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THE USE OF ISOTOPIC CORRELATIONS
IN VERIFICATION
(SAFEGUARDS APPLICATION)

CONTRACT NUMBER ACDA/WEC-216

PREPARED FOR
U.S. ARMS CONTROL AND DISARMAMENT AGENCY

PREPARED BY
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by

R. A. Schneider, K. B. Stewart, and D. E. Christensen

FEBRUARY 10, 1972

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THE USE OF ISOTOPIC CORRELATIONS IN VERIFICATION

I. INTRODUCTION

Previous work⁽¹⁻¹¹⁾ has shown the potential value of heavy element isotopic correlations to international safeguards. Potential applications are centered on that part of the fuel cycle which extends from the output of fuel fabrication through the input to reprocessing. This paper addresses two specific applications. These are:

- 1) Verification of the plutonium to uranium ratio of spent low enriched uranium fuels.
- 2) Confirmation of information which is stated about the spent fuels prior to dissolution at reprocessing, including tests for substitution.

Other applications include the use of isotopic correlations in evaluating the quality of the analytical measurements that are made on reprocessing plant dissolver solutions. The correlations can also serve as a diagnostic tool for deducing additional information about fuel history. These other applications are discussed in previous publications⁽¹⁻¹¹⁾ and other papers⁽¹³⁾ presented at this Panel.

The purpose of this report is to describe the technical bases and conditions for the valid application of heavy element isotopic correlations in international safeguards. Further and equally important aims are to describe the (1) information requirements, (2) inspection and verification activities, and (3) evaluation of results.

As an aid to the reader, the fundamental points are covered in the first 16 pages. This is followed by a detailed discussion of technical applications which includes specific examples, information needs, suggested procedures, and methods of evaluation.

II. TECHNICAL BASES AND CONDITIONS OF APPLICATION

In discussing the safeguards verification of materials accounting data, several terms require introduction. These include:

- Technical Reliability - The precision and accuracy or the exactness of a given method to measure the quantities or compositions of materials or to calculate such properties from other measured data and physical constants.
- Validity of Verification - The ability of a given verification procedure to truly affirm the integrity and quality of materials accounting data.
- Reliance - The extent to which a safeguards authority relies upon the results of a given verification procedure or activity in making tests of significance.

A. Verification of Plutonium Input to Reprocessing

The major safeguards impact of isotopic correlations stems from their potential use in verifying the Pu/U ratio of spent low enriched uranium fuels. In this application, isotopic correlations are used in conjunction with the Pu/U ratio method⁽⁷⁾ in verifying the total plutonium input to the reprocessing plant. Here the primary verification of total plutonium is by the Pu/U ratio method. Plutonium input to the reprocessing plant is computed by applying individual or the weighted Pu/U ratio(s) to the shipper's value (fabrication value corrected for burnup) for total uranium. Conventionally, the Pu/U ratio is obtained by the direct measurement of the Pu/U ratio of input dissolver solutions. The isotopic correlations are used to verify or substantiate individual or the weighted

Pu/U ratio of an input campaign. Alternatively, isotopic correlations may be used to estimate the Pu/U ratio from historical relationships and the measured uranium and plutonium isotopic composition of the spent fuel. The isotopic correlations of interest in this application are those relating the plutonium concentration (Pu/U ratio) of a particular spent fuel to the uranium or plutonium isotopic composition, e.g.,

$$\text{Pu/U} = k(^{235}\text{D})$$

$$\text{where D} = \text{wt. \% } ^{235}\text{U initial} - \text{wt. \% } ^{235}\text{U final}$$

$$\text{or } \text{Pu/U} = m(^{240}\text{Pu} / ^{239}\text{Pu}) + b$$

where k, m, and b are constants.

Valid application of the Pu/U ratio method in verification requires 1) that the uranium content of fabricated fuel be accurately known and verified, 2) that the burnup of total uranium can be fairly accurately determined and verified, and 3) that the Pu/U ratio itself be verified. Validity of the method also requires that effective containment and surveillance procedures be applied after fuel fabrication primarily for detecting any additions of uranium. In addition, the reprocessing plant head-end process must allow use of the ratio method.

The technical reliability (as opposed to validity of verification) for the use of the Pu/U ratio method is well established for low enriched uranium fuels; i.e., the capability to measure the total U in fabricated UO_2 fuels, the capability to measure or calculate total U burnup, and the capability to measure the

Pu/U ratio of dissolver solution are well established. A similar reliability is expected for the Pu/U ratio method for the case of plutonium thermal recycle fuels which contain mostly uranium ($> 96\% \text{UO}_2$).

The ratio method under the conditions previously described may be used as a primary method of verification, independent of support from the isotopic correlations. The main benefit, however, which can arise from application of isotopic correlations lies in their use to substantiate, reinforce, and validate the Pu/U ratio. Depending on the sampling plan used to independently measure Pu/U ratios, the correlations can be used to reinforce direct independent measurements or they can be a necessary part of the verification process. For fractional sampling plans they can be necessary to the verification in protecting against gross atypical errors (or falsifications) occurring on a few of the input batches. In this case, the correlations minimize this maximum risk by verifying the collective ratio data of a series of input batches.

In a fractional sampling plan approach, the Agency would directly measure the Pu/U ratio and the uranium and plutonium isotopics on a few input batches from a reactor lot. From these measurements it would form correlations between Pu/U and the isotopes, e.g.,

$$\text{Pu/U} = K(^{235}\text{U})$$

It would then compare those relationships with similar ones formed from plant operators data on all the batches (assuming $Pu/U = K(^{235}D)$ to hold for all batches). If no differences are noted between the Agency and the operator, the problem of gross atypical errors (or falsification) reduces to errors in isotopic composition. That is (under conditions where the correlations hold) the correct relationships between Pu/U and isotopic composition could not have been reported without both the Pu/U ratio and the isotopic compositions being in error. To protect against such errors the Agency monitors the isotopic compositions across the plant, e.g., the weighted isotopic composition of input equals the weighted isotopic composition of product when computed over reasonable periods of time. The Agency could also use composite samples to provide collective verification of the Pu/U ratio and isotopic compositions.

The technical reliability of using isotopic correlations to verify the Pu/U ratio is dependent on many factors. These include the characteristics of the spent fuel, the scope, exactness, and accuracy of historical information, the reprocessing plant head-end process, and the batching sequence used for dissolving and measuring the spent fuel. By contrast, the extent of application in verification depends on the degree of reliance placed on the correlations for verification of the Pu/U ratio as opposed to the reliance placed on direct measurements by the Agency. Various degrees of reliance have been suggested.^(4,8,12) These range from a minimum level of

using the correlation as an "addition-to" to reinforce or substantiate, to a maximum level of reliance where the Pu/U ratio is estimated (or verified) solely from established relationships and measured isotopic data. Some workers⁽¹²⁾ recognize the robustness added by isotopic correlations; others⁽⁸⁾ recognize the practical ease of verifying ratios and isotopic compositions as opposed to verifying element concentrations.

The balance between the reliance placed on isotopic correlations in verification and the reliance placed on direct Agency measurements is dictated by particular circumstances. For a well characterized spent fuel, a high degree of reliance on the correlation techniques is suggested. In other less characterized and more varied situations, less reliance is suggested.

B. Confirmation of Information

During the irradiation of low enriched uranium, the initial isotopic composition of uranium is changed as well as the isotopic composition of the plutonium produced in that process. Graphs of the ratios of the various uranium and plutonium isotopes from zero exposure to the end-of-life are a portrayal of the burnup (and buildup) paths of those isotopes. The isotopic compositions observed in spent fuel dissolver solutions represent the end point of those burnup paths. Extrapolation backwards along the burnup paths from end-of-life to zero exposure yields the initial isotopic composition of the spent fuel. Through such extrapolation techniques, it is possible to confirm the initial

isotopic composition of the fuel with some degree of exactness. Correlations useful for this purpose include Pu/U versus final ^{235}U , Pu/U versus final ^{236}U , and published or measured alpha values for ^{235}U . These, of course, are not a substitution for direct verification at the time of fabrication.

The measured values on dissolver solutions of spent fuel for the Pu/U ratio and uranium and plutonium isotopic composition may be used to confirm with a high degree of exactness shippers' values for the Pu/U ratio, and isotopic compositions. The isotopic relationships themselves tend to be characteristic of a given reactor type and can be used to distinguish one reactor type from another.

Another area of safeguards concern is the possibility of substitution taking place after fuel fabrication. In this regard, the isotopic correlations and Pu/U ratio data can provide a degree of "after the fact" quantitative assurance that certain forms of substitution did not take place. In general, the use of all isotopic data makes substitutions that alter isotopic compositions difficult to conceal. The substitution of natural or depleted uranium for low enriched uranium would be detected through confirmatory tests of initial enrichment and comparison to historical values for relationships involving the initial enrichment. Substitution of a few rods which are identical but unirradiated into all irradiated assemblies would be detectable by relationships involving the plutonium isotopes such as Pu/U versus $^{240}\text{Pu}/^{239}\text{Pu}$. In this situation, the plutonium isotopes would retain their exposure dependent composition, e.g.,

a high $^{240}\text{Pu}/^{239}\text{Pu}$ ratio, whereas the Pu/U ratio and ^{235}U depletion would be reduced. This comparison would require "true" historical values as a basis for testing.

Other tactics such as the substitution of entire fuel bundles of a lower exposure for ones of higher exposure would be extremely difficult to detect by isotopic data alone. Reported reactor data would also have to be altered to the same extent.

Other confirmatory type tests which arise from use of isotopic correlations include 1) tests for the selective removal of plutonium (e.g., precipitation or plating) prior to the input accountability tank, and 2) tests for the introduction of recycle material into head-end tanks. The most important test is the test for the selective removal of plutonium. In this situation, comparisons to "true" historical values would detect plutonium removal within the technical reliability of pre-established values for Pu/U versus ^{235}D or Pu/U versus $^{240}\text{Pu}/^{239}\text{Pu}$.

C. Supporting Requirements

The full use of isotopic correlations in verification depends on certain supporting activities, process features, and availability of information. With the exception of inspection and evaluation activities, many of the conditions for (at least partial) use of the isotopic correlations currently exist. This conclusion is based on currently operating or proposed fabrication plants, reactors, and reprocessing plants and upon current measurement practices and availability of

supporting information.

The main process requirement at reprocessing for the use of the Pu/U ratio method and for the isotopic correlations is that the input dissolver solutions be truly representative, e.g., do not contain significant quantities of extraneous nuclear materials such as recycle. Recycle materials can be handled without adverse effect in some situations, by taking specific actions. A second requirement is that it be possible to know the order of dissolution of fuel assemblies and also be possible to roughly relate input accountability batches to the particular fuel bundles from which the batch is made up.

The more important information to be obtained from or reported by plant operator include 1) the total U, total Pu, and U and Pu isotopic compositions of fuel bundles both before and after irradiation, 2) the Pu/U ratio and uranium and plutonium isotopic compositions of input dissolver solution batches, 3) head-end waste losses, and 4) the uranium and plutonium isotopic compositions of product batches. In case, complete isotopic data are not readily available, the power of the technique is reduced. The general order of importance of the isotopic compositions (decreasing importance) is for uranium: ^{235}U , ^{236}U , ^{238}U , ^{234}U ; and for plutonium: ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , and ^{238}Pu .

The correlation techniques have their maximum use in the verification of the Pu/U ratio when it is possible to apply them to successive lots of fuel from the same reactor. The validity with which they may be applied in this type of application is greatly enhanced when the irradiation conditions are well known. Thus, knowledge of those factors which influence the correlations (such as fuel design, cladding, fuel to moderator ratio (including void fraction for BWR's), initial enrichment, burnable poison level, operating mode etc.) allow accumulated data to be applied with high validity. In many cases, it is sufficient to know only that the conditions which influence the correlations are the same for successive reactor lots. However, in other cases when it is known quantitatively that certain conditions have changed, it is possible from theoretical considerations to predict with some exactness the effect of such changes.

Reactor statements for the end-of-life composition are important to the application of the correlation techniques. The reactor statements for total uranium serve as the shipper's value for total U in the application of the Pu/U ratio method. Shipper's statements for total Pu, ^{235}U , ^{236}U , the plutonium isotopic composition, and exposure are extremely useful in safeguards pre-planning activities. From those data, "predicted" correlation values can be computed and optimum sampling plans for Agency verification can be established in advance.

If isotopic data are also available on feed and product of uranium enrichment plants and from plants receiving product from the reprocessing plants, this ring of isotopic data makes the falsification of isotopic data a near impossibility, even under modest levels of verification.

III. SUMMARY AND CONCLUSIONS

A review was made of the work done to date on isotopic correlations of potential safeguards value. Particular attention was given to those correlations involving the uranium and plutonium isotopes and the Pu/U ratio of spent low enriched uranium fuels. Two applications which have potential value in Agency verification activities were considered in detail. These are:

- The use of isotopic correlations in supporting the Pu/U ratio method of verifying the plutonium input to reprocessing.
- The use of isotopic correlations and associated dissolver solution data in confirming safeguards information about spent fuels which is stated prior to receipt at the reprocessing plant. This aspect includes tests for substitution.

The following overall conclusions have been reached for the case of low enriched uranium fuels. It is believed that the above applications have been sufficiently demonstrated to be included in Agency verification activities. The technical basis and conditions of applications for use of the Pu/U ratio method are well established. The isotopic correlations of plutonium and uranium have also been demonstrated both experimentally and theoretically. The technical

basis of supporting isotopic techniques such as the "across-the-plant" balance for isotopic composition at chemical reprocessing is also well established.

Several important questions must be resolved in regard to the application of isotopic correlations in safeguards. These are:

- The degree of reliance which should be placed on their use in verification.
- The nature of the information which will be required of plant operators for full use of the techniques and the manner in which that information is transmitted or made available to the Agency.
- The nature of the inspection procedures and activities required for use of the Pu/U ratio method and the correlations.
- The data evaluation procedures and staff capabilities which would be required for practical implementation.

This report addresses the questions of reliance and data evaluation. For the other points, it endeavors to provide a descriptive basis to assist the Agency in establishing information requirements and inspection activities.

The question of reliance is treated in two parts. First is the reliance placed on the Pu/U ratio method for verifying the total plutonium input to reprocessing. Second is the reliance placed on isotopic correlations as a means of independently estimating or verifying the Pu/U ratio of reprocessing plant input batches.

A major concern about the Pu/U ratio method is the validity of using the shipper's value for total U as the basis for converting the weighted or individual Pu/U ratios to total plutonium. The main conditions of validity are met when international independence exists or when the Agency is able to verify the total uranium content of fabricated fuel to the desired degree of exactness and maintain that verification through containment and/or surveillance until reprocessed. The concern in the verification of fabricated fuels is that the total uranium not be understated at fabrication, e.g., overload the rods.

It is apparent that for low enriched uranium fuels, the Agency can always use the shipper's value for total U to some advantage. Whether it uses that value as a primary means of verification or simply as a check, depends on its evaluation of validity (as discussed above).

Various degrees of reliance have been suggested for use of isotopic correlations to verify the Pu/U ratio. Suggestions range from a minimum level of their use as an "addition to" to a maximum level of use as the sole means of independently estimating or verifying the Pu/U ratio. The level of use depends on the relevant circumstances. The minimum level of using the correlations as a means of substantiating and reinforcing measurements made by the Agency will add to the robustness of the verification process. The maximum level of use is based on the situation where the correlations for a given fuel have been characterized to a point approaching the certainty of physical constants. The authors favor a balanced approach where the correlations are used in conjunction with some Agency measurements for the Pu/U ratio and isotopic compositions.

The balance between reliance on direct Agency measurements and reliance on the correlations would be dictated by the particular situation. For well characterized fuels, fewer Agency measurements would be required. For more varied situations the Agency's direct measurement plan would be increased. However, in all cases where applicable, the shipper's total U value and all appropriate correlations would be used to provide boundary conditions for fractional sampling plans. Only the degree of reliance and number of direct measurements would be varied. The use of isotopic correlations and data to confirm information and test for substitution would be applied "routinely" within statistical limits of uncertainty.

Information requirements for use of the Pu/U ratio and isotopic correlations are given in Tables VII and VIII on page 35. Information requirements are listed according to two priorities - Essential and Supporting. Essential information is that necessary for application of the techniques whereas information which reinforces or supports the essential information is listed as supporting information. A further subdivision of essential information is given in the text (pages 34,37) which describes the scope of application as a function of the availability of information.

As a general rule, the scope of application as well as the reliance which can be placed on the isotopic techniques increases with the scope of the information base. This aspect is particularly important in using historical data. For example, the confidence with which previously measured values for certain correlations could be applied to future situations would depend on the Agency's knowledge of irradiation conditions which affect the correlations e.g. cladding, fuel-to-moderator ratio, etc.

Inspection and verification activities are outlined on page 37 of the text. Since the Agency already plans to verify the output from low enriched uranium fabrication, the main impact on verification activities would be in activities for verifying the input to reprocessing. The use of the Pu/U ratio method of verification reduces the dependence of verification on plant input volume measurements and on absolute solution concentrations. The requirement for the presence of the inspector does not appear to be greatly changed. However, the activities and time schedule of head-end inspection activities would be somewhat different.

The use of isotopic correlations will require a greater emphasis on isotopic measurements by the Agency. Their use will also place greater emphasis on the data evaluation aspect of verification, particularly on a greater application of reactor physics.

The use of isotopic correlations assumes the Agency has the necessary technical procedures and staff expertise for data evaluation. It is generally felt, that one to two qualified staff members with proper calculational support could handle the work load envisioned for the early years of NPT. It is further assumed that advances in the state of the art as well as additional experimental data would be furnished as available by the member nations.

The general approach to evaluating the results of verification is given in the discussion on verification. A more detailed discussion of the Evaluation of Results is given in a separate section beginning on page 39.

To date, experimental and theoretical work has centered on the use of isotopic correlations for low enriched uranium fuels. With conditions

which permit use of the Pu/U ratio method, the correlation techniques can play an important role in the verification of total Pu input to reprocessing. The Pu/U ratio method also appears applicable to the case of plutonium thermal recycle fuels which are made up largely of uranium (> 96% UO_2). However, as yet, sufficient work has not been done to provide a firm basis for evaluating the potential value of isotopic correlations and isotopic data to such fuels. Future work should be directed to this case. Other future work should be directed to expected new fuel cycles such as the ^{233}U cycle and the fast breeder cycle.

IV. DISCUSSION

The discussion section is divided into four parts. In order of presentation, these are:

- Verification of Pu Input to Reprocessing
- Information Requirements
- Verification and Inspection Activities
- Evaluation of Results and Confirmation of Information.

A. Verification of Input to Reprocessing

Pu/U Ratio Method

When the Pu/U ratio method is used for verification of the total plutonium input to reprocessing, the shipper's value for total uranium is used for the total uranium input. The conditions of validity for using the shipper's value for total uranium were discussed previously concerning the need to assure the integrity of total uranium values. The technical reliability of the total uranium content of spent low enriched uranium fuel has been previously reported (11). Evidence of the long-term agreement expected between reactors and a reprocessing plant is shown in Table I for uranium

receipts at the Nuclear Fuels Services Plant at West Valley, New York. As shown by the data, the long term agreement is excellent ($+0.05\%$) and the standard deviation of the S-R difference is about that expected for measurement uncertainties (0.56% versus 0.54%).

The main process condition required for use of the Pu/U ratio and also the isotopic correlations is that the input dissolver solutions have the same Pu/U ratio and isotopic composition as the spent fuel from which they were derived. This is normally the case for head-end processes without recycle. If the acid in which the spent fuel is dissolved contains nuclear materials from the inner portions of the plant, corrections are required for such additions. Recycle of nuclear materials to the input accountability tank must also be taken into account. Appreciable quantities ($>2\%$) of recycle nuclear material, even with proper accountability, tend to complicate use of the technique. This is true also for conventional verification procedures. In general, verification procedures whether by the Pu/U ratio method or the volume-concentration method are easier to apply for the case of no recycle than the case of recycle to head end.

In comparing the shipper's total uranium to reprocessing total uranium, head-end wastes such as hull losses or chemical decladding losses must be taken into account. The actual comparison is between the shipper's value and the sum of reprocessing plant input batches plus wastes. For the chop-leach process, hull losses are of the order of $0.1 - 0.2\%$ of input. Chemical decladding losses are somewhat larger, ranging from $0.1 - 1.0\%$ of input depending on the particular fuel.

Table I

NUCLEAR FUELS SERVICES DATA FOR TOTAL URANIUM SHIPPER-RECEIVER
DIFFERENCES BETWEEN REACTORS AND CHEMICAL REPROCESSING

<u>Campaign Number</u>	<u>Shipper-Receiver Difference, % of Input</u>
1	-0.05
2	-0.53
3	+0.18
4	-0.05
5	+0.59
6	-0.94
7	-0.40
8	+0.18
9	+0.50
10	+0.04
11	-0.45
12	-0.16
13	+0.40
14	-0.26
15	+1.19
16	+0.90
Accumulative	+0.05% of total U receipts

1. Observed standard deviation of S-R = 0.56%
2. Expected standard deviation of S-R due to measurement uncertainties = 0.54%.
3. Total input $\cong$ 530 metric tonnes U.

When chemical decladding is used, the chemical attack is on the outside of the fuel elements tending to dissolve nuclear materials at the surface or of a higher burnup. As a result, the Pu/U ratio of cladding waste is often higher than the corresponding dissolver solution. When chemical decladding losses are small ($\sim 0.1\%$), that effect can be neglected. However, when decladding wastes are greater than about one percent, the solutions should be checked to see if a correction should be applied.

When recycle or cladding waste corrections for the Pu/U ratio are not required, the quantitative expression for total plutonium input to the reprocessing plant by the Pu/U ratio method is.

$$\frac{\text{Total Pu}}{\text{Input}} = (\text{total U shipper})(\text{weighted Pu/U})$$

where: total U shipper is the fabrication value corrected for burnup.

and

$$\text{Weighted Pu/U} = \frac{U_1 R_1 + U_2 R_2 + U_3 R_3 + \dots}{U_1 + U_2 + U_3 + \dots}$$

Where: U_i = total U in an input accountability batch in weight units.

R_i = the measured Pu/U of the corresponding input batch.

The total uranium is conventionally measured by the volume-concentration method. The weighting factors (U_1 , U_2 , etc.) can be written as $V_1 C_1$, $V_2 C_2$, etc. As the expression for the weighted ratio shows, proportional biases in either volume or uranium concentration would appear in both the numerator and denominator, hence cancel out. It

is also apparent, that random errors would also tend to be self-cancelling. It has been shown previously⁽⁷⁾ that only large (20%) and carefully selected alternate falsifications of the weighting can appreciably bias the weighted ratio. In addition, such biasing is only possible when the Pu/U ratios of the input batches are quite different. For all practical purposes, the operators' total uranium in each input batch may be used (without fear of falsification) in the weighting process. It can be shown that use of the "across-the-plant isotopic balance," and the shipper's value for total U in the fuel bundles, makes such large falsifications without detection very unlikely.

During the irradiation process the total uranium content of the fuel is changed at the rate of about 0.15% per thousand MWD/tonne U. For fuel exposed to 20,000 MWD/tonne U, the total change is about 3%. Normally, the reactor operators stated values for final uranium are sufficiently exact. The Agency can both verify stated burnup values and check their technical reliability from its own measurements of the corresponding dissolver solutions. The equation proposed by Moeken and Bokelund (4) is very useful for this purpose. (See page 45.)

Isotopic Correlations

The process conditions for use of isotopic correlations are the same as for the use of the ratio method. The main requirement is that the input dissolver solutions be a true representation of the dissolved spent fuel. The condition of freedom from contamination from recycle also applies here.

The isotopic correlations are used to verify the measured Pu/U ratio. They may be used to test both plant operator data as well as measurements made by the Agency. The theoretical and experimental basis for the correlations is covered in detail in previous publications and other papers presented at this panel. (See References)

The application of isotopic correlation in verification can be classified into two general cases according to time and into two general cases according to consistency. These are:

- Application with historical data
- Application without historical data
- Application when high consistency exists
- Application when consistency is low.

When historical data have been accumulated which highly characterize a particular fuel, then these values may be applied to future similar fuels from the same reactor. (Assuming of course that controlling irradiation conditions are the same). The use of historical data to check successive discharges from the same reactor is illustrated in Table II for Yankee Rowe Reactor. The Yankee fuel was stainless steel clad and of the same design. Reactor lots processed (10-20 tonnes U) at the chemical plant differed only in the initial enrichment and exposure. Two features of the empirical correlations are illustrated in Table II. First is the constancy of Pu/U versus ^{235}D

for material of the same initial enrichment and design for a PWR. Second is the empirically derived expression relating the term $\frac{\text{Pu/U}}{^{235}\text{D}}$ to the initial enrichment. For this, near ideal case, the general verification procedure for the Pu/U ratio would consist of monitoring the "across-the-plant isotopic composition" (see page 28) and in directly measuring one or two samples and a composite sample for bias check. Each batch reported by the operator would be tested by forming the correlation between Pu/U and ^{235}D . If all data check within statistical limits of the historical values and within direct Agency measurements of input and product samples, the Pu/U ratio of input to the campaign would be verified.

The case of no historical data but a high degree of consistency is illustrated by the data in Table III. In this, near ideal case, advantage is taken of the consistency of the data over a wide range of exposure, e.g., the Pu/U varies widely but the correlations are nearly constant. The general verification procedure as outlined just previously is used. However, in the case of no historical data the Agency places all of its reliance for bias control on its own directly measured data for Pu/U ratios and isotopic compositions. Each batch reported by the operator is tested to see if it differs from the majority of the data. If no significant differences are found on the bias test, across-the-plant isotopic composition, individual batch correlations tests, and composite samples, the Pu/U ratio of the reactor lot is verified.

Table II

COMPARISON OF PLUTONIUM RESULTS BY EMPIRICAL

 ^{235}U DEPLETION METHOD TO MEASURED VALUES

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

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

Table III
EXAMPLE OF Pu/U AND ISOTOPIC CONSISTENCY
YANKEE ROWE CORE V DATA

Raw Data

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

Pu/U - Isotopic Correlations

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

Where: $^{235}\text{D} = \text{wt. \% } ^{235}\text{U initial} - \text{wt. \% } ^{235}\text{U final}$

$\Delta^{236}\text{U} = \text{wt. \% } ^{236}\text{U final} - \text{wt. \% } ^{236}\text{U initial}.$

The most difficult case for application of isotopic correlations is the case of a low degree of consistency between input batches. Inconsistency is predicted for some cases. These include differences in the initial enrichment, differences in fuel design, and for BWR's differences in moderator void fraction.

The case of low consistency and no historical data is illustrated in Table IV for the Humboldt Bay boiling water reactor. This reactor lot included fuels of different original enrichments, different designs, as well as different void fractions (steam to water ratios). As a guide in establishing a fractional independent measurement plan, the expected consistency for Pu/U versus ^{235}D is computed from reactor statements. This is shown in the last column in Table IV. The actual measured values are shown in the adjacent column. Of the twenty batches, six have (dotted area) $\frac{\text{Pu/U}}{^{235}\text{D}}$ values which are lower than the majority of values for the same initial enrichment due to the fact that they were irradiated under lower void conditions than the other assemblies. This fact was not clearly established prior to reprocessing.

To verify the reported Pu/U ratios shown in Table IV by a fractional measurement plan, two plans could be considered. First is a direct fractional measurement plan which could be developed on the basis of exposure, fuel design, and initial enrichment. That plan is shown by the batches marked with an asterisk. Another approach is to use a purely random measurement plan. A 30% Agency measurement plan is suggested for a "mixed lot" without history. In the example, the designed plan (the starred batches) would confirm the low values with #29 and the higher values with #13, #22, #20, #23, and #18. For a purely random plan of 30%, there is a 92% chance of including at least one of the low batches in the plan. In evaluating the data reported by the plant, the safeguards authority would be concerned

Table IV
CHEMICAL REPROCESSING PLANT⁽¹⁾ INPUT DATA
FOR HUMBOLT BAY BWR REACTOR

Input Batch Number	²³⁵ U Initial Wt. %	²³⁵ U Final Wt. %	²³⁵ D Wt. %	Measured Values		Predicted Values	
				Pu/U grams/tonne U	Pu/U ²³⁵ D	Pu/U grams/tonne U	Pu/U ²³⁵ D
29*	2.578	1.919	0.659	2756	4182	3091	4427
28	2.578	1.874	0.704	2820	4006	3179	4391
25	2.578	1.860	0.718	2924	4072	3340	4408
26	2.578	1.847	0.731	3133	4286	3400	4412
27	2.578	1.837	0.741	3288	4437	3541	4464
17	2.578	1.810	0.768	3455	4498	3657	4424
4	2.578	1.764	0.814	3920	4816	3890	4475
16	2.578	1.751	0.826	3938	4762	3954	4527
14	2.578	1.709	0.869	4018	4624	4126	4415
15	2.578	1.706	0.872	4079	4677	4147	4440
13*	2.578	1.683	0.895	4236	4733	4203	4465
22*	2.310	1.250	1.060	5195	4901	5281	4848
21	2.310	1.223	1.087	5219	4801	5376	4815
20*	2.310	1.167	1.143	5398	4723	5517	4787
5	2.557	1.706	0.851	3931	4620	4054	4475
12	2.567	1.655	0.912	4234	4642	4251	4444
23*	2.402	1.441	0.961	4577	4675	4706	4713
2	2.433	1.423	1.010	4796	4748	4830	4628
19	2.276	1.108	1.168	5576	4774	5641	4789
18*	2.258	1.087	1.171	5741	4898	5820	4814

(1) Nuclear Fuels Services Plant, West Valley, New York

(2) Low Void Fraction

* Fractional Independent Measurement Plan Based on Physics Considerations.

about the possibility that the low values (batches 17, 25, 26, 27, 28, 29) represented an understatement of the Pu/U ratios. Direct measurement of one of the six low values would have partially confirmed the six low values. At this point in the evaluation process, several options are open. One would be Agency measurement of all low values. Another option would be measurement of a composite sample to confirm the entire lot. However, if full information were available, confirmation could come from isotopic data on the higher isotopes of Pu rather than additional Agency measurement. This type of confirmation was done for the Humboldt Bay Campaign. (13b) From core positions, power history, and the use of spectrum dependent plutonium isotopes, it was possible to confirm that the low values were due to differences in void fractions. The assemblies with low values for $\frac{\text{Pu/U}}{^{235}\text{D}}$ were those from outer regions of the core and had lower power generation (on the average) than the other assemblies.

Although the situation encountered in the previous example may not occur frequently, the principles involved are useful. A more common case of between batch variation for a given reactor lot is the case of mixed initial enrichments. Here the same general principles apply. Direct measurements comparisons would be used to test for bias, and the correlations would be applied to each individual input batch. In the case that historical data did not exist for the particular fuel, a pre-determined fractional measurement plan is favored. The aim here would be to have a direct comparison on each group of similar input batches e.g. $\frac{\text{Pu/U}}{^{235}\text{D}}$ expected to be the same. When historical data exist so that expected values for the correlations could be predicted, then the fractional measurement level could be reduced.

Another effective method of verifying the collective ratio data is by the use of composite samples. In the composite sample approach, the correlations may be used to advantage, particularly the expression Pu/U versus ^{235}D . As is apparent from the data shown in the tables the expression, $\frac{\text{Pu/U}}{^{235}\text{D}}$, varies to a much lesser extent than the Pu/U ratio; ranging from essentially a constant for the case of the Yankee data up to a standard deviation of about six percent for the Humboldt Bay data. As a consequence, the value of $\frac{\text{Pu/U}}{^{235}\text{D}}$ determined from a composite sample is less prone to proportioning errors than an estimate Pu/U or the weighted isotopic composition. The degree of robustness would depend on the process variance of those values. However, the correlations do provide a "built-in" check of composite sample results for Pu/U and isotopic composition, that is, the proportioning errors should produce the same error in the Pu/U ratio of the composite as in the isotopic composition of the composite. Thus, if the Pu/U of the composite sample is lower than the weighted Pu/U of the plant, then the ^{235}U value of the composite sample should be proportionately higher than the weighted ^{235}U of the plant.

Across-The-Plant Isotopic Composition

A basic assumption in the application of isotopic correlations in input verification is that the isotopic compositions of the batches not included in the fractional sample taken for independent measurement are not biased. One form of protection is the composite sample approach. Another practical means is to monitor the "across-the-plant" isotopic composition. The reprocessing plant does not

separate or mutate the isotopes. With the exception of the small effect of variable waste losses, the weighted isotopic composition of input should almost exactly equal the weighted composition of product, when beginning and ending inventories are taken into account. For clean-plant-to-clean-plant operations at a reprocessing plant, the effect of inventory is small. Historical data of weighted feed and product isotopic compositions are shown in Table V for the Nuclear Fuels Service Plant at West Valley, New York. These data have not been corrected for either waste losses or beginning and ending inventory. However, those effects are small. The data closely represent the agreement expected from measurement error alone. The actual agreement on most isotopes is seen to be within a few tenths of a percent.

It is evident that a total bias (or falsifications) of input isotopic data of more than about one percent would be apparent from verifying isotopic data product materials. For an outside safeguards group, product materials are more convenient to monitor than the highly radioactive feed solutions. In addition, product batches are often larger than feed batches, thereby reducing the effort expended in monitoring the same amount of material.

The sensitivity with which isotopic composition unbalance may be distinguished from measurement uncertainty is discussed in the Section on Evaluation of Results.

Table V

NUCLEAR FUEL SERVICES DATA FOR THE WEIGHTED
ISOTOPIC COMPOSITION OF FEED AND PRODUCT

Campaign Number		Uranium Isotopes, Wt. %		Plutonium Isotopes, Wt. %				
		<u>^{235}U</u>	<u>^{236}U</u>	<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{241}Pu</u>	<u>^{242}Pu</u>	<u>$^{239} + ^{241}\text{Pu}$</u>
2	Feed	0.937						99.0
	Product	0.938						99.0
1	Feed	0.931						99.0
	Product	0.934						98.9
3	Feed	0.821						91.16
	Product	0.819						91.21
4	Feed	1.006						79.94
	Product	1.006						79.82
5	Feed	2.509						86.47
	Product	2.510						86.39
6	Feed	0.712						87.12
	Product	0.714						87.13
7	Feed	0.733						87.79
	Product	0.734						87.72
13	Feed	2.708	0.398	75.087	14.154	8.585	1.356	83.672
	Product	2.704	0.399	75.190	14.148	8.525	1.340	83.714
15	Feed	0.704						
	Product	0.704						
17	Feed	2.724	0.480	72.397	14.913	9.942	1.681	82.339
	Product	2.722	0.479	72.301	14.972	9.954	1.696	82.255
23	Feed	1.593	0.174	75.805	17.173	5.561	1.102	81.366
	Product	1.601	0.174	75.964	17.066	5.520	1.093	81.484

The isotopic composition balance also has the power to detect gross non-proportional errors in input volume measurements, sampling, and concentration measurements. This is seen by the expression for the weighted enrichment of the input batches making up an input campaign:

$$\text{Weighted } ^{235}\text{U} \text{ of Input} = \frac{U_1 \text{ } ^{235}\text{U}_1 + U_2 \text{ } ^{235}\text{U}_2 + U_3 \text{ } ^{235}\text{U}_3 + \dots}{U_1 + U_2 + U_3 + \dots}$$

where:

U_i = total uranium in an input batch in weight units.

$^{235}\text{U}_i$ = Weight percent ^{235}U in the corresponding input batch.

Since the total uranium is normally determined as the product of volume and uranium concentration, the weighting factor includes the tank volume and sample concentration.

As was noted earlier, one verification concern is to insure that the weighting factors are not selectively biased to change the weighted Pu/U ratio. This is a second order effect, but gross selective falsifications could yield a total U input which agrees with the shipper's value and yet bias the Pu/U ratio. (This subject is discussed in detail in Reference (7)). This biasing tactic is only practical when the process variance is large. However, in this case the Pu/U ratio generally varies inversely with the final ^{235}U . The result is that if the weighting factors are biased to understate the Pu/U ratio, then the uranium isotopic composition is biased through the same weighting factor but in the opposite direction. The ability to detect such a scheme is shown in Table VI. In the example, only the total U in the input batch is selectively miss

stated and other values are true. The effect is to bias the weighted Pu/U ratio low. Concurrently, the weighted enrichment is biased high, the effect is nearly proportional to the degree that the Pu/U ratio is biased. In the example, the biasing of weighting factors would cause a difference of about 1.2 relative percent between the stated isotopic composition of feed and the observed isotopic composition of product. A difference of this magnitude is about 3-4 times greater than the expected measurement uncertainties.

For the example shown in Table VI where there is a relationship between Pu/U and isotopic composition, certain forms of sampling error (or falsification) would be detected. For example, the substitution of sample #1 for #6 would produce a gross difference between the stated input isotopic composition and the true isotopic composition. The substitution of the two closest samples #5 for #6 would also cause a small but significant difference.

Table VI

EFFECT OF SELECTIVELY BIASING WEIGHTING FACTORS
ON THE WEIGHTED ISOTOPIC COMPOSITION OF INPUT

<u>Input Batch</u>	<u>True U Kg</u>	<u>Biased U Kg</u>	<u>True ²³⁵U Wt. %</u>	<u>True Pu/U grams/tonne U</u>
1	1000	1100	3.176	4968
2	1000	1100	2.796	7104
3	1000	1100	2.675	7930
4	1000	900	2.471	8863
5	1000	900	2.254	9794
6	1000	900	2.046	11119
Total	6000	6000		

<u>True Weighted ²³⁵U, Wt. %</u>	<u>Biased Weighted ²³⁵U, Wt. %</u>	<u>True Weighted Pu/U, Grams/Tonne U</u>	<u>Biased Weighted Pu/U, Grams/Tonne U</u>
2.5697	2.6009	8296.3	8133.4

<u>Ratio of True/Biased ²³⁵U</u>	<u>Ratio of Biased/True Pu/U</u>
0.988	0.980

B. Information Requirements - Low Enriched U Fuels

The information which would be required from plant operators for full use of the isotopic correlations in verification or confirmation is listed in Tables VII and VIII. The information which is essential for full use is listed in Table VII. Information which supports or reinforces the essential information is listed on Table VIII. The information listed in Table VII is subdivided into priority #1 and priority #2 information. Priority #1 information would allow full use of the correlations for verification and confirmation at the current state of the art. Information in priority #2 is a listing of additional isotopic data which are normally included when complete isotopic data are measured or calculated. Priority #2 data would permit maximum use of uranium and plutonium isotopic correlations, including possible future advances in the state of the art.

The supporting information listed in Table VIII completes the ring of isotopic data for the low enriched uranium fuel cycle. Its main value to the correlative and confirmatory tests described in this report is that of supporting and reinforcing the more primary data. For example, the plutonium fabrication input data can be used to confirm the output of the reprocessing plant.

The relationship between availability of information and degree of specific implementation may be described qualitatively for each type of application. When only knowledge of ^{235}U content is available at fuel fabrication output, then the use of dissolver solution ^{236}U values to confirm initial enrichments is weakened. The relationship between Pu/U and $\Delta^{236}\text{U}$ is also largely lost. If the chemical reprocessing plant isotopic data is limited to ^{235}U , the power to detect certain

Table VII
ESSENTIAL INFORMATION FOR USE OF ISOTOPIC
CORRELATIONS IN VERIFICATION AND CONFIRMATION
LOW ENRICHED URANIUM FUELS

<u>Fuel Fabrication Data</u>	<u>Chemical Reprocessing Data</u>	<u>Reactor Data</u>
<u>Priority #1</u>	<u>Priority #1</u>	<u>Priority #1</u>
1) <u>Each Fuel Assembly</u> Total U Weight ^{235}U Wt. % ^{236}U Wt. %	1) <u>Each Input and Product Batch</u> Total U Weight Total Pu Weight ^{235}U Wt. % ^{239}Pu Wt. % ^{236}U Wt. % ^{240}Pu Wt. % ^{241}Pu Wt. % ^{242}Pu Wt. %	1) <u>Each Spent Fuel Assembly</u> Total U, Total Pu, ^{235}U , Exposure.
2) Fuel element design (1) information, e.g. dimensions cladding, burnable poison, etc. If graded bundle, rod pattern in bundle by individual enrichment giving nominal enrichment of rods.	2) Order of dissolution by assembly. 3) Correspondence between dissolution and input batches.	2) <u>Reactor Design Characteristics</u> Type, moderator, fuel design data - cladding, dimensions, within assembly enrichment, lattice pitch.
<u>Priority #2</u>		<u>Priority #2</u>
1) <u>Each Fuel Assembly</u> ^{238}U Wt. % ^{234}U Wt. %	4) Input tank and dissolver heel volumes per batch. 5) Hull or chemical decladding losses per dissolution batch. 6) Normal accountability data.	3) Operational history for core, assembly history, core position, charge and discharge times.
2) <u>Each Input Batch</u> Total U Weight ^{235}U , ^{236}U , ^{238}U , ^{234}U	<u>Priority #2</u> 1) <u>Each Input and Product Batch</u> ^{238}U Wt. % ^{238}Pu Wt. % ^{234}U Wt. %	1) <u>Each Spent Fuel Assembly</u> ^{236}U , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu

(1) Information may be available from either fabricator or reactor utility company.

Table VIII
SUPPORTING INFORMATION FOR USE OF ISOTOPIC
CORRELATIONS IN VERIFICATION AND CONFIRMATION
LOW ENRICHED URANIUM FUELS

<u>Isotope Separation</u> <u>Plant Data</u>	<u>Uranium Conversion</u> <u>Plant Data</u>	<u>Plutonium Fabrication</u> <u>Plant Data</u>
1) <u>Each Input and Product Batch</u>	1) <u>Each Input and Product Batch</u>	1) <u>Each Input Batch</u>
Total U Weight	Total U Weight	Total Pu
^{235}U Wt. %	^{235}U Wt. %	^{239}Pu Wt. %
^{236}U Wt. %	^{236}U Wt. %	^{240}Pu Wt. %
^{238}U Wt. %	^{238}U Wt. %	^{241}Pu Wt. %
^{234}U Wt. %	^{234}U Wt. %	^{242}Pu Wt. %
		^{238}Pu Wt. %

forms of substitution or recycle is lost. The use of the plutonium isotopic data to confirm or diagnose void effects is lost. If reactor information which details the conditions affecting the values of the correlations is not available, historical data cannot be applied with confidence. In addition, it would not be possible to confirm predictable changes between successive reactor discharges. Nor would it be possible to predict new values for the correlations from historical value established under other conditions, e.g., different cladding or fuel design. For Pu recycle fuels, the plutonium isotopic composition of each fuel assembly would also be priority #1 fabrication information.

C. Verification and Inspection Activities

It is assumed that the Agency has independent reliable data on the output from uranium fuel fabrication. Under that assumption, the main impact of the techniques discussed in this report is on inspection activities for the input area of the reprocessing plant. Product verification activities would not be materially affected. Measurement verification plans would include isotopic measurements of product batches by the Agency. The following general inspection and verification plan is suggested for the Pu/U ratio and the supporting isotopic correlation techniques:

Suggested General Verification Activities for Total Pu Input Verification

1. Preprocessing Planning (By Central Technical Staff)

Obtain from the reprocessing plant operator the tentative schedule for the various reactor discharges to be processed (for the next few years). From previously obtained fabrication data and

current data from the corresponding reactors, determine the range of consistency to be expected for the Pu/U ratio and isotopic correlations. From these and a general knowledge of core ordering practices for reprocessing, develop the following general verification criteria:

- i) From the consistency expected for the Pu/U ratio, determine the level of surveillance required for input volume or weight measurements to assure the validity of accepting operators weighting factors (total U in each batch).
- ii) From the consistency expected for the correlations and the extent and exactness of historical data, prepare a tentative fractional independent measurement plan. If the reactor lot is expected to be highly consistent in the correlations, e.g. $\frac{\text{Pu/U}}{^{235}\text{D}} = K$, a purely random measurement plan is suggested. If the reactor lot is a mixture, prepare a guideline for a designed fractional plan which can be applied when the actual dissolution order is known.
- iii) Forward technical guidelines and tentative Agency measurement plans to the field inspectors.

2. Inspection Activities

It is assumed that inspection activities such as checking fuel receipts and inventories in the storage basin, surveillance and checking of dissolution order, waste measurements for hulls or chemical decladding, etc. will be carried out routinely. The following activities are emphasized for use of the Pu/U ratio-correlation method.

- i) Obtain and check the order of dissolution of assemblies and approximate amounts of each assembly contained in the input

accountability batches.

- ii) Check stated reprocessing plant values for total U in each batch against shipper's weight for U dissolved and batch volume and concentration statements.
- iii) Observe sampling procedures and obtain Agency samples of each input batch.
- iv) From actual dissolution order, tentative remeasurement schedule and technical guidelines, prepare sample dilutions, composite samples etc. for independent Agency measurements.
- v) Apply Agency measurement schedules to plutonium and uranium product materials. Prepare individual and composite samples for Agency measurements.
- vi) Obtain any information required batch sizes, isotopic data, heel sizes etc., that is not routinely reported by the plant operator. Detailed guides for information gathering will have been previously established.
- vii) Submit all appropriate data, sample identifications, etc., for evaluation.

D. Evaluation of Results and Confirmation of Information

For the verification procedures suggested in this report, it is assumed that the evaluation of results and the confirmation of information would be performed by a central technical staff. The following general procedure is envisioned:

1. Results of Total Pu Input Verification

From the total data gathered from plant operators, field inspectors, and the safeguards laboratories, perform statistical tests of significance for the following: (See Statistical Methods, Part 3)

i) Total U Shipper-Receiver Difference

Test the shipper-receiver difference for total U of the entire reactor lot against expected measurement uncertainties. Test cumulative shipper-receiver differences against long-term expectations.

ii) Pu/U Ratio and ^{235}U wt% Shipper-Receiver Differences

Test each individual batch (or groups) for Pu/U and ^{235}U wt% differences against expected uncertainties in reactor predictions. Test for entire input lot and cumulative to date.

iii) Paired Comparison Tests

Perform paired comparison (D statistic) tests on all Agency measurement data on input batches. This would include Pu/U ratios, isotopics and concentrations when quantitative dilutions were made. Make similar paired comparisons on product isotopic and concentration data where appropriate to test isotopic data for the "across-the-plant" isotopic composition.

iv) Across-Plant-Isotopic Composition

From operators' data on the isotopic composition of input and product batches and estimates of beginning and ending inventory, compute the weighted isotopic composition of input + beginning inventory and the weighted isotopic composition of product + ending inventory. Test the difference in the weighted isotopic composition across-the-plant against expected measurement uncertainties. Note, if uranium IV is used as a process reductant, the beginning

and ending inventory totals and the enrichment of the uranium used as a process reductant must be taken into account.

v) Isotopic Correlations

Using isotopic data for the unirradiated fuel and the listing of specific assemblies contained in a particular input batch, form the correlations relating Pu/U to isotopic composition on each input batch, including whenever possible, $\frac{\text{Pu/U}}{^{235}\text{U}}$, $\frac{\text{Pu/U}}{\Delta^{236}\text{U}}$ and

$\text{Pu/U} = m(^{240}\text{Pu}/^{239}\text{Pu}) + b$ on each batch with operator's data. Compare consistency and mean values with historical data if available. Compare to similar relationships obtained from the samples also analyzed by the Agency. If consistency is low due to a "mixed core" or difficulty in relating initial enrichment to a particular input batch, form the weighted correlations for the entire reactor lot. Use the weighted Pu/U and weighted isotopic composition of the operator. Compare to historical values. Compare to weighted values (weighted by total U in batch) from Agency measurements of individual samples and to the composite sample.

vi) Evaluation of Results

If no significant differences have been noted for total U, Pu/U, the "across-the-plant" isotopic balances, isotopic correlations, and all paired comparisons of isotopic and concentration data, the total plutonium input has been verified.

vii) Estimation of Maximum Differences

From the previous difference tests, estimate possible over or understatements of input as follows:

a. Pu Shipper-Receiver

Multiply the total U shipper-receiver difference by operator's weighted Pu/U ratio.

b. Pu/U Ratio Difference

From paired comparison data on the Pu/U ratio, convert the average difference to grams Pu for entire input lot. Use as a proportional total bias estimate.

c. $\frac{\text{Pu/U}}{^{235}\text{D}}$ Difference

From operator's input data for the entire reactor lot and fabrication data on initial ^{235}U , compute the weighted $\frac{\text{Pu/U}}{^{235}\text{D}}$. Compare to weighted (weight by total U $\times$ ^{235}D) value of $\frac{\text{Pu/U}}{^{235}\text{D}}$ from individual Agency samples, composite samples and historical data. Convert to relative percent difference and determine if the difference would represent an over or understatement of input.

d. "Across-the-Plant Enrichment Difference" Applied to Input

Determine relative difference in the across-the-plant enrichment from iv). Apply to operator's weighted $\frac{\text{Pu/U}}{^{235}\text{D}}$ and determine difference in weighted Pu/U and in total Pu input.

The estimates of difference given in vii; a, b, c, and d, represent estimates of possible over or understatement of input. These estimates should be used in the evaluation of MUF. The Pu shipper-receiver estimate in "a" would represent the plutonium difference equivalent to uranium difference. If that difference represented a significant overstatement of input, then it could explain a positive MUF. The paired comparison difference in "b" would represent the observed bias between Agency measurements of the Pu/U ratio and the operator's measurements. The variance of the "D" statistic would represent an estimate of the combined measurement precision. The difference estimates for "c" and "d" represent the collective verification of total input under the assumption that the fractional remeasurement plan did not include atypical errors (or falsifications) on a few of the batches.

2. Confirmation of Information and Tests for Substitution

The verified operator's dissolver solution data for Pu/U and uranium and plutonium isotopic compositions are used as the basis for confirming information stated about the fuel prior to dissolution and in testing for substitution. Three basic approaches are used. Confirmation of initial fuel isotopic composition is done by either extrapolation back to zero exposure or by comparison to "true" historical data involving initial isotopic values. Confirmation of end-of-life statements is simply a comparison of reactor statements with chemical plant measured values. Tests for substitution are in part covered by the confirmation of initial composition and in part by comparison to history. The general procedure is as follows:

i) Confirmation of Initial Isotopic Composition

From operator's data for batches of the same initial enrichment and having a high degree of consistency in Pu/U versus ^{235}U and ^{236}U , form the linear regressions:

a. $\text{Pu/U} = \alpha_1(^{235}\text{U final}) + \beta_1$

b. $\text{Pu/U} = \alpha_2(^{236}\text{U final}) + \beta_2$

solve for β_1 and β_2 at Pu/U = 0. If the lines are over a reasonable range of values (e.g., 50% relative difference), the confidence limits on beta should be the order of 2-5% relative. If the exposure range is limited, use initial and final values for ^{236}U and published (or previously measured) values for ^{235}U alpha (fission to capture ratio) and estimate the initial enrichment. When data covers a reasonable range of exposure, other graphical plots or regression lines are useful for confirmation. These include:

$^{236}\text{U final}$ versus $^{235}\text{U final}$

$^{236}\text{U final}$ versus $^{240}\text{Pu}/^{239}\text{Pu}$

$^{235}\text{U final}$ versus $^{240}\text{Pu}/^{239}\text{Pu}$

ii) Confirmation of End-of-Life Shipper's Values

From operator's data on individual input batches or groups of batches (where matching of assemblies is difficult), make a tabular comparison of predicted versus measured for all available predicted values, e.g., Pu/U, $^{235}\text{U wt\%}$, $^{236}\text{U wt\%}$, and Pu isotopics. Also form predicted correlations and

measured correlations for $\frac{\text{Pu/U}}{235\text{D}}$, $\frac{\text{Pu/U}}{\Delta 236\text{U}}$, and $\text{Pu/U} = m(^{240}\text{Pu}/^{239}\text{Pu}) + b$

wherever possible. Compare predicted versus measured for all paired comparisons to see if reasonable agreement exists, e.g., 5% relative difference. Compare to previously obtained measured values for the correlations and to the worldwide data base for that class of reactors and initial fuel enrichment. The tabular representation of predicted versus measured is a portrayal largely of the quality of all predicted values and the quality of agreement. The comparison to historical values and the worldwide data base serves as confirmation that the fuel is from the stated reactor and that the stated irradiation conditions are true (within the uncertainties of the values). The comparison with historical data is also an implicit check on certain forms of substitution or plutonium removal.

iii) Confirmation of Total U Burnup

The equation suggested by Moeken and Bokelund⁽⁴⁾ is useful to test reactor statements total uranium burnup against dissolver solution measurements. Using verified operator's data for the weighted Pu/U and weighted uranium and plutonium isotopic compositions and initial enrichment values, determine

$\frac{U_{\text{initial}}}{U_{\text{final}}}$ for input lot using the equation below and compare to reactor statements.

$$\frac{U_{\text{initial}}}{U_{\text{final}}} = \frac{1 + (1 + 1.886 w_0 + 2.085 w_1 + 8.95 w_2) \text{Pu/U} - 0.9081 w_5}{1 - 0.9081 w_5^0}$$

where w_0 = final ^{240}Pu weight fraction
 w_1 = final ^{241}Pu weight fraction
 w_2 = final ^{242}Pu weight fraction
 w_5 = final ^{235}U weight fraction
 w_5^0 = initial ^{235}U weight fraction

iv) Tests for Substitution

Tests for possible substitution after fabrication include tests for 1) substitution of natural or depleted uranium for low enriched uranium and 2) the substitution in all irradiated assemblies of a few rods of material of the same initial enrichment but unirradiated.

a. Tests for Substitution of Natural or Depleted Uranium

1. Compare $\frac{\text{Pu/U}}{^{235}\text{D}}$ with historical values.
2. Compare previous tests for initial composition with stated initial values.

If substitutions using natural U were made for 5% of the fuel in a typical PWR assembly of 2.70 wt% final ^{235}U , the value of $\frac{\text{Pu/U}}{^{235}\text{D}}$ would be reduced by about 10%. In this case, the

substitution would decrease the final enrichment or increase depletion and decrease the Pu/U ratio.

b. Substitution of Unirradiated Material of the Same Initial Enrichment (in all fuel assemblies)

1. Compare $\frac{\text{Pu/U} + b}{^{240}\text{Pu}/^{239}\text{Pu}}$ with historical values for the same fuel for entire reactor lots of about the same value of Pu/U.

The substitution essentially reduced Pu/U by the fraction substituted without appreciably changing the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio. Thus, a substitution of 5% would reduce

$$\frac{\text{Pu/U} + b}{^{240}\text{Pu}/^{239}\text{Pu}} \text{ by } 5\%.$$

3. Statistical Methods

i) $\hat{D}$ (Difference Statistics)

Many of the statistical tests which are suggested herein are based on the $\hat{D}$ (difference) statistic. The main reason for this is that product variation, biases and systematic errors (where biases are distinguished from systematic errors which result from calibration) are subtracted out in the between differences variance estimate

$$s_d^2 = \frac{\sum_1^n (d_i - \bar{d})^2 / (n-1)}{\frac{\sum_1^n d_i^2 - \left(\sum_1^n d_i \right)^2 / n}{n-1}} \quad (3.1)$$

Thus, in a test of the average difference $\bar{d}$ divided by $s_{\bar{d}} = s_d / \sqrt{n}$, the presence of an effect which makes the categories different will be reflected in the numerator but not in the denominator and the test's discriminatory power will be enhanced. The i th difference is $d_i = x_{1i} - x_{2i}$, where x_{1i} and x_{2i} are from two categories of measurements which are to be compared, e.g., shipper-receiver, operator-inspector. Then the statistic

$$\hat{D}_1 = \frac{\bar{d}}{s_{\bar{d}}} = \frac{\bar{x}_1 - \bar{x}_2}{s_d/\sqrt{n}} = \frac{\sum_{i=1}^n x_{1i}/n - \sum_{i=1}^n x_{2i}/n}{s_d/\sqrt{n}}$$

is distributed as Student's t with $n-1$ degrees of freedom if the null hypothesis $H_0: E(d_i) = 0$ is true, and if the differences are normally and independently distributed.

$t_{\beta}(n-1)$ is the 100β percentile of the Student's t distribution with $n-1$ degrees of freedom. If the sample statistic $\hat{D}_1$ lies in the critical region, the null hypothesis is rejected.

The alternative hypotheses which are of interest and their corresponding critical regions at the $100(1-\alpha)\%$ level are

alternative hypothesis	critical region
$H_1: E(\hat{D}_1) < 0$	$\hat{D}_1 < t_{\alpha}(n-1) = -t_{1-\alpha}(n-1)$
$H_1: E(\hat{D}_1) > 0$	$\hat{D}_1 > t_{1-\alpha}(n-1)$
$H_1: E(\hat{D}_1) > 0$	$\hat{D}_1 < -t_{1-\alpha/2}(n-1) : t_{1-\alpha/2}(n-1) < \hat{D}_1$

If the precisions, defined as the variance of reproducibility are essentially known due, for example, to an accumulation of historical results, then

$$\hat{D}_2 = \frac{\bar{x}_1 - \bar{x}_2}{\sigma_d/\sqrt{n}}$$

is distributed as a normal variable, $\mu = 0$, $\sigma = 1$, under the null hypothesis $H_0: E(d_i) = 0$, where $\sigma_d^2 = \sigma_1^2 + \sigma_2^2$ and

(σ_1^2, σ_2^2) are the known variances. The critical regions, corresponding to the different alternative hypotheses are formed in a manner similar to those above using the percentiles of the $N(0,1)$ distribution to establish the critical regions for $\hat{D}_2$.

ii) Cumulative Differences

The distinction here from the previous material is simply the manner in which the data originate. One has data for the present period and for the previous $(m-1)$ periods, say. If σ_d^2 has been estimated for each of the previous $m-1$ periods one may estimate σ_d^2 by

$$s_d^2 = \frac{\sum_{i=1}^m (s_{di}^2) (n_i - 1)}{\sum_{i=1}^m n_i - m}$$

with $\sum_{i=1}^m n_i - m$ degrees of freedom, or one may calculate the σ_d^2 estimate using all the data together for the m periods and using a common mean difference. The distinction here is really one in choosing the procedure which is easiest to apply. The overall average difference is tested by $\bar{d}/s_{\bar{d}}$ where

$$\bar{d} = \bar{x}_1 - \bar{x}_2 = \frac{\sum_{i=1}^m n_i (\bar{x}_{1i} - \bar{x}_{2i})}{\sum_{i=1}^m n_i} = \frac{\sum_{i=1}^m \sum_{j=1}^{n_i} (x_{1ij} - x_{2ij})}{\sum_{i=1}^m n_i}$$

$$\text{and } s_{\bar{d}} = s_d / \sqrt{\sum n_i}$$

where x_{1ij} and x_{2ij} refer to the values for the j th sample in the i th period.

One may also proceed as follows:

$$\bar{d}' = \frac{\sum_{i=1}^m \bar{d}_i}{m}$$

with standard deviation estimate

$$s_{\bar{d}'} = s_d \sqrt{\frac{1}{m} \sum_{i=1}^m \frac{1}{n_i}}$$

and $\bar{d}'/s_{\bar{d}'}$ is distributed as Student's t under the null hypothesis of no difference between categories.

If the differences are ordered with time as may happen with shipper-receiver differences, one may be interested in detecting a change in the state of control and in estimating when the change occurs. Techniques such as CUSUM have been found to be very effective in this situation.

iii) Linear Regression

The plots of the relationships between Pu/U , ^{235}D , $\Delta^{236}\text{U}$, and the various Pu and U isotopic tend to be linear if the ranges of the relationships are reasonable. There are several cases which arise in isotopic correlations.

a. Model 1: $E(y) = bx$, $\sigma_y \propto E(y)$

This model indicates that the line goes through the origin and the standard deviation of y is proportional to the expected value of y . Q , the sum of the weighted residuals,

$$Q = \sum_{i=1}^n \left(\frac{y_i - bx_i}{\sigma_{y_i}} \right)^2$$

is minimized when $\hat{b} = (\sum_{i=1}^n y_i/x_i)/n$, and the variance of the estimate of b is estimated as

$$s_{\hat{b}}^2 = \frac{s^2(y_i/x_i)}{n}$$

In other words, under the models for the expected value and the variance of y , the minimum variance unbiased estimate of b is simply the average of the ratios y_i/x_i and the variance estimate of the estimate of b is simply the variance of the individual ratios y_i/x_i divided by n . This situation is one of the bases for the use of the statistics

$$R_5 = (Pu/U)/^{235}\text{D}, \text{ and } R_6 = (Pu/U)/\Delta^{236}\text{U}$$

as types of optimum estimators where $y = Pu/U$ and $x = ^{235}\text{D}$ for R_5 and $x = \Delta^{236}\text{U}$ for R_6 .

The predicted $\hat{y}_i$ value is estimated as $\hat{b}x_i$ with estimated variance $\hat{s}_{\hat{y}_i}^2 = x_i^2 \hat{s}_b^2$. The residual variance is estimated as $s_{r_i}^2 = s^2 - \hat{s}_{\hat{y}_i}^2$. An individual residual $r_i = y_i - \hat{y}_i$ can be checked against s_{r_i} to see if it represents an outlier. r_i/s_{r_i} is approximately distributed as Student's t with $n-1$ degrees of freedom under $H_0: E(r_i) = 0$. The tests for the individual residuals are, however, not independent for several reasons, 1) the residuals are somewhat correlated, and 2) the same estimate of s^2 is used in all the tests.

b. Model 2: $E(y) = a + bx$, $\sigma_y^2 = \text{constant}$

This model, which is occasionally employed here, is fully treated in most statistical textbooks.

iv) Across-Plant-Isotopic Composition Comparisons

In order to make an understated Pu/U value at the separations input point consistent with the inherent isotopic weight percent relationships, the reported isotopic weight percents which $\begin{pmatrix} \text{decrease} \\ \text{increase} \end{pmatrix}$ with exposure must be $\begin{pmatrix} \text{increased} \\ \text{decreased} \end{pmatrix}$ appropriately. Such falsifications can, within statistical limitations, be detected in the separations facility's product. The campaign weight percent of the isotope jE for the element E at the separations facility input is estimated by

$$\hat{j}_E = \frac{\sum_{i=1}^m \hat{E}_i \hat{j}_{E_i}}{\sum_{i=1}^m \hat{E}_i}$$

over the m batches in a campaign where $\hat{j}_{E_i}$ is the estimated weight percent of the isotope j_E of the element E in the i th batch and $\hat{E}_i$ is the estimate of the amount of the element E in the i th batch where E is either Pu or U. If the batches all have the same amount E , and the measurements are such that the relative measurement variance of the estimate of the isotope j_E dominates that of E_i , then the relative variance of the estimate $\hat{j}_E$ is approximately

$$V\hat{j}_E = \frac{V\hat{j}_{E_i}}{m}$$

where $V\hat{j}_{E_i}$ is the relative measurement variance of the isotopic weight percent. If all the values E_i are precise but proportionally biased then the expected value of $\hat{j}_E$ is

$$E(\hat{j}_E) = \frac{\sum_{i=1}^m (pE_i) \hat{j}_{E_i}}{\sum_{i=1}^m (pE_i)} = j_E,$$

since the proportionality factor cancels out, and thus,

$$V_{jE}^{\wedge} = \frac{V_{jE_i}^{\wedge}}{m}$$

as before. If the jE_i values are variable with a non-negligible process variance, then obviously the situation is more complicated. It would seem, however, that the measurement variance of jE_i would be dominating and that the other terms, the measurement variance of E_i , the process variance of E_i and jE_i would enter as second-order effects. The formula derived for this situation

$$V_{jE}^{\wedge} = \frac{V_E^{\wedge} V_{jE'}^{\wedge} + V_{jE}^{\wedge} (1 + V_{E'}^{\wedge})}{m}$$

where V denotes relative variance, a caret denotes measurement variance and a prime denotes process variance, shows these considerations to be the case. This formula is taken from (7) and extensive simulation results of such a variable ratio agree with expected results as given by the formula.

A similar test would apply to product so that the statistical test of the across-the-plant difference in the campaign weight percent of the isotope jE is

$$\frac{(\hat{j}_{E.})_{\text{feed}} - (\hat{j}_{E.})_{\text{product}}}{\sqrt{\left(\hat{j}_{E.} \sqrt{V_{j_{E.}}}\right)_{\text{feed}}^2 + \left(\hat{j}_{E.} \sqrt{V_{j_{E.}}}\right)_{\text{product}}^2}} = \frac{d(\hat{j}_{E.})}{s_d(\hat{j}_{E.})}$$

The number of degrees of freedom has to be estimated by the Satterthwaite approximation. However, if the measurement variance of the isotope weight percent from either the feed or product is dominating then the number of degrees of freedom in $s_d(\hat{j}_{E.})$ is approximately equal to the number of degrees of freedom in this dominating error source. If the relative variances of the isotopic weight percent measurement sources are the two dominating components in $s_d(\hat{j}_{E.})$ then the degrees of freedom for $s_d(\hat{j}_{E.})$ is approximately

$$f' = \frac{s_d^4(\hat{j}_{E.})}{\frac{\left[\hat{j}_{E.}^2 (1 + V_{E.}) V_{j_{E.}}/n\right]_{\text{feed}}^2}{df_{\text{feed}}} + \frac{\left[\hat{j}_{E.} (1 + V_{E.}) V_{j_{E.}}/n\right]_{\text{product}}^2}{df_{\text{product}}}}$$

where the degrees of freedom for feed and product are the degrees of freedom available for estimating measurement variances of the isotopic weight percents. Naturally, values such as n , $V_{E.}$, and $V_{j_{E.}}$ are not the same for feed and product values.

vi) The Effect of Biases

If j_E is a measurement with a constant bias δ , then

$$E(\hat{j}_E) = \frac{\sum_{i=1}^m (j_{E_i} + \delta)}{\sum_{i=1}^m j_{E_i}} = \frac{\sum_{i=1}^m j_{E_i}}{\sum_{i=1}^m j_{E_i}} + \delta$$

so that a bias in the isotopic weight percent measurement leads to the same bias in the campaign isotopic weight percent estimate. If the bias in the isotopic weight percent measurement is relative then

$$E(j_E) = \frac{\sum_{i=1}^m (p j_{E_i})}{\sum_{i=1}^m j_{E_i}} = \frac{p \sum_{i=1}^m E_i j_{E_i}}{\sum_{i=1}^m E_i j_{E_i}}$$

so that a relative bias in the isotopic weight percent measurement leads to the same relative bias in the campaign isotopic weight percent estimate. A relative bias in measuring E_i obviously cancels out and a small constant bias in the measurement of E_i would tend to have little effect. A calibration error which affects the measurement of E_i , for example, would have little effect if the error is small. The following formula for the relative differential of j_E under the influence of a constant bias of k in E_i

$$\frac{d(j_E)}{j_E} = -\left(\frac{k}{\bar{E}}\right) \left[1 - \frac{\sum_{i=1}^m E_i j_{E_i}}{\sum_{i=1}^m j_{E_i}} \right] = -r \left[1 - \frac{\sum_{i=1}^m E_i j_{E_i}}{\sum_{i=1}^m j_{E_i}} \right]$$

illustrates this. The proportional bias r in E_i is multiplied by a value which is very close to zero. This implies that proportional errors in E_i or constant errors in E_i which are not large will have little effect on the estimates of the isotopic weight percents at the feed or product parts of the reprocessing facility.

vii) The Choice of Sample Sizes

Suppose one wishes to test whether the input values to a facility are understated using differences $d_i = x_{1i} - x_{2i}$ where x_{1i} and x_{2i} are, respectively, the operator's and the inspector's values. Then one wishes to test $H_0: E(d_i) = D_0$ against $H_1: E(d_i) = D_1 < D_0$. If, on the other hand, one wishes to test whether the x_{1i} values are overstated, as for example in a facility's product, the alternative hypothesis is $H_1: E(d_i) > D_0$. Only the latter case will be considered here since the extension to the former case is obvious. One of the main problems in this area is the determination of a sample of sufficient size to induce the desired power of discrimination.

The following assumes that the variance of the differences is constant and essentially known, and the differences are independently and normally distributed. If a test of a null hypothesis $H_0: E(d_i) = D_0$ against an alternative hypothesis $H_1: E(d_i) = D_1 > D_0$ is to have α and β probabilities of type I and type II errors, the following relationship involving the sample size n

$$D_0 + T_{1-\alpha} \frac{\sigma_d}{\sqrt{n}} = D_1 + T_{\beta} \frac{\sigma_d}{\sqrt{n}} = D_1 - T_{1-\beta} \frac{\sigma_d}{\sqrt{n}}$$

must hold. Then upon solving for n ,

$$n = \sigma_d^2 \left(\frac{T_{1-\alpha} + T_{1-\beta}}{D_1 - D_0} \right)^2$$

This formula is well known. One difficulty with this approach, however, is that if there are systematic errors induced by calibration, these errors which act as biases become one of the assignable causes which are responsible for observed, significant, differences when in fact their genesis lies in a random error (and, thus, acceptable) situation which has been propagated into a systematic-type effect. One would like to distinguish this calibration type of systematic error from that which is induced by biases (intentional or unintentional) where in contradistinction here the biases signify a departure from true values, and are thus unacceptable.

If the variance of the calibration-induced bias is known, then under controlled conditions one may say $E(b) = 0$, so that calibrations errors can vary between $0 - T_{1-\alpha/2} \cdot \sigma_b$ to $0 + T_{1-\alpha/2} \sigma_b$ by chance alone. When a t test indicates significance it is desirable that the test detects an effect which is beyond the systematic errors induced by calibration. One approach here is to estimate

σ_d^2 , the random error component by (3.1). Then σ_b^2 is obtained from other sources. Set

$$D_0 + T_{1-\alpha} \sqrt{\frac{\sigma_d^2}{n} + \sigma_b^2} = D_1 - T_{1-\beta} \sqrt{\frac{\sigma_d^2}{n} + \sigma_b^2}$$

so that

$$n = \frac{\sigma_d^2}{\left(\frac{D_1 - D_0}{T_{1-\alpha} + T_{1-\beta}} \right)^2 - \sigma_b^2}$$

This formula is valid if $|D_1 - D_0| > \sigma_b (T_{1-\alpha} + T_{1-\beta})$.

The α and β risks here are those risks which obtain under many recalibrations over many periods. For a period in which the systematic error is constant, one cannot construct a variance estimate out of one observation. For an individual period then the risks will, in general, be greater or less than α and β . The formula also indicates that when systematic error is present, the ultimate power which the test can have is limited by the systematic error component. If the alternative hypothesis is too close to the null hypothesis when systematic error is present it is simply not possible to obtain small α and β values.

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13. Accompanying Papers Prepared for this Panel

- a. "Experience with the Use of Isotopic Correlation Techniques to Verify the Plutonium Content of Spent Reactor Fuels", D. E. Christensen and R. A. Schneider.
- b. "Application of Isotopic Correlation Safeguards Techniques to Verify the Plutonium Content of Humboldt Bay Spent Fuel", D. E. Christensen.
- c. "Use of Isotopic Composition Data to Improve Reactor Calculation Techniques", N. E. Carter and D. L. Prezbindowski.
- d. "Theoretical Viewpoint of Present and Future Applications of Isotopic Correlations to Safeguards", D. L. Prezbindowski.
- e. "Use of a Simplified Analytical Model to Determine Reasons for Constancy of the Ratio $\text{Pu/U/U}^{235}\text{D}$ ", D. L. Prezbindowski.