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**THEORY EXPERIMENT CORRELATION OF BURNUP DATA
FROM Al-1.8 wt% Pu FUEL (INITIALLY 6 at .% Pu ²⁴⁰)**

L. J. PAGE

JULY, 1967

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THEORY EXPERIMENT CORRELATION OF BURNUP DATA
FROM A1-1.8 wt% Pu FUEL (INITIALLY 6 at.% Pu²⁴⁰)

By

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Reactor Lattice Physics Section
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July, 1967

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INTRODUCTION

A large amount of experimental data has recently been obtained for 19 rod clusters of plutonium-aluminum alloy fuel irradiated in the Plutonium Recycle Test Reactor (PRTR) at Pacific Northwest Laboratory. The fuel was initially 1.82 wt% plutonium which contained 6.25 at.% Pu²⁴⁰. The experimental data covered the range from initial concentration up to 50% depletion of the initial plutonium.

Analytical calculations of isotopic concentrations, cross section ratios, and reactivity values were undertaken with two main objectives. The objectives were (1) to determine how well the calculational results are verified by experimental evidence and (2) to obtain information on theoretical techniques necessary to reproduce the experimental results.

THEORETICAL MODEL

The PRTR is a heavy water moderated and cooled reactor composed of a hexagonal array of 19-rod clusters. A typical cell and the concentric ring approximation used in the study are shown in Figure 1. The power generated in the cluster was assumed to be 1 MW. The temperature of the fuel and coolant was kept constant at 600 °F and the moderator at 150 °F which were the D₂O temperatures

recorded while the elements were being irradiated in the PRTR.

For the calculations the range of neutron energies was divided into four groups with boundaries at 0.683, 2.38, 1.17×10^4 and 1.0×10^7 eV. Flux and volume averaged cross sections were computed under the assumption of transport theory using program THERMOS⁽¹⁾ for the lower two energy ranges. Average cross sections over the two high energies were obtained using the homogeneous-media slowing-down theory code, HRG^{(2)*}. The four energy group cross sections were used in the transport-theory code, Program S⁽³⁾ to calculate isotopic concentrations, cross section ratios, and eigenvalues (K_{∞}). The region boundaries used in the THERMOS and Program S calculations are listed in Table I. Two separate calculations were made using program THERMOS. The group structure for one calculation consisted of 30 energy groups between zero and 0.683 eV. In the second calculation, 26 energy groups were spaced between 0.683 and 2.38 eV; mainly around the

**Since this study was made, changes have been made to the code. The most significant change is in the calculation of the effective cross section*

$\sigma_{eff} = \frac{RI}{\int \phi dE}$ where RI is the resonance integral. The version used assumes $\phi(E) = 1/E$ and the later version uses a $\phi(E)$ compatible with the NR or NR1A approximation.

FIGURE 1. PRTR Mark I-H Fuel Element and Reactor Cell Geometry

TABLE I. Description of Pu-Al Cell

THERMOS		Program S	
Radii	Region	Radii	Region
0.0	fuel	0.0	fuel
0.62700		0.62700	clad
	coolant + clad	0.71600	coolant
		1.08151	clad
1.19564	fuel	1.19564	fuel
1.94632		1.94636	clad
	coolant + clad	2.06049	coolant
		2.46156	clad
2.58197	fuel	2.58197	fuel
3.37403		3.37403	clad
	clad + coolant + process tube	3.49444	coolant
		4.12800	process tube
4.36300	process tube + moderator, etc.	4.51900	moderator + shroud tube + gas gap
10.66900		10.66900	

1.056 eV resonance of Pu^{240} . In the latter calculation four groups were spaced between zero and 0.683 eV to properly handle the thermal neutron calculation. The fine group structure for HRG is 34 one-quarter lethargy groups between 2.38 and 1.17×10^4 eV and 27 one-quarter lethargy groups between 1.17×10^4 and 1.0×10^7 eV. All fine group cross sections were obtained from the BNW Master Library.⁽⁴⁾ The Brown St. John scattering model⁽⁵⁾ was used for all materials in the cell calculations except for D_2O in the energy range zero to 0.683 eV where the Honeck-Nelkin⁽⁶⁾ model was used.

Since it was known from previous experience^(7,8,9) that the plutonium cross section evaluated by Leonard⁽¹⁰⁾ better described the experimental burnup data, these data were used exclusively throughout the entire calculation. Doppler broadened cross sections for the 1.056 eV resonance of Pu^{240} at 600 °F were used in the calculations as computed with program GROUSS⁽¹¹⁾ and data from the BNW Master Library.

The four pseudo elements suggested by Nephew⁽¹²⁾ were used to account for the fission products of plutonium. Since these four pseudo elements do not include the very high absorption cross section elements -- Xe^{135} , Sm^{149} , Sm^{151} , Eu^{155} , and Gd^{155} , they were treated separately. Spontaneous decay of Pu^{241} , Xe^{135} and Eu^{155} was accounted for by allowing Pu^{241} to decay to the first pseudo product; Xe^{135} to decay to a dummy material with zero cross section; and Eu^{155} to decay to Gd^{155} .

The average cross sections were computed several times during the burnup study using programs THERMOS and HRG since program S has no provision for changing the cross sections internally as a function of time. When this study was begun, there was no evidence as to how often the cross sections should be recalculated. Average cross sections were computed for zero, ten, forty and eighty days of full power (1 MW) operation and the calculation was terminated at 120 days of operation. All evidence points to the fact that these time steps were adequate.

ISOTOPIC CONCENTRATIONS

Since the vast majority of experimental data is in the form of isotopic concentrations as determined by mass spectrometer analysis, primary importance is put on correlation of these data. Concentrations of the plutonium isotopes are shown in Figures 2, 3, and 4 as a function of percent depletion of the initial plutonium for each of the three fuel regions of the 19-rod cluster. Burnup is defined as the amount of plutonium (all isotopes) fissioned or decayed divided by the initial amount of plutonium. All experimental numbers and errors came from data in References 7, 8 and 9. The experimental uncertainty quoted on burnup values ranges from 4.2% at low depletion to 2.1% at 50% depletion. The uncertainties on the experimental concentrations are 1% for Pu^{239} , 1 1/2% for

FIGURE 2. Isotopic Concentrations for the Center Rod of a Pu-Al Cluster

FIGURE 3. Isotopic Concentrations for Fuel in the Middle Ring of a Pu-Al Cluster

FIGURE 4. Isotopic Concentrations for Fuel in the Outer Ring of a Pu-Al Cluster

Pu^{240} , 2% for Pu^{241} , and 2½% for Pu^{242} . When these uncertainties are taken into account, the calculated values agree with the experimental values for Pu^{239} and Pu^{240} over the entire burnup range studied. The experimental and calculated values for the other isotopes agree up to approximately 33% burnup.

At 50% burnup the theoretical values of the concentration of Pu^{241} are about 7% below the experimental values while the theoretical values for Pu^{242} are about 15% below the experimental values. If these discrepancies had originated because of the frequency at which the cross sections were calculated the calculated values would show a trend of agreeing better after the cross sections were recalculated. The disagreement in concentrations would get worse until the next cross section calculation. As shown in Figures 2 through 4 this is not the case since the calculated values gradually diverge from the experimental values. It is believed that the concentration errors in Pu^{241} and Pu^{242} may be explained by inadequate calculations of the Pu^{240} absorption cross section $\bar{\sigma}_a^{240}$ in the resonance energy range. As seen in Figures 2, 3 and 4 the calculated concentration of Pu^{240} , while still within experimental uncertainty, is larger than the experimental values. A larger value for the average $\bar{\sigma}_a^{240}$ cross section in the energy range 0.683 to 2.38 eV would lower the concentration of Pu^{240} and raise those of Pu^{241} and Pu^{242} . Also, recent evaluations⁽¹³⁾ have shown that the 2200 m/sec

value of $\frac{\sigma_c^{241}}{\sigma_f^{241}}$ is 0.379 instead of

0.303 which was used in this calculation. This change would raise the concentration of Pu^{242} but would leave the concentration of Pu^{241} the same since σ_a^{241} remained unchanged. Further work is planned to help understand these discrepancies by using more recent data for $\sigma(E)^{241}$ and 30 barns for the 2200 m/sec value of σ_c^{242} instead of the 19 barns used in this study.

CROSS SECTION RATIOS

Three sets of effective cross section ratios were computed. They are

$$\frac{\hat{\sigma}_c^{239}}{\hat{\sigma}_f^{239}} = \hat{\alpha}^{239}, \quad \frac{\hat{\sigma}_a^{240}}{\hat{\sigma}_a^{239}}, \quad \text{and} \quad \frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$$

Of particular interest is the ratio $\hat{\alpha}^{239}$ since various values have been reported for the 2200 m/sec value. The values of $\hat{\alpha}^{239}$ obtained in the Sher evaluations⁽¹⁴⁾⁽¹⁵⁾ are about 10% greater than the Leonard value and previous experience^(8,9) has shown that the Leonard value of $\hat{\alpha}^{239}$ reproduces the burnup data. Since the previous work was done only over the thermal energy range and compared to experimental data averaged over all energies, more verification was necessary. The results of the correlation of $\hat{\alpha}^{239}$ for the middle fuel ring is shown in Figure 5, and the results for the outer ring are shown in Figure 6. The experimental data for the single center rod was included in the middle ring data and, thus, is not shown separately.

FIGURE 5. Cross Section Ratios vs Exposure for Middle Fuel Ring

FIGURE 6. Cross Section Ratios vs Exposure for Outer Fuel Ring.

All experimental data is taken from Reference 9. The data is slightly different from that reported in References 7 and 8 because of the value for $\hat{\sigma}_a^{241}$ which was assumed for the analysis. The quoted experimental uncertainty is shown on the experimental points.

It is shown in the figures that the calculated cross section ratios agree quite well with the experimental values. In particular all the calculated values of $\hat{\sigma}_a^{239}$ agree within the experimental uncertainty in both the middle and outer fuel ring. Both calculations and experiment show no

difference in the ratios, $\frac{\hat{\sigma}_c^{239}}{\hat{\sigma}_f^{239}}$ $\frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$ for the outer or middle fuel ring although the experimental and

calculated results for $\frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$ are 20% different. Recent work has shown⁽¹⁶⁾ that the value of

$\frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$ deduced from the burnup data

is obtained with approximately a factor of ten less sensitivity than the other ratios. For example assuming the calculated value of

$\frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$ is the correct experimental one

(i.e. a 20% increase), values which are only 2% higher result from the experimental data for the other ratios. The corresponding concentrations

are as good a fit to the experimental data as the one in which the

$\frac{\hat{\sigma}_a^{241}}{\hat{\sigma}_a^{239}}$ is allowed to vary. The

expected self-shielding effect of Pu^{240} is demonstrated by both the experimental and theoretical ratios of

$\frac{\hat{\sigma}_a^{240}}{\hat{\sigma}_a^{239}}$.

In an attempt to adequately calculate the effects of the 1.056 eV Pu^{240} resonance, program THERMOS was used over this range. The last cross section calculation was made with a Pu^{240} -to- Pu^{239} atom ratio equal to 0.365 for the outer ring. At this point the average Pu^{240} absorption cross section was about 19% too low in the outer ring, 8% too low in the middle ring, and 7% too low in the inner ring to predict the experimental Pu^{241} concentration at the next time step.

Throughout the entire calculation cross sections were also computed using program HRG in the energy range of 0.683 to 2.38 eV. As stated earlier, values from Program THERMOS were used in the Program S calculations for this energy range. A comparison of the HRG and THERMOS data is made in Table II. At the first two time steps with low Pu^{240} concentration the result from the codes differ by 15% and the THERMOS values agree with that necessary to predict accurately the concentration of Pu^{241} . At high Pu^{240} content (>20%) the results from the codes differ by approximately 30% in their value of $\hat{\sigma}_a^{240}$ (energy

TABLE II. Comparison of Cross Sections in the Energy Range 0.683 to 2.38 eV

Time, days	Atom Ratio, $\text{Pu}^{240}/\text{Pu}^{239}$	σ_a^{240} , barns		Difference, %	Necessary σ_a^{240} for data, barns
		HRG	THERMOS		
0	0.067	3.7598+3	3.2801+3	14.6	$\sim 3.3+3$
10	0.088	3.4662+3	2.9696+3	16.7	$\sim 3.0+3$
40	0.195	2.9552+3	2.2746+3	30.0	$\sim 2.6+3$
80	0.365	2.4850+3	1.8829+3	32.0	$\sim 2.3+3$

TABLE III. Comparison of Cross Sections as a Function of Fuel Region in the Energy Range 0.683 to 2.38 eV

Time, days	σ_a^{240} , barns			Source of Information
	Inner Rod	Middle Ring	Outer Ring	
0	3.0354+3	3.2183+3	3.8898+3	THERMOS
0	3.0354+3	3.2183+3	3.8898+3	Data (estimate)
10	2.7086+3	2.8918+3	3.5967+3	THERMOS
10	2.7086+3	2.8918+3	3.5967+3	Data (estimate)
40	1.9810+3	2.1462+3	2.8918+3	THERMOS
40	1.9810+3	2.1462+3	$\sim 3.4+3$	Data (estimate)
80	1.6027+3	1.7568+3	2.5116+3	THERMOS
80	$\sim 1.7+3$	$\sim 1.9+3$	$\sim 3.1+3$	Data (estimate)

and volumetrically averaged) and the result necessary for the data lies between the two.

The change of σ_a^{240} with position and time is shown in Table III. Also shown are the estimated σ_a^{240} values that would predict the experimental data. It is seen from the extreme spatial dependence of σ_a^{240} that even if the spatially averaged value from HRG had agreed with the data (Table III), it could not be used because it does not accurately describe any fuel regions.

REACTIVITY

The eigenvalue K_∞ is a measure of the change in reactivity that occurs because of changes in concentrations

during irradiation. The calculated K_∞ values as a function of time are listed in Table IV and plotted in Figure 7. The change in K_∞ due to the difference between the Pu^{241} and Pu^{242} experimental data and the theoretical data at the last experimental point is shown in Table V. The net reactivity effect is to underestimate K_∞ by 5.8 mk. Such a change in K_∞ is less than the uncertainty of a calculation of this type. Since the 5.8 mk discrepancy comes at the very end of the burnup range being studied, it is concluded that such errors in the concentrations of Pu^{241} and Pu^{242} would be insignificant in reactivity calculations.

*FIGURE 7. Reactivity Versus Exposure for
a Pu-Al Cluster Generating 1 MW Average
Power*

TABLE IV. Reactivity as a Function of Time for a Pu-Al Cluster

Time, days	Reactivity, K_{∞}	Time, days	Reactivity, K_{∞}
0	1.66125	50	1.43647
5	1.56769	60	1.40397
10	1.55187	70	1.37073
20	1.51945	80	1.33610
30	1.49042	90	1.30396
40	1.46122	100	1.26218
		110	1.21774

TABLE V. Reactivity Changes Due to Errors in the Concentrations of Pu²⁴¹ and Pu²⁴²

Percent Burnup	Fuel Region	Isotope	Concentrations, atoms/barn-cm		% Difference	ΔK_{∞}
			Theory	Experiment		
46.7	Inner	Pu ²⁴¹	4.48-5	4.91-5	8.8	-2.732-4
46.7	Inner	Pu ²⁴²	8.30-6	1.00-5	17.0	2.851-6
48.0	Middle	Pu ²⁴¹	4.70-5	5.04-5	6.7	-1.437-3
48.0	Middle	Pu ²⁴²	9.50-6	1.00-5	5.0	5.385-6
48.7	Outer	Pu ²⁴¹	5.28-5	5.64-5	6.4	-4.195-3
48.7	Outer	Pu ²⁴²	1.03-5	1.21-5	14.9	4.191-5

Measurements of reactivity as a function of exposure have been made⁽¹⁷⁾ but the results are not now in a form usable for comparison with this calculation.

CONCLUSIONS

There are two primary conclusions which can be made as a result of this correlation study. First, the cross sections as presented⁽¹⁰⁾ by Leonard do adequately predict experimental isotopic concentrations and effective cross section ratios for plutonium aluminum fuel as concluded earlier from the experimental data. Analytical results agreed with experimental results

up to approximately 33% burnup; and it is felt that the discrepancies for larger values of burnup may be attributed to the fact that THERMOS as used in this calculation is not adequate for cross section calculations involving the Pu²⁴⁰ resonance.

The second conclusion concerns the analytical methods used for the correlation. The HRG-THERMOS-Program S series of calculations produced a difference of 5.8 mk in K_{∞} due to differences between the computed and experimental concentrations of Pu²⁴¹ and Pu²⁴². Since the reactivity difference came at about 48% burnup and was zero below 33% burnup, the concentration

uncertainty is probably negligible for most reactivity calculations. It was also found that use of program THERMOS in the region of the Pu^{240} resonance allowed the computation of the experimental concentration of Pu^{241} up to the point where the atom ratio $\frac{\text{Pu}^{240}}{\text{Pu}^{239}}$ was 0.20. At larger values, the average Pu^{240} absorption cross section is computed too small using THERMOS and too large using Program HRG.

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