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ESTIMATION OF RESONANCE EFFECTS IN FUEL BURNUP ANALYSIS

A.R. Olsen J.W. Prados

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MARCH 1967

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ESTIMATION OF RESONANCE EFFECTS IN FUEL BURNUP ANALYSIS

A. R. Olsen

J. W. Prados

ABSTRACT

This report describes a computer program, RCROSS, which calculates time-averaged thermal flux, resonance-to-thermal flux ratios and atomic percent heavy metal burnup for uranium fuels. The input data required are the irradiation time, the preirradiation and post-irradiation uranium isotopic analyses for the fuel, and literature values for the various neutronic constants.

INTRODUCTION

The number of fissions occurring in a reactor fuel irradiation test specimen must be known accurately if one is to make a rational evaluation of the fuel's performance. Consequently, it is desirable that the experimenter responsible for postirradiation examinations utilize several independent methods of analysis to calculate the amount of fissioning that has occurred in a specimen. These analyses act as checks to reduce the uncertainties which result from limitations in experimental and computational accuracy of the common burnup determination techniques.¹ However, in many cases, the burnups calculated from these various analyses show disagreement. It is the purpose of this report to describe a computational procedure which we have found useful in resolving apparent discrepancies.

Three distinct burnup analyses were employed in the evaluation of the burnup in pyrolytic-carbon-coated fuel particles exposed in the first

¹R. L. Stover and G. K. Moeller, Methods for Determining Fuel Burnup, MIT-OR-6 (July 1961).

capsule (ORNL 43-89) in the "X-basket" series² of irradiations in the ETR. These were fluence measurement by activation of ⁵⁹Co in a stainless steel monitor tape, radiochemical analysis for the fission products, ¹³⁷Cs and ¹⁴⁴Ce, and mass spectrographic determination of the change in isotopic composition of the uranium in the UO₂ fuel kernels.

In the initial calculations of burnup, prior to using the computer code developed here, no corrections for resonance-neutron effects were made in the monitor tape cross sections since the ratio of resonance-to-thermal flux was not available for the particular ETR position employed. Also, the resonance-to-thermal flux ratio can be expected to vary with reactor operation and experiment loading arrangements. The resulting burnup values calculated from monitor tape activation were 50 to 100% higher than the values obtained from the fission-product and uranium isotopic determinations.

There is a distinct possibility that the latter two methods could be in error because of cesium or cerium loss to capsule components during irradiation or because of sample contamination during remote handling in the preparation of analytical samples. Consequently, it was necessary to find some means of evaluating the resonance effect on the monitor-tape cobalt and on uranium isotopic cross sections in order to establish a higher degree of confidence in the burnup levels calculated.

CALCULATIONS AND DISCUSSION

At the suggestion of J. Halperin, we developed a procedure to calculate a time-averaged resonance ratio. This calculation would utilize the uranium isotopic analyses, the basic reaction equations, and the best published values for thermal cross sections, resonance integrals, and "non-1/v" corrections. This computed resonance ratio can then be used to correct the cobalt and uranium cross sections to permit a more accurate calculation of fluence and burnup from the monitor-tape activation results.

²J. H. Coobs, H. C. McCurdy, and O. Sisman, GCRP Semiann. Progr. Rept. Mar. 31, 1966, ORNL-3951, pp. 34-53.

Standard mathematical relations are employed to describe the production and consumption of uranium isotopes by neutron capture and fission during irradiation. These are:¹

$$N_{25} = N_{25}^0 e^{-\phi t(\sigma_{5a})} \quad (1)$$

$$N_{28} = N_{28}^0 e^{-\phi t(\sigma_{8a})} \quad (2)$$

$$N_{26} = N_{26}^0 e^{-\phi t(\sigma_{6a})} + N_{25}^0 \sigma_{5c} \left[\frac{e^{-\phi t(\sigma_{5a})}}{\sigma_{6a} - \sigma_{5a}} + \frac{e^{-\phi t(\sigma_{6a})}}{\sigma_{5a} - \sigma_{6a}} \right] \quad (3)$$

where,

N_{25}^0 = initial amount of ^{235}U (atoms),

N_{28}^0 = initial amount of ^{238}U (atoms),

N_{26}^0 = initial amount of ^{236}U (atoms),

N_{25} = final amount of ^{235}U (atoms),

N_{28} = final amount of ^{238}U (atoms),

N_{26} = final amount of ^{236}U (atoms),

ϕ = average thermal neutron flux (neutrons $\text{cm}^{-2}\text{sec}^{-1}$),

t = irradiation time (seconds),

and where all σ values represent effective cross sections:

$$\sigma = \sigma_o(g) + \frac{\phi_r}{\phi} I \quad (\text{barns}) = (\text{cm}^2 \times 10^{-24}) ,$$

the σ values are subscripted with an (a) for absorption, a (c) for capture, and an (o) for 2200 m/sec value.

g = neutron-temperature correction for "non-1/v" dependence of thermal cross section (barns),

I = resonance integral (barns), and

ϕ_r = resonance flux (neutrons $\text{cm}^{-2} \text{sec}^{-1}$).

The absolute initial and final values of the "N's" cannot be obtained from the mass spectrographic analyses of uranium isotopes, since these yield only relative amounts of each isotope present. However, by dividing Eqs. (2) and (3) by Eq. (1), we obtain

$$\frac{N_{28}}{N_{25}} = \frac{N_{28}^0}{N_{25}^0} \left[e^{-\phi t (\sigma_{28a} - \sigma_{25a})} \right], \quad (4)$$

and

$$\frac{N_{26}}{N_{25}} = \frac{N_{26}^0}{N_{25}^0} e^{-\phi t (\sigma_{26a} - \sigma_{25a})} + \frac{\sigma_{25c}}{\sigma_{26a} - \sigma_{25a}} \left[1 - e^{-\phi t (\sigma_{26a} - \sigma_{25a})} \right], \quad (5)$$

which contain only the ratios of isotopic contents. These ratios can be obtained directly from the pre- and postirradiation mass spectrographic analyses.

The 2200-m/sec (σ_0) cross sections, the resonance integrals (I) and the non-1/v corrections (g) are available in the literature.^{3,4} This leaves only the average flux (ϕ) and the resonance ratio $R = \phi_r/\phi$ as two unknowns, and these are determined uniquely by simultaneous solution of Eqs. (4) and (5).

The nature of Eqs. (4) and (5) precludes explicit solution for the unknowns ϕ and R , and an iterative procedure must be employed. We have written a computer program, RCROSS, which performs these iterations automatically to yield values of average flux, ϕ , and resonance ratio, R ,

³John R. Stehn et al., Neutron Cross Sections, BNL-325, vol III, 2nd ed., Supplement 2, Z = 88 to 98 (February 1965).

⁴C. H. Westcott, Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra, CRRP-787 (AECL-670) (Aug. 1, 1958).

for each set of pre- and postirradiation isotopic analyses supplied. In addition, the program computes ^{235}U depletion and total uranium burnup, using the following relations derived in ref. 1.

$$\frac{F_D}{100} = 1 - \left[\frac{Y_O + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}}{Y + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}} \right] \frac{\sigma_{5a}}{\sigma_{5a} - \sigma_{6a}} \quad (6)$$

and

$$\text{Bu} = y_{\text{O}}^{25} F_D \left(\frac{\sigma_{5a} - \sigma_{5c}}{\sigma_{5a}} \right) \quad (7)$$

where

F_D = % depletion of ^{235}U ,

Bu = % of initial uranium atoms fissioned,

$$Y_O = \frac{N_{25}^O}{N_{26}^O}, \quad Y = \frac{N_{25}}{N_{26}}, \quad y_{\text{O}}^{25} = \frac{N_{25}^O}{N_{\text{U}}^O}$$

N_{U}^O = initial amount of uranium (atoms), and the σ values are all the calculated effective values including resonance corrections.

The final working equations and Fortran statements for this program, together with a sample input data sheet and printed output, are presented as Appendix A.

The program was applied to the analytical results from coated-particle irradiation experiment ORNL-43-89, which had previously shown internal disagreement. As seen in Table 1, our new procedure gave quite reasonable agreement between the burnup calculated by each method and helped to identify the few questionable analytical results. This agreement permitted us to establish with reasonable assurance the burnup

Table 1. Selected Burnup Analyses for X-Basket 1

Container Number	Atom % Heavy Metal Fissioned					ϕ_r/ϕ	Advertised $-\phi t$ Peak Burnup ^c
	Cobalt Flux Monitors		Isotopic	¹³⁷ Cs	¹⁴⁴ Ce		
	(a)	(b)					
3	19.9	12.2	(9.2) ^d	12.0	e	0.689	
6	24.1	15.4	13.2	12.5	12.5	0.470	
11	32.1	20.5	19.8	19.6	e	0.269	
14	33.7	21.5	20.7	(16.1) ^d	19.1	0.277	
15	33.9	21.6	e	e	e	e	28.3
23	28.5	18.2	18.1	17.1	e	0.289	
26	23.0	14.7	13.6	13.1	12.7	0.454	

^aValues calculated from monitor tapes with an uncorrected 37-barn cross section for ⁵⁹Co → ⁶⁰Co.

^bValues calculated from monitor tapes using a $\phi_r/\phi = \text{constant} = 0.278$.

^cThe burnup value reported here was calculated from the peak ϕt values reported by the ETR operations.

^dThe parentheses indicate analytical results that are deemed questionable.

^eNot analyzed.

levels for the fuel in all 28 containers in the experiment. The same techniques will be used on the future experiments in the X-basket series and are currently being used in other coated-particle irradiation test evaluations.

APPENDIX

Details of Program RCROSS

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A.1. Working Equations

$$\sigma_{5a} = \sigma_{o5a} g_{5a} + R I_{5a}$$

$$\sigma_{5c} = \sigma_{o5c} g_{o5c} + R I_{5c}$$

$$\sigma_{6a} = \sigma_{o6a} g_{6a} + R I_{6a}$$

$$\sigma_{8a} = \sigma_{o8a} g_{8a} + R I_{8a}$$

$$\psi = \left(\frac{Y}{X_O} \right) \frac{\sigma_{5a} - \sigma_{8a}}{\sigma_{5a} - \sigma_{6a}}$$

$$T = \frac{Y - Y_O \psi}{\psi - 1}$$

$$R = \frac{(\sigma_{o5} g_{5a} - \sigma_{oc} g_{6a}) T - \sigma_{o5c} g_{5a}}{I_{5c} - (I_{5a} - I_{6a}) T}$$

$$\tau = \frac{1}{\sigma_{5a} - \sigma_{8a}} \ln \left(\frac{X}{X_O} \right) = \frac{1}{\sigma_{5a} - \sigma_{6a}} \ln \frac{Y + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}}{Y_O + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}}$$

$$\phi = \tau/t$$

$$X = y_{28}/y_{25} ; Y = y_{26}/y_{25}$$

$$F_D = 100 \left\{ 1 - \left[\frac{Y_O + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}}{Y + \frac{\sigma_{5c}}{\sigma_{5a} - \sigma_{6a}}} \right] \frac{\sigma_{5a}}{\sigma_{5a} - \sigma_{6a}} \right\} \text{fractional depletion of } {}^{235}\text{U}.$$

$$\text{Bu} = y_O^{25} F_D \left(\frac{\sigma_{5a} - \sigma_{5c}}{\sigma_{5a}} \right) \text{ atom \% heavy metal fissioned.}$$

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A.2. FORTRAN Statements

```

PROGRAM RCROSS
DIMENSION R(80),PSI(80),UPS(80),TI(7)
READ(50,100)N $ NDATE=IDATEF(DUMMY)
READ(50,100)MAX,TOL
READ(50,101)C05A,G5A,A5A,C05C,G5C,A5C,C06,G6,A6,C08,G8,A8
CC5A=C05A*G5A $ CC5C=C05C*G5C $ CC6=C06*G6 $ CC8=C08*G8 $ D01J=1,N
READ(50,103)TI
READ(50,102)Y025,Y026,Y028,Y25,Y26,Y28,T $ WRITE(51,200)TI,NDATE
WRITE(51,205)Y025,Y026,Y028,Y25,Y26,Y28
X0=Y028/Y025 $ Y0=Y026/Y025 $ X=Y28/Y25 $ Y=Y26/Y25
PSI0 = X/X0 $ UPS0 = (Y-Y0*PSI0)/(PSI0-1.)
R(1)=((CC5A-CC6)*UPS0-CC5C)/(A5C-(A5A-A6)*UPS0)
DO 2 I=1,MAX
C5A=CC5A+A5A*R(I) $ C5C=CC5C+A5C*R(I) $ C6=CC6+A6*R(I)
C8=CC8+A8*R(I) $ PSI(I)=(X/X0)**((C5A-C8)/(C5A-C6))
UPS(I)=(Y-Y0*PSI(I))/(PSI(I)-1.)
R(I+1)=((CC5A-CC6)*UPS(I)-CC5C)/(A5C-(A5A-A6)*UPS(I))
EPSI=ABS(R(I+1)-R(I))/R(I+1) $ IF(EPSI-TOL)3,2,2
2 CONTINUE
WRITE(51,201)MAX $ WRITE(51,202)(R(I),I=1,MAX)
3 SR=C5C/(C5A-C6) $ TAU=LOGF((Y+SR)/(Y0+SR))/(1.E-24*(C5A-C6))
FLUX = TAU/(8.64E4*T)
B=C5A/(C5A-C6) $ DEPL=100.*(1.-((Y0+SR)/(Y+SR))**B)
BU=Y025*DEPL*(C5A-C5C)/C5A
OWRITE(51,203)C05A,G5A,A5A,C5A,C05C,G5C,A5C,C5C,C06,G6,A6,C6,C08,
1G8,A8,C8,X0,X,Y0,Y,R(I+1),TAU,T,FLUX
1 WRITE(51,204)DEPL,BU
CALL EXIT
100 FORMAT(110,F10.0)
101 FORMAT(6F10.0/6F10.0)
102 FORMAT(8F10.0)
103 FORMAT(7A8)

```

A.2. FORTRAN Statements (continued)

```

200 FORMAT(1H1,7A8,5X,5HDATE ,A8/1H0/1H0/)
2010FORMAT(1H0,37HTHIS CALCULATION DID NOT CONVERGE IN ,12,21H TRIES.
      1 R-VALUES ARE)
202 FORMAT(8E13.3)
2030FORMAT(1H0,10HSIG025A = ,F5.1,5X,7HG25A = ,F5.3,5X,7HI25A = ,F5.1,
      15X,12HSIGEFF25A = ,F5.1/1H0,10HSIG025C = ,F5.1,5X,7HG25C = ,F5.3,
      25X,7HI25C = ,F5.1,5X,12HSIGEFF25C = ,F5.1/1H0,9HSIG026 = ,F5.1,6X,
      36HG26 = ,F5.3,6X,6HI26 = ,F5.1,6X,11HSIGEFF26 = ,F5.1/1H0,9HSIG028
      4 = ,F5.1,6X,6HG28 = ,F5.3,6X,6HI28 = ,F5.1,6X,11HSIGEFF28 = ,F5.1/
      51H0,5HX0 = ,F8.6,7X,4HX = ,F8.6,7X,5HY0 = ,F8.6,4X,4HY = ,F8.6/1H0
      6,4HR = ,F8.6,5X,7HDOSE = ,E9.3,      6H N/CM2,5X,7HTIME = ,F6.2,5H
      7DAYS/1H0,10HAV FLUX = ,E9.3,10H N/CM2-SEC)
2040FORMAT(1H0/1H0,14HBURNUP RESULTS/1H0,5X,30HPER CENT DEPLETION OF U
      1-235 = ,F5.2/1H0,5X,50HPER CENT OF INITIAL HEAVY METAL ATOMS FISSI
      2ONED = ,F5.2)
2050FORMAT(1H0,22HINITIAL ATOM FRACTIONS/1H0,5X,8HU-235 = ,F7.5,5X,
      18HU-236 = ,F7.5,5X,8HU-238 = ,F7.5/1H0,20HFINAL ATOM FRACTIONS/
      21H0,5X,8HU-235 = ,F7.5,5X,8HU-236 = ,F7.5,5X,8HU-238 = ,F7.5)
      END RCROSS

```

A.3. Sample Input Data Sheet

650 DATA SHEET

REQUEST		SAMPLE INPUT DATA		JOB TITLE		WRITTEN BY		DATE							
				RCRΦ55		J W PRADOS		3/9/66							
1 - 10		11 - 20		21 - 30		31 - 40		41 - 50		51 - 60		61 - 70		71 - 80	
NO. OF CASES → 3															
MAXIMUM NO. OF ITERATIONS 40		CONVERGENCE TOLERANCE ON TRIAL & ERROR CALCULATION													
678.2 ← σ _{05A}		.96 ← g _{5A}		418. ← I _{5A}		101. ← σ _{05C}		.96 ← g _{5C}		144. ← I _{5C}		CROSS SECTIONS, "NON-1/U" CORRECTIONS, AND RESONANCE INTEGRALS			
6. ← σ ₀₆		1. ← g ₆		400. ← I ₆		2.73 ← σ ₀₈		1. ← g ₈		280. ← I ₈					
X-BASKET NΦ. 1		ΦP-3		← TITLE											
.9271 ← y ₀ ²⁵		.00223 ← y ₀ ²⁶		.0608 ← y ₀ ²⁸		.8963 ← y ₀ ²⁵		.0288 ← y ₀ ²⁶		.0639 ← y ₀ ²⁸		72.63			
X-BASKET NΦ. 1		ΦP-11		← TITLE											
.9271 ← y ₀ ²⁵		.00223 ← y ₀ ²⁶		.0608 ← y ₀ ²⁸		.8609 ← y ₀ ²⁵		.0552 ← y ₀ ²⁶		.0720 ← y ₀ ²⁸		72.63		← FULL-POWER DAYS	
X-BASKET NΦ. 1		ΦP-23		← TITLE											
.9271 ← y ₀ ²⁵		.00223 ← y ₀ ²⁶		.0608 ← y ₀ ²⁸		.8673 ← y ₀ ²⁵		.0501 ← y ₀ ²⁶		.0706 ← y ₀ ²⁸		72.63			
INITIAL ATOMIC FRACTIONS						FINAL ATOMIC FRACTIONS									

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A.4. Sample Output Data Sheet

X-BASKET NO. 1, OP-3

DATE 03-15-66

INITIAL ATOM FRACTIONS

U-235 = .92710 U-236 = .00223 U-238 = .06080

FINAL ATOM FRACTIONS

U-235 = .89630 U-236 = .02880 U-238 = .06390

SIG025A = 678.2 G25A = .960 125A = 418.0 SIGEFF25A = 938.1

SIG025C = 101.0 G25C = .960 125C = 144.0 SIGEFF25C = 195.8

SIG026 = 6.0 G26 = 1.000 126 = 400.0 SIGEFF26 = 280.6

SIG028 = 2.7 G28 = 1.000 128 = 280.0 SIGEFF28 = 195.0

X0 = .06558 X = .071293 Y0 = .002405 Y = .032132

R = .686608 DMSE = 1.436 + 20 N/CM2 TIME = 72.63 DAYS

AV FLUX = 2.288 + 13 N/CM2-SEC

BURNUP RESULTS

PER CENT DEPLETION OF U-235 = 12.60

PER CENT OF INITIAL HEAVY METAL ATOMS FISSIONED = 9.24

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