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USE OF ISOTOPIC COMPOSITION DATA TO IMPROVE
REACTOR CALCULATIONAL TECHNIQUES

CONTRACT NUMBER ACDA/WEC-216

PREPARED FOR
U. S. ARMS CONTROL AND DISARMAMENT AGENCY

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MARCH 20, 1972

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Presented to the IAEA Panel on the Use of Isotopic Composition Data in Safeguards at Vienna, Austria, April 10-14, 1972

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ABSTRACT

Before the availability of isotopic data from chemical reprocessing of spent nuclear fuels, burnup code normalization was done using data from small burnup samples. Chemical reprocessing plant isotopic data are an additional source of burnup data reliably measured to permit normalization of calculational code processes to real data. By applying isotopic correlation techniques to this source of data and to data from burnup calculations, it appears that procedures are available a). to detect code input errors, b). to detect calculational biases in both U and Pu isotopes and c). to diagnose calculational weaknesses. A means whereby the maximum use of dissolution data is realized is given.

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I. INTRODUCTION

Collection of measured isotopic data and analysis of the data to observe isotopic relationships are important means of providing independent verification of the measured composition of spent reactor fuel. The techniques are an example of maximum utilization of isotopic data that are presently being measured at various portions of the nuclear fuel cycle. Analyzed collectively, isotopic data provide a system of independent checks and balances to detect or deter diversion of nuclear materials.

The techniques can be useful in meeting the needs of the reactor operator as well. Some of the needs of the reactor operator in the area of isotopic composition data are as follows:

- a) to know the measured "as built" isotopic concentrations of fabricated fuel assemblies,
- b) to calculate the final isotopic content of irradiated fuel assemblies
- c) to ascertain the accuracy of the isotopic measurements of the reprocessed spent fuel, and

d) to ascertain the accuracy of the calculated isotopic concentrations.

The knowledge of the measured "as built" isotopic concentrations of fabricated fuel assemblies is important to the reactor operator for a number of reasons; for example, they serve as input to the burnup codes used in calculating the final fuel content.

The reactor operator who is attempting to improve calculated final isotopic fuel content and to verify measured values will find an evaluated collection of measured data useful. The more extensive the collection, the better the statistics of the collection become. Several qualities peculiar to the measured isotopic data from spent fuel dissolver batches makes them of particular use to the reactor operator. First, the data are a volumetric average of the isotopic concentrations throughout the fuel assemblies included in the dissolver batch. The volumetric averaged data are especially useful to supplement small sample measured data since small sample data are sensitive to the local variations in the neutronic environment in a fuel assembly. The sensitivity of the small sample data to local neutronic variations causes a scatter in the data which somewhat obscures the general transmutation or burnup paths representative of the fuel assembly as a whole that the reactor operator seeks to iden-

tify. Secondly, the accuracy of the measurements of the batch dissolver data is generally better than that of the small sample data because of the large amount of fuel dissolved in a batch.

The result from the above two qualities is that isotopic data measured at a chemical reprocessing plant evidence well defined burnup paths. In addition, isotopic ratios including those which are invariant with exposure and with other reactor characteristics provide a means of detecting errors in both calculated and measured data alike.

The purpose of this paper is to discuss the ways in which a reactor operator could improve his prediction of reactor core inventory by judicious use of the isotopic composition data provided by the reprocessing plant. Examples illustrating the ideas using calculated data are given. A large part of the data presented were submitted by Yankee Atomic Electric Company (YAEC) to Nuclear Fuels Services, Inc., as part of a statement of material to be processed for Yankee Rowe core V⁽¹⁾ and use of these calculated data are hereby acknowledged.

II. EXAMPLES OF CALCULATIONAL DATA APPLIED TO ISOTOPIC CORRELATIONS

The reactor operator normally includes gram quantities of the individual isotopes as part of the output of his calculational scheme. By addition to his calculational computer codes of a limited number of computer state-

ments concerned with program output, gram quantities of each fuel isotope can be transformed into easily observed isotopic relationships as a normal part of the computer output. With the relationships readily available, the following advantages will result:

- First, the calculated relationship values serve as a check on the calculations to detect input errors.
- With the calculated values of the relationships available for examination, additional understanding and insight into the neutronic and calculational processes affecting the relationships are gained. The insight gained can be confirmed by actual chemical reprocessing plant measurement by requesting an appropriate fuel assembly dissolution order. This can be facilitated by deciding on a given order before the assemblies are shipped to a reprocessing plant (and thereby avoid fuel shuffling in storage areas).
- Once the measured isotopic data are available from the reprocessing plant, comparisons of calculated and measured relationship values can be made. Detailed comparisons can include isotope by isotope comparisons with the possible result that cross section and physics model deficiencies could be isolated and corrected to improve the accuracy of the subsequent calculations of the content of spent fuel assemblies.

By applying the concept, the reactor operator performs much the same type of isotopic correlations as used for safeguards but the techniques are adjusted to his needs.

A. Detection of Input Errors

To illustrate the above suggestions, calculated values of $(\text{Pu}/\text{U})/^{235}\text{D}$ for Yankee Rowe core V fuel assemblies are shown in Table I. These values were determined from reactor code generated data submitted as part of a statement of materials to be processed. The dissolution of Yankee Rowe cores V and VI should be considered a special case rather than a routine dissolution campaign since special care was taken during dissolution to maintain the physical identity of fuel assemblies in each batch of dissolved fuel until accountability samples were obtained from the accountability tank. However, the principles to be explained can be applied to all cores.

The first column of Table I lists the assemblies in the same order that they were tabulated by YAEC with the calculated $(\text{Pu}/\text{U})/^{235}\text{D}$ values for each in the second column. All assemblies except the last two were of the same initial enrichment, 4.1 weight percent ^{235}U , and were stainless steel clad.⁽¹⁾ The calculated exposure for the 4.1 fuel assemblies ranged from 9,697 to 21,442 MWD/MTU* while the $(\text{Pu}/\text{U})/^{235}\text{D}$ calculated values varied from 5228 to 5496. Some of the 4.1 assemblies had different irradiation histories and these

* Megawatt Days per Metric Ton Uranium

TABLE I. SUMMARY OF (Pu/U)/²³⁵D FOR YANKEE ROWE CORE V ASSEMBLIES

Assembly No. 1	$\frac{\text{Pu/U}}{^{235}\text{D}}$ 2	Assembly No. 3	$\frac{\text{Pu/U}}{^{235}\text{D}}$ 4	Batch No. 5	$\frac{\text{Pu/U}}{^{235}\text{D}}$	
					6*	7*
A 156	5262	A 158**	5228	1	5228	5375
A 157	5496	A 162**	5228		-2.7%	
A 158**	5228	A 263-C	3736	2	3736	5765
A 160	5253	A 264-C	3736		-35%	
A 161	5301	A 182	5347	20	5347	5447
A 162**	5228	A 184	5347		-1.8%	
A 166	5314	B 197	5347			
A 167***	5240	B 216	5347			
A 169***	5240	A 166	5315	6	5304	5446
A 173	5253	A 186	5315		-2.6%	
A 174	5292	B 193	5315	10	5304	5416
A 182	5347	B 219	5315		-2.1%	
A 183	5262	B 192	5292			
A 184	5346	B 221	5292			
A 186	5315	A 174	5292	6		
A 189	5496	A 226	5292			
B 192	5292	A 161	5301	8	5273	5471
B 193	5315	A 225	5270		-3.6%	
B 194	5258	B 210	5270	14	5266	5478
B 195	5258	B 222	5270		-3.9%	
B 196	5252	B 217	5262			
B 197	5347	B 220	5262			
B 199	5496	A 156	5262	8		
B 200	5252	A 183	5262			
B 201***	5240	A 160	5253	4	5373	5431
B 204***	5240	A 173	5253		-1.1%	
B 210	5270	B 196	5252	12	5373	5378
B 214	5496	B 200	5252		-0.1%	
B 216	5347	B 199	5496			
B 217	5262	B 214	5496			
B 219	5315	A 157	5496	4		
B 220	5262	A 187	5496			
B 221	5292	B 194	5258	22	5258	5440
B 222	5270	B 195	5258		-3.3%	
A 225	5270	A 167***	5240	18	5240	5375
B 226	5292	A 169***	5240		-2.5%	
A 263-C	3736	B 201***	5240			
A 264-C	3736	B 204***	5240			

* Calculated values are in column 6 and measured values are in column 7.

** Two assemblies discharged with core IV, August 1965.

*** Four assemblies charged November, 1963. Assemblies A263-C and A264-C charged November, 1965, and all others were charged September, 1964. All these assemblies were discharged October, 1966.

differences are indicated in the footnotes to Table I. The last two assemblies listed in the first column were of an initial enrichment of 2.9 weight percent ^{235}U and were clad with Zircaloy. The calculated exposure and $(\text{Pu}/\text{U})/^{235}\text{D}$ values were 12,262 MWD/MTU and 3736, respectively.

The exceedingly low $(\text{Pu}/\text{U})/^{235}\text{D}$ values for the 2.9 assemblies were noted to be in error. The $(\text{Pu}/\text{U})/^{235}\text{D}$ value increases with decreasing initial enrichment and while the change in cladding from stainless steel to Zircaloy will cause a decrease in $(\text{Pu}/\text{U})/^{235}\text{D}$, the values for the last two assemblies were much too low. A probable cause for the low values may have been an error in calculational input for ^{235}U for these assemblies. The observation that the calculated $(\text{Pu}/\text{U})/^{235}\text{D}$ values were low was confirmed during the dissolution campaign when the two 2.9% assemblies were dissolved as a separate batch; i.e., batch #2. The measured values of $(\text{Pu}/\text{U})/^{235}\text{D}$ for this batch was 5765 which was more consistent with the rest of the fuel and theory. The calculated values were in error and would most likely have been corrected by checking the calculational input for an error. Thus, tabulating relationship values provided a ready check of calculational input.

Additional checks on the calculated results were obtained by rearranging the order of the assemblies according to the

estimated exposure. Using the order by assembly number, variations with exposure were difficult to observe. Listing the assemblies according to exposure in calculational output is a simple but effective procedure which is illustrated by columns 3 and 4 where the assembly numbers and estimated $(\text{Pu}/\text{U})/^{235}\text{D}$ values are again shown. A decreasing trend of the $(\text{Pu}/\text{U})/^{235}\text{D}$ values with increasing exposure is evident for assemblies of the same initial enrichment (4.1 wt% ^{235}U) that were discharged together, that is, starting with assembly A-182 of column 3 on down. The trend is highly consistent until four assemblies (B199, B214, A157, A187) having higher exposures also exhibit a higher $(\text{Pu}/\text{U})/^{235}\text{D}$ value, 5496. Generally, the values appear in groups of four due to quarter core symmetry considerations. As an example of gaining understanding and insight into neutronic and calculational processes affecting $(\text{Pu}/\text{U})/^{235}\text{D}$, an answer as to why these four particular values are high is of value. Is the input at fault or do the values actually represent physical and neutronic effects? A second point would be to try and verify by actual measurement at the reprocessing plant that these four assemblies had a higher value by dissolving them in one batch, if possible.

Column 5 of Table I indicates dissolution batch numbers. The assemblies that were dissolved together in batches are indicated by the horizontal lines. Although the order of the

dissolution had been fixed according to purposes of a safe-guards experiment⁽¹⁾ in the case of core V assemblies, the order was not strictly according to increasing exposure as estimated by YAE. For example, batches 4, 6 and 8, were made up of four assemblies that did not follow consecutively in column 3. Thus, the batch numbers appear in column 5 opposite sets of 2 rather than groups of 4. The calculated $(Pu/U)/^{235}D$ values for each batch are given in column 6 (the values were determined from summing U, Pu and ^{235}U values where the sums included the contributions of the individual assemblies, see reference 1).

The calculated values can be compared to the measured values which are shown in column 7 (the values are not repeated for the batch numbers that are listed twice). The percent differences between calculated and measured values for each batch are shown in column 6. It will be noted that the four assemblies in question, B-199, B-214, A-157 and A-187, were not dissolved together as one batch but were dissolved in two batches, 4 and 12, with two assemblies of the 5496 value per batch. The calculated $(Pu/U)/^{235}D$ values for these two batches was 5375 instead of 5496 due to the mixing with other assemblies of a lower value. Nevertheless, the calculated values for batches 4 and 12 were still the highest calculated values for the 4.1 fuel batches. However, the calculated increase

was not evident from the measured values. Thus, the high $(Pu/U)/^{235}D$ calculated values based on computer codes were not confirmed by measurement. It appears to be profitable to look more closely at the calculations to find an explanation for the high values thereby gaining further insight into calculational processes.

Another point of importance was that the measured values of $(Pu/U)/^{235}D$ are seen to vary less than the calculated values for all 4.1 fuel dissolution batches ranging only from 5375 to 5478. Also, the measured values for batches 4 and 12 are seen to be the lower values rather than the higher values. Again, calculational processes may be better understood through an effort to match the constancy of the measured results with calculations.

B. Detection of Calculational Biases

Comparisons of calculations of reactor core isotopic inventory versus measured inventory have demonstrated several typical biases that seem to appear in great frequency. In addition to the bias evident between measured and calculated $(Pu/U)/^{235}D$ values, several examples are given below.

1. Calculation of ^{235}U

Although the calculated total U versus measured total U has been shown to agree favorably,^(1, 2, 3) the comparisons of U isotopic values do not show the same consistency.⁽¹⁾

Listed in Table II are the calculated and measured ^{235}U values for 15 NFS campaigns. The overall bias is -0.83%. Many of these campaigns are for NPR fuel, which is somewhat different from the material typical of the commercial nuclear industry. If one looks at the campaigns of commercial reactor fuel only, the bias is -3.66%. Thus, it would appear that standard calculational techniques yield an overburn of ^{235}U , which in turn influences the calculated plutonium isotopics. A logical step to improve the agreement between calculations and measurements would be to reduce the calculational bias for ^{235}U .

2. Biases in U and Pu Isotopes

Typically a reactor operator will determine his estimated core isotopic inventory using three-dimensional computer codes such as FLARE⁽⁴⁾ or PDQ-7⁽⁵⁾. Comparisons of the calculated results to measured batch data have shown biases in isotopics, even though the total U and, in certain cases, the total Pu have agreed well with measured values⁽¹⁾. The results from a FLARE type calculation done at Battelle-Northwest are used in this section for the purpose of illustration. The biases are readily observed when calculated and measured isotopic data are plotted together as show in Figure 1. The successive calculated data points represent calculational results at various exposures for the four assemblies of batch 18 listed in Table I. In the figure, the $^{242}\text{Pu}/^{240}\text{Pu}$ isotopic ratio is shown as a function of the $^{236}\text{U}/^{235}\text{U}$ isotopic ratio.

TABLE II. SHIPPER-RECEIVER DIFFERENCES FOR ^{235}U OF FUELS
 PROCESSED AT NFS⁽³⁾

NFS Campaign Number	Shipper ^{235}U Grams	NFS ^{235}U Grams	S-R % D
1	184644	184714	-0.038
2	269088	268387	+0.26
3	386651	383260	+0.88
4*	488704	503435	-3.0
5*	1235452	1248670	-1.1
6	190583	189190	+0.58
7	193071	191590	+0.77
8	309500	313870	-1.4
9	277820	274710	+1.13
10	397000	386880	+2.6
12	356000	339570	+4.76
13*	509320	529890	-3.99
15*	140960	151650	-7.37
16*	268760	277543	-3.2
17*	244118	252143	-3.28
		σ_0 =	3.00%
		$\bar{X}$ =	-0.83%
		$ X $ =	2.29%

* Commercial Fuel

FIGURE 1. COMPARISON OF YANKEE ROWE CORE V DISSOLUTION BATCH DATA AND CORE V CALCULATED DATA

It is important to note that the measured batch data and calculated data for Yankee Rowe core V describe distinct burnup paths and that the burnup paths are significantly different. The different burnup paths are indicative of the biases between calculated and measured isotopic data.

One of the goals of a burnup calculation is to calculate accurately the isotopic content of spent fuel in addition to the totals for uranium and plutonium. Although calculations give absolute isotopic concentrations and absolute values of exposure, the chemical plant data provide absolute isotopic concentrations but exposures are not measured. However, by changing absolute measured isotopic concentrations to relative values in weight percents, these values can be used to check the relative amounts of isotopic concentrations from calculations. In other words, calculated and measured data can be compared to determine if they are on the same "burnup path" as illustrated by Figure 1.

The calculational procedure to determine isotopic composition is to first use an operations analysis code to calculate the exposure distribution of a core. The calculation is accomplished using measured and calculated core parameters such as

power level, heat transfer, fluid flow, control rod patterns, accumulated exposure, etc. Given the three dimensional exposure distribution, the differential burnup equations are solved which gives the isotopic burnup paths and the desired isotopic data either directly by a burnup code or indirectly by tabular methods based upon a burnup code. It is at this point that calculation can be compared to measurement for the purpose of obtaining agreement between the calculated and measured burnup paths.

With the calculated burnup paths matching the measured burnup paths, it is more likely that the correct isotopic compositions will result from calculations. However, as long as biases exist between measured and calculated burnup paths, the process of calculating isotopic values is limited by the uncertainty in the operational analysis code calculation and the uncertainty in the burnup code calculation. Matching the measured and calculated burnup paths leaves the uncertainty in the operations analysis code which may then be dealt with free of the uncertainty of the burnup path calculation.

III. HOW TO MAXIMIZE THE USE OF DISSOLUTION DATA

As shown in previous examples, calculational predictions can be confirmed by measured chemical reprocessing plant input data. The usefulness of these data can be increased by being selective about fuel assembly dissolution order. Fixing an optimum dissolution order, which will provide maximum measured information pertaining to core burnup calculations, before the assemblies are shipped to a reprocessing plant is a logical first step. Shipping the assemblies according to the desired order and requesting storage at the chemical plant in the same order avoids fuel shuffling at the chemical plant while assuring that the cost of specifying assembly dissolution order is very small.

The main emphasis in specifying dissolution order is not to be able to compare absolute amounts of uranium and plutonium as measured versus calculated per input batch. The dissolution batches more than likely will not represent one or several whole assemblies and their contents due to the heels left in tanks as the dissolved fuel solutions are pumped from dissolver to measuring tanks. The main emphasis is upon comparing calculated and measured relative amounts, that is isotopic weight percents, Pu/U values, and isotopic relationships such as $(\text{Pu}/\text{U})/^{235}\text{D}$. The process of comparison can be further aided by plotting data relationships to compare burnup paths as seen in Figure 1. Even though a particular batch may

not represent whole assemblies, the assemblies that it does represent can all be from similar core positions, thereby permitting batch data to be correlated to core position.

During the development of isotopic correlations, the occasion to order the dissolution sequence of spent fuel assemblies has arisen three times for different reasons. In two cases, YANKEE ROWE cores V & VI⁽¹⁾ and HUMBOLDT BAY⁽⁶⁾, the ordering was done to assure minimum mixing of different initial enrichment fuels as well as to assure a large range of exposures of the dissolution batches at input. In the third case, YANKEE ROWE core VIII (for which the measured data are not available as yet) the ordering was done to provide a reasonably uniform ²⁴⁰Pu weight percent of all input batches and thus provide a uniform isotopic composition of all product batches. This experience has shown that specifying a certain dissolution order can greatly increase the usefulness of dissolution data and that this specification of order can be accomplished at very little extra effort to the reactor operator or the reprocessor.

IV. CONCLUSIONS

Isotopic correlation techniques as developed under the safeguards program are applicable to a reactor operator and it is to his advantage to apply them. The application is strictly in the framework of theory-measurement correlations

to permit normalization of calculational code data to real data. Before the advent of spent fuel data from reprocessing, code normalization was done using data from small burnup samples. These data are rather sparse. Input batch data obtained from measurements at a chemical reprocessing plant are thus an additional source of burnup data at a very moderate cost which represent a source that is reliably measured. The batch data are particularly applicable for testing the reactor operator's burnup models because they represent a volumetric average of the burnup in the reactor core. The batch data can be used to diagnose shortcomings in the calculational methods which could then be corrected by code and cross-section improvement.

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