of four-4.1 fuel assemblies to three-4.94 fuel assemblies as the campaign continued. The physical mixing requires that

the product data lie between the burnup paths of the 4.1 and 4.94 fuels and also that these points should lie closer to the 4.1 fuel's burnup path. This is the case, as seen from Figure 6, and the effect of the 2.9 fuel on the first uranium product batch was to move this data point even closer to the 4.1 fuel burnup path. Thus the product uranium isotopic data do show consistency.

At the same time it is difficult to determine whether or not any one uranium product sample is inconsistent by plotting the uranium product isotopes. They can be plotted only on one graph of Table II (No. 1) and, as indicated, there are reasons for them to scatter as they do.

As an example of the consistency of plutonium isotopic data, $^{242}\text{Pu}/^{241}\text{Pu}$ data as a function of $^{240}\text{Pu}/^{239}\text{Pu}$ data are shown in Figure 7 for the input batch samples and the plutonium product samples. The ^{241}Pu has been decay corrected to date of discharge for each input batch sample in order to compare the ^{241}Pu that resulted from cores V and VI. However, since the plutonium product samples are mixtures of cores V and VI, the correction for ^{241}Pu decay is not straightforward. An average cooling time weighted by the amount of 4.1 and 4.94 plutonium in each product batch is needed. Since this information is not possible to measure, a decay correction has been estimated and both the corrected and uncorrected product data are shown in Figure 7.

The first observation concerning the input batch sample data shown in Figure 7 is that a variation of the initial ^{235}U enrichment does not have a large effect on changing the burnup path of the plutonium isotopes. Before the shape of the curves shown was drawn by inspection, all the graphs of Table II were examined for data consistency so that the two curves shown

Figure 7 Plutonium Isotopic Composition Data from Input Batch Samples and Product Batch Samples for Yankee Cores V and VI. The Product Data are Shown Plotted both for Uncorrected ^{241}Pu and Corrected ^{241}Pu .

are consistent with all the information available. It was expected that a third curve would indicate the burnup path of the 2.9 fuel sample. However, the 2.9 data point lies near the 4.94 fuel burnup path and it is not possible to conclude that a third burnup path should be indicated with only one data point available.

Three samples have been flagged as being high or low and the isotope or isotopes that appear inconsistent are noted on the graph for each sample. With the exception of those samples noted, input batch sample data points consistently lie within $\pm 1\%$ of the curves shown indicating the consistency of these plutonium isotopic composition data.

The plutonium product isotopic compositions also reflect consistency since they describe a smooth curve. Another point of interest is that the estimated correction for ^{241}Pu decay was fairly accurate because the corrected product data generally do plot between the 4.1 plutonium and the 4.9 plutonium input data. Never-the-less, the uncorrected product data are seen to trace a path parallel to the corrected input batch data. The product data can be plotted on three graphs of Table II (No. 5, 6, and 7) and based on these graphs, none of the plutonium product samples isotopic data are considered inconsistent.

The consistency between uranium and plutonium isotopic data can be checked by plotting any of the graphs of Table II that involve uranium and plutonium isotopes. For example, $^{241}\text{Pu}/^{240}\text{Pu}$ as a function of $^{235}\text{U}/^{238}\text{U}$ is appropriate. However, a plot of $^{240}\text{Pu}/^{235}\text{U}$ as a function of $^{241}\text{Pu}/^{238}\text{U}$, Figure 8, which involves the same four isotopes in a different format is more useful because the resulting burnup path traced by these parameters is nearly linear and the initial values are zero.

Figure 8 Uranium and Plutonium Isotopic Composition Data from Input Batch Samples for Yankee Cores V and VI.

Consistency checks are made easier when such parameter properties are available by data manipulation. The curves shown were drawn by inspection and the consistency of the data indicate that the uranium isotopes match the plutonium isotopes except for sample 1 where it has been noted that ^{241}Pu is low. (There are no matching uranium and plutonium product data).

The uranium and plutonium isotopic data for input batch samples and product samples thus appear consistent whether plotted separately or together, except for the data noted. Data manipulation can be used to advantage to form other isotopic parameters for input batch isotopic data such as a double ratio, $^{240}\text{Pu}/^{235}\text{U}/^{241}\text{Pu}/^{238}\text{U}$, since the values of this particular parameter are nearly constant as a function of exposure (the parameter represents the slope of the curves shown in Figure 8 and the slope of a straight line is a constant). A total of 35 double ratios are possible if we make the restrictions that each isotope is introduced linearly (not ^{240}Pu squared, for example) and the same 4 isotopes are not repeated in a different sequence (for example, $^{241}\text{Pu}/^{240}\text{Pu}/^{235}\text{U}/^{238}\text{U}$ would not be considered). The double ratio parameters can also be plotted either as a function of single ratio parameters, $^{236}\text{U}/^{235}\text{U}$ for example, or as a function of a double ratio parameter. The choice of two parameters for plotting was based on the range of the parameters from lowest exposure to highest exposure and two parameters having the same range were plotted, one as a function of the other. For example, $^{240}\text{Pu}/^{239}\text{Pu}$ had the same range as $^{242}\text{Pu}/^{240}\text{Pu}/^{236}\text{U}/^{238}\text{U}$ for both the 4.1 and the 4.94 fuel batches. The resulting plot is linear, Figure 9, where the slope, S_1 , is given by

$$S_1 = \frac{^{240}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{240}\text{Pu}}{^{242}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}} \quad (3)$$

Figure 9 $^{240}\text{Pu}/^{239}\text{Pu}$ as a Function $^{242}\text{Pu}/^{240}\text{Pu}/^{236}\text{U}/^{238}\text{U}$.
Data Parameters Indicate Possible Ratios of Isotopes from
Data Manipulation and Results when such Parameters Are Plotted.

(Slope parameters involve isotopic parameters squared.) The value of S_1 for each input batch is listed in Table III. Other parameters are also identified in Table III which when plotted describe a linear or nearly linear burnup path. The input batch samples are listed in the order of decreasing exposure and separated according the initial enrichment. This format is used to observe any smooth variation of the slope as a function of exposure, which would mean that the burnup path involved is not truly linear. The averages and plus or minus 1 standard deviations, $C \pm \sigma$, are shown for the slopes that appear to be constant.

It is noted from Table III that the different slope parameters involve from 2 to 7 isotopes. A convenient test of data consistency, in addition to the graphs of Table II, is a tabulation of constant parameters such as those listed in Table III because inconsistent data will be noted by an unexpected change in a given parameter. For example, in column A, sample 24 is low and sample 1 is high which is consistent with the information given in Figure 6. All the remaining values are consistent within plus or minus 2 standard deviations, $\pm 2\sigma$, of the average values. As a general rule, remeasurement of a sample for more accurate values of the isotopes shown in column A should be done if the sample value is outside $\pm 2\sigma$. Thus, samples 1 and 24 should be remeasured for better ^{242}Pu values.

The parameters of column B are those shown in Figure 8. The values of the slope show the smooth variation as a function of exposure expected when the resulting burnup path is not truly linear. The expected smooth variation is also another means of testing for consistency, and it is noted that sample 1 is the only sample that departs from the trend. In this case, the ^{241}Pu was indicated to be low.

TABLE III
TABLE OF DATA PARAMETERS WHOSE RESULTING BURNUP PATTERNS ARE DESCRIBED BY A STRAIGHT (OR NEARLY STRAIGHT) LINE.
NUMERICAL VALUES OF THE SLOPE PARAMETERS OFFER A CONVENIENT TEST OF MEASUREMENT ACCURACY.

ENRICHMENT	BATCH NUMBER	SLOPE																
		A		B		C		D		E		F		G				
		$\frac{^{240}\text{Pu}}{^{242}\text{Pu}} \cdot \frac{^{240}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{242}\text{Pu}}{^{240}\text{Pu}} / \frac{^{236}\text{U}}{^{238}\text{U}}$	$\frac{^{240}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{238}\text{U}}{^{235}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{238}\text{U}}$	$\frac{^{241}\text{Pu}}{^{242}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{242}\text{Pu}}{^{241}\text{Pu}}$	$\frac{^{242}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{242}\text{Pu}}{^{241}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}}$ VS $\frac{^{239}\text{Pu}}{^{238}\text{U}}$	$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{241}\text{Pu}}{^{239}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{238}\text{U}}$ VS $\frac{^{241}\text{Pu}}{^{240}\text{Pu}} / \frac{^{239}\text{Pu}}{^{238}\text{U}}$
4.94	13	0.00955	48.48	0.905	1.56	0.01023	5118.	1.59										
	21	0.00950	48.41	0.899	1.53	0.01015	4980.	1.57										
	15	0.00960	48.10	0.909	1.52	0.00994	4975.	1.62										
	5	0.00963	47.92	0.909	1.51	0.00988	4978.	1.65										
	11	0.00954	46.88	0.919	1.56	0.00985	5016.	1.59										
	7	0.00944	46.99	0.895	1.55	0.01010	5023.	1.59										
	19	0.00942	46.68	0.915	1.57	0.00991	5018.	1.63										
	9	0.00944	46.92	0.904	1.56	0.00997	4999.	1.60										
	17	0.00953	46.60	0.918	1.56	0.00986	5019.	1.60										
	24	0.00907*	46.42	0.865*	1.54	0.01024	4922.	1.56										
	3	0.00946	46.23	0.894	1.54	0.00975	4883.	1.59										
		0.00951 ± 0.00008		0.907 ± 0.009		0.00999 ± 0.00016	4994 ± 60											
4.1	16	0.00783	66.91	0.840	1.71	0.01015	5417.	1.61										
	18	0.00780	63.14	0.860	1.76	0.01018	5307.	1.66										
	22	0.00783	59.78	0.886	1.83	0.01026	5440.	1.56										
	12	0.00785	60.24	0.869	1.80	0.01029	5378.	1.64										
	4	0.00798	59.28	0.886	1.80	0.00999	5431.	1.60										
	8	0.00774	58.34	0.870	1.84	0.01009	5471.	1.52										
	10	0.00767	58.58	0.865	1.85	0.01018	5416.	1.54										
	14	0.00787	58.68	0.870	1.85	0.01011	5478.	1.52										
	6	0.00777	58.00	0.875	1.84	0.00992	5446.	1.53										
	20	0.00793	56.64	0.928*	1.86	0.00920*	5447.	1.50										
	1	0.00810*	64.36	0.765*	1.89	0.00991	5375.	1.40										
		0.00783 ± 0.00009		0.869 ± 0.014		0.01011 ± 0.00013	5419 ± 49											
2.9	2	0.00592	102.21	0.672	2.07	0.01141	5765.	1.58										
-	26	0.00953	46.92	0.905	1.58	0.01003	4981.	-										

*value not used in average.

The parameters shown in columns C and D are additional parameters that, when plotted, describe a linear function, column C, or a near linear function, column D. However, column C is included to indicate that random measurement errors can be amplified to aid detection. For example, errors of ^{241}Pu are amplified by the fact that the ^{241}Pu of the slope is squared. The result is particularly noticeable for samples 20 and 1.

Column D is an example where the slope involves only 2 isotopic parameters. However, in ratioing one plutonium isotope to one uranium isotope, data other than the isotopic data are introduced, and these data are the uranium and plutonium concentrations measurements. That is,

$$\frac{^{239}\text{Pu}}{^{236}\text{U}} = \frac{^{239}\text{Pu wt\%}}{^{236}\text{U wt\%}} \cdot \frac{\text{Pu (grams)}}{\text{U (grams)}} \cdot \frac{236.04}{239.05} \quad (4)$$

where 236.04 and 239.05 are the atomic weights for ^{236}U and ^{239}Pu respectively. These measurements are introduced into isotopic ratio parameters such as the slopes shown in column D and E of Table III whenever a ratio of plutonium to uranium isotopes occurs in the slope parameter. The fact that the slope of column D does change as a function of decreasing exposure is evident from the 4.1 fuel data values where the values increase. The increasing trend is not as obvious with the 4.94 fuel data values and the reason will be considered after discussing the parameters in column E.

The slope of column E should be written

$$\frac{^{242}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{240}\text{Pu}}{^{241}\text{Pu}} \cdot \frac{^{236}\text{U}}{^{235}\text{U}} \cdot \frac{^{239}\text{Pu wt}\%}{^{238}\text{U wt}\%} \cdot \frac{\text{Pu (grams)}}{\text{U (grams)}} \cdot \frac{238.05}{239.05} \quad (5)$$

and we see that this slope includes the Pu/U ratio also. The plotting parameters shown in column E are of particular interest for two reasons. First, the slope includes all 7 isotopes that are being considered in the data evaluation plus the uranium and plutonium concentration measurements. Secondly, the values of the slope do not appear to be dependent on initial enrichment for the 4.94 and the 4.1 fuel (the value of the 2.9 fuel is high but without additional data points, it is not known whether or not the value is correct). This presents a convenient check of most of the accountability data for consistency (^{234}U and ^{238}Pu are not included). All the random effects of measurement are reflected in the values of column E and one would then expect to obtain a value of 0.0100 ± 0.0002 for each set of batch measurements when the measurements are combined as shown in equation 5. Sample 20 would be rejected on this basis due to ^{241}Pu and ^{242}Pu while random effects in the Pu/U values make sample 24 high but only 2% high rather than the 4% expected due to ^{242}Pu as discussed. Sample 13 is high and sample 3 is low. The values of samples 3, 13 and 24 are within two standard deviations, 2σ, of the average, however, and the reason for the values being high or low is evident when the Pu/U data are considered separately.

Figure 10 presents the Pu/U ratio as a function of ^{235}U depletion, ^{235}D , where

$$^{235}\text{D} = ^{235}\text{U initial wt}\% (\text{BOL}) - ^{235}\text{U final wt}\% (\text{EOL}). \quad (6)$$

(BOL and EOL meaning beginning and end of life)

Figure 10 Ratio of Plutonium Measured to Uranium Measured as a Function of ^{235}U Depletion for Yankee Cores V and VI

It is noted that these parameters describe a straight line burnup path, the slope of which changes with initial enrichment. The values of the slope for each batch are shown in column F of Table III (units are grams plutonium per metric ton uranium over ^{235}U depletion). All values are generally within $\pm 2\sigma$ of the averages shown where all the samples values of both the 4.94 and 4.1 fuel were used to determine the averages. If samples 24 (2% high), 3 (2% low) and 18 (2% low) are deleted from the averages, the resulting 2σ deviations would not include these samples and, as a general rule they should be remeasured. However, based on data yet to be discussed, it is felt that the σ values shown in column G are appropriate and remeasurement is not necessary.

The results of data examination of Figure 9 and column G as related to the data considered in column D and E indicates that the Pu/U ratio causes the values of sample 13 to be high in columns D and E. Sample 24 is high due to ^{242}Pu but not 4% high due to a low Pu/U value. Thus we see that data manipulation of uranium and plutonium isotopic and concentration measurements leads to many convenient data parameters by which measurement accuracy can be tested. In the case of normal plant measurement data, it is evident from the evaluation presented that we are extracting information at the 1 to 2% level. To do this, measurement accuracy has to have been at least $\pm 1\%$ or better. It is emphasized that data evaluation methods based on the requirements that

- The plotted data points must lie on a near a smooth curve drawn through the data by inspection,

- o for a given constant parameter determined by data manipulation, the individual sample values should not vary outside $\pm 2\sigma$ of the average,
- o and for parameters that vary as a function of exposure, the sample values should not show unexpected changes,

have been successful in isolating inconsistent data, especially in the case of 4.1 and 4.94 input batch data. It will be of interest to see if duplicate measurements done for each sample supports the information gained thus far.

B. Duplicate Data Measurements By NBS, II and NFS

As indicated earlier, normal plant measurement data are considered the basic data set. Measurements made by NBS, II and NFS on duplicate samples have been evaluated in the same manner as the normal plant data. The results are presented by comparing these data sets to the basic data set following the evaluation sequence outlined in the previous section as much as possible.

Two additional sets of the uranium and plutonium isotopic composition data are available, one measured by NBS and the other measured by II. A comparison of the uranium isotopic data, $^{236}\text{U}/^{238}\text{U}$ as a function of $^{235}\text{U}/^{238}\text{U}$, is presented in Figures 11a, 11b and 11c (including product data) where Figure 11a contains NBS data measurements, Figure 11b contains II data measurements and Figure 11c contains normal plant measurements (same as Figure 6). Figures 11a and 11b are transparent and by laying one over the other to coincide with Figure 11c, it is evident that all input batch measurements trace the same burnup paths. Remeasurements of sample 2(2.9 fuel) indicate that the burnup path shown in Figure 6 from normal plant uranium isotopics is correct. It is also evident that the scatter of the data about the burnup paths increases for II data as compared to NBS and normal measurement data. This was expected since the analyses performed by NBS were designed to achieve maximum precision whereas the II measurements were made with sufficient precision to accomplish the objective; namely, to confirm that the total concentrations of ^{235}U , ^{239}Pu and ^{241}Pu were within a fixed percent of the values reported by the processors. The required precision of the measurements of minor isotopes was less than that of the required isotopes since their contribution to the total error was small. Comparison of these two sets of data yields an excellent example of the range of variability that could normally be expected in such measurements (an important consideration in safeguards applications).

The percent difference between normal plant data and the respective NBS and II data is shown in Table IV where the percent difference has been determined by

$$\text{Percent Difference} = \left[\left(\frac{\text{NBS, II or NFS Measurement}}{\text{Normal Plant Measurement}} \right) - 1.0 \right] 100. \quad (7)$$

The isotopic data expressed as weight percents were used in equation 7.

The input batch samples are again grouped according to enrichment and listed in order of decreasing exposure.

Sample 11, NBS input batch data, has the largest percent differences from corresponding normal plant data for each uranium isotope (considering input batch samples only) and in Figure 11a, it is the only data point that does not plot on the straight line for 4.94 fuel. Without the data graphs and the manipulation of data, one would tend to believe the measurements by NBS and II since they support each other rather than the normal plant measurements for sample 11. However, the consistency of the plotted data indicates that the two remeasurements are more inconsistent than the original measurement and data evaluation methods are as important as remeasurement. In the case of uranium product sample number 2, the opposite is true because data evaluation methods are limited in identifying consistent product data. Thus, where there are enough samples, data evaluation methods do provide important information and duplicate measurements can be done in support of the information learned. Where there is only one sample or scatter in sample data is consistent with mixing in a reprocessing plant, duplicate measurements are important. (Duplicate measurements of product batches will be done as a normal result of transferring product material to the fuel fabricator, for example.)

Figure 11 a,b, c Uranium Isotopic Composition Data from NBS Measurements (a),
 II Measurements (b), and Normal Plant Measurements (c).

TABLE IV
COMPARISON OF NBS AND II ISOTOPIC COMPOSITION MEASUREMENTS
AND NBS AND II U AND Pu CONCENTRATION MEASUREMENTS TO NORMAL PLANT MEASUREMENTS

²³⁵ U			²³⁶ U		²³⁸ U		²³⁹ Pu		²⁴⁰ Pu		²⁴¹ Pu		²⁴² Pu		Pu / U	
NBS	II		NBS	II	NBS	II	NBS	II	NBS	II	NBS	II	NBS	II	NBS	II
INPUT BATCH DATA																
13	+0.02	+1.0	+0.2	+1.5	-0.002	-0.04	-0.06	-0.07	-0.1	-0.2	+0.7	+0.7	-0.4	-0.2	-1.1	+0.08
21	+0.4	+1.2	+0.2	+1.1	-0.01	-0.05	0.0	+0.02	-0.02	-0.02	+0.4	-0.09	-0.6	-1.5	-0.3	-1.3
15	-0.2	+0.5	-0.2	+0.04	+0.006	-0.01	-0.03	+0.01	-0.4	-0.5	+0.5	+0.1	-0.4	-1.1	+0.2	+0.6
5	-0.3	-0.05	-0.4	-2.0	+0.01	+0.01	-0.08	-0.2	+0.05	+0.5	+0.6	+0.3	-0.09	-0.6	-0.3	+1.0
11	-0.6	-1.0	-0.9	+0.2	+0.02	+0.02	+0.08	-0.2	-0.4	+0.4	+0.2	-0.3	-0.4	+5.8*	-0.5	+0.6
7	-0.07	+1.3	-0.3	+1.2	+0.004	-0.05	+0.09	+0.4	-0.09	-0.05	+0.7	-1.4	-2.0	-1.6	-0.3	-1.4
19	+0.04	+0.5	+0.6	+0.3	-0.005	-0.02	+0.03	+0.08	-0.1	-0.05	+0.2	-0.3	-1.0	-2.2	-0.3	+0.08
9	+0.4	+0.7	+0.2	-0.6	-0.01	-0.02	-0.1	+0.2	+0.01	+0.01	+1.0	-0.02	-0.3	-0.8	-0.4	-1.0
17	+0.1	+0.5	+0.2	+0.7	-0.006	-0.02	-0.4	+0.1	-0.1	-1.6	+0.4	-1.7	-0.05	-3.7	-0.6	+0.02
24	+0.04	-0.3	+0.1	-0.2	-0.001	+0.009	+0.02	+0.05	-0.04	-0.1	+0.5	+0.3	-4.7*	-5.1*	+0.9	+0.1
3	-0.03	+0.4	+0.06	+0.7	+0.003	-0.02	-0.09	-0.2	+0.07	+0.7	+0.9	+1.1	-0.4	+0.1	+1.1	+1.6
16	-0.3	-0.2	+0.04	+0.1	+0.006	+0.003	-0.07	+0.3	-0.1	-0.8	+0.6	-1.0	-0.2	-2.3	+0.4	-0.04
18	+0.3	+1.3	+0.5	+1.2	-0.009	-0.03	-0.02	-0.2	-0.2	-0.06	+0.6	+1.2	-0.2	0.0	0.0	-0.1
22	-0.3	+0.2	-0.2	-0.5	+0.008	+0.03	-0.02	-0.04	-0.1	+0.05	+0.6	+0.5	-0.3	-1.1	-0.2	-1.0
12	-0.08	+0.7	0.0	+0.5	+0.002	-0.02	-0.08	-0.02	-0.1	+0.01	+0.9	-0.06	-0.1	-0.6	-0.3	-0.3
4	-0.3	+1.1	-0.2	0.0	+0.008	-0.001	+0.03	-0.03	-0.3	-0.1	+0.4	+0.4	-0.8	-0.9	-0.9	-0.4
8	-0.1	+0.2	+0.3	+0.2	+0.002	-0.007	+0.06	-0.08	-0.02	+0.5	+0.6	+0.5	-2.3	+0.6	-0.4	-0.1
10	+0.02	+0.4	+0.03	-1.0	-0.0005	-0.005	-0.05	-0.3	-0.2	-0.1	+0.8	+0.4	-0.6	-0.6	-0.6	+0.4
14	-0.04	+0.7	+0.4	+0.7	-0.0002	-0.02	-0.01	-0.1	-0.07	-0.3	+0.7	+0.2	-1.8	-2.5	-0.9	-1.4
6	-0.4	-0.6	-0.2	-0.4	+0.01	+0.02	+0.1	+0.1	-0.2	-0.2	+0.1	-0.5	-2.0	-0.6	+2.6*	0.0
20	-0.05	-0.03	0.0	-0.3	+0.002	+0.002	+0.5	+0.4	+0.2	+0.5	-2.1	-2.3	+1.3	+0.3	-0.04	-1.1
1	+0.1	+0.6	+0.3	-0.9	-0.003	-0.02	+0.2	+0.1	-0.2	+0.2	-1.0	-1.2	-9.1*	-10.0*	+0.9	+2.7*
2	+0.2	-1.1	+0.5	+0.09	-0.006	-0.02	+0.02	+0.4	+0.1	-0.1	+0.5	-0.3	-13.4*	-14.1*	-0.9	-0.2
26	+0.1	-0.4	+0.2	+0.5	-0.005	+0.005	+0.01	+0.02	-0.2	-0.7	+0.2	-0.2	-0.5	-2.1	--	-0.2
PRODUCT DATA																
1	-0.2	+0.6	-0.3	+0.3	+0.006	-0.02	-0.07	-0.1	+0.09	-0.2	+0.8	+0.8	+0.8	+0.5		
2	-1.7*	-1.3	-0.6	+0.3	+0.06	+0.04	+0.01	-0.001	-0.04	-0.2	+0.1	-0.2	-0.6	-1.9		
3	-0.2	+0.6	-0.08	0.0	+0.006	-0.02	-0.1	+0.08	-0.02	-0.6	+1.1	+0.5	+0.5	-0.8		
4	-0.2	+0.04	-0.1	-0.5	+0.004	0.00	-0.07	-0.2	-0.06	-0.08	+0.7	+1.7	-0.01	-0.2		
5	-0.2	-0.2	-0.07	-0.7	+0.005	+0.008	-0.04	-0.08	-0.08	-0.4	+0.5	+1.1	+0.07	+0.1		
6							-0.04	-0.05	-0.05	-0.3	+0.3	+0.7	+0.4	+0.1		
7							+0.09	+0.04	-0.3	-0.3	-0.01	+0.4	-0.4	-1.3		
8							-0.02	+0.06	+0.01	-0.2	+0.2	+0.03	-0.3	-1.1		
σ _D	0.2	0.7	0.3	0.7	0.01	0.01	0.1	0.2	0.1	0.4	0.6	0.9	0.8	1.0	0.6	0.8
$\bar{X}$	-0.07	+0.3	+0.01	+0.09	+0.004	-0.008	-0.003	+0.01	-0.09	-0.1	+0.4	+0.04	-0.5	-0.9	-0.2	-0.2
σ _X	0.04	0.02	0.06	0.1	0.002	0.002	0.02	0.03	0.02	0.08	0.01	0.2	0.2	0.2	0.1	0.2

*Value not used in average.

Further examination of the columns for uranium isotopes of Table IV brings up the question as to what can be considered random variations in the 3 sets of measurements and whether or not any biases between data sets are present. The spread in the differences, σ_D^* , between the normal plant measurements and NBS or II measurements and the mean difference or bias, $\bar{X}$, with reference to plant data are summarized at the bottom of table IV. The standard deviation of the mean, $\bar{\sigma}_X^*$, is also shown. The bias is considered real when $\bar{\sigma}_X$ is small (at least by a factor of 2) compared to the value of the bias. The different mass spectrometer machines have different measurement characteristics and it is important that a safeguards program have available a central machine or other means which can be used for measurement of duplicate samples and serve to control bias between machines.

The NBS and II plutonium isotopic ratio data are compared in Figure 12a, 12b and 12c where $^{242}\text{Pu}/^{241}\text{Pu}$ data are shown as a function of $^{240}\text{Pu}/^{239}\text{Pu}$ data. Columns of Table IV under the headings of plutonium isotopes show the percent differences where weight percents in equation 7 have been compared. The figures are in the same order with respect to NBS data, II data and normal plant data as indicate for the uranium isotopes. All ^{241}Pu data, both weight percents and atom ratios, have been decay corrected to date of fuel discharge from the reactor. Significant differences between data points are evident for samples 1, 2, 20 and 24. However, the burnup paths being traced by NBS data and II data appear to be nearly identical to the burnup paths described by normal plant data for 4.1 and 4.94 samples.

*

$$\sigma_D = \sqrt{\frac{1}{n-1} \sum (X_i - \bar{X})^2}$$

$$\bar{\sigma}_X = \sigma_D / \sqrt{n}$$

X_i = difference between measurements
for one sample

$$\bar{X} = \sum \frac{X_i}{n}$$

Figure 12a,b,c Plutonium Isotopic Composition Data from NBS Measurements (a),
 II Measurements (b), and Normal Plant Measurements (c).
 Product Data are Shown both for Uncorrected ^{241}Pu and for Corrected ^{241}Pu .

Samples 1, 20 and 24 were identified earlier by data evaluation methods as having inconsistent ^{241}Pu and/or ^{242}Pu data (Figure 7). Table IV supports this information for samples 20 and 24. The ^{242}Pu for sample 24 by normal plant measurement is higher by approximately 5% than either NBS or II measurement. The ^{241}Pu is approximately 2% higher and ^{242}Pu is approximately 1% lower for sample 20. However, there is a 10% difference in ^{242}Pu measurements for sample 1 and ^{241}Pu measurements for sample 1 differ only 1% making the sample 1 data point appear even more inconsistent for NBS and II measurements. Also, the slope values where correspond to column E, Table III, are not consistent, 0.00917 for NBS data and 0.00957 for II data. Therefore, it is concluded that all three ^{242}Pu measurements are inconsistent for sample 1 or additional information is needed to explain which measurements might be consistent.

In the case of sample 2, the data point lies in a position to indicate a separate burnup path for 2.9 fuel in Figure 12a and 12b. Large differences are shown in Table IV for the ^{242}Pu value of the sample and with the resultant change in ^{242}Pu , the slope values corresponding to column E, Table III, 0.00982 for NBS data and 0.00995 for II data, appear consistent. Therefore, the NBS and II data are considered to be correct.

The remaining differences less than 2% that appear in the ^{241}Pu and ^{242}Pu columns of Table IV for either NBS data or II data are accepted as being within the bounds of consistency unless the combined differences, where the differences are added by

$$^{241}\text{Pu} \% \text{ difference} - ^{242}\text{Pu} \% \text{ difference}, \quad (8)$$

are greater than $\pm 3\%$. The plotted points do not show significant change in Figure 12a and 12b when the percent differences are in the range of 2 to 4%.

The spread in differences and biases are also listed for the plutonium isotopes in Table IV. The ^{241}Pu bias shown for NBS data and II data is not entirely due to measurement bias since the decay correction can introduce a bias due to the half life of ^{241}Pu which appears in the decay correction.

Two additional sets of uranium and plutonium concentration measurements are also available, see Table I. One set was measured by what is referred to as the mixed spike isotopic dilution technique by NFS at the plant laboratory for all input batch samples. The other set was measured by II and measurements methods included using the mixed spike techniques for input batch samples and other methods as noted in Table I to measure product samples (the product samples are not compared). The 3 sets of input batch measurements for uranium and plutonium concentrations are compared in Figures 13a, 13b and 13c by showing the Pu/U ratio as a function of ^{235}U depletion. Normal plant ^{235}U measurements are used to form ^{235}U depletion for Figures 13a and 13c where the mixed spike measurement data are presented in Figure 13a and normal plant measurement data are shown in Figure 13c (same as Figure 10). Figure 13b presents the concentration measurements made by II where ^{235}U data measured by II are used to form ^{235}U depletion.

The burnup paths as described by data points from each set are nearly identical. The largest percent differences of any Pu/U ratio as compared to normal plant data and as listed in Table IV occur for sample number 6, NFS mixed spike data (+2.6%) and for sample number 1, II data (+2.7%). Both of these data points appear inconsistent in Figure 13a and Figure 13b, respectively. (Sample 13, Figure 13b, appears even more inconsistent than normal plant data, but this is due to ^{235}U instead of Pu/U data). All other

Figure 13a,b,c Pu/U Ratio Data from NFS Measurements (a), II Measurements (b), and Normal Plant Measurements (c), as a Function of ²³⁵U Depletion.

Pu/U value differences are within $\pm 1.6\%$ with the average difference being in the range of 0.5 to 0.6 percent. The additional information gained by re-measurement generally supports the conclusion based on the consistency evaluation of Pu/U values from normal plant data. For example, the Pu/U value of sample 3 was indicated to be 2% low and is seen to be higher for the NFS mixed spike and II data and thus appears more consistent. The parameters that involve the Pu/U ratio in Table III columns D, E and F, are also more consistent (not shown) for sample 3. Sample 18 was also indicated to be 2% low, but the additional data measurements agree with the normal plant value. It is felt that even more remeasurements will only give the present value. Thus rather than delete samples 13, 3 and 18 from the average of the Pu/U ratio over ^{235}U depletion shown in column F of Table III, they are retained because remeasurements indicate that agreement may be only to within $\pm 2\%$.

C ^{137}Cs DATA MEASUREMENTS

Measurement of the concentration of a fission product is a normal procedure in a burnup experiment, but is not done in the course of obtaining accountability data at a reprocessing plant. The fact that fission product data can be obtained was demonstrated by measuring ^{137}Cs at NFS in the plant laboratory. Since the procedures to measure ^{137}Cs are not routine, it was not expected that measurement accuracy would be as good as with normal plant measurements. Never-the-less, measurement accuracy was reasonable as shown by the consistency of ^{137}Cs data in Figure 14. The ^{137}Cs was measured in disintegrations per minute per milliliter and are shown converted to disintegrations per min per gram of measured uranium (d/m/gm U.). They are plotted as a function of $^{235}\text{U}/^{238}\text{U}$ atom ratio data. The ^{137}Cs data also describe burnup paths which are indicated by the lines drawn by inspection. The consistency of the data is further illustrated in column G of Table III where the values of ^{137}Cs (d/m) per gram of measured plutonium are shown. This parameter increases with increasing exposure as evidenced by the values for the 4.1 fuel. Based on the information presented, it appears reasonable to assign a one standard deviation of $\pm 2\%$ to ^{137}Cs measurements.

The ^{137}Cs data are of particular use in deriving cross section ratios from the accountability data as discussed in Section IX. The average exposure of each input batch was also determined from ^{137}Cs data and is presented in the following section.

Figure 14. Ratio of ^{137}Cs (Disintegration Per Minute) to Grams of Uranium (GMU) as a Function of $^{235}\text{U}/^{238}\text{U}$ Data.

V. ESTIMATED VERSUS MEASURED DATA

The data evaluation of Pu/U values has indicated that the uranium and plutonium concentration measurements by normal plant methods were consistent. However, Pu/U values are relative and do not specify whether uranium is high or plutonium is low when the Pu/U ratio appears low. It is useful to measure the concentration of a fission product to provide information concerning the consistency of uranium and/or plutonium concentrations in addition to the consistency of their ratio. The procedures to extract this information will not be presented, however, in view of the fact that fabricator values of initial uranium content of each fuel assembly and estimated values of the post irradiation uranium and plutonium content of each fuel assembly are available. These data, as shown in Table A-I, are used to provide information concerning the absolute values of uranium and plutonium content of each dissolver batch plus uranium and plutonium totals from cores V and VI. (Also, data evaluation methods require greater accuracy, $\pm 1\%$, for fission product measurements than were realized with the ^{137}Cs measurements).

As a first estimate of the total uranium content of cores V and VI, one could assume that the total amount fabricated, U-fab, was available for reprocessing. The next estimate of total uranium would be the fabricated amounts corrected for burnup, U-est, as calculated by a burnup code. In column A, Table V, the correction for burnup is shown to be approximately 4% of the fabricated amounts at the highest exposure and 1.6% at the lowest exposure. The burnup correction for the total uranium from cores V and VI is 3.2%. Column B compares the fabricated amounts to the measured amounts, U-meas, for each input batch by showing the percent difference between the

TABLE V

COMPARISON OF TOTAL URANIUM AS FABRICATED AND AS ESTIMATED
AND TOTAL PLUTONIUM AS ESTIMATED TO TOTAL URANIUM AND PLUTONIUM AS MEASURED

	A.	B.	C.	D.	E.	F.	G.	H.	I.
	<u>U-fab.</u> <u>U-est</u>	<u>U-fab</u> <u>U-meas.</u>	<u>U-est</u> <u>U-meas</u>	<u>Pu</u> <u>U</u>	<u>Pu From</u> <u>U-fab</u>	<u>Pu From</u> <u>U-est</u>	<u>Pu(U-fab)</u> <u>Pu-meas</u>	<u>Pu(U-est)</u> <u>Pu-meas.</u>	<u>Pu-est.</u> <u>Pu-meas.</u>
					kgs	kgs			
13	+3.96	+3.92	-0.04	0.01098	9.011	8.668	+3.91	-0.05	-3.54
21	+3.98	+2.65	-1.29	0.01053	8.642	8.311	+2.65	-1.28	-0.25
15	+3.90	+2.72	-1.14	0.01046	8.568	8.246	+2.72	-1.14	-0.80
5	+3.88	+4.24	+0.35	0.01044	8.568	8.248	+4.23	+0.34	+0.29
11	+3.57	+4.04	+0.45	0.00984	8.068	7.790	+4.08	+0.49	+0.53
7	+3.58	+3.80	+0.22	0.00983	8.060	7.782	+3.71	+0.26	+0.54
19	+3.37	+3.16	-0.20	0.00975	8.009	7.749	+3.21	-0.23	-3.49
9	+3.42	+3.32	-0.10	0.00970	7.953	7.690	+3.33	-0.09	-1.71
17	+3.46	+3.47	+0.01	0.00968	7.937	7.672	+3.45	0.00	-0.66
24	+3.14	+2.16	-0.94	0.00909	4.978	4.827	+2.11	-0.98	-2.69
3	+2.92	+2.96	+0.04	0.00859	7.043	6.844	+2.94	+0.03	-1.42
16	+3.99	+3.76	-0.21	0.01112	12.154	11.700	+3.77	-0.10	-1.50
18	+3.38	+4.60	+1.18	0.00979	10.709	10.359	+4.56	+1.14	+0.10
22	+3.14	+2.90	-0.23	0.00886	4.845	4.698	+2.87	-0.25	+3.31
12	+3.04	+3.07	+0.02	0.00874	9.548	9.266	+3.10	+0.05	+2.60
4	+3.04	+1.22	-1.76	0.00864	9.457	9.178	+1.27	-1.71	+1.76
8	+2.79	+3.74	+0.93	0.00798	8.733	8.496	+3.72	+0.90	+4.16
10	+2.55	+2.48	-0.07	0.00793	8.657	8.442	+2.47	-0.07	-1.47
14	+2.74	+2.49	-0.24	0.00793	8.657	8.426	+2.50	-0.24	+3.84
6	+2.54	+3.55	+0.98	0.00776	8.489	8.278	+3.54	+0.96	+1.52
20	+2.25	+4.39	+2.10	0.00710	7.764	7.593	+4.34	+2.04	+1.76
1	+1.62	+2.55	+0.91	0.00497	2.718	2.675	+2.57	+0.94	+4.23
2	+2.41	+8.31	+5.76	0.00596	3.020	2.949	+8.36	+5.81	+7.50
				CAMPAIGN TOTALS					
	+3.16	+3.37	+0.21	0.00900	181.589	175.887	+3.36	+0.11	+0.31

$$\text{Percent Difference} = \left[\left(\frac{\text{U-fab, U-est, Pu(U-fab), etc.}}{\text{U-meas or Pu-meas}} \right) - 1.0 \right] 100$$

fabricated and measured uranium determined according to the equation shown beneath Table V. These difference values are seen to range from 1.2% to 8.3%, and the difference between the total fabricated uranium and measured uranium is 3.4%. The 8.3% difference occurs for batch 2, the 2.9 zircaloy clad fuel, and it is not known what the exact reason for the discrepancy is. One possible reason for such a large discrepancy is the possibility that 30 or more rods could have been removed from the 2.9 fuel assemblies for destructive examination before the assemblies were sent to NFS. Column C compares U-est to U-meas for each batch. Except for batch 2, the largest difference is 2.1%, and the difference between U-est and U-meas for the total uranium for cores V and VI is 0.2%. Thus, there are two levels at which the uranium content of irradiated fuel assemblies can be considered, (a) the fabricated amount which will be approximately 3% high for slightly enriched fuels and (b) the fabricated amount corrected for burnup which is seen to be in good agreement with the amounts measured.

The total uranium estimated to have been sent to NFS was 19571.0 kg. The total uranium as measured by NFS from batch samples taken from the accountability tank was 19530.4 kg. Thus there was estimated to be 40.6 kg (0.21%) more uranium sent to the reprocessing plant than the reprocessor measured as received. Two thirds of this difference is accounted for by the 2.9 fuel assemblies dissolved as batch 2. Excluding this batch, the estimated and measured total uranium agree very closely (to within 0.07%). The recovered uranium was measured to be 19251 kg or a loss of 279.4 kg (1.43%) across the reprocessing plant. We now ask, is this loss compatible with uncertainties in the measurement process?

It is possible to estimate the uncertainty in the measurement process if the "true" amount of uranium that went into each batch is known. The "true" amount is not known exactly but it can be computed quite accurately. Since about 3% of the uranium is lost during the irradiation of the fuel and this can be calculated to better than one part in ten, the total uranium in a given batch should be estimated to better than 0.3% when the initial amount of unirradiated uranium is known. Column C of Table V shows the percent difference between the uranium estimated by calculations and the uranium measured for each batch. In many cases this difference is considerably larger than the 0.3% uncertainty of the calculations and therefore must be assumed to be the result of uncertainties in the measurement process.

If we assume that all (instead of most) of the difference observed in column C is due to uncertainties in the measured data, an estimate of the measurement uncertainty can be made using standard techniques. Excluding the sample from batch 2 and assuming that all of the remaining batches are of about the same size, the average difference of the $n = 22$ measurements is

$$\bar{C} = \frac{1}{n} \sum C_i = 0.044\%. \quad (9)$$

The measurement uncertainty for a single sample is

$$\sigma = \sqrt{\frac{\sum C_i^2 - n (\bar{C})^2}{n - 1}} = 0.87\% \quad (10)$$

and the measurement uncertainty in the average difference, $\bar{C}$, is

$$\sigma_{\bar{C}} = \sqrt{\frac{\sigma^2}{n}} = 0.186\% \quad (11)$$

Even though the measurement uncertainty associated with a single sample taken from the accountability tank is nearly one percent (actually 0.87%), the measurement uncertainty of the total uranium from all batches is only 0.186% (i.e., the accountability measurements total 19530.4 ± 36.3 kg).

If we assume that the uranium product samples have the same single sample uncertainty as do those from the accountability tank, the measurement uncertainty for the total uranium in the 5 product batches is

$$\sqrt{0.186^2/5} = 0.39\% \text{ (i.e., the product batch measurements total } 19251 \pm 75 \text{ kg.)}$$

The probability that the accountability and product total uranium measurements are statistical variations from the same number is less than one part in a thousand. Therefore, a small amount (279.4 ± 83.3 kg or $1.43 \pm 0.43\%$) of the uranium was not recovered.

The plutonium content can also be determined at levels (a) and (b), pg 62, using the measured Pu/U values, column D, Table V, which have been shown to be consistent. Multiplying the Pu/U ratio by the fabricated amounts of uranium, the values of plutonium content, Pu(U-fab) in column E are obtained and illustrates level (a). Since the fabricated amounts were 3% high, it is expected that the plutonium amounts of column E should be high also, and they are as shown in column G. The (b) level at which plutonium content can be determined is illustrated by the values shown in column F where the Pu/U values have been multiplied by U-est to obtain Pu(U-est). These values are shown to be in good agreement with the measured plutonium amounts as shown by the percent differences of column H, except for batch 2. The percent difference for the total plutonium of cores V and VI is 0.2%. Thus, the fabricated uranium content and the burnup corrected uranium content are very useful to fix the absolute amounts of uranium and plutonium measured.

A third level exists at which to determine and compare plutonium content for each batch and that is to use the estimated plutonium, Pu-est, of Table A-I. These values are calculated using burnup codes and are not the same as the values mentioned for levels (a) and (b) because they do not involve the measured Pu/U ratio. The percent difference from measured plutonium content for each batch is shown in column I of Table V. It is noted that although larger percent differences exists on a batch by batch basis, the total estimated plutonium agrees to within 0.3% of the measured plutonium for cores V and VI. This result appears to be fortuitous in light of the information to be presented later concerning ^{235}U as estimated.

The measurement uncertainty associated with total plutonium can not be estimated in the way that the uranium was done. This is because we do not possess an accurate prediction of the plutonium generated during the core irradiation. The difference between the plutonium estimated by calculation and plutonium measured as shown in column I is generally larger than the uranium difference shown in column C. Since the methods used to measure plutonium content are the same as those used to measure uranium, one would expect the measurement uncertainty for plutonium to be about the same as for uranium. If this is so, The larger differences associated with column I as opposed to column C must be attributed to uncertainties associated with the calculated plutonium estimates. The plutonium dissolved was measured to be 175.69 kg. whereas the amount recovered was 171.48 kg. making a difference of 4.21 kg or 2.40%. Statistical arguments associated with the measurement process would not seem to account for this loss.

As mentioned previously, the measured uranium in batch 2 is much lower than that estimated by calculation. Column I shows that this is also true for plutonium. The probability that this is due to a statistical fluctuation in the measurement process is less than one part in 10^8 . The cause for this discrepancy is unknown but it could be explained if some of the rods were removed before it was sent to NFS for reprocessing. Something is definitely amiss for this batch.

Other estimated quantities are available from Table A-I for comparison, the ^{235}U for example. The total uranium ($^{234}\text{U} + ^{235}\text{U} + ^{236}\text{U} + ^{238}\text{U}$) agreement for each batch was seen to be good but the agreement between estimated ^{235}U and measured ^{235}U is not as good. The estimated and measured ^{235}U for each batch are shown in Table VI along with the percent difference between them. For only 2 batches, 18 and 20, was the measured ^{235}U less than the estimated ^{235}U . The average difference, excluding batch 2, was 3.5% and a bias of -3.3% exists between the estimated and measured values. This indicates that, although the total uranium was estimated correctly, the isotopic composition of the uranium was not done as well. It is probable that the estimated plutonium isotopic composition (not available) also reflects the deficiencies that uranium isotopic compositions do.

The ^{235}U values are an indication of the heat produced, through fission of ^{235}U , and the measured ^{235}U indicates that not as much heat was produced as was estimated. The ^{137}Cs data confirm this observation and the ^{137}Cs data converted to MWd/MM* via appropriate conversion constants are also shown in Table VI. The spread in the difference between estimated

* Megawatt Days/Metric Ton Metal

TABLE VI

COMPARISON OF ESTIMATED ^{235}U AND ESTIMATED EXPOSURE TO MEASURED ^{235}U AND ^{137}Cs DATA

	Uranium - ^{235}U			Exposure		% Dif.
	Estimated	Measured	% Dif.	Est.	Meas.	
13	20.795	22.029	-5.60	25708	23232	+10.7
21	20.706	22.547	-8.17	26521	21968	+20.7
15	20.956	22.586	-7.22	25270	22498	+12.3
5	21.110	22.340	-5.51	25127	22810	+10.2
11	22.286	23.434	-4.90	23053	20792	+10.9
7	22.255	23.525	-5.40	23101	20769	+11.2
19	23.192	23.824	-2.65	21722	21169	+2.6
9	22.917	23.766	-3.56	22034	20662	+6.6
17	22.758	23.830	-4.50	22286	20659	+7.9
24	16.132	16.553	-2.54	20134	18964	+6.2
3	25.111	25.290	-0.71	18576	18322	+1.4
16	20.810	21.557	-3.47	25607	23738	+7.9
18	23.820	23.576	+1.03	21442	21558	-5.4
22	12.485	13.130	-4.91	19697	18484	+6.6
12	25.781	26.229	-1.71	19047	19133	-.4
4	25.867	27.180	-4.83	19047	18405	+3.5
8	26.996	27.836	-3.02	17049	16233	+5.0
10	27.953	28.077	-0.43	15845	16405	-3.4
14	26.908	28.245	-4.73	17044	16185	+5.3
6	28.042	28.258	-0.76	15845	15974	-.8
20	29.685	29.285	+1.37	13946	14395	-3.1
1	16.783	16.937	-0.91	9697	9483	+2.3
2	6.340	8.730	-27.38	12262	—	—
Totals	509.690	528.803	-3.61	BIAS		6.34 +5.59

TABLE VII
Independent Measurements Bounding a Commercial Chemical
Processing Plant

In many cases, plutonium isotopic compositions and other uranium isotopes are also measured and are available to the safeguards agency. However, for our first example, only the constituents listed in Table VII are pertinent. As shown, the chemical plant is tied at both the feed and product ends by independent measurements for total uranium. The uranium enrichment is also tied at both ends, although the tie is not as apparent. The uranium enrichment of chemical plant feed is predicted by the reactor operator (or his technical agent) from the initial enrichment and exposure. Because of the uncertainties involved in such predictions, that independent tie is good to only about three percent relative. The best verification of the chemical plant's input measurement of enrichment is provided by the uranium product receiver. This is shown by the dotted line extending from the product receiver to the chemical plant uranium product area and then through to the input measurement area. For normal operation, there is essentially no change in enrichment across the chemical plant. Consequently, verification of uranium product enrichment is equivalent to verification of the enrichment of uranium input. High waste losses of feed of a different than average enrichment or a similar holdup of material can cause some distortion. However, for the most part of the net effect of waste losses, holdup, and recycle on the across-the-plant enrichment is small.

From the measurement uncertainties listed in Table VII, it is seen that the major loophole in the independent measurement of the chemical plant feed is in the predicted values for plutonium from reactors. However, as shown, the independent boundary for total uranium and weight percent ^{235}U is good, e.g., equivalent to chemical plant measurements. Because of the reliability of independent statements of total uranium input and initial ^{235}U enrichment, methods to use that knowledge are to the advantage of a safeguards program. Of particular advantage would be a relationship between plutonium content, uranium content and ^{235}U depletion, ^{235}D . In this case, a cross-check has been developed from minimum accountability data.

For irradiations under identical conditions, there is a relationship between ^{235}D and plutonium content as indicated by Figure 10 and column F, Table III. However, that a single linear equation might represent that relationship for an entire core of the same enrichment was not evident until chemical plant data became available on gross quantities of spent power fuels. The relationships between plutonium formation and the uranium and plutonium isotopic composition of spent fuels from early Yankee reactor cores were previously reported in Reference (4). Since that time, the additional chemical plant data have become available on material from Yankee Cores V and VI, and all the data are considered.

The constancy of gross (one tonne U) sample data in terms of $\frac{\text{grams Pu/tonne U}}{^{235}\text{D}}$, is shown in Table VIII for input batch data from the chemical processing plant for Yankee cores I thru IV (same as column G, Table III for cores V and VI). As the data indicate, the relationship for a given original enrichment appears to have a constancy which

TABLE VIII

Relationship Between Pu Content and ^{235}U Depletion
for Gross Samples of Yankee Spent Fuel

Core I		NFS Data for Cores I-VI				
Input Batch	^{235}U Wt %	^{235}U Wt %	^{235}U D(1)	g Pu tonne U	g Pu/tonne U ^{235}U	% Dev.
16	3.406	2.373	1.0341	5989	5792	+ 2.30
3	3.406	2.404	0.9996	5858	5860	+ 3.50
4	3.406	2.431	0.9736	5558	5709	+ 0.83
6	3.406	2.502	0.9013	5132	5694	+ 0.57
1	3.406	2.511	0.8946	5221	5936	+ 3.07
5	3.406	2.518	0.8840	4919	5545	- 1.71
13	3.406	2.538	0.8666	4834	5579	- 1.46
15	3.406	2.541	0.8631	4910	5689	+ 0.48
2	3.406	2.542	0.8626	4810	5577	- 1.50
8	3.406	2.597	0.8101	4537	5600	- 1.09
7	3.406	2.620	0.7832	4457	5691	+ 0.51
12	3.406	2.737	0.6697	3794	5665	+ 0.05
9	3.406	2.754	0.6489	3576	5511	- 2.67
14	3.406	2.786	0.6364	3499	5498	- 2.90
10	3.406	2.908	0.4969	2858	5751	+ 1.57
11	3.406	2.950	0.4538	2530	5574	- 1.55
Ave.					5662	1.61%
Core II & III						$\sigma = 1.96\%$
31	3.406	2.108	1.2997	7395	5690	- 1.59
30	3.406	2.111	1.2949	7544	5826	+ 0.76
29	3.406	2.278	1.1258	6554	5822	+ 0.70
22	3.406	2.329	1.0731	6176	5755	- 0.46
17	3.406	2.362	1.0418	6044	5801	+ 0.33
26	3.406	2.388	1.0166	5839	5743	- 0.67
27	3.406	2.398	1.0088	5870	5819	+ 0.64
28	3.406	2.401	1.0047	5767	5740	- 0.72
25	3.406	2.402	1.0042	5791	5767	- 0.26
24	3.406	2.417	0.9907	5766	5820	+ 0.66
23	3.406	2.420	0.9885	5749	5816	+ 0.59
21	3.406	2.461	0.9470	5454	5759	- 0.39
19	3.406	2.502	0.9031	5242	5804	+ 0.38
32	3.406	2.618	0.7875	4554	5784	+ 0.04
Ave.					5782	0.585%

$$(1) \quad ^{235}\text{U}_r = \text{wt}\% \, ^{235}\text{U}_o - \text{wt}\% \, ^{235}\text{U}_f$$

where U_o = original enrichment at fabrication

U_f = final enrichment at chemical processing

$$\sigma = 0.70\%$$

Table VIII (Con't)

Core IV

Input Batch	$^{235}\text{U}_0$ Wt%	$^{235}\text{U}_f$ Wt%	^{235}D	$\frac{\text{g Pu}}{\text{tonne U}}$	$\frac{\text{g Pu/tonne U}}{^{235}\text{D}}$	% Dev.
43	4.10	2.261	1.839	9006	4897	- 8.46*
42	4.10	2.438	1.6620	8064	4852	- 9.24*
35	4.10	2.462	1.6380	7944	4850	- 9.28*
38	4.10	2.490	1.6100	7818	4856	- 9.16
39	4.10	2.606	1.4927	8132	5448	+ 1.90
36	4.10	2.637	1.4630	7850	5366	+ 0.37
41	4.10	2.643	1.4570	7756	5323	- 0.43
33	4.10	2.675	1.4266	7510	5265	- 1.52
40	4.10	2.706	1.3928	7516	5396	+ 0.94
34	4.10	2.721	1.3778	7268	5275	- 1.33
37	4.10	2.726	1.3766	7364	5349	+ 0.06
Ave.					5346	0.94%

Core V

16	4.10	2.046	2.0524	11119	5417	- 0.03
18	4.10	2.254	1.8454	9794	5307	- 2.06
22	4.10	2.471	1.6291	8863	5440	+ 0.39
12	4.10	2.475	1.6246	8738	5378	- 0.75
4	4.10	2.514	1.5900	8636	5431	+ 0.23
8	4.10	2.639	1.4588	7982	5471	+ 0.96
10	4.10	2.636	1.4641	7930	5416	- 0.05
14	4.10	2.652	1.4477	7930	5478	+ 1.09
6	4.10	2.675	1.4252	7762	5446	+ 0.05
20	4.10	2.796	1.3043	7104	5447	+ 0.52
1	4.10	3.175	0.9243	4968	5375	- 0.81
					5418	0.67

Core VI

13	4.94	2.789	2.1457	10981	5118	+ 2.49
21	4.94	2.820	2.1142	10530	4980	- 0.27
15	4.94	2.832	2.1022	10459	4975	- 0.38
5	4.94	2.837	2.0973	10440	4978	- 0.31
11	4.94	2.973	1.9612	9836	5016	+ 0.45
7	4.94	2.978	1.9564	9827	5023	+ 0.59
19	4.94	2.992	1.9438	9754	5018	+ 0.49
9	4.94	2.995	1.9403	9699	4999	+ 0.11
17	4.94	3.007	1.9289	9681	5019	+ 0.51
24	4.94	3.088	1.8477	9094	4922	- 1.44
3	4.94	3.176	1.7596	8592	4883	- 2.22
Ave.					4994	0.84

σ = 1.21%

* Not used in average since fuel assemblies of different initial enrichments were dissolved together.

approaches that of the experimental errors associated with the batch measurements. (The batch data are ordered according to decreasing exposure). Standard deviations of individual terms within a particular enrichment range from 0.70 relative percent to a maximum of about 2.0 relative percent. This compares to an experimental error for a single batch measurement of the term $\frac{\text{grams Pu/tonne U}}{^{235}\text{D}}$ of about 0.80-1.40 relative percent standard deviation. The random error associated with the factor relating plutonium to depletion consists of the random error associated with the plant Pu/U ratio measurement, the plant measurement of wt% ^{235}U , and any process variance associated with the stated original enrichment.

The individual core data are summarized in Table IX. The means for the term $\frac{\text{grams Pu/tonne U}}{^{235}\text{D}}$ have been adjusted for cores 1 and 2 to correspond to manufacturers' records for original enrichment. Two of the means for material of supposedly the same enrichment and cladding are statistically different. Although the difference is not large (~1.9%) it does indicate the presence of systematic error or shifts in systematic error. The difference may also be due to real differences in irradiation conditions and absorption characteristics. However, the data are sufficiently close to provide the basis of a useful cross-check for plutonium content.

The change in the factor relating measured ^{235}D to plutonium content listed in Table IX is shown graphically in Figure 15. The points indicated by a box, $\square$, are points obtained from input batches of a single initial enrichment. The points indicated by a dot, $\bullet$, are for an entire chemical plant campaign. These points (dots) are essentially mechanical mixtures of several enrichments. They also lie near the

TABLE IX

SUMMARY OF INDIVIDUAL CORE AND CAMPAIGN DATA
RELATING PLUTONIUM CONTENT TO ^{235}U DEPLETION

NFS Campaign	Input Batches	$^{235}\text{U}_\text{O}$ Wt % (1)	$^{235}\text{U}_\text{F}$ Wt %	^{235}D	grams Pu per tonne U	Average gram Pu/tonne U (2) ^{235}D	% σ (3) Individual	% σ Mean
Core I	16	3.4042	2.6014	0.8028	4552	5760	1.96	0.50
Core II & III	14	3.4058	2.3821	1.0237	5922	5785	0.70	0.20
Core IV	6	4.1010	2.6787	1.4223	7607	5348	1.26	0.52
Core V	11	4.1000	2.5515	1.5485	8388	5417	0.92	0.28
Core VI	11	4.9345	2.9494	1.9851	9923	4999	1.21	0.36
<u>Campaign Mixtures</u>								
Cores I-IV	41	3.5245	2.5090	1.0155	5729	5642	--	--
Cores V & VI	23	4.4315	2.7076	1.7239	8996	5218		

(1) Obtained from recently obtained complete manufacturers records.

(2) Weighted average in terms of total uranium in each batch.

(3) Relative percent standard deviation for term, $\frac{\text{grams Pu/tonne U}}{^{235}\text{D}}$.

FIGURE 15. Straight Line Function Relating Measured Plutonium Content, Uranium Content and ^{235}U Depletion to the Initial ^{235}U Enrichment Shown Graphically

line joining the points of single enrichments when the core (campaign) includes just two initial enrichments. The empirical least squares equation of the straight line is as follows:

$$\frac{\text{grams Pu/tonne U}}{^{235}\text{D}} = 7351 - 478.2 (^{235}\text{U}_0) \quad (12)$$

The equation does not include the point marked with a triangle for an initial enrichment of 2.9 weight percent ^{235}U . This point is only one single measurement. It is also a doubtful point since the cladding for the 2.9 material was zircaloy rather than the stainless steel cladding of all other fuel rods.

The use of the plutonium content versus depletion to check plutonium input measurements is shown in Table X. The technique may be used to check individual input batches or the entire input to a campaign. To check individual input batches the initial enrichment must be known from consideration of the dissolving order and batch sizes involved. The plutonium content of a core of mixed enrichment may be checked approximately by equation (12) using the average initial and average final enrichment of the core. There is, however, a small calculation bias introduced since the product of the means is generally not equal to the mean of the products. An exact calculation can be made by using all of the shipper and chemical plant input data. However, for future Yankee cores which are expected to be of a near uniform initial enrichment either equation (12) or a point calculation may be used with good reliability.

TABLE X

COMPARISON OF PLUTONIUM RESULTS BY EMPIRICAL
 ^{235}U DEPLETION METHOD TO MEASURED VALUES

Yankee NFS Campaign	$^{235}\text{U}_\text{O}$ Wt %	$^{235}\text{U}_\text{f}$ Wt %	^{235}D	$\frac{\text{grams Pu/tonne U}}{^{235}\text{D} (1)}$	Empirical Method grams Pu/tonne U	Measured grams Pu/tonne U	% Difference
Core I	3.4042	2.6014	0.8028	5723	4595	4552	+ 0.94
Cores II & III	3.4058	2.3821	1.0237	5723	5858	5922	- 1.08
Core IV	4.0278	2.5864	1.4414	5425	7819	7812	+ 0.09
Cores V&VI	4.4315	2.7076	1.7239	5232	9019	8996	+ 0.26
All Campaigns to date - Weighted Average (2)					6651	6650	+ 0.02

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(1) $\frac{\text{grams Pu/tonne}}{^{235}\text{D}}$ obtained by equation $\frac{\text{g Pu/TU}}{^{235}\text{D}} = 7351 - 478.2 (^{235}\text{U}_\text{O})$. For mixed cores see page 78.

(2) Weighted according to total uranium in each campaign.

Further application of the technique is illustrated in Table XI. In method #1, (the worst case) the safeguards group knows only two points on the enrichment curve and makes a simple linear extrapolation from cores I thru IV to cores V and VI. The procedure is then reversed and the safeguards team knows only the two enrichments of cores V and VI and attempts to cross-check cores I thru IV. Method #2, which is based on all the data, uses the empirical equation which actually fits the enrichment versus $\frac{\text{grams Pu/tonne U}}{235_D}$ curve. This, of course, is as good as the chemical plant data and is on the average in nearly perfect agreement.

The historical data needed for application of the method can be obtained from the following traditional accountability data:

1. Initial enrichment and total U from fabrication.
2. Chemical plant data for total Pu and total U or the weighted Pu/U ratio.
3. Chemical plant data for final enrichment.

The main safeguards value of such relationships as the one discussed is that they are independent of the chemical plant in that they can be formed and verified by data obtained from other parts of the fuel cycle. They, of course, depend initially on chemical plant data or data from special spent fuel measurement programs. As yet, there is not sufficient evidence to indicate that a general equation can be developed to cover different claddings, reactor types or even different reactors of the same general type. It is evident at this time, that a valuable safeguards data

TABLE XI
COMPARISON OF EMPIRICAL TO MEASURED VALUES FOR
PLUTONIUM CONTENT OF YANKEE FUEL

^{235}U Depletion Method

NFS Campaign	^{235}U wt% O	Empirical Methods		Measured by		% Difference #1 - #3	% Difference #2 - 3
		#1 Pu,g/TU	#2 Pu,g/TU	Chem. Plant #3 Pu,g/TU			
Core I	3.4042	4629	4595	4552		+ 1.7	+ 0.94
Cores II&III	3.4058	5901	5858	5922		- 0.35	- 1.08
Core IV	4.0278	7860	7819	7812		+ 0.61	+ 0.09
Cores V&VI	4.4315	8908	9019	8996		- 0.98	+ 0.26

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Pu,g/TU = grams Pu/tonne U

Method #1 is based on a simple linear extrapolation from two points on the enrichment curve. Cores I thru IV were estimated from data from cores V and VI and cores V and VI were estimated from data from cores I thru IV.

Method #2 is based on use of all chemical plant data available to date and uses the true enrichment versus the term,
 $\frac{\text{grams Pu/tonne U}}{^{235}\text{U Depletion}}$ as the linear equation below

$$\frac{\text{grams Pu/tonne U}}{^{235}\text{D}} = 7351 - 478.2 (^{235}\text{U}_\text{O})$$

where $^{235}\text{D} = \text{wt}\% ^{235}\text{U}_\text{O} - \text{wt}\% ^{235}\text{U}_\text{f}$

base can be established simply by accumulating the proper data. The value of the data can be greatly enhanced if a planned program is carried out at the chemical processing plant so that input accountability batches can be related to specific fuel elements or at least to specific groups of fuel elements.

Another example of an independent cross check utilizing a maximum of accountability data is to use equation 5. Solving the equation for plutonium, we have

$$\text{Pu} = 0.01 \left[\frac{^{241}\text{Pu}}{^{242}\text{Pu}} \frac{^{241}\text{Pu}}{^{240}\text{Pu}} \frac{^{235}\text{U}}{^{236}\text{U}} \frac{^{238}\text{U wt\%}}{^{239}\text{Pu wt\%}} \frac{239.05}{238.05} \right] \text{U (grams)} \quad (13)$$

The term is independent of initial enrichment, however, it is applied in much the same manner as discussed above using all the measured isotopes for uranium and plutonium (except ^{234}U and ^{238}Pu) either per batch or on a per core and campaign basis.

VII PARAMETERS THAT CHARACTERIZE THE YANKEE REACTOR

The information presented in Section V thru VII has been based on accountability data from Yankee cores V and VI. The evaluation methods of plotting and manipulating the data have identified useful graphs and parameters which were used to check the consistency and reliability of the data. When a future Yankee core is processed, it is only logical that the same plots and parameters would be used to check the data from that core as well. The plots and numerical values of the several data parameters are characteristic of the Yankee reactor and are a part of the reactor's "fingerprint."

The shape of the burnup paths shown in Figures 5 thru 9 are not as characteristic of a reactor as is the location that the burnup paths occupy on a given plot. When data from different reactors (i.e., reactors of different design such as a pressurized water reactor, PWR, or a boiling water reactor, BWR) are plotted, the data paths of the cores generally have similar shapes but are found to occupy different locations on the plot. Thus, differences in path location are more important than differences in path shape for establishing the fingerprint of a reactor. Differences in the numerical values of the same data parameter from different reactors are also important for establishing the fingerprint. Plots and data parameters that show the greatest differences between reactors are the most useful for distinguishing between data from different reactors and therefore provide the best method of fingerprinting reactors. Thus, plotting and manipulation of accountability data from different reactors will identify those plots and parameters which most readily characterize or fingerprint the reactors.

Differentiation between reactors of the same basic design (i.e., one PWR versus another PWR) may not be as straightforward. However, if differences do exist, they will most likely be reflected in the accountability data. Data evaluation methods are still useful to identify reprocessed fuel or to identify that a change in design has occurred. Therefore, plots and parameters as determined by data evaluation methods constitute a basic approach in defining a reactor's fingerprint. Another method is discussed in the following section.

VIII. LEAST SQUARES ANALYSIS OF YANKEE CORES V AND VI ACCOUNTABILITY DATA

Information is contained in the measured accountability data other than just the content of each isotope that is present. This extra information can be obtained by investigating the relationships that govern isotopic changes within the fuel. Plutonium isotopes are created from ^{238}U by a transmutation process governed by the general transmutation equation:

$$\frac{dN^i}{dt} = \hat{\sigma}_c^{i-1} N^{i-1} \phi - \hat{\sigma}_a^i N^i \phi - \lambda^i N^i; i = {}^{239}\text{Pu}, {}^{240}\text{Pu}, {}^{241}\text{Pu}, {}^{242}\text{Pu} \quad (14)$$

where N^i is the isotopic concentration of the i^{th} isotope, $\hat{\sigma}_c^{i-1}$ is the spectrum averaged capture cross section for the $i-1$ isotope, $\hat{\sigma}_a^i$ is the spectrum averaged absorption cross section for the i^{th} isotope, ϕ is the neutron flux, λ^i is the decay constant of the i^{th} isotope, and t is the time. These equations can be manipulated and integrated in such a way that the flux and time variables are eliminated. What remains is a relation between the isotopic concentrations (the N^i) and ratios of the spectrum averaged cross sections (the $\hat{\sigma}^i$). These relations are developed in References (10-12). Thus, the isotopic concentration measurements of the accountability data contain cross section information within them. By fitting the relations containing the N^i and the cross section ratios to the measured N^i , information about the cross section ratios can be obtained. The accepted method of fitting these relations to the data is by the method of least squares. A computer code called DBUFIT-I⁽¹²⁾ is used to perform the fitting process. This code adjusts the cross section ratios until the best least squares fit to the data is obtained. The resulting cross section ratios govern (through the transmutation relations) the isotopic changes that occur during the exposure of the fuel in the reactor. Thus, the cross

section ratios provide a "fingerprint" of the exposed fuel.

The least squares fitting technique described above is normally applied to data that is a little different than accountability data. In the past, the isotopic concentration data has been obtained from small "incremental" samples cut from selected positions within the reactor, e.g., see Reference 11. By selecting the incremental samples from locations with similar (neutron) spectral properties, the isotopic concentration data describe a single transmutation path and therefore, can be adequately described by a single set of cross section ratios, as is required by the least squares fitting process. The accountability data obtained during the reprocessing of Yankee cores V and VI are different in that they are not collected from a single spectral region within the core but are an average from all regions within the several fuel bundles dissolved in each batch. This raises the question of whether accountability data can be legitimately described by a single set of cross section ratios. Although in the strictest sense, data of this type should not be so analyzed, the fact that (a) the accountability data traces an averaged burnup path through the incremental data (see Figure 1) and (b) each accountability measurement represents the same kind of averaging process (namely, an average over entire fuel bundles), allows us to analyze these data with the least squares technique outlined above. However, the cross section ratios obtained from such an analysis of accountability data must be interpreted as averaged values which apply to entire fuel bundles rather than to a specific point within the reactor. This interpretation in no way hampers using the cross section ratios as a fingerprint of the core.

Before the least squares analysis can be applied, the measured data must be converted to isotopic concentrations (i.e. the N^i of the transmutation equations). The raw data measured at NFS are plotted in Figures 6-10. In preparation for analysis with the DBUFIT-I least squares analysis code, the data are converted to atoms per initial gram of uranium for the isotopes ^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu according to the equations given on pages 3 and 4 of Reference 11. The number of ^{239}Pu fissions and the exposure in thousands of megawatt days per metric ton of uranium (GWd/MTM) are also calculated for each measured sample by making use of the ^{137}Cs data after they were corrected for in core and out of core decay. The measurement uncertainties associated with these data are also required. Since uncertainty information was not provided with the set of measurements that were used for this analysis, uncertainties were assigned to the data on the basis of the scatter in the plotted data and past experience with somewhat similar sets of incremental data. In addition to the batch sample data measured at NFS, data for the zero exposure point (i.e. unirradiated fuel containing no plutonium) has been included with the data used in the least squares analysis.

The least squares fits to the data for cores V and VI are shown in Figures 16-21. The least squares analysis curves occur in sets of three. The central curve is the best least squares fit to the data and the outside curves represent the one standard deviation uncertainty assigned to the central curve. The cross section ratio results are presented in Table XII and Figures 22 and 23.

The ^{236}U versus ^{235}U analysis is the easiest to perform. It determines the ^{235}U capture to fission cross section ratio. The initial ^{236}U concentra-

Figure 16 ^{236}U vs ^{235}U
Core V Data and Least Squares Fit

Figure 17 ^{236}U vs ^{235}U
Core VI Data and Least Squares Fit

Figure 18 Pu Isotopes vs ^{235}U
Core V Data and Least Squares Fit

Figure 19 Pu Isotopes vs Exposure
Core V Data and Least Squares Fit

Figure 20 Pu Isotopes vs ^{235}U
Core VI Data and Least Squares Fit

Figure 21 Pu Isotopes vs Exposure
Core VI Data and Least Squares Fit

Figure 22 $\hat{\sigma}_a^{49}/\hat{\sigma}_a^{25}$ vs ^{239}Pu
 ^{239}Pu Absorption to ^{235}U Absorption Cross Section Ratio

Figure 23 $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{25}$ vs ^{240}Pu
 ^{240}Pu Absorption to ^{235}U Absorption Cross Section Ratio

tion was unknown for core V. It was estimated to be 1.02×10^{18} atoms/initial gram of uranium, but the least squares fit (see Figure 15) indicates that 0.79×10^{18} atoms/initial gram of uranium is a better value. The initial ^{236}U concentration was known for core VI and Figure 16 shows that the least squares fit passes nicely through this point.

The least squares fit to the ^{238}U - ^{239}Pu - ^{240}Pu - ^{241}Pu - ^{242}Pu data from core V is shown in Figure 18. The ^{238}U data, which are nearly constant with exposure, are not shown. The same data plotted versus exposure instead of ^{235}U are shown in Figure 19. Similar plots for the core VI data are shown in Figures 20 and 21. The resulting cross section ratios are shown in Table XII and Figures 22-23.

TABLE XII

<u>Cross Section Ratios</u>			
<u>Ratio</u>	<u>Symbol</u>	<u>Core V</u>	<u>Core VI</u>
Capture to Fission for ^{235}U	$\alpha^{25} = \sigma_c^{25}/\sigma_f^{25}$	$0.2555 \pm .0027$	$0.2640 \pm .0027$
Capture in ^{238}U to Absorption in ^{235}U	$\sigma_c^{28}/\sigma_a^{25}$	$0.02447 \pm .00023$	$0.02558 \pm .00026$
Capture to Fission for ^{239}Pu	$\alpha^{49} = \sigma_c^{49}/\sigma_f^{49}$	$0.648 \pm .020$	$0.871 \pm .034$
Absorption in ^{241}Pu to Absorption in ^{235}U	$\sigma_a^{41}/\sigma_a^{25}$	$3.455 \pm .034$	$3.010 \pm .033$

In general, the least squares lines adequately describe the data. However, when the cross section ratios of core V are compared with those of core VI, several discrepancies occur. For example, α^{49} for core VI should be much the same as that for core V, but it is considerably higher. There are several assumptions built into the present version of DBUFIT-I

which are not met by the Yankee data. This probably accounts for the discrepancies mentioned above. Even though the cross section ratio results from the present version of DBUFIT-I are of questionable value, there is reason to believe that an improved least squares code which will yield reliable values for the cross section ratios can be developed. The most pressing need is to relax the restrictions on the form of the cross section ratios. In the current version of DBUFIT, they are required to be constant as a function of exposure or to vary inversely with isotopic concentration with the resulting weakness that the cross section ratio becomes infinite at zero concentration (e.g., see Figures 22 and 23). Further development of the DBUFIT code designed to overcome this defect which strongly affects the cross section ratio results from ^{235}U enriched fuels such as Yankee cores V and VI should go a long way toward establishing a fingerprint of cross section ratios within the framework of an analytical model.

IX. SMUDGING OF THE FINGERPRINT

The isotopic measurements made upon the dissolved fuel and the many combinations that can be formed from these measurements make up a fingerprint of the spent fuel. If any of the design or operating variables of the reactor are changed, the fingerprint may be altered (i.e., smudged). The effects of changing some of the more important design and operating variables upon the fingerprint have been studied.

Calculations were performed to determine if the mode of nuclear plant operation noticeably affects the fingerprint. Two situations were studied. The first assumed that the plant was operated as a base load (i.e., full power) and the second assumed the plant operated as a load follower (i.e., variable power).

The results of a previous analytical survey⁽¹³⁾ were also used to determine the effects of fuel and core design changes upon the fingerprint. The effects of changing fuel enrichment, fuel rod size, spacing between fuel rods, and clad type on the fingerprint were examined.

A. The Effect of the Mode of Core Operation on the Fingerprint

The mode in which commercial nuclear plants operate depends on the particular utility system in which the plant resides. In some environments, the plant will operate as the base load whereas in others, it will operate as the load follower. Since the mode may smudge the fingerprint, a preliminary evaluation of the effect was made.

A few calculations have been performed for Yankee core V. These consisted of two evaluations of a given cycle for 5000 MWd/MTM. In the first evaluation, the power level was held constant and a given rod pattern was used. In the second evaluation, the same cycle was analyzed, only the

the power level was allowed to change in a square wave manner from full power to half power by using a different control rod pattern. The width of the square waves were about 1500 MWd/MM. All other parameters were held constant for the two cycles. The constraints of realistic values of the minimum critical heat flux and core criticality were used in order to simulate an actual operating situation.

The calculations were performed with a calculational scheme much like the ISOCHECK approach.⁽¹⁴⁾ Briefly this consisted of using the BATTELLE-REVISED-FLARE code⁽¹⁵⁾ for the core simulation with input data derived from previous ISOCHECK calculations performed on this core.

The control rods used in Yankee (as in most reactors) are, to the first order, absorbers of mainly thermal neutrons. Therefore, it is expected that the different control rod patterns would have the largest effect on those isotopes for which thermal absorptions are the largest part of the overall absorption rate. To compare the calculated results, the data for those fuel assemblies that were grouped in dissolution batches 10, 16, 18 and 22 (4.1 fuel) are presented. The individual assembly data were summed to represent the actual dissolution batches. The fuel assemblies of batches 10, 18, and 22 were discharged with core V while those of batch 16 were recycled to core VI and achieved higher exposure than considered here. Two additional calculated data groups of 4.9 fuel were also summarized but these data do not represent any of the dissolution batches. The 6 calculated data sets were then plotted by program ACTSUR in the same formats as the actual accountability data and these results are shown in Figures 24 and 25.

FIGURE 24. Uranium and Plutonium Isotopic Variations Due to Changes in Mode of Reactor Operation. Data Points Shown Represent Dissolution Batches 10, 16, 18 and 22 as Determined by Burnup Calculations. (a and b are Exaggerated)

FIGURE 25. Variations in Pu/U Ratio and ^{235}U Depletion Due to Changes in Mode of Reactor Operation. Data Points Shown Represent Dissolution Batches 10, 16, 18 and 22 as Determined by Burnup Calculations.

Figure 24 presents the calculated uranium and plutonium isotopic data in the same format as Figures 6 and 7 for both the base load and load following modes of operation (the calculations used zero ^{236}U at zero exposure). Changes in the isotopic concentrations due to mode operation can be illustrated by considering the batch representing the highest calculated exposure (batch 18 at a calculated exposure of 19000 MWd/MTM). Both the ^{235}U and the ^{236}U changed due to mode operation, the ^{235}U decreased an amount a (-2.3%) and ^{236}U increased an amount b (+3.1%). Both a and b are indicated in Figure 24. The calculated weight in grams of each isotope are compared by the percents listed. However, the change in operation mode did not change the burnup path of ^{235}U and ^{236}U , just the position at which the data point lies on the burnup path. The ^{235}U and ^{236}U changes have little affect on the total uranium. The major contribution to total uranium, ^{238}U , changed only -0.06% and thus the total uranium changed -0.1%.

The calculated plutonium isotopes for batch 18 also changed, +1.4% for ^{239}Pu , +4.9% for ^{240}Pu , +5.7% for ^{241}Pu and +9.9% for ^{242}Pu , such that the atom ratios $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{242}\text{Pu}/^{241}\text{Pu}$ changed +3.4% and +3.9%, respectively. The 3.4% and 3.9% changes are indicated in Figure 24 by c and d, respectively. The total plutonium changed by +2.5%. The data points representing the base load operation and load following operation, however, lie on the same burnup path which was drawn through the base load operation data points by inspection. This is not the case with all the calculated data points. In particular, the batch 16 data point representing load following operation does vary from the base load operation mode burnup path. The departure from the path is due to ^{241}Pu and

^{242}Pu , principally ^{242}Pu , which was determined by examining all the graphs of Table II. Thus, operation mode does affect the burnup paths of ^{241}Pu and ^{242}Pu .

The burnup path of the Pu/U ratio vs ^{235}U depletion does not change due to operation mode changes as shown by Figure 25. A change in ^{235}U depletion occurred, indicated by e, and a change in the Pu/U ratio, indicated by f. The change in the Pu/U ratio was principally due to plutonium but the changed data point still lies on the same burnup path.

Thus, though the burnup paths of the higher plutonium isotopes are power level dependent, the paths involving the uranium isotopes, Pu/U ratio and ^{239}Pu and ^{240}Pu isotopes are not. The departure from the burnup path which involved ^{241}Pu and ^{242}Pu was not drastic, that is greater than 3%, and it is felt that the net result of variations in reactor operation will be to introduce more scatter of ^{241}Pu and ^{242}Pu accountability data about their burnup paths.

B. The Effect of Fuel and Core Design Changes on the Fingerprint

The data used to study the effect of fuel and core design changes on the fingerprint were taken from the report to the International Atomic Energy Agency (IAEA) entitled "Theoretical Analyses of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors."⁽¹³⁾ The results reported in this document were calculated using the ALTHAEA code⁽¹⁶⁾ for reactors typical of the 1000 MWe class. They are the Diablo Canyon plant for a pressurized water reactor (PWR) and the Browns Ferry plant for a boiling water reactor (BWR). The results for both ^{235}U and plutonium enriched fuels are included in the survey. Only the PWR results for a ^{235}U enriched fuel are reproduced here since they are sufficient

Although predictive codes are not required by the safeguards plan, they may be useful to it in a number of ways. Various burnup codes can be used to predict the burnup of the fuel bundles as required in step 1 or to calculate the uranium remaining in a spent bundle as suggested in step 4. The value of the plan as a safeguards tool is that

- c It is an accurate method of determining fuel inventory.
- c It is already available and can be rapidly implemented.
- c It requires minimal funding in excess of what is already required.
- c It causes minimal interference in the operation of the processing plant.

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REFERENCES

1. L.C. Schmid, et. al., "Experimental and Theoretical Methods Used for Burnup Analyses at Battelle-Northwest," Proceedings of International Conference on Physics Problems in Thermal Reactor Design, June 27-29, 1967.
2. "Reprocessing of Dresden Reactor Fuel at Nuclear Fuel Services, Inc., West Valley Facility," (Compiled by Fuel Reprocessing Branch Savannah River Operations Office, U.S. AEC) SRO-123 (Oct. 14, 1968).
3. "Reprocessing of Yankee Reactor Fuel at Nuclear Fuel Services, Inc., West Valley Facility," (Compiled by Fuel Reprocessing Branch Savannah River Operations Office U.S. AEC) SRO-124, (Oct. 21, 1968).
4. R.J. Nodvik, et. al., "Evaluation of Fuel Accountability Methods Applied to Yankee Fuel," WCAP-3896-1, Westinghouse Electric Corporation (June, 1968).
5. D.E. Christensen and R.A. Schneider, "Consistency Evaluation of Accountability Data for a Chemical Processing Plant, BNWL-CC-2346 PEV. (Nov. 1969).
6. "Evaluation of Minor Isotope Safeguard Techniques (MIST) in Reactor Fuel Reprocessing - Field Test FT-62," (Prepared by Field Operations, Weapons Evaluation and Control Bureau, US Arms Controls and Disarmament Agency) (Jan. 27, 1969).
7. C.C. Peak, U.S. AEC, Private Communication (Aug. 6, 1969) (from a letter to Col. E. P. Gaines, ACDA/FO).
8. R.J. Nodvik, "Evaluation of Mass Spectrometric and Radiochemical Analysis of Yankee Core I Spent Fuel," WCAP-6068, Westinghouse Electric Corporation (March, 1966).
9. J.A. Corbett, Westinghouse Electric Corporation, Private Communication (Sept. 11, 1969) (from a letter to Col. E.P. Gaines, ACDA/FO).
10. R.P. Matsen, "A Technique for the Determination of Ratios of Effective Cross Sections from Reactor Fuel Burnup Data," Nucl. Sci. Eng., 32, p. 267, (1968).
11. R.P. Matsen "An Analysis of Yankee-Rowe Burnup Data," BNWL-1122 (July 1969).
12. R.P. Matsen, "DBUFIT-I A Least Squares Analysis Code for Nuclear Burnup Data," BNWL-1396. (May 1970).
13. "Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors," (Compiled by Fuel Cycle Analysis Section at Battelle-Northwest for the Division of Nuclear Power Reactors of the International Atomic Energy Agency) (Aug. 1968).

REFERENCES (Continued)

14. M.F. Valerino and Z.R. Rosytoczy, "Analytical and Experimental Methods of Determining Heavy-Isotope Content of Operating Fuel Elements," CEND-540. (1965).
15. N.E. Carter, W.L. Purcell, R.H. Holeman and W.C. Wolkenhauer, "Battelle Revised Flare (BRF-1)," BNWL-1150, Reactor Physics Quarterly Report (August, 1969).
16. W.W. Porath, D.E. Deonigi, and E.T. Merrill, "Some Novel Plutonium Fueling Methods for Thermal Reactors, BNWL-183, Part IV- "The Calibration of High Speed Burnup Code for Nuclear Fuel Surveys," (Nov., 1965).

APPENDIX A

APPENDIX ADATA TABLES

The processing of irradiated fuel assemblies from Yankee cores V and VI has resulted in numerous groups of data. Certain groups of data, Table A-I and Table A-II, were collected as a result of normal fuel accounting procedures while the remaining data, of which Table A-III and Table A-IV are part, were collected by the additional requirements of FT-62 and the Yankee Atomic Electric Company. The data tabulated in Tables A-I thru IV are the data considered in the body of the report and do not represent all the data collected during the experiment.

Accompanying the shipment of Yankee fuel to NFS was a statement of material to be processed and an attachment to that statement tabulated the data that are presented in Table A-I. In addition to these data (fuel assembly number, enrichment, etc), Table A-I shows the order of dissolution and dissolver batch number for the fuel assemblies. The ordering of the dissolution sequence was done prior to the processing of the fuel and to our knowledge this was the first time that ordered fuel assemblies had been processed at NFS. Batch totals both for uranium as charged in the reactor and for uranium and plutonium as estimated at discharge are shown as well as totals for the campaign. These data are referred to in the body of the report as the pre-processing data.

The results of data measurements done and reported by NFS are tabulated in Table A-II. These data include both the normal accountability data (under the heading of normal plant measurements) and data from additional measurements which included uranium and plutonium concentration measurements and ^{137}Cs measurements. The data shown are tentative and may be subject to

TABLE A-1
 TABULATION OF PRE-PROCESSING DATA OF YANKEE FUEL ASSEMBLIES
 FROM CORES V AND VI SHOWING THE ORDER OF DISSOLUTION AND DISSOLVER BATCH NUMBERS

Batch No.	Element No.	Date Charged	Weight Charged		Weight Percent 235U	Date Discharged	Estimated Exposure Wd/MTM	Total Exposure		Total Pu	
			Total U kg	235 U kg				Total U kg	235U kg	Total U kg	235U kg
1.	A-158	9/64	273.477	11.212	4.1	8/65	9,697	269.104	8.391	269.104	1.381
	A-162	9/64	273.501	11.213	4.1	8/65	9,697	269.128	8.392	269.128	1.381
			546.978	22.425				538.232	16.783	538.232	2.762
2.	A-263	11/65	251.243	7.292	2.9	10/66	12,262	245.271	3.109	245.271	1.498
	A-264	11/65	255.440	7.414	2.9	10/66	12,262	249.468	3.231	249.468	1.498
			506.683	14.706				494.739	6.340	494.739	2.996
3.	A-244	11/65	273.825	13.512	4.94	3/68	18,576	266.079	8.395	266.079	2.249
	B-255	11/65	273.053	13.474	4.94	3/68	18,576	265.308	8.358	265.308	2.248
	B-259	11/65	273.053	13.474	4.94	3/68	18,576	265.308	8.358	265.308	2.248
			819.931	40.460				796.695	25.111	796.695	6.745
4.	A-157	9/64	273.403	11.209	4.1	10/66	19,274	265.251	6.512	265.251	2.398
	A-160	9/64	273.463	11.212	4.1	10/66	18,821	265.481	6.405	265.481	2.353
	A-173	9/64	273.812	11.226	4.1	10/66	18,821	265.830	6.419	265.830	2.353
5.	A-187	9/64	273.861	11.228	4.1	10/66	19,274	265.709	6.531	265.709	2.398
			1094.539	44.875				1062.271	25.867	1062.271	9.502
	A-229	11/65	273.825	13.512	4.94	3/68	24,842	263.724	7.101	263.724	2.728
6.	A-240	11/65	273.825	13.512	4.94	3/68	24,863	263.716	7.097	263.716	2.730
	B-250	11/65	273.053	13.474	4.94	3/68	25,679	262.640	6.912	262.640	2.786
			820.703	40.498				790.080	21.110	790.080	8.244
6.	A-166	9/64	273.432	11.210	4.1	10/66	15,788	266.670	7.023	266.670	2.078
	A-174	9/64	273.601	11.218	4.1	10/66	15,902	266.789	7.001	266.789	2.084
	A-186	9/64	273.617	11.218	4.1	10/66	15,788	266.855	7.031	266.855	2.078
A-226		9/64	273.246	11.204	4.1	10/66	15,902	266.434	6.987	266.434	2.084
			1093.896	44.850				1066.748	28.042	1066.748	8.324

Table A-1 (Cont'd)

Batch No.	Element No.	Date Charged	Weight Charged		Weight Percent 235U	Date Discharged	Estimated Exposure MWD/MTM	Total Exposure		Total Pu	
			Total U kg	235 U kg				Total U kg	235U kg	Total Pu kg	Total Pu kg
7.	A-228	11/65	273.825	13.512	4.94	3/68	22,497	264.616	7.566	2.556	
	B-247	11/65	273.053	13.474	4.94	3/68	22,646	263.788	7.497	2.567	
	B-258	11/65	273.053	13.474	4.94	3/68	24,163	263.210	7.192	2.681	
8.			819.931	40.460				791.614	22.255	7.804	
	A-156	9/64	273.638	11.219	4.1	10/66	17,468	266.196	6.672	2.232	
	A-161	9/64	273.685	11.196	4.1	10/66	16,631	265.973	6.819	2.153	
	A-183	9/64	273.325	11.206	4.1	10/66	17,468	265.883	6.659	2.232	
	A-225	9/64	273.738	11.223	4.1	10/66	16,631	266.626	6.846	2.153	
9.			1094.386	44.844				1064.678	26.996	8.770	
	A-233	11/65	273.825	13.512	4.94	3/68	22,798	264.501	7.503	2.579	
	B-245	11/65	273.053	13.474	4.94	3/68	21,605	264.180	7.718	2.490	
	B-253	11/65	273.053	13.474	4.94	3/68	21,698	264.145	7.698	2.496	
10.			819.931	40.460				792.826	22.919	7.565	
	B-192	9/64	273.141	11.198	4.1	10/66	15,902	266.329	6.981	2.084	
	B-193	9/64	272.897	11.189	4.1	10/66	15,788	266.135	7.002	2.078	
	B-219	9/64	272.761	11.183	4.1	10/66	15,788	265.999	6.996	2.078	
	B-221	9/64	272.931	11.191	4.1	10/66	15,902	266.119	6.974	2.084	
11.			1091.730	44.761				1064.582	27.953	8.324	
	A-237	11/65	273.825	13.512	4.94	3/68	24,163	263.982	7.230	2.680	
	B-254	11/65	273.053	13.474	4.94	3/68	22,497	263.844	7.528	2.556	
	B-262	11/65	273.053	13.474	4.94	3/68	22,497	263.844	7.528	2.557	
12.			819.931	40.460				791.670	22.286	7.793	
	B-196	9/64	273.433	11.211	4.1	10/66	18,821	265.451	6.404	2.353	
	B-199	9/64	273.197	11.201	4.1	10/66	19,274	265.045	6.504	2.398	
	B-200	9/64	273.074	11.196	4.1	10/66	18,821	265.092	6.389	2.353	
	B-214	9/64	272.712	11.181	4.1	10/66	19,274	264.560	6.484	2.398	
			1092.416	44.789				1060.148	25.781	9.502	

TABLE A-II
RESULTS OF DATA MEASUREMENTS DONE BY NFS
ON INPUT BATCH SAMPLES AND PRODUCT BATCH SAMPLES

INPUT ACCOUNTABILITY DATA (24)										
NORMAL PLANT MEASUREMENTS						ADDITIONAL MEASUREMENTS				
Sample number	Enrichment*	U ISOTOPES*				U (g/g)	Total U (grams)	Mixed Spike U (g/g)	137Cs	
		234	235	236	238				dpm/ml x 10 ¹⁰	Days Cooled
1	4.1	0.0214	3.1755	0.2239	96.5623	245.48	533370.	245.51	1.537	1533
2	2.9	0.0197	1.8690	0.2188	97.8925	112.15	467802.	113.29	0.967	1123
3	4.94	0.0131	3.1750	0.4250	96.3869	179.90	796337.	179.59	2.284	588
4	4.1	0.0184	2.5136	0.3590	97.1090	222.64	1081296.	220.63	2.761	1123
5	4.94	0.0130	2.8373	0.4990	96.6508	182.50	787352.	183.64	2.906	588
6	4.1	0.0187	2.6748	0.3253	96.9811	236.78	1056390.	237.72	2.525	1123
7	4.94	0.0130	2.9782	0.4682	96.5407	178.72	789877.	179.83	2.576	588
8	4.1	0.0189	2.6388	0.3313	97.0109	237.37	1054889.	238.95	2.587	1123
9	4.94	0.0126	2.9943	0.4593	96.5333	188.23	793613.	188.64	2.696	588
10	4.1	0.0187	2.6359	0.3282	97.0171	233.66	1065294.	234.30	2.563	1123
11	4.94	0.0124	2.9734	0.4552	96.5490	167.82	788115.	168.91	2.418	588
12	4.1	0.0179	2.4754	0.3601	97.1466	237.57	1059897.	238.76	3.042	1123
13	4.94	0.0124	2.7880	0.5043	96.6945	178.08	789749.	180.07	2.873	588
14	4.1	0.0187	2.6523	0.3296	96.9994	236.86	1065114.	236.95	2.555	1123
15	4.94	0.0124	2.8324	0.4968	96.6584	190.08	797490.	190.09	2.966	588
16	4.1	0.0163	2.9467	0.4454	97.4916	225.25	1053376.	226.57	3.671	588
17	4.94	0.0118	3.0057	0.4593	96.5227	181.53	792471.	182.75	2.600	588
18	4.1	0.0175	2.2545	0.3993	97.3287	225.00	1045784.	225.46	3.231	1123
19	4.94	0.0122	2.9907	0.4596	96.5376	175.30	796311.	175.24	2.565	588
20	4.1	0.0201	2.7958	0.2999	96.8843	244.81	1047457.	245.35	2.353	1123
21	4.94	0.0124	2.8203	0.4946	96.6727	183.23	799544.	183.49	2.796	588
22	4.1	0.0180	2.4709	0.3589	97.1522	175.32	531435.	175.96	2.183	1123
24	4.94	0.0131	3.0868	0.4430	96.4571	164.15	536060.	162.99	2.152	588
25	-	0.0130	3.0595	0.4437	96.4840		1388.			
						Total	19530411			
URANIUM PRODUCT DATA (5)										
1	0.0165	2.7377	0.3619	96.8839	4237000.					
2	0.0168	2.8262	0.3908	96.7617	4113000.					
3	0.0170	2.6500	0.4120	96.913	4284000.					
4	0.0150	2.6790	0.4180	96.888	3838000.					
5	0.0150	2.6790	0.4180	96.888	2779000.					
				Total	19251000.					
					*wt %					

*wt %

TABLE A-II (Cont'd)

NORMAL PLANT MEASUREMENT									Additional Measurements	
Sample Number	Enrichment*	Pu Isotopes*					Pu (g/l)	Total Pu (grams)	Days Cooled	Mixed Spike Pu (g/l)
		237	238	240	241	242				
1	4.1	0.2890	85.0504	10.2940	4.0105	0.3561	1.222	2650.	1381	1.230
2	2.9	0.4132	77.0680	14.8107	6.6196	1.0886	0.668	2787.	959	0.668
3	4.94	0.7263	76.9673	13.1553	8.0899	1.0612	1.551	6842.	427	1.558
4	4.1	0.6701	75.6664	14.2846	8.1138	1.2651	1.928	9338.	965	1.889
5	4.94	1.0372	72.9158	14.6825	9.7586	1.6059	1.905	8220.	431	1.911
6	4.1	0.5770	77.9612	13.2569	7.2199	0.9850	1.838	8199.	967	1.894
7	4.94	0.9674	74.5809	14.0261	9.0534	1.3722	1.756	7762	433	1.762
8	4.1	0.6162	77.5050	13.4530	7.3824	1.0434	1.895	8420.	971	1.899
9	4.94	0.8561	75.1180	13.8339	8.8894	1.3024	1.827	7697.	437	1.823
10	4.1	0.5419	77.5738	13.4712	7.3680	1.0451	1.846	8448.	974	1.847
11	4.94	0.8865	74.7847	13.9596	9.0369	1.3323	1.650	7752.	442	1.653
12	4.1	0.6909	75.2167	14.5248	8.2264	1.3412	2.076	9261.	983	2.080
13	4.94	1.0769	72.5647	14.8030	9.8849	1.6705	1.954	8672.	449	1.956
14	4.1	0.5385	77.7337	13.4348	7.2783	1.0148	1.873	8446.	985	1.862
15	4.94	1.0239	72.9417	14.6952	9.7332	1.6060	1.986	8341.	451	1.992
16	4.1	1.2277	69.3089	16.3367	10.8526	2.2741	2.509	11712.	454	2.546
17	4.94	0.8486	75.0776	13.8645	8.9127	1.2966	1.758	7672.	456	1.761
18	4.1	0.9210	72.6915	15.4591	9.1778	1.7506	2.204	10242.	993	2.208
19	4.94	0.8592	75.0471	13.8537	8.9315	1.3086	1.709	7767.	461	1.704
20	4.1	0.6838	79.3513	12.5320	6.6510	0.7819	1.739	7441.	1000	1.741
21	4.94	1.0391	72.8453	14.7559	9.7289	1.6308	1.929	8419.	469	1.925
22	4.1	0.6940	75.1394	14.5352	8.2871	1.3442	1.555	4710.	1005	1.556
24	4.94	0.7886	75.9508	13.5383	8.4845	1.2378	1.493	4875.	471	1.496
26	-	0.7888	75.8790	13.619	8.5181	1.195	-	13.	-	-
							Total	175686		
PLUTONIUM PRODUCT DATA (8)										
1		0.685	76.804	13.656	7.737	1.118		18678		
2		0.705	76.209	13.775	8.121	1.190		21435		
3		0.728	76.257	13.734	8.103	1.177		26533		
4		0.775	75.216	14.193	8.490	1.326		24418		
5		0.949	73.273	14.795	9.360	1.624		24605		
6		0.931	73.267	14.902	9.273	1.626		19317		
7		0.769	75.802	13.862	8.301	1.266		19955		
8		0.838	74.664	14.279	8.825	1.394		19536		
							Total	171477		

*Wt%

change. However, at this point, there is little probability that any of the data will be changed. The total number of samples analyzed are shown enclosed in parenthesis for input samples and product samples. Sample 26 represents flush material for the campaign (batches 23 and 25 do not exist). The uranium and plutonium concentrations (gms/liter) were not reported and ^{137}Cs was not measured for this sample. The approximate cooling times since fuel discharge are included in Table A-II and were used to decay correct ^{241}Pu and ^{137}Cs to date of discharge.

The results of data measurements done and reported by NBS are tabulated in Table A-III. Duplicate samples of each input and product batch, samples c and d, were sent to NBS from NFS. Measurements were made on 8 of the c and d input samples and on 5 of the c and d uranium product samples. Only the c sample of the remaining input and plutonium product samples were analyzed. NBS did not measure the uranium and plutonium concentrations, only the uranium and plutonium isotopic compositions. The approximate cooling times to correct ^{241}Pu decay to date of discharge are also tabulated in Table A-III.

The results of data measurements done jointly by Teledyne Isotopes and Ledoux & Company for the Yankee Atomic Electric Company are tabulated in Table A-IV. The plutonium isotopic composition was measured twice for sample 20 and this is the reason for showing a sample 20a and a sample 20b.

Interlaboratory comparisons of standard samples between many laboratories or comparisons of a number of burnup samples between 2 laboratories have occurred in the past. In the case of processing of Yankee cores V and VI, 4 laboratories (including NBL, whose data are not reported here) have made

independent measurements on 24 input samples, 5 uranium product samples and 8 plutonium product samples. The number of independent measurements on the same material serves to emphasize the importance of independent measurements in a safeguards program and to point to the independent measurements as an unique feature of the FT-62 experiment.

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