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Isotope Correlations for Safeguards Surveillance
and Accountancy Methods*

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Isotope Correlations for Safeguards Surveillance and Accountancy Methods

Isotope correlations corroborated by experiments, coupled with measurement methods for nuclear material in the fuel cycle have the potential as a safeguards surveillance and accountancy system. The ICT allows the verification of: fabricator's uranium and plutonium content specifications, shipper/receiver differences between fabricator output and reactor input, reactor plant inventory changes, reprocessing batch specifications and shipper/receiver differences between reactor output and reprocessing plant input. The investigation indicates that there exist predictable functional relationships (i.e. correlations) between isotopic concentrations over a range of burnup. Several cross-correlations serve to establish the initial fuel assembly-averaged compositions. The selection of the more effective correlations will depend not only on the level of reliability of ICT for verification, but also on the capability, accuracy and difficulty of developing measurement methods. The propagation of measurement errors through the correlations have been examined to identify the sensitivity of the isotope correlations to measurement errors, and to establish criteria for measurement accuracy in the development and selection of measurement methods.

1. INTRODUCTION

The production and depletion of plutonium and uranium isotopic composition in a fuel assembly with burnup are a measure of the irradiation history of the fuel in a power reactor. The feasibility of utilizing fission product and heavy element isotopic ratios has been explored by many investigators [1-5] for verifying the burnup of fissile material input to a reprocessing plant. Throughout the major portion of the fuel cycle, the fissile surveillance and accountability measures are basically in the form of item accountability (fuel powder containers, pellets, elements, and assemblies). The isotopic measurement of the fuel assemblies input to the reprocessing phase of the fuel cycle is the primary direct determination of whether an anomaly exists in the fuel management of nuclear material. The nuclear materials accountancy gap (4 or more years) which exists between the fabrication plant output and the input to the reprocessing plant can be minimized by using isotope correlation

techniques (ICT) at the dissolver stage of the reprocessing plant. The ICT would allow the verification of: fabricator's uranium and plutonium content specifications, shipper/receiver differences between fabricator output and reactor input, reactor plant inventory changes, reprocessing batch specifications and shipper/receiver differences between reactor output and reprocessing plant input.

2. ICSAM ANALYTICAL DEVELOPMENT OF ICOTOPE CORRELATIONS

Experimental data on LWR reprocessed fuel indicate that there exist predictable functional relationships between isotopic concentrations over a range of reactor operating conditions and burnup. The validity of extrapolating the correlation functions much beyond the range of the burnup of the current data bank is uncertain and should be established by computation and confirmed with precisely tailored experiments. In part, preliminary efforts in this direction have been initiated in Europe [6-10]. This paper presents the US/DOE/OSS isotope correlation for surveillance and accountancy methods (ICSAM) program to extend the applicability of ICT beyond this limitation for an operating LWR fuel cycle. The ICSAM program has been structured into three phases: (a) the analytical development of ICT for actual power reactor fuel cycles, (b) the development of a dedicated portable ICT minicomputer system for in-field implementation, and (c) the experimental program for measurement of U,Pu isotopics in representative spent fuel-rods of the initial 3 or 4 burnup cycles of the Commonwealth Edison Zion 1 and 2 PWR power plants [11]. The ICSAM experimental program is tailored to establish the computation methods and the applicability of measurement methods to ICT in terms of computation and measurement error propagation.

The selection of the more safeguards effective correlations will depend not only on the level of reliability of ICT for verification, but also on the capability and difficulty of developing measurement methods. The sensitivity of ICT analysis to measurement errors is analyzed in order to identify limitations of the technique within the present measurement capabilities, and to identify measurement improvements that would increase the sensitivity of this technique.

2.1. Reactor System

The Commonwealth Edison Zion 1 and 2 are pressurized water reactors (PWR) with an initial loading configuration of 193 assemblies comprised of three (2.25, 2.79, and 3.29 w/o ^{235}U) enrichment zones of zircalloy-4-clad fuel elements as shown in Fig. 1. The second cycle core reloading schedule consists of the removal of all but 6 enrichment zone 1 assemblies, the shuffling of zone 2 and zone 3 assemblies into the center portion of the core, and the replacement of zone 3 assemblies with fresh zone 4 3.06 w/o ^{235}U enriched fuel assemblies in the periphery of the core. This method of loading fuel assemblies is then maintained for the subsequent cycles with most of zone 2 assemblies

discharged after the 2nd cycle and with fresh fuel of enrichment 2.79 w/o being positioned in the outer core region. Selected fuel assemblies of zone 2 and zone 3 enrichments contain borosilicate burnable poison rods (BPR) for initial reactivity control and are removed from core during the normal sequence of assembly shuffling in fuel cycles 2 and 3. The fresh fuel assemblies do not contain BPR's. Consequently, over the lifetime of the reactor, the fuel assemblies are primarily those without BPR's. The reactor control is maintained by varying the concentration of the soluble boron in the coolant.

2.2. Isotope Correlation Computations

The primary computation of the isotopic concentration as a function of the Zion 2 reactor power history was performed using 2-group cross-sections and a detailed 3-D reactor model with azimuthal quarter core symmetry and axial symmetry with 8 axially segmented depletion regions. Since any particular correlation could generate different curves depending upon the type and positioning of the fuel assembly, and in order to understand the general trend and the scatter of the available measured isotopic composition data bank, the calculation for the first cycle of the Zion 2 power plant was performed with each fuel assembly as a depletion block [12]. The EPRICELL [13] code was used to generate burnup-dependent cross-sections and the PDQ-7 code [14] to compute neutron flux and perform transmutation calculations for 18 heavy-metal isotopes. The calculation was structured in 18 depletion time periods of approximately 60 days with reactor power and soluble boron concentration averaged over each period to cover the 1060-day duration of the cycle.

Of the 14 correlations proposed in the literature, the correlation ^{235}U ^{241}Pu vs. ^{239}Pu ^{240}Pu is found to be too non-linear to be useful. A partial listing of some of the correlations found effective in this study are presented in Table I including correlations involving the burnup parameter directly. It is found that for a given PWR all assemblies with a unique combination of enrichment zone and number of burnable poison rods (BPRs) generate one coincident curve. Some correlations are found to generate a single curve for assemblies of all enrichments and numbers of BPRs, e.g. correlation No. 5, ^{240}Pu vs. ^{239}Pu ($1 - ^{239}\text{Pu}$) and correlation No. 10, $(^{239}\text{Pu})^2 \times (^{240}\text{Pu})^2$ vs. ^{240}Pu . The ^{236}U vs. ^{235}U correlation shown in Fig. 2 represents the superpositioning of the correlation curves determined for assemblies of one enrichment. The significance of this result is that extrapolation to zero burnup could yield the initial enrichment of a measured depleted isotopic composition with minimum uncertainty introduced by the BPRs. It also indicates that spatial and neutron spectral effects are minimal. It is anticipated that at the end of the fuel cycle corresponding to the respective enrichment zones, the burnup of all the assemblies would cluster as in the zone 1 case but at a higher burnup level. In Fig. 3 all 8 axial segments of the 3-D calculation for each of the zone 1 assemblies generate one coincident curve

for each correlation and in this case the curve is found to be coincident with the correlation curves of each assembly where the axial segments have been homogenized over the core height. The chemical dissolution in the reprocessing operation axially integrates the isotopic concentrations over the height of the assembly.

The correlation $\text{Pu/U vs. } ^{239}\text{Pu } ^{242}\text{Pu}/(^{240}\text{Pu})^2$, Fig. 4, is found to generate a single coincident curve for the 20 axially homogenized assemblies of zone 1 but this curve deviates from the isotope correlation attained for the axial segments or equivalent pellets. This latter correlation is representative of that attained from the point-reactor model. This appears to introduce a significant limitation of the point reactor model and simplified transmutation. The deviation is found to be based primarily on the transmutation lag between the outer axial segments of the assembly and the central segments of the assembly. The experimental implication of this behavior is that the isotope correlations showing this behavior can be determined by dissolving a full assembly but not by dissolving pellets or axial segments separately.

3. SELECTION ATTRIBUTES OF PWR ASSEMBLY ISOTOPE CORRELATIONS

The correlation functions exhibiting linear behavior, or well-behaved slope, appear to be most effective for determining initial composition by extrapolating to zero burnup. Although the isotope correlations were found to be non-linear regardless of the computational model, most of the correlations did exhibit well behaved slopes to allow extrapolation. The selection attributes of isotope correlations would include the following considerations: dependence/independence on enrichment, dependence/independence on burnable poison rods, varying or slightly varying slope, sensitivity/insensitivity to composition anomalies, sensitivity/insensitivity to measurement errors.

3.1. Sensitivity of ICT to Anomalies in Nuclear Material Flow

The sensitivity of the ICT to composition anomalies in an enrichment zone 1 assembly was studied by simulating a substitution or dilution equivalence of one unirradiated or natural uranium assembly for a spent fuel assembly in a six-assembly dissolver batch. The dilution may represent the partial interchange of fuel pins in the assemblies. The dissolver composition point in the ^{235}U vs. ^{236}U correlation is presented in Fig. 5. The results indicate that in the case of the unirradiated assembly substitution, the dissolver composition would be superposed on the curve but displaced along the curve to a lower burnup position. The consequence of this is that this correlation is not sensitive to an anomaly where declared or recorded burnup is understated. However, an anomaly or outlier in the correlation effected by a natural uranium substitution appears to be detectable. Most correlations were found to be sensitive to this type of substitution.

3.2. Sensitivity of ICT to Measurement Errors

The sensitivity of the correlations to the propagation of isotopic measurement errors was investigated for errors at the 2% level and were assumed to apply to each of the isotopes. The effect of measurement errors are included in Fig. 5 and shows that the resolution is adequate to discern a one-sixth natural uranium substitution. This result was found to hold for all the correlations sensitive to anomalies in the nuclear material content. As discussed in the ICT study for LMFBR fuel cycles [15-17], the investigation of the effect of measurement errors on correlations serves two purposes: 1) It identifies the correlations more insensitive to measurement errors and as such could be used in optimizing the set of functions for cross-correlations, and 2) It establishes measurement methods design criteria for development and selection of measuring instruments.

4. GRAVIMETRIC METHOD

The verification of the quantitative estimate of the plutonium input to the reprocessing plant obtained by the Pu/U or Pu/U+Pu ratio method (Gravimetric Method) is not sufficient to detect certain anomalies in the nuclear material flow. The gravimetric method must be supplemented by the ICT to establish the integrity of the batch as planned for reprocessing, and the consistency between the Pu/U ratio and burnup.

The gravimetric method serves only as a reference measurement of the plutonium input to the reprocessing plant against which subsequent material accountancy measures can be compared for material balance throughout the remainder of the reprocessing plant. In Table II are listed the isotopic concentrations and the relevant Pu/U and burnup data for the case of the one-sixth unirradiated fuel dilution of a reprocessing batch. The one-sixth dilution column represents a misstatement of composition in a manner which would give the appearance of consistency between the isotopic concentrations, the Pu/U ratio and the burnup. In the last column are listed measured data independently verified. The data indicate that the consistency between the Pu/U ratio and actual burnup can be established only by the relative plutonium isotopic concentrations. The Pu/U ratio alone does not appear to verify the actual burnup even if the uranium isotopics are measured with great accuracy in the dissolution tank, as indicated by the example scenario depicted in Fig. 5. The Pu/U ratio may be tampered by changing the plutonium content of the batch and subsequently reporting the burnup inferred from the altered Pu/U instead of the actual burnup. The integrity of the reprocessing batch may be verified by utilizing the plutonium isotopic concentrations to determine the actual burnup consistent with these isotopics and then estimating the Pu/U ratio directly from the correlation between the Pu/U ratio and burnup. This correlation has been found to be well behaved, i.e., almost linear. The PWR ICT studies has identified a linear correlation of the function $(Pu+U)/U_0$ vs. burnup, with U_0 being the initial

uranium content in the fresh assembly as shown in Fig. 6. This correlation is found to be independent of initial enrichment and the number of burnable poison rods. This suggests that a more effective measure of the plutonium content in the batch solution may be obtained by the relationship

$$Pu = \left(\frac{Pu}{U+Pu} \right)_{\text{meas.}} \times \left(\frac{Pu+U}{U_0} \right)_{\text{correl.}} \times U_0,$$

where the $(Pu+U)/U_0$ is determined from the correlation with burnup which in turn is established from correlations based on the plutonium isotopic concentrations.

5. PORTABLE ICT COMPUTER

In order to verify the consistency among initial enrichment, discharge burnup, Pu/U or $Pu/U+Pu$ ratio, and uranium-plutonium isotopic composition, and in effect verify a material balance, a preliminary algorithm of cross-correlation (using measured U, Pu isotopic concentrations in different correlations) has been programmed for the portable ICT computer as a completely self-contained system for in-field implementation by a safeguard inspector.

5.1. Procedure for Utilizing ICT

An overall scheme for utilizing ICT would involve the following requirements: 1) Reprocessing Input Data: reactor system identification (class or type, output power), assembly design specifications, fabricator's initial measure of uranium/plutonium quantity and isotopic composition, assembly identification of batch, in reactor positioning sequence and related dates of operations, reactor operator's statement of burnup, and reactor operator's statement of uranium/plutonium isotopic content. 2) Isotope Correlation Data: establish isotope correlation data banks, historic data correlations for reactor class, calculated data correlations for reactor class, establish consistency of correlations between historical and calculational data for specific reactor class, input correlation data into ICT portable computer system, and input assembly specifications data into ICT portable computer system. 3) Reprocessing Plant Data: measured uranium and plutonium elemental and isotopic concentrations per liter of dissolver solution. 4) Data Interpretation using the ICT Portable Computer system: generating the relevant quantities Pu/U ratio and isotopics on a w/o basis from input data, applying consistency algorithms to identify existence of anomalies, using algorithms designed to emphasize correlations sensitive to variations from planned fuel management programs, and programming computer output summaries at several levels of detail for in-field assessment.

5.2. Isotope Cross-Correlation Algorithm

The objectives of an isotope cross-correlation algorithm are 1) To determine burnup and initial enrichment of the reprocessing

batch using the U and Pu isotope concentration measurements and isotope correlations for verification of reported burnup and initial enrichment; 2) To establish that the measured U and Pu isotope concentrations define a point on the isotope correlation curves for verification of the absence of anomalies in the batch, in the internal consistency of the measured concentrations, and in the Pu/U ratio; 3) To use the measurement errors in the isotope concentrations and the variances in the correlations obtained from the computations/data bank for establishing a level of confidence in objectives 1 and 2; and 4) To effect the cross-correlation on a completely self-contained portable ICT computer for in-field near-real-time implementation and assessment.

A preliminary ICT cross-correlation algorithm involves the following procedures: 1) Use correlation 3 and measured value of ^{236}U , determine a value for ^{235}U . Call this ^{235}U value its outlier; 2) Use correlation 8 and the measured value of ^{235}U , determine a ^{240}Pu value. Use correlation 8 and the outlier value of ^{235}U , determine another value of ^{240}Pu . The outlier of ^{240}Pu is that value which is farther from its measured value; 3) Use correlation 1 and the two values of ^{235}U (measured and outlier), determine two values of Pu/U. The outlier of Pu/U is that value which is farther from its measured value; 4) Use correlation 4 and the two values of ^{235}U , determine the outlier value of ^{239}Pu ; 5) Use correlations 12, 13, 14, and Pu/U, ^{240}Pu (measured values and then outlier values) and solve simultaneously to determine ^{239}Pu , ^{241}Pu , ^{242}Pu outlier values; 6) Use correlations 12, 13, 14, and Pu/U, ^{239}Pu (measured values and then outlier values) and solve simultaneously to determine ^{240}Pu , ^{241}Pu , ^{242}Pu outlier values; 7) Use correlation 12, and ^{239}Pu , ^{240}Pu , ^{242}Pu (measured values and then outlier values), determine Pu/U ratio; and 8) Use correlation 14, and ^{240}Pu , ^{241}Pu , ^{242}Pu (measured values and then outlier values), determine Pu/U ratio. The ICT computer output format is listed in Table III. For each section of Table III, the isotopic concentrations listed are: the undiluted (i.e. data bank value), the measured value (independently verified), and the value determined from measurement/outlier data and isotopic cross-correlations. In the absence of an anomaly in the material flow, the measured and outlier values should be consistently in agreement.

The preceding algorithm is just a representative sample of the algorithms which could be designed to emphasize those cross-correlations that are sensitive to many expected operationally (reactor plant and reprocessing plant) dependent variations from the planned fuel management programs.

The ICT portable computer outputs have been designed to print out summaries at several levels of detail (depending on the information the inspector may want) for in-field assessment of the batch integrity. The data outlined in Tables II and III including the summary statement would be two such levels of detail.

6. ICSAM EXPERIMENTAL PROGRAM

The ICSAM experimental program objectives are to:

- 1) Establish computation methods in isotope production and depletion of an operating power reactor over 4 power cycles;
- 2) Establish a sound technical base for isotopic correlation functions over actual operating range of conditions and burnup cycles; and 3) Establish the applicability of measurement methods to ICT which have a near-real-time basis and adaptable for independent verification measurements.

7. SUMMARY STATEMENT

The current ICT study of the Commonwealth Edison Zion 2 PWR [12] and the reference LMFBR system, [15-17] indicates that the point reactor model with simplified transmutation equations does not reflect the full assembly isotopics obtained from the detailed three-dimensional (3-D) computation model. The detailed 3-D reactor model, with each fuel assembly a depletion block, indicates that the isotope correlations for the full assembly (axially homogenized) deviates from the isotope correlations attained for pellets or axial segments. The deviation is found to be based primarily on the transmutation lag between the outer axial segments of the assembly and the central sections of the assembly.

Some of the experimental data in the current data bank being used in ICT studies have been based on small sample sizes (pellets) and consequently explain some of the discrepancy and data spread. There is agreement between computations and reprocessing data regarding the curvature (non-linearity) of the correlations.

The ICT is found to be sensitive to anomalies in nuclear material flow resulting from one-sixth substitution or dilution equivalence. The propagation of measurement errors on the correlations and the respective sensitivities to isotopic composition anomalies have been examined and found to be consistent with current measurement methods. There may be a limited safeguards usefulness of the current gravimetric method. However, a modification of the current gravimetric method has the potential of establishing the plutonium content in the dissolver solution consistent with the actual burnup. It normalizes the reprocessing plant input/output to the external fuel cycle facilities, i.e., fuel fabrication and power reactor. The gravimetric method should be supplemented by the ICT to establish the integrity of the planned reprocessing batch and the consistency between the Pu/U ratio and burnup. The significance of this study is the reaffirmation that the ICT can be developed into the most effective safeguards technique that is currently available for the overall nuclear fuel cycle.

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ZONE	ENRICHMENT W/O	NO. OF ASSEMBLIES	NO. OF ASSEMBLIES IN 1/4 CORE
1	2.248	65	20
2	2.789	64	19
3	3.292	64	17

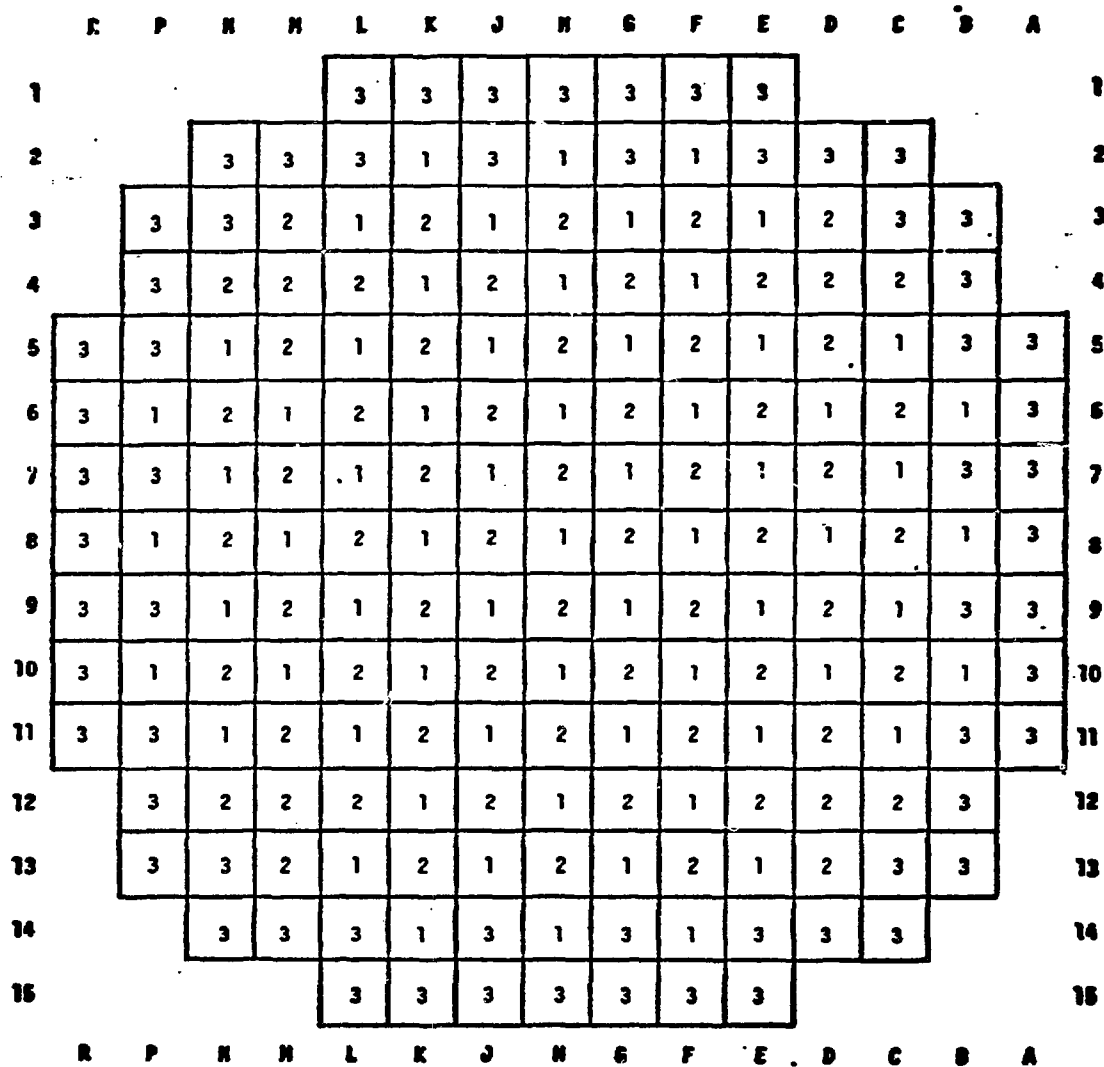


FIG. 1. Fuel Assemblies in Cycle 1 of ZION 2

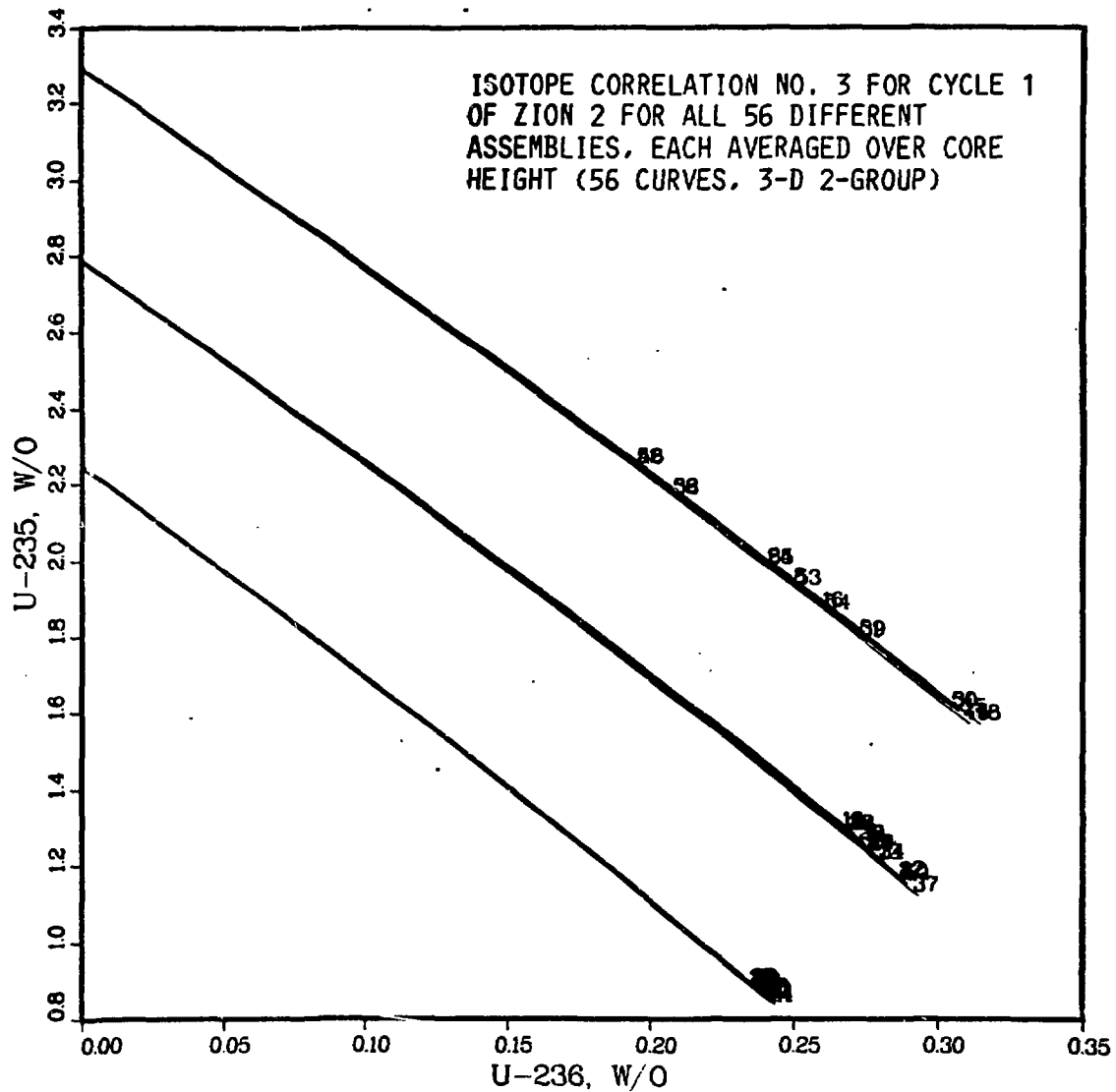
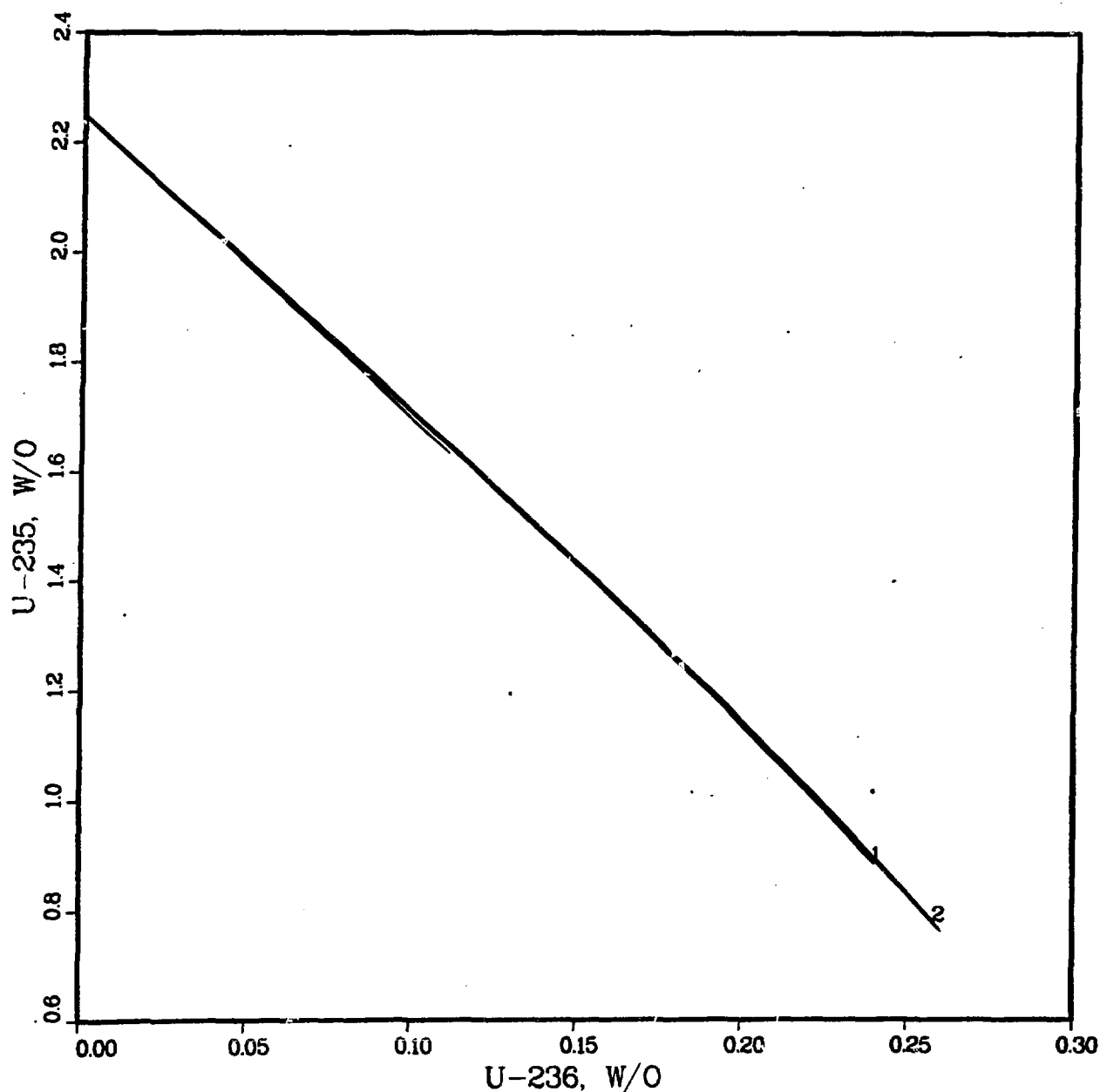


FIG. 2. Isotope Correlation No. 3 for Cycle 1 of ZION 2

ISOTOPE CORRELATION NO. 3 FOR CYCLE 1 OF ZION 2
FOR ALL AXIAL SEGMENTS OF ALL ZONE 1 ASSEMBLIES
(180 CURVES, 3-D 2-GROUP CALCULATION)



- 1 = EACH ASSEMBLY HOMOGENIZED OVER CORE HEIGHT (20 CURVES)
2 = THE 8 AXIAL SEGMENTS OF EACH ASSEMBLY (160 CURVES)

FIG. 3. Isotope Correlation No. 3 for Cycle 1 of ZION 2

ISOTOPE CORRELATION NO. 12 FOR CYCLE 1 OF ZION 2
FOR ALL AXIAL SEGMENTS OF ALL ZONE 1 ASSEMBLIES
(180 CURVES, 3-D 2-GROUP CALCULATION)

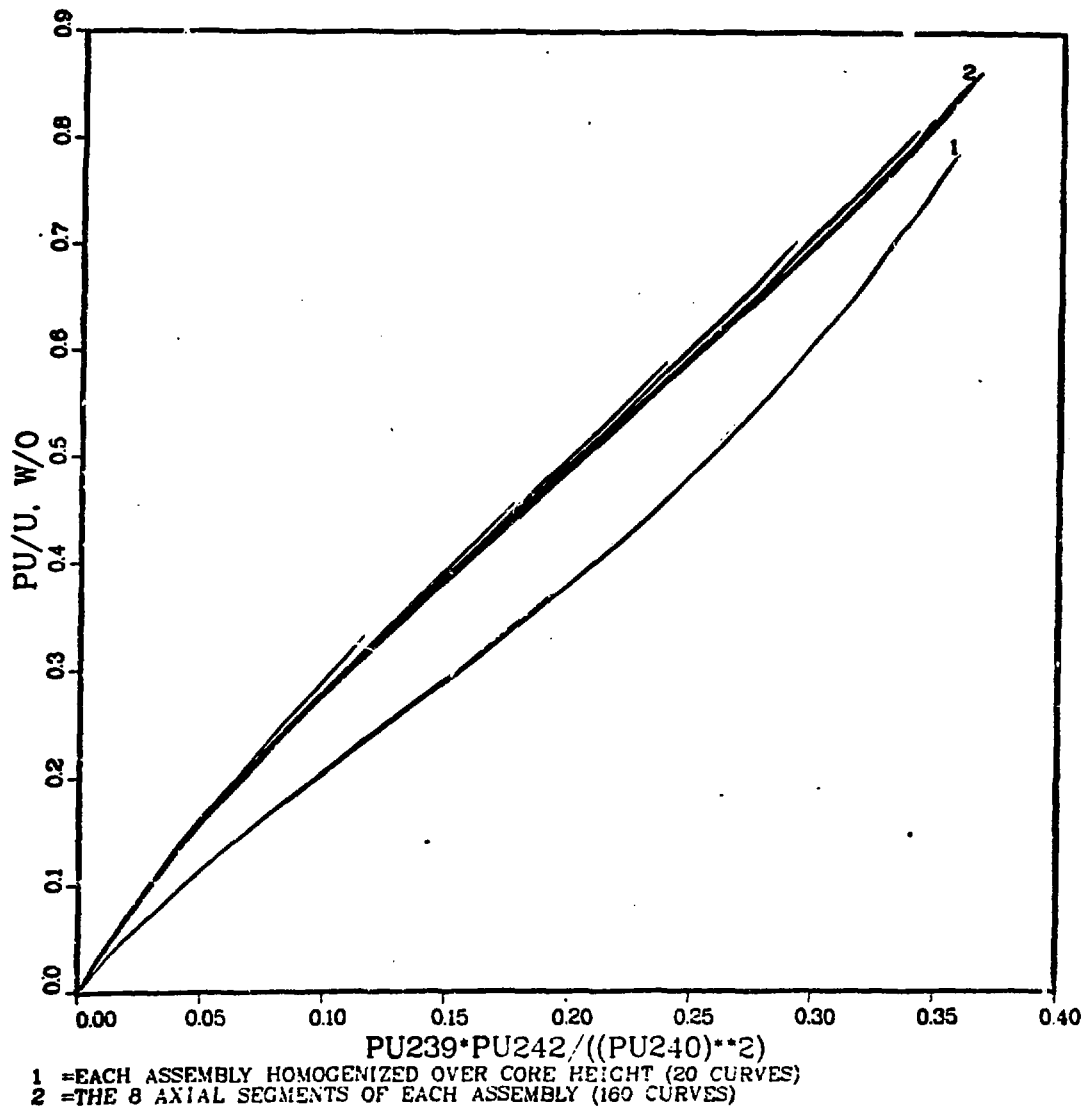
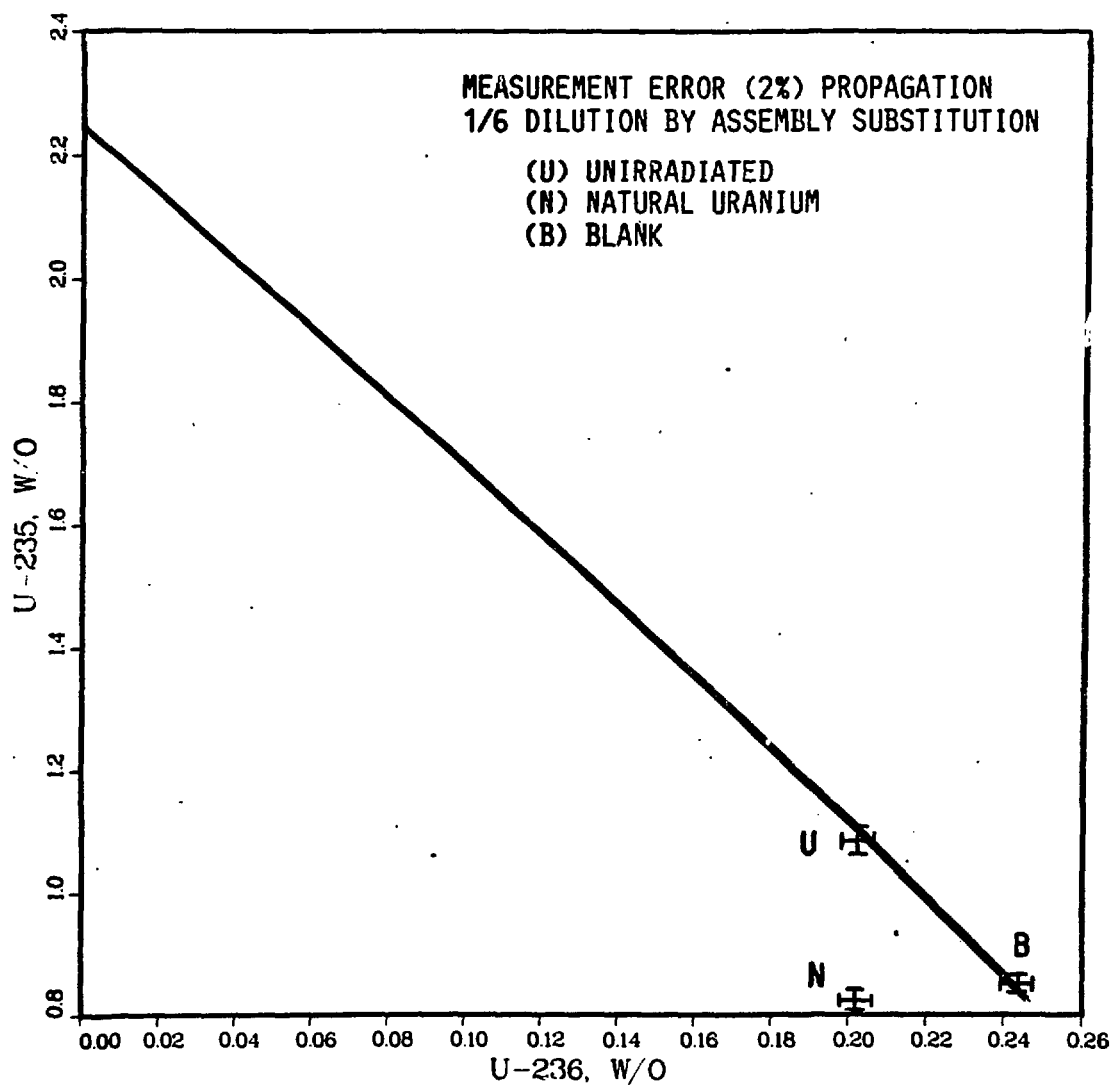


FIG. 4. Isotope Correlation No. 12 for Cycle 1 of ZION 2



	X	% DIFF.	Y	% DIFF.
NO SUBST	2.430×10^{-1}		8.463×10^{-1}	
UNIRR SUB	2.016×10^{-1}	-1.704×10^1	1.085×10^0	2.822×10^1
NATUR SUB	2.016×10^{-1}	-1.704×10^1	8.234×10^{-1}	-2.704×10^0
BLANK SUB	2.430×10^{-1}	0.000	8.463×10^{-1}	0.000

FIG. 5. Measurement Error (2%) Propagation

BURNUP CORRELATION NO. 17 FOR CYCLE 1 OF ZION 2
 FOR ALL AXIAL SEGMENTS OF ALL ZONE 1 ASSEMBLIES
 (180 CURVES, 3-D 2-GROUP CALCULATION)

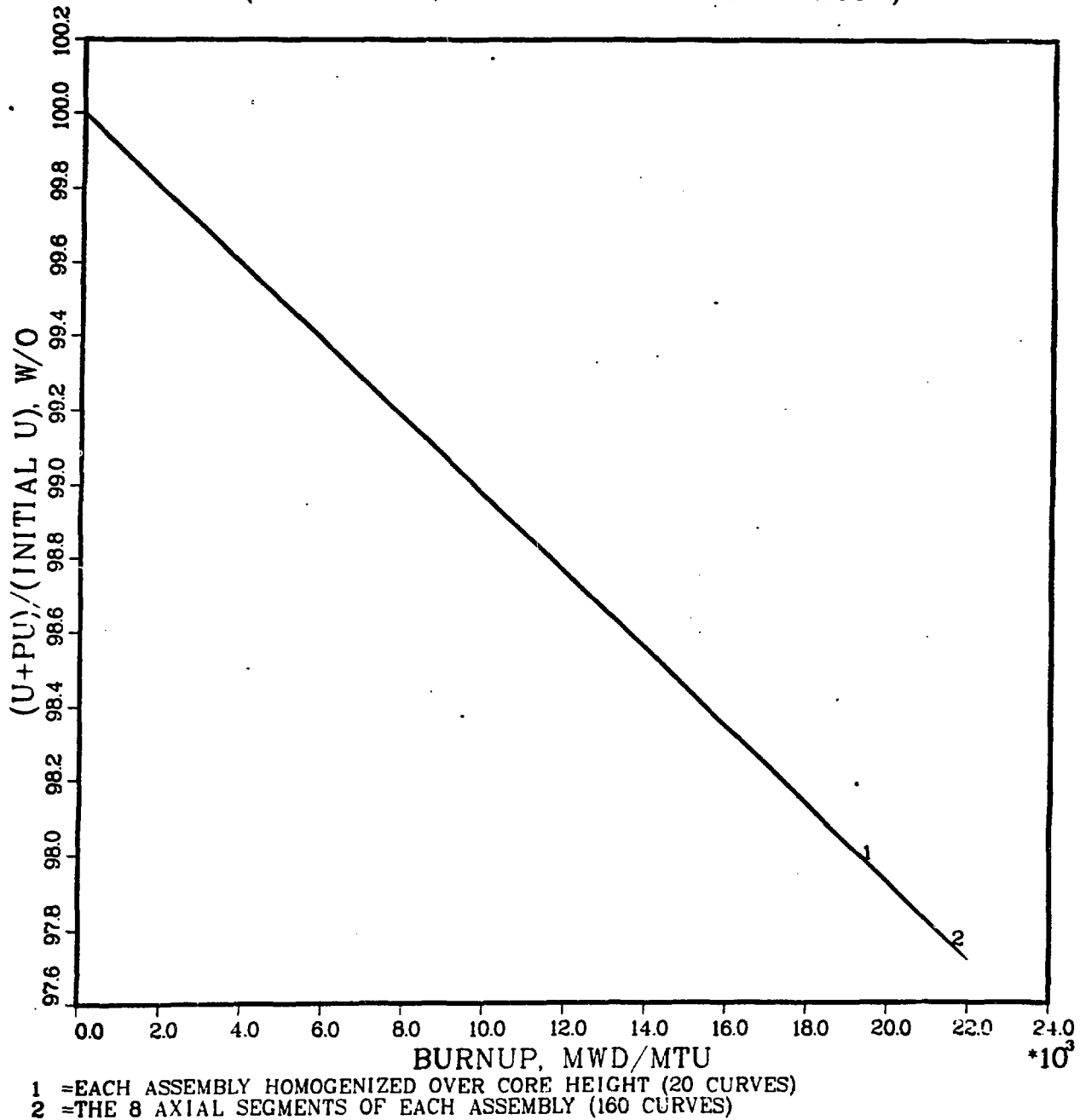


FIG. 6. Burnup Correlation No. 17 for Cycle 1 of ZION 2

TABLE I
TYPICAL ISOTOPE CORRELATIONS
UNITS OF WEIGHT FRACTION

CORRELATION
NUMBER

3. ^{236}U vs ^{235}U
4. ^{235}U vs $(^{239}\text{Pu})^2$
5. ^{239}Pu (1 - ^{239}Pu) vs ^{240}Pu
8. DEP [$(^{235}\text{U})^2$] vs ^{240}Pu
9. (1 - ^{239}Pu) vs $(^{239}\text{Pu})^2$ (1 - ^{239}Pu) / $(^{235}\text{U})^2$
12. ^{239}Pu , $^{242}\text{Pu}/(^{240}\text{Pu})^2$ vs Pu/U
14. $(^{241}\text{Pu} + ^{242}\text{Pu})/^{240}\text{Pu}$ vs Pu/U
17. BURNUP VS (U + Pu)/U
22. BURNUP VS Pu/U
24. BURNUP VS ^{236}U
25. BURNUP VS ^{239}Pu
26. BURNUP VS ^{240}Pu

TABLE II

EXAMPLE SCENARIO TO DEMONSTRATE THE USE OF ICT TO DETECT ANOMALIES
IN SNM FOR SAFEGUARDS: SUBSTITUTE ONE ASSEMBLY IN SIX WITH
UNIRRADIATED FUEL (OR ONE ROD IN SIX) AND PREDICT PU,U ISOTOPES IN
UNDILUTED AND DILUTED ZONE I SPENT FUEL SOLUTION.

ISOTOPICS/ ELEMENTAL/ BURNUP	NO SUBSTITUTION PREDICTED DATA	1/6 SUBSTITUTION PREDICTED DATA	% DIFFERENCE	SOLUTION MEASURED DATA
U-235	0.889%	1.12%	+25.9	1.12%
U-236	0.24	0.20	-16.7	0.20
PU-239	63.03	69.3	+ 9.9	63.03
PU-240	22.61	19.53	-13.6	22.61
PU-241	11.07	9.16	-17.3	11.07
PU-242	2.89	1.72	-40.4	2.89
PU/U	7838 G/TON	6501 G/TON	-17.1	6501 G/TON
BURNUP (MWD/T)	19420	14420	-25.7	-

CONCLUSION: FROM SOLUTION MEASURED DATA AND ICT:

PU ISOTOPICS ARE INCONSISTENT WITH PU/U, BURNUP, AND U ISOTOPICS

AN ALGORITHM AND COMPUTER OUTPUT

CORRELATION OF ISOTOPES FROM MEASURED VALUES
FIRST VALUE IS UNDILUTED, SECOND IS MEASURED, THIRD IS FARTHEST OUTLIER

U-235 (1) .8886
1.1205
1.1432

PU-240 (2)	22.6105	PU/U (3)	7838.	PU-239 (4)	63.0271
	22.6105		6501.		63.0271
	19.5323		6637.		69.3213
FROM U-235(1)		FROM U-235(1)		FROM U-235(1)	
CORRELATION 8		CORRELATION 1		CORRELATION 4	

PU-242 (7)	2.8922	PU-242 (10)	2.8922
	2.8922		2.8922
	1.7276		1.7229

PU-242 (14)	2.8922	PU-242 (15)	2.8922
	2.8922		2.8922
	1.7329		1.7290
FROM PU-240(2), U-235(1),		FROM PU-240(2), U-235(1),	
&PU-241(6) CORRELATION 7		&PU-241(9) CORRELATION 7	

MEASURED PU/U: 6501.

U-236	PU-239	PU-240	PU-241	PU-242	PU-240&PU-241
14421.	19420.	19420.	19420.	19420.	19420.
CORRELATION 24	CORRELATION 25	CORRELATION 26	CORRELATION 27	CORRELATION 28	CORRELATION 29

SUMMARY STATEMENT: FROM SOLUTION MEASURED DATA AND ICT, PU ISOTOPICS ARE INCONSISTENT WITH PU/U, U ISOTOPICS, AND BURNUP