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ABSTRACT

Nondestructive gamma-ray analyses (NDA) for fission products and mass-spectrometric analyses for five uranium isotopes in two samples of fuel removed from the Peach Bottom reactor have been compared with corresponding quantities calculated from reactor operating data using the ORIGEN code to form the basis for material balance calculations in head-end reprocessing steps. Satisfactory agreement between ORIGEN results and measured uranium isotopic distributions was found with the standard ORIGEN inputs.

Many fission products of interest, including ^{89}Sr, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{127}Te*, ^{129}Te*, ^{131}I, ^{134}Cs, and ^{144}Ce, have half-lives that are less than about 2 years. Comparisons of ORIGEN calculations and NDA values for ^{95}Zr, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{144}Ce, and ^{154}Eu demonstrated the need to include realistic reactor operating data, including shutdown times, in the ORIGEN input parameters.

1. INTRODUCTION

Quantities of activation products (such as ^{14}C), actinides, and fission products formed in samples and fuel rods irradiated in the Peach Bottom reactor[1] may be calculated by use of the ORIGEN[2,3] code and various parameters. Nuclear data libraries used in these calculations are frequently updated[3,4] to improve the accuracies of calculated values; these may be used to provide cross checks for mass balances calculated from analytical data obtained from head-end reprocessing studies and from nondestructive gamma-ray scanning.[5] Tobias[6] has reported on some calculations for driver fuel and on the sensitivity of calculated values to the variation of several of the input parameters.

The usual calculations performed with ORIGEN are based on broad averages such as effective full-power days (EFPD). This approximation to reality is adequate for calculating reactor core inventories for nuclides with relatively long half-lives. However, for specific small fuel samples and for many nuclides of interest[5] [(e.g., ^{89}Sr, ^{91}Y, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{127}Te*(109d), ^{129}Te*(33.4d), ^{131}I, ^{134}Cs, and ^{144}Ce)], inventories calculated on the basis of EFPD will be biased with respect to true inventories because radioactive decay in the time corresponding to the difference between true calendar days and EFPD is neglected. This bias can be eliminated by use of irradiation data, including actual irradiation times and power levels during operating intervals and shutdown-time intervals. Additional improvement may be obtained from more detailed knowledge of fluxes at the specific rod locations in the reactor core during the successive irradiation intervals.

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The ORIGEN code requires input data obtained from flux and temperature distributions in the Peach Bottom reactor, as well as the various nuclear libraries that pertain to a high-temperature gas-cooled reactor. The terms THERM, RES, FAST, RITH, RING, RIF, etc., as defined by Bell,[2] are calculated from available information and are described in the next section.

2. PREPARATION OF ORIGEN INPUT DATA

Tobias[6] has made calculations of ORIGEN input parameters for fuel element E06-01 in the Peach Bottom reactor; these are summarized in Table 1 and in the following equations in terms of the four-group fluxes listed by Wallroth.[1]

From the definitions:

$$X \equiv \varphi_4 * Q/W,$$

$$Y \equiv \varphi_1 * P/R,$$

and

$$Z \equiv (\varphi_1 - Y) + \varphi_2 + \varphi_3 + (\varphi_4 - X),$$

we may obtain the input values of THERM, RES, and FAST for ORIGEN as follows:

$$\text{THERM} = \sqrt{\pi/4 * 293.16/T},$$

where

$$T = \text{sample (in reactor) temperature,}^\circ\text{K};$$

$$\text{RES} = Z/(X * U);$$

$$\text{FAST} = 1.45 * Y/X.$$

The lethargy interval, U, conforms to Bell's definition of RITH. The definitions of ORIGEN terms SIGF, SIGNG, RING, and RIF are also used in the Tobias calculations; terms σ_{fi} and σ_{ci} are average cross sections for fission and capture, respectively, of the fissile nuclide (^{232}U, ^{233}U, or ^{235}U) in the flux band i (i = 1, 2, 3, 4). However, cross sections of each of the nuclides ^{232}U, ^{233}U, and ^{235}U were calculated for this study as follows:

$$\text{SIGF} = \sigma_{f4} * \text{THERM},$$

$$\text{SIGNG} = \sigma_{c4} * \text{THERM}.$$

Finally,

$$\text{RING} = U * [\sigma_{c1}(\varphi_1 - Y) + \sigma_{c2}\varphi_2 + \sigma_{c3}\varphi_3 + \sigma_{c4}(\varphi_4 - X)]/Z,$$

$$\text{RIF} = U * [\sigma_{f1}(\varphi_1 - Y) + \sigma_{f2}\varphi_2 + \sigma_{f3}\varphi_3 + \sigma_{f4}(\varphi_4 - X)]/Z.$$

The four-group cross sections used were from Table 2 of ref. 6.

The current study uses a modified definition of SIGF, SIGNG, RING, and RIF, which results in the same final reaction rate for use in the ORIGEN calculations; however, different

Table 1. Fluxes, lethargy intervals, and other parameters pertaining to Peach Bottom fuel

Flux	Upper energy, E_u (eV)	Lower energy, E_l (eV)	Lethargy interval [$\Delta u = \ln (E_u/E_l)$]
φ_1	14.96E+6	8.65E+4	5.1530 ≡ R
φ_2	8.65E+4	1.76E+1	8.5000 ≡ S
φ_3	1.76E+1	2.38	2.0008 ≡ V
φ_4	2.38	0.001[a]	7.7749 ≡ W

$P \equiv \ln (14.96E+6/1.0E+6) = 2.7054$

$Q \equiv \ln (0.5/0.001) = 6.2146$

$U \equiv \ln (1.0E+6/0.5) = 14.5087$

[a]We use a lower limit of 0.001 eV, instead of 0. as listed by Wallroth,[1] for the lower energy of neutrons in the fourth flux group.

intermediate numbers apply. The modified definitions used in this study are given in Sect. 2.2.

At present, values for flux φ_4 are available for the individual rods at only a few times, as shown in Table 2. These have been used to provide fluxes for use in the ORIGEN code. For example, at the three real times of 356, 845, and 1142 days after July 15, 1970, the values of φ_4 for sample FTE-4-3-5-7 were 3.374×10^{13}, 4.294×10^{13}, and 4.530×10^{13}, respectively.[1] Multiplying each of these by the ratio $\ln(0.5/0.001)/\ln(2.38/0.001)$, which equals 0.79932, gives the three different values for flux used in these ORIGEN calculations.

2.1 Calculation of ORIGEN Input Values from Peach Bottom Parameters

Information provided by Wallroth has been used in desk-calculator calculations of input parameters required by ORIGEN; for convenience, it has also been used as the basis of a computer program (Appendix A) that generates both cards and magnetic tape information required for ORIGEN. The code to utilize the Peach Bottom reactor information uses fluxes, as in hand calculations, to determine THERM, RES, and FAST, and to determine the fluxes as a function of time. For determining the fluxes as a function of time, it is necessary to know the absolute value of φ_4 during each time period of the irradiation, including shutdown times when $\varphi_4 = 0$. The time-averaged ratios of φ_1/φ_4, φ_2/φ_4, and φ_3/φ_4 are also needed for each time period. In addition to the flux and temperature information, the program requires the composition of the sample, appropriate titles, and a list of decay times for which calculations are to be made. The irradiation, shutdown, and decay times may be presented as days (or hundredths of a day) following some reference time, or simply as calendar dates. In either case, the ORIGEN printout includes a table listing the corresponding days used in the ORIGEN calculations.

2.2 Reactor Dependence of the Code

The code assumes the input flux spectrum and cross sections given in Tables 1 and 2 of ref. 6. The ORIGEN code defines the cross sections of the main components (Table 2 of ref. 6) using only one of the factors (THERM) and a zero for resonant and fast cross sections. The thermal cross section is defined as:

$$\Sigma\sigma_i\varphi_i/(\text{THERM} * \text{Flux})$$

ORIGEN immediately collapses the three-group cross sections using THERM, RES, and FAST as weighting factors. Thus, after collapsing the cross sections and multiplying by the flux, the ORIGEN code will obtain a reaction rate of $\Sigma\sigma\varphi$. The end result is the same as that obtained by Tobias.

Table 2. Irradiation, power, and flux data for samples FTE-4-3-1-8 and FTE-4-3-5-7

Trever time point[a]	Real time[b] (days)	Average reactor power[a] [MW(t)]	Real exposure time (days)	Effective full-power days[a] (EFPD)	Fluxes for samples FTE-4- [neutrons cm^{-2} sec^{-1} (1.0E-13)]			
					φ_4[c]		ORIGEN calculations[e]	
					3-1-8	3-5-7	3-1-8	3-5-7
7	356.1				3.164	3.374		
		94.2	56.4	46	(3.164)[d]	(3.374)[d]	2.53	2.70
8	412.5							
		93.4	55.6	45	(3.164)[d]	(3.374)[d]	2.53	2.70
9	468.1							
		0.	22.0	0	0	0	0	0
10	490.1							
		103.2	47.0	42	(3.164)[d]	(3.374)[d]	2.53	2.70
11	537.1							
		0.	162.0	0	0	0	0	0
12	699.1							
		91.0	145.9	115	(3.164)[d]	(3.374)[d]	2.53	2.70
13	845.0				4.038	4.294		
		0.	46.0	0			0	0
14	891.0							
		91.7	80.6	64	(4.038)[d]	(4.294)[d]	3.23	3.43
15	971.6							
		103.2	51.5	46	(4.038)[d]	(4.294)[d]	3.23	3.43
16	1023.1							
		0.	20.0	0	0	0	0	0
17	1043.1							
		106.4	98.8	91	(4.260)	(4.530)	3.41	3.62
18	1141.9				4.260	4.530		

[a]Data from ref. 7.
[b]Since July 15, 1970.
[c]Data from ref. 1.
[d]Assumed to be unchanged until a different value is given.
[e]See text.

3. PRELIMINARY RESULTS AND COMPARISONS

Preliminary results of ORIGEN calculations are shown for uranium isotopic compositions in Table 3 with the pertinent analytical data. Calculated and mass spectrometric values of uranium isotopic compositions appear to be in good agreement for samples FTE-4-3-1-8 and FTE-4-3-5-7. The word "good" is more qualitative than quantitative at present.

In like manner, fission-product activities are presented in Table 4. These fall into three groups: (1) sample FTE-4-3-5-7, for which there is good agreement between ORIGEN calculations and the results of the nondestructive (NDA) gamma-ray scanning for seven gamma-emitting isotopes; (2) sample FTE-4-3-1-8, for which there is a consistently low ratio NDA/ORIGEN; and (3) the two nuclides ^{152}Eu and ^{155}Eu, for which there is no agreement between ORIGEN calculations and the gamma-ray scanning for either fuel sample.

The ratios of the values of NDA/ORIGEN approximate unity for ^{95}Zr, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, and ^{154}Eu. This is considered to be a satisfactorily close agreement. The actual average of these seven values for sample FTE-4-3-5-7 is 1.09; the standard deviation is 0.19. The significantly lower values of the seven ratios, NDA/ORIGEN, for sample FTE-4-3-1-8 (average = 0.36, standard deviation = 0.15) are still under investigation by wet chemical techniques (the subject of a later report). The ^{152}Eu and ^{155}Eu discrepancies are explained by the low counting rates of these isotopes after a 2-year decay period.

Preliminary calculations were made to estimate the "residual nitrogen"[a] content of the fuel rods from ^{14}C analyses. The amount of N_2 present during irradiation ("residual nitrogen") may be estimated by comparison of the ORIGEN results with the ^{14}C contents of the burner off-gases during the reprocessing studies (Table 5). The production of ^{14}C from ^{14}N predominates over that from ^{13}C. Based on the ratio of ^{14}C found experimentally to that produced from 1.0 g of N_2 (per ORIGEN) we estimate that the "residual N_2" was 435 ppm in rod 4-3-1-8, and 1640 ppm in rod 4-3-5-7, based on total heavy-metal content. The ^{14}C values are valid only for fuel rods (i.e., matrix graphite and pyrolytic carbon). Block graphite was not analyzed in this study.

4. DISCUSSION

While some of the calculated results shown in Tables 3 and 4 suggest that the input data for two fuel samples from the Peach Bottom reactor yield relatively accurate ORIGEN results, at least two types of input data need further investigation, namely flux and the value of RES.

The fluxes used in ORIGEN (shown in Table 2) are based on only three sets of values for φ_1, φ_2, φ_3, and φ_4, although the power is seen to be different for each of the seven different nonzero power levels. A more detailed flux-time history obtained from reactor operating data appears likely to provide further improvement in ORIGEN calculations.

The second variable requiring additional investigation is RES. A series of calculations with the ORIGEN program, in which only RES was varied, showed that the quantities of ^{95}Zr, ^{106}Ru, ^{125}Sb, ^{137}Cs, and ^{144}Ce (direct fission products) were not sensitive to the magnitude of

[a] It is likely that the fluidizing gas for the final LTI coating was N_2, although it is not clear if definitive data for these very early fuels can be retrieved at this time.[7]

Table 3. Uranium isotopic compositions and burnup

Nuclide	Input	Composition (wt %)			
		FTE-4-3-1-8		FTE-4-3-5-7	
		ORIGEN	Mass spectrometer	ORIGEN	Mass spectrometer
^{232}U	0.00	7.6E-04	-	4.3E-04	-
^{233}U	0.00	14.6	12.73	7.70	8.38
^{234}U	1.50	1.96	1.49	1.81	1.08
^{235}U	93.14	71.1	72.49	76.3	74.9
^{236}U	0.51	7.26	7.39	8.29	8.23
^{237}U	0.00	6.2E-10	-	7.4E-10	-
^{238}U	4.85	5.15	5.89	5.65	7.41
^{240}U	0.00	5.1E-29	-	9.4E-29	-
Burnup, MWd(t)/MT					
Fissile stream		536,000		558,000	
Fertile stream		10,200		11,300	
Power density, MW/MT					
Fissile stream		716		746	
Fertile stream		13.3		14.7	

Table 4. Comparisons of fission-product activities, measured (NDA) and calculated (ORIGEN)[a]

Fission product	FTE-4-3-1-8			FTE-4-3-5-7		
	ORIGEN	Gamma scan (NDA)	NDA/ORIGEN	ORIGEN	Gamma scan (NDA)	NDA/ORIGEN
^{85}Kr	1.26E+11			1.19E+11		
^{90}Sr	1.01E+12			1.00E+12		
^{95}Zr	8.08E+10	3.11E+10	0.39	7.67E+10	1.02E+11	1.33
^{95}Nb	1.73E+11			1.65E+11		
^{106}Ru	3.89E+11	1.13E+11	0.29	3.97E+11	5.00E+11	1.27
^{110}Ag	6.59E+09			6.96E+09		
^{125}Sb	3.67E+10	1.78E+10	0.49	3.33E+10	2.89E+10	0.87
^{134}Cs	5.43E+11	1.04E+11	0.19	5.81E+11	5.11E+11	0.88
^{137}Cs	1.00E+12	3.15E+11	0.32	9.95E+11	1.19E+12	1.20
^{144}Ce	4.47E+12	1.14E+12	0.26	4.39E+12	5.00E+12	1.14
^{152}Eu	1.03E+08	4.44E+09	43.	9.83E+07	1.11E+10	113.
^{154}Eu	2.85E+10	1.78E+10	0.63	3.10E+10	2.89E+10	0.93
^{155}Eu	6.64E+09	1.11E+11	17.	6.88E+09	2.06E+11	29.9

[a]Activities are in distintegrations per minute for the whole sample.

Table 5. Carbon-14 activities and calculated nitrogen contents for samples FTE-4-3-1-8 and FTE-4-3-5-7

	FTE-4 sample	
	3-1-8	3-5-7
Weight in sample, U+Th, g	5.74	2.93
C, g	4.44	4.29
^{14}C yield calculated from reaction		
$^{13}C(n,\gamma)$, dpm/g C	1.083E+5	1.146E+5
$^{14}N(n,p)$, dpm/g N	1.245E+10	1.316E+10
Measured ^{14}C activity, dpm/g C	7.11E+6	1.49E+7
Estimated nitrogen content, g N/MTHM[a]	435	1640
g N/MTC[b]	562	1120

[a]MTHM is metric tons of U+Th in the sample charged to the reactor.
[b]MTC is metric tons of carbon in the sample.

RES. However, the quantities of ^{134}Cs and ^{154}Eu (activation products) are very sensitive to the value of RES. Further improvements in methods to calculate RES should yield improved estimates for the activation products.

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APPENDIX A. COMPUTER CALCULATION OF ORIGEN INPUT DATA FROM PEACH BOTTOM PARAMETERS

The code to convert Peach Bottom reactor parameters to ORIGEN input data has been written in two versions. One version is for time-sharing use (PDP-10 version); it prompts the user for information. The time-sharing version allows free format input with consecutive input numbers separated by commas. The other version is for batch processing with card input (IBM 360 version); it assumes a standard order for the information. The batch version uses the formats listed below. The order in which the information is requested (or read) is the same for either version.

A. Reads: IO — Format: I5

IO is the unit number to be used for output of the ORIGEN deck. For the IBM 360, this should correspond to a DD card referencing the card punch or a disc file. For the PDP-10 at ORNL, a value of 1 to 4 or 10 to 29 results in output to a file FORnn.DAT where nn is the two digit representation of IO.

B. Reads: TEMP — Format: F10.0

TEMP - Temperature in °C

C. Reads: PHI1, PHI2, PHI3, PHI4 — Format: 4F10.0

These are the time averaged values of φ_1, φ_2, φ_3, and φ_4, respectively. Since only the ratios are important, common factors (such as 10^{13}) may be omitted.

D. Reads: ISTRT, ITYPE — Format: I10,I5

ISTRT is the time at which irradiation starts.

ITYPE describes the format of ISTRT and all other times.

If ITYPE = 1, all times are input as calendar dates. The first two digits from a six-digit time or the first four digits from an eight-digit time are interpreted as being the year. The next two digits are the month and the last two digits are the day of the month. The algorithm for determining the number of days between two dates considers leap years. As an added refinement, the algorithm considers century years as being leap years only when divisible by 400 if four-digit years are used. It is important that all dates have the same number of digits for the year (i.e., either 2 or 4) for any given case.

If ITYPE = 2, all times are assumed to be given in hundredths of a day. If ITYPE = 3, all times are assumed to be in days.

E. Reads: IIRR(I), FLX(I) — Format: I10, F10.0

IIRR(I) is the end of the time period in which the flux φ_4 has the value FLX(I). Successive times are input until a nonpositive time occurs. If desired, any common factor of the flux values (e.g., 10^{11}) can be listed as the flux corresponding to the time, which is nonpositive, since all previous values of the flux are multiplied by this number (if it is greater than one). All the times must be of the form referred to by ITYPE.

F. Reads: IDEC(J) Format: I10

IDEC(J) is the time (J) to be listed in the ORIGEN output as a decay time after reactor discharge. Times are read until a nonpositive value occurs. All must be defined as referenced by ITYPE.

G. Reads: CASE Format: 10A4

CASE is a 40-character case name used in constructing titles for ORIGEN output. The titles are constructed by adding "IRRADIATION TIMES" or "DECAY TIMES" as appropriate.

H. Reads: BASIS Format: 10A4

BASIS is a 40-character subtitle which appears in the ORIGEN output after the word "BASIS." BASIS and CASE are usually identified with a set of concentrations rather than with an irradition history.

I. Reads: NO, MASS, GATOM Format: I2, I3, F10.3

Cards (or lines) of this type are read until a card (or line) with NO = 0 is encountered. These values are the input composition. NO is the atomic number; MASS is the atomic mass; and GATOM is the charge in gram-atoms. The code distinguishes between materials of construction and actinides by examining the atomic number. If the material is a material of construction (i.e., NO < 85), a naturally occurring element can be indicated by making MASS = 0.

J. Reads: MORE Format: I5

(MORE corresponds to NGO in the ORIGEN manual). There are two possible valid choices for MORE. If MORE is zero the code assumes that everything about the next case may be different and the next input is of type A. If MORE is negative the next case will have different titles and charge composition only. The next input is of type G and inputs of type A through F are assumed to be the same. Positive values for MORE are not allowed. If a positive value is input by mistake, the code will behave as if MORE is negative, but the next case (if there is one) will not be run successfully by ORIGEN.

There are two normal ways for the program to stop. When IO is read (type A) the program checks for an end-of-file indication. When input of type C is read the program stops if PHI1 = 0.

Running the ORIGEN Deck

The output deck is a complete set of ORIGEN input. It can be run at ORNL with the following control cards:

Job card
Class card (L = 20, suggested)
/*ROUTE PRINT LOCAL (for PDP-10 submission)
// EXEC ORIGEN
//GO.FT05F001 DD *
Deck of ORIGEN input
/*
//

If no materials of construction are needed, that part of the data library can be omitted by including the card

//GO.FT25F001 DD DUMMY

just before the DD card for unit 5. If, during any execution of this program, the last value of MORE is zero, the deck may be followed by any other complete ORIGEN deck. As many decks as desired may be stacked in this way.

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