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HEAVY ELEMENT ISOTOPIC ANALYSIS OF UO_2 FUEL IRRADIATED IN THE VBWR

REPORT NO. 1

By
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GENERAL ELECTRIC
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Report #1

M. R. Hackney
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I. INTRODUCTION

Task C of the AEC Fuel Cycle Program (contract AT(04-3)-189, P.A. No. 11) provides for the study of long term reactivity characteristics of fuels irradiated in the Vallecitos Boiling Water Reactor (VBWR).

The uranium depletion and plutonium production have been determined experimentally by measuring the isotopic composition of the fuel. The measured quantities have been compared to calculated values in order to test the adequacy of the analytical model including the basic nuclear cross sections. Although direct measurement of basic cross sections is not feasible through experiments of this type, an excellent evaluation can be made of the reliability of currently used analytical methods in determining the nuclear performance of reactor fuel. As a result of the very successful results of this program, it is recommended that it be extended to increased fuel exposure, and that similar programs be undertaken in other operating reactors.

II. SUMMARY

Slightly enriched UO_2 fuel, irradiated in the Vallecitos Boiling Water Reactor (VBWR), with exposures ranging from 100 MWD/Ton to 3200 MWD/Ton has been analyzed for heavy element isotopic composition and compared to computed data.

The primary objective of this program (Task C-I) is to obtain improved data on the changes in nuclear characteristics with burnup of uranium oxide fuel in a boiling water reactor. This information is important in both evaluating the economics of a given reactor design and also in providing a sounder physics basis for improving reactor designs to minimize the resulting fuel costs.

Uranium oxide pellets, with an enrichment of 2.8 atom percent, were analyzed at several axial positions along the fuel rod, spanning the void (steam fraction) range of 0-30%.

The fuel exposure was determined by measuring the concentration of four fission products. The uranium content was determined by isotopic dilution mass spectrometry using U-233 as the spike, and the isotopic composition was simultaneously determined. The Pu-239/U-238 atom ratio was determined by a combination of gross alpha counting, alpha spectroscopic and mass spectrometric analyses. The higher plutonium isotope concentrations were obtained from the mass spectrometric analysis.

The isotopic composition for each pellet was computed, utilizing a general fuel cycle depletion code. Results of the analysis of the comparison of the measured and computed data indicate:

1. The total amount of plutonium computed is consistently lower than that implied from the measurement by approximately 10%.
2. The percentage difference between the measured and computed data increases slightly with exposure.
3. One rod was irradiated near a control rod which was approximately 25% inserted. As expected, since no control rod effects were

included in the calculation, the measured data in that region of the rod shows a greater plutonium production per MWD/T than computed.

Physical effects which might explain the small, but apparently consistent, differences between the measured and computed data have been postulated. It is concluded that the observed differences are the result of a substantial underestimate of void fraction and small uncertainties in fuel exposure and cross sections.

The calculational techniques used in this analysis are concluded to be relatively good for predicting the isotopic composition of oxide fuel in a boiling water reactor over this exposure range and for these operating conditions.

This study is continuing for extended exposures.

III. DESCRIPTION OF ELEMENT

Two Fuel Cycle elements of 2.8% enrichment with a water-to-fuel volume ratio of 4.0, designated 17J and 18J, are being used for this program. Each element contains 16 UO_2 fuel rods 0.375" in diameter with 0.022" Zr clad. The active fuel length is composed of 62 UO_2 pellets which are 0.60" in length. The UO_2 density is assumed to be 95% theoretical density. Each element contains five 0.313" diameter SS thimbles with a wall thickness of 0.016" which allows flux measurements to be made in five locations within the element (see sketch for location of thimble).

Wires for these measurements are inserted in 0.250" diameter SS guide tubes, which are positioned in the stationary thimbles. This arrangement allows insertion and withdrawal of the flux wires during reactor operation.

Figure 1 shows the location of these two elements in a typical VBWR core configuration. The VBWR contains many different types of fuel elements as a result of the many test programs underway. In an effort to irradiate the test elements for this program in as homogeneous a medium as possible, the surrounding elements are of a similar type, with the exception of the control rods. The top entry VBWR control rods have

CONTROL RODS

FIGURE 1 TYPICAL VBWR CORE CONFIGURATION

fuel followers, such that when the control rods are withdrawn fuel elements are inserted in the control rod position. Special care is taken to ensure that these test elements are always oriented in exactly the same way, such that the flux gradient across the element is unchanged.

IV. PRE-IRRADIATION MASS SPECTROMETRIC ANALYSIS

Prior to irradiation of these elements, a mass spectrometric analysis of ten sample pellets was made for U-234, U-235, and U-236 atom concentration. These pellets were randomly chosen from the same fuel lot (lot # JO3HB) used for fabrication of the fuel rods in the two elements. Spare rods were fabricated from this fuel lot and are stored for use as replacement rods. This allows careful control of the fuel used throughout the experiment. Results of this pre-irradiation isotopic analysis are shown below.

<u>Pellet #</u>	<u>Atom Percent</u>		
	<u>U-234</u>	<u>U-235</u>	<u>U-236</u>
1	.0175	2.802	.0209
2	.0165	2.805	.0212
3	.0168	2.803	.0203
4	.0180	2.796	.0220
5	.0160	2.802	.0201
6	.0170	2.802	.0210
7	.0170	2.803	.0208
8	.0159	2.802	.0205
9	.0162	2.802	.0209
10	.0161	2.795	.0208
average atom %	.0167 $\pm$.0005	2.801 $\pm$.002	.0209 $\pm$.0004

The errors are specified at the 95% confidence limit for the average.

It is very encouraging that there is essentially no difference between these ten pellets and that the precision of measurement is very high. The results from this analysis were used as reference data in the calculations; i.e., initial uranium concentrations.

V. IRRADIATION PROCEDURE

Irradiation of these elements in the VBWR began in November, 1961. Figure 2 shows the rod identification for each element during the first irradiation. The following rods have been removed for isotopic analysis.

1. After an average exposure of approximately 500 MWD/T, rods # R1H1 and R2H7 were removed from element 18J for isotopic analysis. These rods were replaced with rods # R5D3 and R5D1 from element 17J which had been irradiated in a similar flux. Replacement rods # R3D31 and R2D2 were then inserted in 17J.
2. After an average exposure of approximately 1000 MWD/T, fuel rods R3D31 and R2D2 were removed from element 17J for isotopic analysis. Replacement rods # R3D34 and R3D32 were then inserted in 17J.
3. After an average exposure of approximately 2000 MWD/T, fuel rod # R2H4 was removed from element 18J and replaced with replacement rod R3D35.

Additional rods will be removed from element 18J after extended exposures and replaced with similar irradiated rods from 17J. Element 17J will be used solely for replacement rods for 18J.

After fuel rod removal, the elements are replaced in the reactor in exactly the same orientation as during the previous irradiation. This plan allows, as nearly as possible, each rod to be irradiated in a constant flux with a constant gradient. All rods in the two elements, with the exception of the four corner rods, are removable. This allows removal of rods from twelve positions within the element during the program.

FIGURE 2 FUEL ROD IDENTIFICATION FOR ELEMENTS 17-J AND 18-J

VI. POWER AND VOID DETERMINATION

A. Gamma Scan of Irradiated Fuel Rod

After removal of the irradiated fuel rods from the element, they are transferred to the Radioactive Materials Laboratory (RML) for gamma scanning and preparation of samples for isotopic analysis.*

Figure 3 shows the gamma scan of the three fuel rods irradiated to average exposures of approximately 500, 1000, and 2000 MWD/T. Each curve is normalized to an average of 1.0. These gamma scans were obtained with a conventional scintillation counting system with the discriminator set to accept pulses from gamma rays having energies in excess of 0.60 MeV; i.e., primarily the 65 day Zr-95 activity. Figures 4, 5, and 6 are presented in order to show the detail in the actual gamma scan distribution. It is seen that each pellet along the rod can be located. The large depressions in the scan were found, by sampling those positions, to be cracked or broken pellets. Care was taken not to select a pellet for isotopic analysis either at or adjacent to this depression. From the normalized gamma scan distribution, it is seen that the peak-to-average activity is reduced from about 1.64 to 1.59 from an average exposure of 500 MWD/T to 1000 MWD/T. The peak activity for the 2000 MWD/T exposure is shifted slightly toward the bottom of the core. This can be explained by the fact that during the latest runs the adjacent control rod was about 25-30% inserted. It is, however, encouraging that the general power shape has changed only slightly for average exposures up to 2000 MWD/T.

B. Determination of Normalized Axial Power Distribution

During irradiation of each of the fuel rods detailed records were kept of the VBWR core power as a function of time. This power, from the daily operating log, was then averaged over each series of operating days between shutdowns, by weighting the power by time. Figures 7, 8 and 9 show these average VBWR core powers vs. time during the irradiation of each of the three fuel rods. A two-dimensional diffusion theory calculation for the entire VBWR core is made for each series of operating days; i.e., each VBWR run.⁽¹⁾ From this calculation the per cent of the total core power in 17J and 18J is obtained. The per cent of the element power in the rod of interest is obtained from a two-dimensional diffusion theory

* All RML work was done under the direction of R. F. Boyle.

FIGURE 3 RELATIVE ACTIVITY FROM GAMMA SCAN OF IRRADIATED FUEL ROD

FIGURE 4 GAMMA SCAN OF IRRADIATED FUEL ROD #1

FIGURE 5 GAMMA SCAN OF IRRADIATED FUEL ROD #2

FIGURE 6 GAMMA SCAN OF IRRADIATED FUEL ROD #3

FIGURE 7 VBWR CORE POWER vs TIME DURING IRRADIATION OF ROD 1

FIGURE 8 VBWR CORE POWER vs TIME DURING IRRADIATION OF ROD 2

FIGURE 9 VBWR CORE POWER vs TIME DURING IRRADIATION OF ROD 3

calculation, where each rod in 17J and 18J is specified discretely. Figure 10 shows the power distribution across the elements. This distribution was assumed to be constant over the entire irradiation period. This shows a difference in power across the element of 20-30%. From this calculation, the average power or power density for each rod of interest is known for each series of operating days. Then, utilizing the gamma scan activity distribution, the average power density for each pellet along the rod is determined for each series of operating days. Figures 11, 12, and 13 show this axial power density distribution for each of the three rods, for each series of operating days. This power density for each pellet is then utilized to determine the axial void and temperature distribution. This power density is not integrated with time to determine the fuel exposure. The measured fission product concentration is utilized for fuel exposure as described in Section VIII.

C. Normalized Axial Void Distribution

During irradiation of each of the fuel rods the pump flow rate was recorded on Recirculation Total Flow Circular Charts. This pump flow rate as a function of time was converted to core flow rate as shown in Figure 14.⁽²⁾ This accounts for the estimated bypass leakage. At very low pump flows, the core flow actually exceeds the pump flow slightly. This results from the natural circulation loop established within the reactor with the bypass leakage paths serving as down comers. During the fuel irradiations the flow was in general 15000 gpm or greater. From this detailed record of core flow vs. time, the average core flow for each series of operating days was determined by weighting the flow by megawatt hours.

An attempt was then made to determine the average flow rate in element 17J and 18J for each series of operating days. Element 18J is not in a channel and is unorificed. Element 17J is in a channel and contains a #6 type orifice. The ratio of element flow to core flow was assumed to be independent of power and core configuration. It is recognized that these assumptions are not totally correct, but these effects cannot be evaluated very precisely. The ratio of element flow to core flow is based on the hydraulic analysis of the core characteristics and knowledge of the pump

17J	15.0 KW	15.9 KW	16.6 KW	17.5 KW
	15.8	16.7	17.6	18.4
	16.5	17.6	18.5	19.2
	17.2	18.4	19.3	20.2
18J	17.9	19.0	20.2	21.2
	18.5	19.7	20.9	21.9
	19.0	20.3	21.5	22.5
	19.4	20.7	21.9	22.9

FIGURE 10
POWER PER FUEL ROD IN ELEMENTS 17-J AND 18-J
VBWR CORE POWER - 36 MW VBWR CORE FLOW RATE - 20,000 gpm

FIGURE 11 AXIAL POWER DENSITY DISTRIBUTION FOR ROD #1

FIGURE 12 AXIAL POWER DENSITY DISTRIBUTION FOR ROD #2

FIGURE 13 AXIAL POWER DENSITY DISTRIBUTION FOR ROD #3

FIGURE 14 CORE FLOW RATE vs PUMP FLOW RATE

flow rate. For reasons unique to this reactor, the element flow rate is very difficult to evaluate precisely. For instance, the hydraulic analysis does not include cross flow, which could be significant in the VBWR.

From this average element flow rate for each series of operating days and from the axial power distribution, the quality is computed as a function of axial position along the element from the following expression:

$$Q = \frac{P - W h_{\text{sub}}}{W h_{\text{fg}}}$$

P	=	power up to point (BTU/hr)
W	=	flow rate (#/hr)
h_{sub}	=	subcooling (BTU/#)
h_{fg}	=	heat of vaporization (BTU/#)

This axial quality distribution is then related to void as shown in Figure 15.⁽³⁾ This curve is based on Larson⁽⁴⁾ data for the higher qualities and then altered in the low quality regions in order to include subcooled boiling. The resulting axial void distribution for each rod for each series of operating days is shown in Figure 16, 17, and 18. The void fraction for each pellet, for each series of operating days, is then used to obtain cross sections as a function of void.

FIGURE 15 STEAM VOID FRACTION vs QUALITY

FIGURE 16 AXIAL VOID DISTRIBUTION FOR ROD 1

FIGURE 17 AXIAL VOID DISTRIBUTION FOR ROD 2

FIGURE 18 AXIAL VOID DISTRIBUTION FOR ROD 3

VII. CALCULATIONAL METHODS AND CROSS SECTION DETERMINATION

The isotopic atom per cent of U-234, U-235, U-236, and U-238, obtained from the pre-irradiation pellet analysis, was used to obtain a set of three group cross sections. Cross sections were obtained for 0, 10, 20, and 30% void and linearly interpolated for other void fractions.

Thermal cross sections are based on the energy dependent spectra and the spatial flux distribution of neutrons with a cylindricized single fuel rod cell. The energy variation of the spectra is based on the Wilkins heavy gas model, which utilizes a homogeneous description of the fuel rod unit cell. The spatial neutron distribution within the single rod cell, comprised of fuel, clad, and moderator, is based on the P-3 spherical harmonics approximation to the transport equation. A multigroup consistent P-1 fundamental mode calculation is used to determine the epithermal few group nuclear parameters and the epithermal spectrum. Geometric interaction and spatial distribution of nuclear events, which affect both the fast fission and resonance capture are accounted for in the analyses. The moderation by hydrogen is calculated directly and the heavy elements are treated by use of the Goertzel-Grueling approximation. The resonance capture is calculated on the basis of a multigroup model in which capture in individual resonances is determined by the infinite mass or narrow resonance approximation. The effective potential scattering is obtained by adding to the potential scattering cross section the effective reciprocal mean chord length of the rod, which includes the Dancoff effect.

The Doppler broadening is calculated from the equation shown below.

$$I = 1.1 + 30 \exp [\alpha(\sqrt{T} - \sqrt{293})] \sqrt{S/M + .077} \quad (5)$$

where: $1.1 = 1/v$ contribution
 $\alpha = 0.00696 - 0.000262 \text{ M/S}$
 $T = \text{fuel temperature}$
 $S/M = \text{effective surface to mass ratio of the } \text{UO}_2$

This equation is based on Arnold and Danneil's⁽⁶⁾ Monte Carlo analysis of the temperature dependence of the resonance integral and a fit to Hellstrands experimental measurements⁽⁷⁾ of the resonance capture of UO_2 rods. This

computed resonance integral as a function of average fuel temperature is shown in Figure 19. Recent analysis indicates that this procedure tends to slightly underestimate the Doppler effect.

Comparison of the calculated resonance capture effects of UO_2 with the measurements of Hellstrand indicates agreement to within about 0.5 barn for a S/M characteristic of this fuel (0.4).

A multidimensional fuel depletion analysis is utilized for depletion and production of isotopes. U-235, U-236, Np-237, U-238, Np-239, Pu-239, Pu-240, Pu-241, Pu-242, Am-243, Cm-244, I-135, Xe-135, Pm-149, and Sm-149 are isotopes which are specifically considered in the fuel depletion analysis. The other important fission products are grouped into five pseudo fission product isotopes which adequately describe the buildup, poisoning and depletion of the fission products. The fission product cross sections are obtained from Reference 8. This analysis is, however, quite insensitive to the fission product cross section. Each of the fissionable isotopes, U-235, U-238, Pu-239, and Pu-241, are provided with separate yields into each of the fission products. The non-linear, inhomogeneous differential equations describing the depletion and production of isotopes are numerically integrated by use of the Runge-Kutta-Gill procedure which is accurate to the fourth order. The resonance capture events are continually re-evaluated throughout exposure of the fuel and specific provisions are made to accurately account for the time and concentration dependent resonance self-shielding of the large, low-energy resonances.

The shielding effect on the resonance integrals is computed from the following equation:

$$I_{\text{eff}} = \frac{I_{\infty}}{\sqrt{1 + \frac{NI\gamma}{S + \sum_p}}}$$

where: I_{∞} = infinite dilution resonance integral
 N = atom concentration
 γ = shielding factor = $\left[\frac{2}{\pi} \frac{E_0}{T} \right]$

FIGURE 19 U-238 EFFECTIVE RESONANCE INTEGRAL vs AVERAGE FUEL TEMPERATURE

$$\begin{aligned}
 S &= \text{mean reciprocal chord length including Dancoff} \\
 &\quad \text{correction} \quad \left[S = \frac{S(1-\delta)}{4V} \right] \\
 \Sigma_p &= \text{potential scattering in fuel}
 \end{aligned}$$

This form for the computed Pu-240 resonance integral as a function of concentration has been compared to the measured resonance integrals of Nichols.⁽⁹⁾ These resonance integrals generally agree to within 5%. The self-shielding of other isotopes was computed in the same manner.

Shown in Tables 1 and 2 are the microscopic absorption and fission cross sections used in the analysis for the uranium and plutonium isotopes. The water temperature assumed is 285°C. The fuel temperature assumed for all resonance integral calculations except U-238 is 640°C.

The U-238 resonance integral as a function of average fuel temperature and void is shown in Figure 19.

TABLE 1
MICROSCOPIC ABSORPTION CROSS SECTIONS

Isotope	σ_{a_1}	$\sigma_{a_2}^*$	σ_{a_3}	σ_{a_1}	$\sigma_{a_2}^*$	σ_{a_3}	I_{∞}^{**}
	0 Void			20% Void			
U-234	0.9219	3.910	61.80	0.9137	3.843	60.87	627.8
U-235	1.694	15.77	372.5	1.698	15.81	366.6	221.7
U-236	0.4988	0.2822	4.120	0.4924	0.2773	4.058	295.0
U-238	0.1268	0.4048	1.595	0.1283	0.4082	1.571	264.2
Pu-239	3.400	30.49	1113.4	3.398	30.39	1130.1	230.4
Pu-240	0.3947	44.50	177.9	0.3981	43.83	176.8	8238.3
Pu-241	3.400	46.76	1133.8	3.398	46.43	1132.1	384.5
Pu-242	0.330	113.8	17.66	0.330	113.8	17.39	997.5

* σ_{a_2} includes the unresolved resonances and the smooth absorption in group 2.

** I_{∞} includes only the resolved resonances.

Group 1 Above 5.53 kev

Group 2 0.625 ev $\rightarrow$ 5.53 kev

Group 3 Thermal

TABLE 2

MICROSCOPIC FISSION CROSS SECTIONS

<u>Isotope</u>	<u>σ_{f1}</u>	σ_{f2}^*	<u>σ_{f3}</u>	<u>σ_{f1}</u>	σ_{f2}^*	<u>σ_{f3}</u>	<u>ν_1</u>	<u>ν_2</u>	<u>ν_3</u>
		<u>0 Void</u>			<u>20% Void</u>				
U-235	1.398	23.62	314.6	1.400	23.49	309.4	2.58	2.458	2.44
Pu-239	2.571	29.95	712.8	2.570	29.83	721.0	2.92	2.91	2.89
Pu-241	2.571	48.01	816.4	2.570	47.51	814.8	3.065	3.05	3.06

* σ_{f2} includes the resolved and unresolved resonances in group 2.

VIII. EXPERIMENTAL METHODS *

The experimental quantities determined in this work are based on the determination of the atom ratio of selected nuclides. In essence the concentration of the nuclides of interest is measured relative to that of U-238 which is assumed to remain constant during this exposure. This is a fairly good assumption for low enrichment UO_2 fuel since even on long exposure (10^4 MWD/T) the loss of U-238 is of the order of 1 per cent or less. In practice, the method of analysis involves the determination of the number of atoms of the nuclide and the U-238 per unit volume of dissolver solution.

In the first section below, the procedures and constants used in the burnup determinations will be given. The second section will be devoted to a discussion on the determination of Pu-239 to U-238 ratio and plutonium isotopic composition.

A. Burnup

The percent fission is defined as the ratio:

$$\text{atom percent fission} = \frac{\text{fissions per unit volume} \times 100}{\text{initial fissionable} + \text{fertile atoms per unit volume}}$$

The fissions per unit volume were obtained by measuring the number of atoms of a particular fission product nuclide (burnup indicator) formed during the irradiation. The initial fissionable plus fertile atoms per unit volume were obtained by measuring the number of U-238 atoms per unit volume. This quantity was then divided by the initial U-238 content (0.9716). Thus the above equation becomes:

$$\text{atom \% fission} = \frac{[\text{F.P.}] \times N_{238}^0 \times 100}{[\text{U-238}] \times \text{f.y.} \times \frac{\sum_{n=1}^i f_n (1 - e^{-\lambda t_n}) (e^{-\lambda t'_n})}{\sum_{n=1}^i f_n t_n}}$$

* The radiochemical separation was performed by G.L.Fox and F.R.Smith.
The mass spectrometric analysis was performed by J.H.Asay and E.L.Kramer.

where: [F.P.] = the fission product content per unit volume
 [U-238] = the U-238 content per unit volume
 N_{28}^0 = the initial U-238 atom fraction
 f.y. = the fission yield

The summation term is the in-pile and out-of-pile correction term where:

n = the number of exposure periods during the irradiation
 f_n = relative power levels of the reactor
 λ = decay constant of fission product
 t_n = in-pile time of period n
 t'_n = out-of-pile time from period n to counting time

In practice, the number of periods that were used for a given irradiation was determined by the half-life of the fission product and the length of irradiation. In Rod 1, one exposure period was used. For Rod 2, two exposure periods were used for the 65 day Zr-95 and one for 285 day Ce-144 and 29.2 y Cs-137. For Rod 3, three exposure periods were used for all these nuclides.

The atom percent fission was converted to MWD/T by multiplying by the factor 8700, which assumes an energy release of 205 MeV per fission.

1. Fuel Dissolution and Sample Preparation

The approximately 1/2 inch section of fuel was cold dissolved out of the cladding in about 50 ml of 8 M HNO_3 containing cesium, cerium, zirconium and barium carriers to retard sorption of these fission product elements onto the glassware. The dissolution occurred over a period of several hours. The cladding was left in the solution several hours after dissolution to ensure complete leaching of all fuel and fission products from the cladding. After mixing the solution, a few drops were pipetted into a volumetric flask and transferred to chemistry from the Radioactive Materials Laboratory (RML).

2. Fission Products and Radiochemistry

The fission products Cs-137, Ce-144, Zr-95 and Ba-140 were chosen as the burnup indicators. The amount of each in a known volume of solution, which was a dilution of the primary solution, was determined by various radiochemical techniques.

The Cs-137, Zr-95 and Ba-140 content was determined by means of the characteristic gamma radiation associated with the decay of each

species. The number of gamma ray events per unit time was determined by scintillation spectroscopy using a multi-channel pulse-height analyzer. The absolute counting rate (4π geometry) was determined by comparison of the samples with a Cs-137 and Co-60 standard having a known number of gamma ray events per minute, after making suitable corrections for photopeak efficiency and peak to total ratios.

The Cs-137 standard was used for the Cs-137 and Zr-95 samples. This standard was known to an accuracy of $\pm 1\%$ by comparison with other Cs-137 samples having a known number of atoms determined by isotope dilution mass spectrometry.⁽¹⁰⁾

The Co-60 standard was used for the Ba-140 samples. The accuracy of this standard is probably $\pm 2\%$.

The gamma ray events were converted to disintegrations per minute by dividing by the gamma ray intensity (γ/β).

The Cs-137 was chemically separated from the fission product-fuel mixture by standard radiochemical methods and precipitated as the chloroplatinate. The chemical yield was determined by weighing the precipitate and dividing by the weight which would be present for 100% yield for the 20 mg cesium carrier* added (before chemistry).

The Zr-95 and Ba-140 were not chemically separated. These were measured from a direct mount of a measured volume of the solution.

The Ce-144 was separated by scavenging by hydroxide precipitation and final precipitation as the oxalate. The chemical yield was determined as described above. The Ce-144 concentration was determined by counting the high energy beta particles from the rapidly equilibrated Pr-144 through 635 mg/cm^2 Al absorber.

A summary of the partial decay chains and the measured radiation is presented in Table 3. In the case of Zr-95, since the Nb-95 half-life is not very different from that of Zr-95, the system was not at equilibrium. Therefore, a correction was made for the Nb-95 contribution to the counting rate.

In Table 4, the values of the constants used in the determination of fissions per unit volume are tabulated. The fission yields used are those for U-235 thermal neutron fission.

TABLE 3

Partial Decay Chains and Measured Radiation of Burnup Indicators

<u>Decay Chain</u>	<u>Burnup Indicator</u>	<u>Measured Radiation</u>
I-137 24.4 sec Xe-137 3.9 min Cs-137 29.2 years Stable Ba-137	Cs-137	The .662 mev gamma ray from Cs-137 decay
Ce-144 285 days Pr-144 17.4 min Nd-144 5 x 10 ¹⁵ year	Ce-144	The 2.98 mev β^+ decay from Pr-144 in equilibrium with Ce-144
Zr-95 65 days Nb-95 35 days Stable Mo-95	Zr-95	The .723 and .760 mev gamma rays from Zr-95 decay and .768 mev radiation from the Nb-95 decay
Ba-140 12.8 days La-140 40.2 hrs Stable Ce-140	Ba-140	The 1.596 mev gamma ray in equilibrium with La-140 decay

TABLE 4

Constants Used for Burnup Indicators

<u>Nuclide</u>	<u>Half-Life</u>	<u>Fission Yield</u>	<u>γ-ray Intensity (γ/β)</u>
Cs-137	29.2 year	0.0615	0.82
Ce-144	285 days	0.056	—
Zr-95	65 days	0.060	0.99
Ba-140	12.8 days	0.064	0.88

3. Uranium

The uranium content per unit volume was determined by isotope dilution mass spectrometry. In this method a known number of U-233 atoms were combined with a measured volume of sample solution. Then, after chemically separating the uranium from the fission products, the uranium isotopic composition, including the U-233 spike isotope was determined. A knowledge of the number of U-233 atoms and the U-233 to U-238 ratio yields the U-238 content per unit volume.

The uranium was separated from the fission products (and plutonium) by converting a measured portion of the nitrate dissolve solution to HCl and passing through a 3 mm by 5 mm Dowex-1 anion column. After a suitable washing volume to remove the fission products and the plutonium, the uranium was eluted with dilute HCl. This was converted back to the nitrate for loading onto the mass spectrometer filaments.

For the analysis of Rod 1, the uranium sample size ranged from 20 to 50 micrograms. A Faraday cage was used to detect the UO_2^+ beam. The current was fed through a vibrating reed electrometer and read on a chart recorder. For the other uranium mass analyses performed, the sample size was reduced to about 100-300 nanograms. The U^+ beam was detected on a more sensitive electron multiplier and the signal fed through the same circuitry as previously described.

4. Discussion

The accuracy of burnup measured by a fission product to uranium ratio is limited by the accuracy with which the two independent measurements are made and by the constants involved. The error involved in the uranium determination is relatively small and is estimated to be $\pm 1-1/2\%$ (1σ). The majority of the error is involved with the radiochemical determination of the fission product and the nuclear constants required to convert counts per minute to fissions. A comprehensive discussion on the errors involved in each step of a Cs-137 determination is given in Reference 11 and will not be discussed further here. A similar treatment can be given for any other fission product. On the basis of this type of analysis and from the agreement

obtained on other samples the accuracy for the fission products is estimated to be $\pm 6\%$, $\pm 7\%$, $\pm 8\%$, and $\pm 10\%$ for Cs-137, Ce-144, Zr-95, and Ba-140, respectively, for the data presented here.

A severe limitation on the accuracy is the half-life. For this reason Ba-140 was used only for the initial series of samples (Rod 1). The Zr-95 half-life began to seriously limit its usefulness for the last rod analyzed. The Ce-144 is relatively long lived but its fission yield is extremely sensitive to the fissioning nuclide. The fission yield for U-235 and Pu-239 is 5.62 and 3.79%, respectively. For the last rod analyzed a correction was made to the U-235 fission yield to account for the Pu-239 fission contribution. This was obtained from the heavy element mass data (to be described in the next section). The Cs-137 usually yields the most accurate burnup data if the assumption can be made that no axial migration has occurred. This is usually a good assumption since most migration is in the direction of the largest temperature gradient which is in a radial direction. However, the Cs-137 data from Rod 2 consistently gave burnup values which were high by a factor of approximately two. Although numerous checks were made all values were consistently high by the same factor. The reason for this was not apparent. Another technique will be employed to check this when the Zr-95 decays to a low enough level.

B. Plutonium Isotopic Number Density

The plutonium in the dissolver solution for mass spectrometric analysis was passed through a small Dowex-1 anion column (3 mm by 5 mm). The plutonium, adsorbed on the column, was washed with 3 M HNO_3 which washed through the fission products and uranium. The plutonium was then eluted with 1 M HCl . This was converted to 0.05 M HNO_3 for loading onto the mass spectrometer filaments. The analyses of the first two rods were made on the double-stage mass spectrometer. For this instrument 50 nanogram samples were used. An electron multiplier, operated in the pulse counting mode, detected the Pu^+ beam.

The other samples were run on the single-stage instrument using the electron multiplier. The errors stated in both the uranium and plutonium are the standard deviation of the scans taken for each mass position.

The Pu-239 to U-238 ratio was obtained by a combination of alpha counting, alpha spectroscopy and mass spectrometry as follows. A measured volume of the sample solution was mounted and counted in a gas flow counter at 2π geometry. This count contained the contribution from all alpha emitters which included the uranium isotopes, Pu-238, Pu-239, Pu-240, Am-241 and Cm-242. The alpha spectrum yielded data on the count ratio of Pu-238 + Am-241 to Pu-239, 240 and Cm-242 to Pu-239, 240. (The energies of the Pu-238 and Am-241 as well as the Pu-240 were not resolved with the 35 keV half-width resolution obtained with the grid chamber.) The contribution of the Pu-238, Am-241 and Cm-242 was then subtracted from the gross alpha count. The uranium count rate (usually small or negligible) was obtained from the uranium mass spectrometer data. The Pu-240 count rate was obtained from the plutonium mass spectrometer data. Subtraction of these various components yielded the counts due to Pu-239 which was converted to atoms of Pu-239 per unit volume through the decay constant. (A half-life of 2.436×10^4 years was used.) An error of $\pm 5\%$ is estimated for the Pu-239/U-238 ratio.

The burnup was also calculated from the heavy element isotopic composition for the last two rods. The fission contribution from U-235 fission (the major source of fission) was calculated from the change in the U-235 and U-236 contents. The Pu-239 fission contribution was calculated from the Pu-240, Pu-241 and Pu-242 contents using a Pu-239 capture to fission ratio of 0.57. The U-238 fission contribution was estimated to be 3%. The error in the exposure based on this method was in the range of 6-8%; the small error is associated with the longer exposure.

A tabulation of all measured data is shown in Tables 5 and 6. The tabulated exposures are average values obtained by weighting the results obtained by the various methods inversely as the square of the error of each determination. All errors are at one standard deviation (1σ).

TABLE 5

MEASURED URANIUM ISOTOPIC COMPOSITION (ATOM %)

<u>Rod # 1</u>	<u>MWD/T</u>	<u>234</u>	<u>235</u>	<u>236</u>	<u>238</u>
Pellet 5*	467 ± 28	0.0179 ± .0009	2.784 ± .016	0.0333 ± .004	97.165
11	678 ± 35	0.016 ± .002	2.74 ± .01	0.0360 ± .001	97.21
16	712 ± 43	0.0174 ± .0002	2.731 ± .004	0.0375 ± .0003	97.214
23	730 ± 44	0.0173 ± .0001	2.717 ± .003	0.0378 ± .0003	97.227
31	598 ± 30	-----	2.75 ± .01	0.032 ± .001	97.22
41	403 ± 20	-----	2.79 ± .01	0.035 ± .002	97.17
52	188 ± 9	-----	2.79 ± .01	0.0254 ± .0005	97.18
58	101 ± 5	-----	2.82 ± .02	0.0254 ± .0008	97.15
<u>Rod # 2</u>	<u>x10³</u>				
Pellet 4	1.03 ± .06	0.0171 ± .0004	2.65 ± .04	0.0452 ± .0007	97.28
10	1.33 ± .08	0.0170 ± .0006	2.657 ± .005	0.0534 ± .0005	97.273
19	1.60 ± .10	0.0170 ± .0002	2.599 ± .006	0.0588 ± .0006	97.325
29	1.34 ± .08	0.0170 ± .0006	2.651 ± .007	0.055 ± .002	97.277
40	0.94 ± .06	0.0173 ± .0004	2.705 ± .009	0.0428 ± .0003	97.234
50	0.51 ± .03	0.0179 ± .0003	2.760 ± .011	0.0332 ± .0002	97.189
<u>Rod # 3</u>	<u>x 10³</u>				
Pellet 4	2.44 ± .12	0.0172 ± .0002	2.48 ± .01	0.0717 ± .0004	97.43
10	3.07 ± .15	0.0164 ± .0002	2.43 ± .01	0.089 ± .003	97.47
19	3.23 ± .16	0.0164 ± .0005	2.41 ± .01	0.0900 ± .0005	97.48
29	2.77 ± .14	0.0173 ± .0005	2.49 ± .01	0.0800 ± .0006	97.42
40	1.74 ± .09	0.0176 ± .0002	2.58 ± .01	0.0599 ± .0005	97.34
50	0.84 ± .04	0.0166 ± .0004	2.70 ± .01	0.0400 ± .0005	97.25

* Pellet number refers to the number of pellets from the bottom of fuel rod. The total number of pellets per rod is 62.

TABLE 6

MEASURED PLUTONIUM ISOTOPIC COMPOSITION (ATOM %)

Rod #	<u>Pu-239/U-238</u>	<u>239</u>	<u>240</u>	<u>241</u>	<u>242</u>
<u>Rod # 1</u>					
Pellet 5*	2.10 ± .105	98.968	1.015 ± .006	0.0173 ± .0004	--
11	3.16 ± .208	98.57	1.392 ± .013	0.0348 ± .001	--
16	3.78 ± .189	98.464	1.499 ± .005	0.037 ± .002	--
23	3.78 ± .189	98.407	1.500 ± .006	0.0425 ± .0006	--
31	3.10 ± .155	98.69	1.278 ± .019	0.033 ± .001	--
41	2.13 ± .106	99.08	0.904 ± .007	0.0160 ± .001	--
52	0.97 ± .049	99.56	0.433 ± .007	0.0050 ± .0003	--
58	0.50 ± .025	99.75	0.246 ± .006	...	--
<u>Rod # 2</u>					
Pellet 4	4.55 ± .228	97.439	2.477 ± .011	0.083 ± .002	--
10	6.35 ± .318	96.54	3.26 ± .03	0.193 ± .002	--
19	7.77 ± .389	95.972	3.759 ± .013	0.269 ± .003	--
29	7.19 ± .360	96.507	3.287 ± .014	0.206 ± .003	--
40	5.36 ± .268	97.636	2.257 ± .008	0.107 ± .004	--
50	2.82 ± .141	98.764	1.206 ± .006	0.030 ± .002	--
<u>Rod # 3</u>					
Pellet 4	10.2 ± .51	94.12	5.40 ± .03	0.471 ± .004	0.0100 ± .0007
10	13.6 ± .68	92.653	6.534 ± .009	0.790 ± .007	0.0228 ± .0005
19	15.2 ± .76	92.02	6.99 ± .02	0.958 ± .006	0.0299 ± .0005
29	13.8 ± .69	93.27	5.98 ± .02	0.729 ± .005	0.0192 ± .0004
40	9.55 ± .478	95.612	4.044 ± .009	0.339 ± .001	0.0054 ± .0005
50	5.43 ± .272	97.79	2.11 ± .01	0.094 ± .001	< 0.0007

* Pellet number refers to the number of pellets from the bottom of the fuel rod. The total number of pellets per rod is 62.

IX. COMPARISON OF MEASURED AND COMPUTED FUEL EXPOSURE

Figure 20 presents the comparison of the measured burnup and that implied from the gamma scan of the irradiated fuel rod as a function of axial position along the rod for the three rods.

This measured data has an uncertainty of $\pm 6\%$. In addition, the pellets sampled from rod 1 have an uncertainty of ± 1 pellet ($\sim 0.60''$) in the axial direction. This uncertainty was eliminated for rods 2 and 3 by removing all pellets and counting to obtain the desired pellet for sampling. These uncertainties are shown by the bars on the graph. The solid line represents that burnup implied from the normalized gamma scan of the irradiated fuel rod. As discussed earlier, this distribution is probably not a precise integrated exposure irradiation, since the activity is more characteristic of the latest exposure period. Additional uncertainties in this burnup distribution result from uncertainties in total reactor power and in the relative power in the sampled rod to total core power.

It is concluded then that the burnup implied from the scan of the irradiated fuel rod is not a precise indicator of the burnup, but does serve as a cross check on the measured burnup and is adequate for determining fuel temperature and voids. Therefore, for comparison of measured and computed plutonium and uranium data, the measured burnup has been used.

FIGURE 20 FUEL EXPOSURE AS A FUNCTION OF AXIAL POSITION ALONG ROD

X. COMPARISON OF MEASURED AND COMPUTED URANIUM ATOM FRACTIONS

Figure 21 shows the comparison of the measured and computed U-235 atom fraction as a function of measured fuel exposure. These data are based on an initial atom per cent of 2.801, as determined from the pre-irradiation analysis. The measured data has an uncertainty of less than 1%. The difference between the measured and computed data is generally no greater than 1% with only one data point exceeding 2%. Therefore, it is concluded that the computed data is generally within the accuracy of the measurement.

The comparison of the measured and computed U-236 atom fraction as a function of measured fuel exposure is shown in Figure 22. The initial concentration determined from the pre-irradiation analysis is 0.0209%. The measured data have an uncertainty of about 1-2%. The computed data is generally lower than that measured by 4-5%. It is then concluded that the computed U-236 atom concentration is within a few per cent of that measured.

The U-234 data is not included since the concentration remains essentially constant at about 1.6×10^{-4} over this range of exposures.

Since the accuracy of the measurement of U-235 and U-236 atom functions is quite good, this should also be a relatively good measurement of alpha-235 for the higher exposures. If α_{25} is defined as $\Delta N_{26} / (\Delta N_{25} - \Delta N_{26})$, this quantity is then computed for both the measured and computed data. α_{25} is computed for only the higher burnup rod; i.e., rod 3 since $\Delta 25$ and $\Delta 26$ involve small differences and the data are not as reliable for the lower exposures. The trend in the data is the same, however.

Shown below is α_{25} from the measured and computed data for rod 3 as a function of axial position along the fuel rod.

FIGURE 21 U-235 ATOM FRACTION AS A FUNCTION OF FUEL EXPOSURE

FIGURE 22 U-236 ATOM FRACTION AS A FUNCTION OF FUEL EXPOSURE

It is seen that the value of α_{25} from the measured data is consistently higher than that from the computed data. This could be due to either a harder spectrum than that used in the calculation or to a larger α in either the thermal or epithermal group than assumed in the calculation. The relatively low α observed at the bottom of the core may be statistically unresolved or could result from enhanced thermal flux resulting from the reflector.

IX. COMPARISON OF MEASURED AND COMPUTED PLUTONIUM ATOM FRACTIONS

Figures 23, 24, 25, and 26 present the comparison of the measured and computed Pu-239/U-238 , Pu-240/Pu-239 , Pu-241/Pu-240 and Pu-242/Pu-241 ratios as a function of measured fuel exposure. The comparison of the measured and computed total plutonium concentration is shown in Figure 27. From these comparisons, it is significant that with the exception of the Pu-242/Pu-241 ratio, the computed data are consistently lower than that implied from the measurement. Although the Pu-242 concentration was measurable for only the higher exposure pellets, i.e., rod 3, it is significant that the Pu-242/Pu-241 data are consistently high. This, of course, implies that the assumed capture cross section of Pu-241 is too high.

If an average difference between the measured and computed atom concentration is defined as $r = \frac{\sum x_i}{n}$, where x_i is (measured-computed)/measured and n is the number of data points, then r for each isotope is:

Pu-239	"	7 %
240	"	9 %
241	"	17 %

A. Discussion of Observed Differences

An effort has been made to determine physical effects which might explain these observed differences. Data is compared in terms of u , the mean square error, which is defined as $\sqrt{\frac{\sum x_i^2}{n}}$, where x_i is (measured-computed)/measured and n is the total number of data points.

One rod was irradiated adjacent to a control rod which was approximately 25% inserted. Since no control rod effects were included in the calculations, the pellets sampled in that region show large discrepancies; i.e., increased plutonium production per MWD/T. Since these differences can be explained by the control rod, data from the pellet near the control rod are discarded.

1. Assume that all discrepancies are due to uncertainties in cross sections used in the calculations. Then if σ_c^{28} and σ_c^{40} are increased 10% and σ_c^{41} is decreased about 10%, the computed data are more nearly that measured. The values of u are shown on page 56.

FIGURE 23 Pu-239/U-238 AS A FUNCTION OF FUEL EXPOSURE

FIGURE 24 Pu-240/Pu-239 AS A FUNCTION OF FUEL EXPOSURE

FIGURE 25 Pu-241/Pu-240 AS A FUNCTION OF FUEL EXPOSURE

FIGURE 26 $\text{Pu-242}/\text{Pu-241}$ AS A FUNCTION OF FUEL EXPOSURE

FIGURE 27 TOTAL PLUTONIUM AS A FUNCTION OF FUEL EXPOSURE

$\bar{u}$ (as computed)		$\bar{u}$ (capture x-sections changed)	
Pu-239	9% (-7%)		6% (+0.7%)
240	11 (-9%)		7 (+0.1%)
241	22 (-17%)		15 (-0.3%)

The number in parenthesis is the value of r as previously defined. Prior to changing the capture cross sections all computed plutonium atom concentrations are low when compared to the measurement. After changing the cross sections about one-half of the data points are higher and one-half are lower than that computed. Although changing the cross sections improves the data, it is not believed that all discrepancies are due to uncertainties in the cross section.

2. Assume that all discrepancies are due to an underestimate of the fuel exposure. Since the measured burnup has an uncertainty of about $\pm 6\%$, assume that the burnup is consistently low by 6% . The comparison of $\bar{u}$ is then as follows:

$\bar{u}$ (as computed)		$\bar{u}$ (burnup increased 6%)	
Pu-239	9% (-7%)		5% (-1%)
240	11 (-9%)		8 (+3%)
241	22 (-17%)		16 (+0.8%)

After this increase in fuel exposure, the Pu-239 is very slightly low, the Pu-240 is slightly high, the Pu-241 is generally half high and half low. Increasing the fuel exposure does appear to improve the computed data, when compared to the measurement, but it is unlikely that the measured burnup is consistently high by as much as 6% .

3. Assume that all discrepancies are due to uncertainties in the computed void fraction. Since neither the core flow rate nor the void as a function of flow rate is known precisely, it is quite likely that the void fraction used in the calculation is not precise. By extrapolating computed data, a void distribution was obtained which would

give the measured Pu-239 atom concentration. The discussion here concerns this analysis for only one rod, but the same analysis applies to each of the three rods. Shown below is the void distribution based on VBWR core flow estimates and that distribution which would give a calculated Pu-239 concentration equal to that measured.

The comparison of u for this rod is then shown below:

u (as computed)		u (increase void)
Pu-239	8% (-8%)	0%
240	13 (-12%)	6% (+0.7%)
241	24 (-22%)	9% (-4.7%)

With this increase in void fraction, the Pu-240 is slightly high and the Pu-241 is generally low. The flow rate required for this extrapolated void distribution is substantially lower than that used in the original calculation. Recent flow measurements made in various VBWR channels indicate significantly less flow in the element than that computed for a pump flow typical of that during irradiation of this rod. This then supports the theory that the void fraction in the calculation is too low since the flow rate appears to be too high. In addition, the method used in obtaining the average void fraction during the irradiation period tends to underestimate the voids slightly.

B. Cross Section Ratios Implied from Computed and Measured Data

For low exposures the isotopic plutonium concentrations may be approximated as follows:

$$N^{49} = N^{28} \sigma_c^{28} \phi t$$

$$N^{40} = N^{49} \sigma_c^{49} \frac{\phi t}{2}$$

$$N^{41} = N^{40} \sigma_c^{40} \frac{\phi t}{3}$$

$$N^{42} = N^{41} \sigma_c^{41} \frac{\phi t}{4}$$

From these expressions, cross sections ratios may then be implied from:

$$\sigma_c^{49} / \sigma_c^{28} = 2 \frac{N^{28} N^{40}}{(N^{49})^2}$$

$$\sigma_c^{40} / \sigma_c^{49} = \frac{3}{2} \frac{N^{49} N^{41}}{(N^{40})^2}$$

$$\sigma_c^{41} / \sigma_c^{40} = \frac{4}{3} \frac{N^{40} N^{42}}{(N^{41})^2}$$

These cross section ratios are then computed for the higher exposure rod using the computed and measured plutonium concentrations. Results are shown in Figure 28. It is noted that, with the exception of $\sigma_c^{41} / \sigma_c^{40}$ the differences between the measured and computed data increase toward the top of the core. This implies uncertainties in the void dependence of the U-238, Pu-240 and Pu-241 cross sections. It also implies that the uncertainty is about the same for the Pu-240 and Pu-241, since the differences are essentially constant with axial position. These data imply that the capture cross section in U-238 and Pu-240 should be increased about 15% and the capture in Pu-241 should be reduced about 15%.

FIGURE 28 CROSS SECTION RATIOS AS A FUNCTION OF AXIAL POSITION ALONG FUEL ROD

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