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Nondestructive Verification and Assay Systems for Spent Fuels

Technical Appendixes

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TABLE A-I
SPENT-FUEL DATA BASE

Reactor and Power	Type and Core	Burnup (MWd/tU) ^a		Reprocessor	No. of Batches Processed
		Mean	Range		
JPDR 12.5 MWe	BWR 1	3 816	1 362 5 449	PNC ^b - Tokai	12
Dresden-1 210 MWe	BWR 4	10 855	8 500 13 400	Nuclear Fuel Services West Valley	18
KRB 250 MWe	BWR 1	13 957	8 700 15 300	Eurochemic - Mol	21
Trino 257 MWe	PWR 1	12 306	9 900 13 400	Eurochemic - Mol	16
Sena 325 MWe	PWR 2	19 877	18 637 20 612	Eurochemic - Mol	12
Trino 257 MWe	PWR 2	20 864	14 800 22 200	Eurochemic - Mol	14
Yankee-Rowe 185 MWe	PWR 8	23 654	21 300 26 700	Nuclear Fuel Services West Valley	11

^aMWd/tU - megawatt day/tonne U; ranges are based on average burnup for accountability tank batch.

^bPower Reactor and Nuclear Fuel Development Corporation.

(²³⁹Pu, ²⁴¹Pu, and ²³⁵U). The relative difference is defined as:

$$\text{Relative Difference (\%)} = \left(\frac{\text{Predicted} - \text{Measured}}{\text{Measured}} \right) 100.$$

Because predicted values of plutonium isotopic abundances were not available for Sena core 2, comparisons of fissile plutonium and total fissile content could not be made for that reactor.

Values of the mean, standard deviation, and range for each parameter and each reactor core are presented in Table A-II. Mean burnup, burnup range, and the number of reprocessing batches also are listed in Table A-II. The results were also pooled by reactor type and for all reactors. The pooled data are presented in Tables A-III-A and B. Because of the variability exhibited by the JPDR data, a set of pooled results eliminating the JPDR values is also included. As expected, elimination of the JPDR data decreases the relative standard

deviations. The lack of predicted plutonium isotopic abundances for Sena core 2 decreases the number of data points used in the pooled comparisons of total fissile and fissile plutonium content.

The pooled data were tested for the presence of relative bias using Student's t test. Comparisons of the calculated t values with the critical values for $\alpha = 0.01$ and a two-tailed test support the following conclusions.

- There is no significant difference between the predicted and measured total uranium content.
- There is a significant positive bias between the predicted and measured total plutonium content.
- There is a significant negative bias between the predicted and measured ²³⁵U content.
- There is a significant positive bias between the predicted and measured fissile plutonium content for the pooled PWR and

TABLE A-II
SPENT-FUEL COMPOSITION
COMPARISON OF PREDICTED AND MEASURED VALUES

Reactor and Power	Type and Core	Burnup (MWD/tU) ^a		No. of Batches Processed	Relative Difference (%) ^{b,c}										Total Fissile	
					U		Pu		U ²³⁵		239Pu + 241Pu					
		Mean	Range		Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range		
JPDR 12.5 MWe	BWR 1	3 816	1 362 5 449	12	1.81 ±14.23	-25.45 35.15	11.16 ±22.34	-24.42 59.98	0.734 ±13.93	-25.49 33.28	10.49 ±22.60	-24.13 59.51	1.11 ±14.03	-25.30 34.30		
Dresden-1 210 MWe	BWR 4	10 855	8 500 13 400	18	-0.058 ±2.02	-5.19 2.12	0.19 ±4.24	-6.05 10.58	-7.14 ±3.19	-11.00 -0.84	-5.25 ±5.07	-11.68 7.44	-6.53 ±3.57	-10.24 1.55		
KRB 250 MWe	BWR 1	13 957	8 700 15 300	21	2.46 ±4.44	-3.06 15.64	5.67 ±5.69	-1.41 18.31	-0.037 ±4.52	-5.68 10.07	3.26 ±5.64	-4.14 16.79	0.81 ±4.39	-4.63 8.16		
Trino 257 MWe	PWR 1	12 306	9 900 13 400	16	0.57 ±2.14	-0.67 8.30	3.74 ±3.14	-3.33 9.43	-1.10 ±2.25	-3.31 5.79	4.39 ±2.97	-2.46 10.17	0.063 ±2.10	-1.72 6.79		
Sena 325 MWe	PWR 2	19 877	18 637 20 612	12	-1.16 ±1.21	-2.86 0.52	-1.62 ±3.82	-9.09 6.18	-6.02 ±3.76	-16.84 -2.93	-	-	-	-		
Trino 254 MWe	PWR 2	20 864	14 800 22 200	14	0.23 ±1.23	-1.38 2.97	7.11 ±2.89	-0.56 10.99	-6.17 ±3.11	-11.36 -1.11	6.68 ±2.89	-0.93 10.62	-2.61 ±2.11	-6.16 1.31		
Yankee-Rose 185 MWe	PWR 8	23 654	21 300 26 700	11	0.54 ±1.74	-3.44 2.81	4.22 ±2.74	-0.60 9.58	-1.17 ±2.85	-5.97 3.03	6.27 ±2.55	1.15 10.69	0.51 ±2.24	-2.92 4.10		

^aMWD/tU - megawatt day/tonne U; ranges are based on average burnup for accountability tank batch.

^b $\frac{\text{Predicted} - \text{Measured}}{\text{Measured}} \times 100$

^cThe ± values are relative standard deviations in per cent.

for the pooled PWR and BWR data (with and without JPDR). Variability in the JPDR data masks possible biases between the predicted and measured data for the BWRs. A negative bias is, in fact, obtained between the predicted and measured fissile plutonium content for the BWRs if the JPDR data are eliminated.

- The pooled data for PWRs and BWRs, excluding the JPDR data, show a significant negative bias for the total fissile content.

IV. DISCUSSION

It is reasonable that for all except the JPDR data the significant differences observed between reactor operator's and reprocessor's values are caused primarily by inaccurate reactor predictions. This is reasonable because the analytical chemistry and mass spectrometric methods should be more accurate and precise than is indicated by the observed differences, and, for all but the JPDR data, there are independent confirmatory measurements that agree well with the reprocessor's data.

Under the hypothesis that most of the observed differences are caused by inaccurate burnup calculations, we restate the conclusions of this study as follows.

- Total uranium content is predicted with an insignificant bias of 1% or less and a variability (1σ) of 2-4%.

- Total plutonium content is predicted with a positive bias of 3-5% and a variability (1σ) of 4-6%.
- Fissile uranium (²³⁵U) content is predicted with a negative bias of 3-4% and a variability (1σ) of 4-6%.
- Fissile plutonium (²³⁹Pu + ²⁴¹Pu) is predicted with a bias of -1 to +6% and a variability (1σ) of 3-7%.
- Total fissile content is predicted with a negative bias of 1-3% and a variability (1σ) of 3-6%.

In most cases the larger values of the ranges quoted for accuracy and precision are associated with BWRs and the lower values with PWRs. Also, there are probable outliers in the data; for example, considering only total uranium and neglecting the JPDR data, a range of ~10% is observed in the PWR data and ~20% in the BWR data. Differences might be even larger for individual fuel assemblies because each reprocessing batch consists of two or more assemblies so that there is some averaging of the assembly data.

The data base used for this study does not include any data for large, modern nuclear power plants because fuel from such plants currently is not being reprocessed. Modern fuels are being designed for higher burnups. The conclusions of this study should be re-examined when burnup data for modern fuels become available.

TABLE A-III
SPENT-FUEL COMPOSITION
POOLED COMPARISONS OF PREDICTED AND MEASURED VALUES

A: Uranium, Plutonium

Reactor Type	Number of Batches	Burnup ^a MWD/tU		Relative Difference (%) ^b					
				U		Pu		235U	
		Mean	Range	Mean	Range	Mean	Range	Mean	Range
BWRs (JPDR, Dresden and KRB)	51	10 476	1 362 15 300	1.42 ±7.42	-25.45 35.15	4.90 ±12.16	-21.42 59.98	-2.36 ±8.19	-25.49 33.28
BWRs (Dresden and KRB)	39	12 525	8 600 15 300	1.30 ±3.72	-5.19 15.64	2.97 ±5.82	-6.05 18.31	-3.32 ±5.31	-11.00 10.07
PWRs	53	18 636	9 900 26 700	0.081 ±1.75	-3.44 8.30	3.52 ±4.37	-9.09 10.99	-3.57 ±3.84	-16.84 5.79
BWRs and PWRs	104	14 634	1 362 26 700	0.74 ±5.36	-25.45 35.15	4.19 ±9.05	-21.42 59.98	-2.98 ±6.36	-25.49 33.28
BWRs and PWRs (excluding JPDR)	92	16 045	8 500 26 700	0.60 ±2.81	-5.19 15.64	3.29 ±5.01	-9.09 18.31	-3.45 ±4.50	-16.84 10.07

B: Fissile Pu and Total Fissile

Reactor Type	Number of Batches	Burnup ^a MWD/tU		Relative Difference (%) ^b			
				239Pu + 241Pu		Total Fissile	
		Mean	Range	Mean	Range	Mean	Range
BWRs (JPDR, Dresden and KRB)	51	10 476	1 362 1 530	1.96 ±13.07	-21.43 59.51	-1.71 ±8.26	-25.30 34.30
BWRs (Dresden and KRB)	39	12 525	8 500 15 300	-0.67 ±6.84	-11.68 16.79	-2.58 ±5.44	-10.24 8.16
PWRs (excluding Sena)	41	18 273	9 900 26 700	5.68 ±2.96	-2.46 10.69	-0.73 ±2.50	-6.16 6.79
PWRs and BWRs (excluding Sena)	92	13 951	1 362 26 700	3.61 ±10.06	-24.13 59.51	-1.27 ±6.36	-25.30 34.30
PWRs and BWRs (excluding Sena and JPDR)	80	15 471	8 500 26 700	2.58 ±6.09	-11.68 16.79	-1.63 ±4.27	-10.24 6.79

^aMWD/tU = megawatt day/tonne U; ranges are based on average burnup for accountability batches.

^b $\left(\frac{\text{Predicted} - \text{Measured}}{\text{Predicted}} \right) 100$.

Based on the results of this study, NDA measurements on spent fuel that are accurate and precise to 5-10% (1σ) or better should be useful for quantitative verification of "typical" spent fuel, that is, fuel with normal irradiation history. Such measurements should also be useful for characterizing "atypical" fuel, that is, fuel with inhomogeneous burnup or unusual irradiation history.

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APPENDIX B

GE-MORRIS EXPERIMENTAL PROGRAM

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Safeguards Assay Group Q-1

I. INTRODUCTION

The effectiveness and operational acceptability of new measurement systems must be demonstrated in actual nuclear facilities. To develop and demonstrate the capabilities of nondestructive measurement systems for spent fuel, prototype systems are being tested at the GE-Morris Operation Spent-Fuel Storage Facility. The initial measurement system was installed at GE-Morris, and the first series of experiments was carried out during the fall of 1980. The results of this first series of measurements are described here.

The measurement system currently installed at GE-Morris is being upgraded for a second series of experiments scheduled for the summer of 1981. Concurrently, based on the results of our laboratory and field experience, we are designing a second prototype system that will incorporate spectral gamma-ray and active neutron measurements. This second system is tentatively scheduled for installation at GE-Morris in mid-1982, following checkout and testing at Los Alamos.

II. EXPERIMENTAL APPARATUS

A spent-fuel measurement system was installed at the GE-Morris Operation Spent-Fuel Storage Facility to evaluate the applicability of passive gamma-ray and neutron measurement techniques to the verification of irradiated fuel assemblies at a spent-fuel storage facility. A schematic of the system initially installed during October 1980 is shown in Fig. B-1. The positioning mechanism (V-shaped device) automatically centers the fuel assembly as the crane moves in to the wall. This geometry was selected because it maintains the same source-to-detector distance for both pressurized-water-reactor (PWR) and boiling-water-reactor (BWR)

fuel assemblies. Figure B-2 shows a representation of a BWR fuel assembly located in the positioning mechanism. The detector pipe is located at the corner of the fuel assembly to ensure a symmetric measurement geometry. The first detector package (Fig. B-3) consisted of three detectors: (1) an ion chamber to measure gross gamma dose, (2) a ^{235}U fission chamber ($\sim 120\text{ mg } ^{235}\text{U}$) to measure the neutron emission rate, and (3) a $\text{Be}(\gamma, n)$ detector to record the high-energy ($>1.66\text{-MeV}$) gamma signature.

Fig. B-1.
Positioning mechanism for prototype
verification and assay system.

DETECTOR TUBE

Fig. B-2.
Relative position of the fuel assembly
and the detector tube.

The Be(γ ,n) detector is shown in Fig. B-4. High-energy gamma rays (>1.66 MeV) interact with beryllium, producing neutrons that are thermalized in the polyethylene and detected using a small ^{235}U fission chamber (~ 35 mg ^{235}U). The principal source of high-energy gamma rays is the 2186-keV gamma ray from the decay of ^{144}Pr . The half-life of ^{144}Pr is only 17.28 min; however, it is in secular equilibrium with its parent ^{144}Ce ($t_{1/2} = 284.5$ days). For fuel assemblies with long cooling times (~ 7 yr), this signature probably would not be detectable. For fuel assemblies with short cooling times, this high-energy signature would be difficult to simulate and therefore would be a good indication that there had been no tampering with the fuel.

III. EXPERIMENTAL RESULTS

The spent-fuel measurement system installed in the unloading pool at GE-Morris is shown in Fig. B-5. Spent-fuel assemblies from four reactors were examined using fission chambers to measure neutron emission rates and ion chambers to measure gamma doses. There were four objectives of these initial measurements.

- (1) Reproducibility: How precise could replicate measurements be made using the V-shaped positioning fixture? Also, could the errors covered by positioning and counting statistics be separated? Which of these two error sources is dominant?

Fig. B-3.
Detector package used in the
GE-Morris field exercise.

- (2) Angular Effects: Are the neutron and gamma-ray measurements at all four corners of the spent-fuel assembly consistent?
- (3) Evaluation of Empirical Correlations: Does the empirical relationship between neutron emission rate and declared exposure apply to fuel assemblies from different reactors? Can the neutron emission rate be corrected for decay of the ^{244}Cm isotope?
- (4) Fuel-Assembly Geometry: What is the effect on measurements of variations in fuel geometry--PWR fuel assemblies with 14 by 14 and 15 by 15 arrays, and BWR fuel assemblies with 7 by 7 arrays?

To address these questions, we measured a total of 17 spent-fuel assemblies, 16 PWR assemblies and 1 BWR assembly, from four power reactors.

Fig. B-4.
Be(γ ,n) detector that is sensitive to only high-energy (>1.66 MeV) gamma rays.

Table B-I summarizes the parameters of the fuel assemblies. The specific fuel assemblies that were measured are described in Table B-II.

A. Reproducibility

The positioning mechanism consisted of a V-shaped holder with two limit switches, one on each side of the V, to indicate when the fuel assembly was in position. Two types of positioning procedures were investigated for assembly number B-C032: (1) measurement when both limit switches indicated contact and (2) measurement when the assembly was placed firmly against the V-shaped

Fig. B-5.
Spent-fuel measurement system installed in the GE-Morris unloading pool.

positioning mechanism. Each of the four corners of B-C032 was measured six times using both procedures (limit switch and fixed). Following each measurement, the fuel assembly was removed from the positioning mechanism and realigned to simulate an independent measurement. Results of these replicate measurements (Table B-III) indicate that better reproducibility is obtained when the fuel assembly is placed firmly against the positioning fixture, as compared with using the limit switches alone to indicate correct positioning. In the optimum procedure the limit switches were used to indicate when the fuel assembly was getting close to the correct position, and final positioning was confirmed by visual inspection. This procedure provided measurement reproducibility of 1-2%.

Following the reproducibility tests, the beryllium sleeve was removed from the Be(γ ,n) detector to ensure that photoneutrons produced in the beryllium did not contribute to the observed counts in the fission chamber. The fission chamber of the Be(γ ,n) detector was left in the detector assembly, so that two simultaneous neutron measurements could be made for each fuel assembly. Additional investigations are being conducted to quantify the effect of the Be(γ ,n) detector on the

TABLE B-I
FUEL ASSEMBLIES EXAMINED AT GE-MORRIS FACILITY

Reactor Designation	Rated Output (Reactor Type)	Number of Assemblies	Fuel Geometry	Fuel Diameter (cm)	Active Length (cm)	Cladding Material	Fuel Rods/Assembly	²³⁵ U Enrichment (%)	Declared Exposure (GWD/tU)	U per Fuel Assembly (kg)
A	510 MWe (PWR)	4	15x15	0.97	309.4	Zr	204	3.02-3.67	10.8-26.0	419.7
B	450 MWe (PWR)	3	14x14	0.93	366	Zr	179	3.4	23.6-40.7	443.7
C	715 MWe (BWR)	1	7x7	1.24	366	Zr	49	2.13	5.1	169.8
D	430 MWe (PWR)	9	14x14	0.97	305	SS	180	3.4-4.0	26.5-32.4	318.5

TABLE B-II
INDIVIDUAL FUEL ASSEMBLIES

Fuel Assembly	Discharge Date	²³⁵ U Enrichment (%)	Declared Exposure (GWD/tU)
A-A019	4/17/70	3.02	19.7
A-C049	4/17/70	3.67	10.8
A-A045	4/17/70	3.02	19.5
A-B012	4/16/71	3.23	26.0
B-C056	3/4/77	3.397	40.7
B-C032	4/6/74	3.400	23.6
B-C052	3/4/77	3.397	32.8
C-DN201	4/72	2.13	5.1
D-A051	12/28/72	3.404	26.7
D-C004	6/21/73	3.865	26.8
D-D001	3/14/75	4.005	31.4
D-C001	6/2/73	3.865	26.5
D-A036	12/28/72	3.404	26.5
D-D020	3/14/75	4.005	26.9
D-A009	12/28/72	3.404	26.8
D-D046	3/14/75	4.005	32.3
D-C020	6/2/73	3.865	32.4

passive neutron measurements for the specific detector design. The fission chamber in the Be(γ,n) detector contained 58 mg 93% ²³⁵U, whereas the larger fission chamber contained 165 mg.

A third technique for positioning the fuel assemblies was evaluated in which the gross gamma activity was monitored using the ion chamber as the fuel assembly was moved into the positioning mechanism. This signal would reach a maximum when the fuel assembly was closest to the detector package. As the fuel-assembly grapple was moved further, the fuel assembly would pivot away from the detector causing the gross gamma signal to drop. This technique was found to compare favorably with the optimum procedure for positioning and eliminated any influence on the measurements arising from different operators.

The next series of measurements was performed to quantify the reproducibility of measurements for various fuel assembly designs. Two 14 by 14 PWR, one 15 by 15 PWR, and one 7 by 7 BWR fuel assemblies were each measured a minimum of six

TABLE B-III
COMPARISON OF MEASUREMENTS USING LIMIT SWITCHES AND FIXED POSITION - FUEL ASSEMBLY B-C032

Orientation	Fission Chamber Limit		Ion Chamber Limit		Be(γ,n) Detector Limit	
	Switch (%)	Fixed (%)	Switch (%)	Fixed (%)	Switch (%)	Fixed (%)
A	5.5	1.0	4.0	0.9	5.5	2.4
B	4.3	0.8	2.6	0.0	4.0	1.9
C	5.0	1.7	4.0	0.8	5.3	2.4
D	5.6	2.3	4.1	0.8	6.5	3.1
Average	5.1 (0.9) ^a	1.5 (0.9) ^a	3.7 (0.3) ^a	0.6 (0.3) ^a	5.3 (1.5) ^a	2.5 (1.4) ^a

^aExpected errors from counting statistics for neutron measurements. The ion chamber measures analog current. Errors quoted for the ion chamber correspond to a one-unit change in the least significant digit.

times at each corner. The fuel assembly was repositioned for each measurement. For the three PWR fuel assemblies, the variation of neutron measurements was 1.1% and the variation of the ion chamber measurements was 0.6% when the optimum (fixed) positioning procedure was used. Also, during this series of measurements, at least four different fuel-handling operators performed the positioning without adversely affecting the measurements. In Table B-IV the estimated variations (1σ) caused by counting statistics are given in parentheses. By comparing these separate components of variability, a significant proportion of the total measurement uncertainty can be explained by the counting statistics.

The B-C032 fuel assembly was selected as our "standard" fuel assembly. It was measured on each of the 5 days during which measurements were performed (Table B-V). An uncertainty of 2-3% was obtained for the neutron measurements and 1.4% for the ion chamber measurements. This fuel assembly will also be measured during future exercises at GE-Morris to determine measurement reproducibility over extended time periods (several months).

TABLE B-IV
REPRODUCIBILITY OF MEASUREMENTS
FOR VARIOUS FUEL-ASSEMBLY GEOMETRIES

Fuel Assembly	Orientation (corner)	Number Measurements	Fission Chamber (s)	Ion Chamber (s)	Fission Chamber ^a (s)
B-C032	A	6	0.69	0.48	0.73
	B	6	0.50	0.19	1.46
	C	6	0.84	0.68	0.59
	D	6	0.35	0.43	1.39
			0.60 (0.86) ^b	0.45	1.04 (1.30) ^b
B-C056	A	12	1.77	1.19	2.80
	B	12	2.56	1.52	3.78
	C	6	1.07	0.46	0.65
	D	12	1.08	0.48	0.77
			1.62 (0.54) ^b	0.91	2.00 (0.81) ^b
A-B012	A	7	1.04	0.44	1.35
	B	6	0.45	0.25	1.46
	C	6	1.15	0.62	1.68
	D	6	1.23	0.76	1.94
			0.97 (0.83) ^b	0.52	1.61 (1.21) ^b
C-DN201	A	6	8.29	1.31	5.88
	B	6	5.52	0.33	5.26
	C	6	10.74	1.05	9.38
	D	6	6.29	1.32	8.11
			7.71 (4.36) ^b	1.00	7.16 (7.27) ^b

^aFission chamber from Be(Y,n) detector.

^bEstimated uncertainty caused by counting statistics.

During this measurement exercise, we monitored the backgrounds for each of the three detectors. The results for 108 background measurements are shown in Table B-VI. As in Table B-IV, the variation caused by counting statistics is of the same magnitude as the actual measured variation.

The experimental measurements show that reproducible measurements can be obtained using the presently designed V-shaped positioning fixture. Counting statistics appear to be the major source of variation in the measurements. After this was established, we proceeded with the investigation of the other objectives.

B. Angular Effects

The second objective was to quantify angular burnup effects by measuring different corners of the spent-fuel assemblies. The results of the reproducibility studies were combined with additional measurements made on all 17 spent-fuel assemblies. The results are given in Table B-VII. The neutron emission rate can vary by 30-40%, depending on the specific corner measured. Similar variability is observed in the ion chamber results.

TABLE B-V
REPRODUCIBILITY OF MEASUREMENTS FOR
STANDARD FUEL ASSEMBLY B-C32

Date	Fission Chamber (counts/s)	Ion Chamber	Fission Chamber ^a (counts/s)
10/30/80	175.9	421	75.8
	174.2	419	76.1
	174.6	416	75.8
	174.7	41	74.1
	174.9	417	75.4
10/31/80	174.2	419	77.4
	174.76 ± 0.6	418 ± 1.8	75.78 ± 1.05
	167.0	411	72.1
	162.8	411	73.4
11/1/80	170.9	416	74.7
	175.6	416	75.3
11/2/80	163.5	399	68.2
	170.2	411	71.9
	170.6	411	71.5
	168.6	411	72.1
11/3/80	172.2	420	76.1
	171.1	420	75.1
	171.3	420	74.3
	Mean	171.3	74.0
	Std Dev	4.0 (2.3%)	5.6 (1.4%)
			2.3 (3.1%)

^aFission chamber from the Be(Y,n) detector.

Because the measurement reproducibility (or precision) is much better than this, the angular effects appear to be real.

The results from these angular effects measurements are presented in Figs. B-6 through B-9 for reactors A, B, C, and D, respectively. The data were corrected for the decay of ²⁴⁴Cm (t = 18.11 yr) because of the differences in cooling times. No correction was included for the ²⁴²Cm decay because its contribution to the measured neutron rates was insignificant. The shortest cooling time was 1340 days for fuel assemblies B-C056 and B-C052. Curium-242 has a half-life of 162.8 days, therefore ²⁴²Cm has decayed by a factor of 0.003.

The maximum variability of the measurements for each of the four corners for the 17 fuel assemblies is given in Table B-VIII. Each of the data sets from the PWR fuel assemblies had a maximum difference of ~40% for at least one fuel assembly. This high variability does not appear to be related to just a single reactor but rather may be a general characteristic of PWR fuel assemblies. The 18% difference found in the BWR fuel assembly was not unexpected because of the cruciform control blades used in BWRs.

TABLE B-VI
BACKGROUND MEASUREMENTS

Date	Time	Number of Measurements	Average Values		
			Fission Chamber (counts/s)	Ion Chamber	Fission Chamber ^a (counts/s)
10/29/80	16:21-16:25	3	1.05	-7.7	
10/30/80	8:11-8:52	6	1.07	-8.0	
	12:20-12:41	5	1.05	-8.0	0.21
	14:21-14:25	2	0.95	-8.0	0.23
	16:36	1	1.00	-8.0	0.18
10/30/80- 10/31/80	16:38-7:53	69	1.02	-8.0	0.21
10/31/80	8:25-8:37	7	1.02	-8.0	0.20
11/1/80	7:47-7:52	3	1.05	-8.0	0.18
	8:26-8:28	2	1.08	-8.0	0.22
	17:36	1	0.92	-8.0	0.27
11/2/80	10:55	1	1.08	-8.0	0.23
	15:07	1	1.07	-10.0	0.20
	16:39	1	0.92	-10.0	0.23
	17:29	1	0.90	-8.0	0.19
	18:29	1	0.96	-8.0	0.15
	19:25	1	1.08	-8.0	0.15
	20:27	1	0.85	-8.0	0.15
11/3/80	10:13-10:14	2	0.82	-8.0	0.24
Weighted average		108	1.02 ± 0.07	-8.0	0.21 ± 0.04
Precision			7%		19%
Ranges			0.85 ~ 1.08	-7.0 to -10.0	0.15 ~ 0.24
Uncertainty caused by counting statistics			10%		22%

^aFission chamber from the Be(γ,n) detector.

TABLE B-VII
NORMALIZED RESPONSES FOR EACH OF THE SPENT-FUEL ASSEMBLIES

Fuel Assembly	Orientation (corner)	Fission Chamber (counts/s)	Ion Chamber	Fission Chamber ^a (counts/s)
A-A019	A	1.00	1.00	1.00
	B	1.02	1.01	1.03
	C	1.02	1.03	1.02
	D	0.99	1.02	0.99
A-C049	A	1.00	1.00	1.00
	B	0.63	0.57	0.62
	C	0.60	0.51	0.59
	D	0.97	0.95	0.94
A-A045	A	1.00	1.00	1.00
	B	1.03	1.01	1.04
	C	1.02	1.04	1.03
	D	0.94	0.98	0.93
A-B012	A	1.00	1.00	1.00
	B	1.04	1.02	1.02
	C	0.98	0.97	0.98
	D	1.00	0.99	0.99

TABLE B-VII (cont.)

Fuel Assembly	Orientation (corner)	Fission Chamber (counts/s)	Ion Chamber	Fission Chamber ^a (counts/s)
B-C056	A	1.00	1.00	1.00
	B	1.03	1.01	1.02
	C	0.98	1.01	0.98
	D	0.91	0.93	0.92
B-C032	A	1.00 (1.00) ^b	1.00 (1.00)	1.00 (1.00)
	B	0.72 (0.72)	0.70 (0.70)	0.74 (0.71)
	C	0.85 (0.84)	0.83 (0.83)	0.86 (0.82)
	D	1.25 (1.22)	1.12 (1.12)	1.24 (1.19)
B-C052	A	1.00	1.00	1.00
	B	0.95	0.85	0.95
	C	1.37	1.22	1.35
	D	1.30	1.24	1.31
C-DN201	A	1.00	1.00	1.00
	B	0.90	0.88	0.89
	C	0.82	0.81	0.75
	D	0.88	0.86	0.87
D-A051	A	1.00	1.00	1.00
	B	0.97	0.96	0.97
	C	1.07	1.03	1.08
	D	1.00	1.01	1.03
D-C004	A	1.00	1.00	1.00
	B	0.77	0.83	0.77
	C	0.90	0.90	0.88
	D	1.21	1.11	1.19
D-D001	A	1.00	1.00	1.00
	B	1.00	0.98	1.00
	C	1.05	1.02	1.06
	D	1.03	1.02	1.03
D-C001	A	1.00	1.00	1.00
	B	1.16	1.09	1.16
	C	0.88	0.92	0.90
	D	0.76	0.80	0.78
D-A036	A	1.00	1.00	1.00
	B	0.94	0.98	0.95
	C	0.89	0.93	0.89
	D	0.95	0.95	0.94
D-D020	A	1.00	1.00	1.00
	B	0.87	0.94	0.89
	C	0.72	0.76	0.74
	D	0.85	0.90	0.87
D-A009	A	1.00	1.00	1.00
	B	0.99	0.99	0.99
	C	1.01	1.00	1.03
	D	0.96	0.97	0.97
D-D046	A	1.00	1.00	1.00
	B	0.92	0.95	0.93
	C	1.00	0.98	0.98
	D	1.05	1.02	1.06
D-C020	A	1.00	1.00	1.00
	B	1.14	1.08	1.11
	C	1.32	1.18	1.28
	D	1.18	1.11	1.15

^aFission chamber from the Be(γ,n) detector.^bNumbers in parentheses are for a second set of data taken at the end of the exercise.

MORRIS EXERCISE - CORRECTED EXPERIMENTAL RESULTS
PWR - 15x15 Assemblies - 510 MWe Rated Power
Exposure Range 10.8-26.3 GWd/tU

Fig. B-6.
Relative neutron emission rates as a function of the four corner positions for fuel assemblies from Reactor A.

MORRIS EXERCISE - CORRECTED EXPERIMENTAL RESULTS
PWR - 14x14 Assemblies - 450 MWe Rated Power
Exposure Range 23.6-40.1 GWd/tU

Fig. B-7.
Relative neutron emission rates as a function of the four corner positions for fuel assemblies from Reactor B.

MORRIS EXERCISE - CORRECTED EXPERIMENTAL RESULTS
BWR - 7x7 Assembly - 715 MWe Rated Power
DN201 - 4.67 GWd/tU

Fig. B-8.
Relative neutron emission rates as a function of the four corner positions for fuel assemblies from Reactor C.

MORRIS EXERCISE - CORRECTED EXPERIMENTAL RESULTS
PWR - 14x14 Assemblies - 430 MWe Rated Power
Exposure Range 28.5-32.4 GWd/tU

Fig. B-9.
Relative neutron emission rates as a function of the four corner positions for fuel assemblies from Reactor D.

TABLE B-VIII

MAXIMUM DIFFERENCES IN THE MEASUREMENTS
AT THE FOUR CORNER POSITIONS

Fuel Assembly	Maximum Difference (%)
A-A019	3
A-C049	37
A-A045	9
A-B012	6
B-C056	12
B-C032	31
B-C052	42
C-DN201	18
D-A051	9
D-C004	36
D-D001	5
D-C001	34
D-A036	11
D-D020	28
D-A009	5
D-D046	12
D-C020	24

TABLE B-IX

INDIVIDUAL IRRADIATION HISTORIES
OF FUEL ASSEMBLIES

Fuel Assembly	Core Position	Average Exposure (Gwd/tU)
C032	F-1	10.80
	D-4	12.77
	Total	23.57
C056	K-10	17.13
	C-5	13.34
	C-5	9.60
	Total	40.07
C052	A-8	11.23
	F-2	12.75
	F-2	8.84
	Total	32.87

The results shown in Fig. B-7 exhibit changes ranging from 12-42% between the corners of the fuel assembly. The irradiation history of each of the fuel assemblies in the reactor were determined and are listed in Table B-IX. A schematic of the reactor core is given in Fig. B-10 with the specific angular orientation of each fuel assembly. In this core, the fuel assemblies cannot be rotated. As can be seen from Table B-VII and Fig. B-10, the corners with the lowest relative neutron rates were always on the periphery of the reactor core. This effect appears to persist even after the fuel assembly is moved to an interior position, as was the history of fuel assembly C-052. From this data, it would appear that the first cycle of a PWR fuel assembly defines a gradient that persists through the irradiation history.

C. Empirical Correlations

Data for the four corners were summed for each fuel assembly and were corrected for the decay of ^{244}Cm . These results are plotted in Fig. B-11 for each of the three PWR data sets. The parameters for the best fits of the power functional relationship are given in Table B-X. The number of fuel assemblies measured was limited, as well

PWR Grid for 14x14 Assemblies

Fig. B-10.

Reactor core in which fuel assemblies C032, C056, and C052 were irradiated.

MEASURED NEUTRON RATE

TABLE B-X

POWER FUNCTIONS FOR PWR MEASUREMENTS^a

Reactor Data Set	Scaling Factor (α)	Exponent (β)	Correlation Coefficient ^b (R^2)
A	7.75 E-12	3.10	0.94
B	2.23 E-14	3.68	0.99
C	1.43 E-15	3.91	0.99
A+B+C	2.71 E-13	3.42	0.98

^aCount rate = $\alpha * (\text{exposure})^\beta$.

^b R^2 is the correlation coefficient and when multiplied by 100 it represents the total variation in per cent about the mean of the regression fit.

as the range of exposures, but the data appear to be consistent with measured and calculated results obtained previously.

D. Fuel Assembly Geometry

We could not determine any significant differences between the PWR fuel assemblies with 15 by 15 and 14 by 14 arrays. Figure B-12 shows a comparison of the results from the 15 by 15 fuel assemblies (C056 and C052) and the 14 by 14 fuel assemblies (C020, D046, and D001). The data are insufficient to draw any specific conclusions; however, it can be noted that the results are similar for fuel assemblies with approximately the same exposures.

E. Conclusions

The primary conclusions that can be drawn to date are:

- (1) Spent-fuel assemblies can be positioned for measurement using the V-shaped positioning mechanism to obtain reproducible results. For neutron measurements the precision is 1-2%, and for gross gamma measurements the precision is better (1%). A significant part of the uncertainty associated with the neutron measurements is associated with the counting statistics.

MORRIS EXERCISE - CORRECTED EXPERIMENTAL RESULTS

Comparison of Assemblies with Similar Exposures

Fig. B-12.

Comparison of results from 15 by 15 (C056 and C052) fuel assemblies with 14 by 14 (C020, D046, and D001) fuel assemblies.

- (2) Angular orientation of spent-fuel assemblies can have a significant effect on passive neutron and gamma-ray measurements with variations of 30-40% found for both types of measurements. The source of these variations appears to be real differences in burnup caused by the position of the fuel assembly in the reactor core.
- (3) Power function relationships appear to relate the measured neutron count rate to the operator-declared values of exposure.

However, only a small number of fuel assemblies were examined from each reactor, and therefore, a general conclusion must be deferred until we have collected additional data.

- (4) We were unable to detect any significant difference between 14 by 14 and 15 by 15 PWR fuel assembly geometries. Again, it must be noted that only a small amount of data has been collected.

APPENDIX C

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APPENDIX D

IRRADIATED-FUEL CALCULATIONS

G. E. Bosler and J. R. Phillips
Safeguards Assay Group Q-1

I. ISOTOPE PRODUCTION AND DEPLETION

Neutron and photon characteristics inherent to irradiated fuel assemblies depend on the production and depletion of various transuranic and fission-product isotopes. Measurement of these characteristics to verify irradiated fuel parameters, such as exposure, cooling time, and fissile content, requires knowledge of the correlation between the measured signal and the parameter being evaluated. Correlations either independent of irradiated fuel conditions or only dependent on initial, well-known parameters, such as ^{235}U enrichment, are desirable. In an effort to better understand important correlations and to examine their possible dependence on irradiated fuel parameters, a calculational study was performed for fuel from the H. B. Robinson Unit 2 reactor. This fuel has been analyzed destructively to determine burnup, exposure, transuranic isotopic composition, and fission-product isotopic composition. Calculations to determine the same information were performed using the EPRI-CELL¹ and EPRI-CINDER^{2,3} codes with a detailed power history for the H. B. Robinson fuel. Comparisons of the calculated and measured data are shown in Table D-I for two H. B. Robinson samples.⁴ Most of the comparisons are reasonably good, except for ^{234}U , ^{238}Pu , and ^{242}Pu . Larger relative differences for these isotopes are not unexpected because the quantity of ^{234}U is very small, the presence of ^{238}U interferes with mass spectrometry measurements of ^{238}Pu , and calculations of the higher actinide isotopes, such as ^{242}Pu , are more uncertain.

Results in Table D-I indicate that the calculational methodology can be used to investigate the effects of changes in basic operating parameters on isotope production and depletion. For

further investigations, a base case using the H. B. Robinson model was established with an initial ^{235}U enrichment of 2.561%, a UO_2 mass density of 90.814% of theoretical density, and a constant power level of 200 W/cm. Irradiation times were calculated for burnups up to 50 GWd/tU. To examine parameter sensitivities, one of the base-case parameters was varied, whereas the other parameters were fixed. Parameter variations included

- (1) initial ^{235}U enrichments of 2.0-4.5%;
- (2) UO_2 mass densities of 90.814 and 94% of theoretical density;
- (3) power levels of 160, 200, and 240 W/cm; and
- (4) irradiation histories with various combinations of 1-yr full power cycles (U) and 1-yr shutdown periods (D).

The principal chains for production and depletion of transuranic isotopes are shown in Fig. D-1. In this figure, horizontal transitions, for example ^{239}Pu to ^{240}Pu , indicate a neutron capture event. Except for the ^{242}Am to ^{242}Pu transition, vertical transitions result from beta decay. For the ^{242}Am to ^{242}Pu transition, the ^{242}Am undergoes an electron capture. Other nuclear events creating chains different from those shown in Fig. D-1 occur. However, most of these events are secondary, producing negligible amounts of material compared to the principal events shown in Fig. D-1.

Table D-II shows that, of the isotopes shown in Fig. D-1, ^{235}U , ^{241}Am , ^{242}Cm , ^{244}Cm , and most of the plutonium isotopes are of primary interest because they are the important sources of neutrons in irradiated fuels. Interpretation of passive neutron signals from irradiated fuel assemblies requires knowledge of the production of these neutron-emitting isotopes.

TABLE D-I
COMPARISON OF MEASURED AND CALCULATED
H. B. ROBINSON-2 ISOTOPIC RATIOS

Quantity	Sample P8A			Sample P8B		
	Measured Value ^a	Calculated Value	Difference (%)	Measured Value ^a	Calculated Value	Difference (%)
Burnup, at. %	2.559	2.560	+0.04	3.221	3.221	0.00
Exposure, GWD/tU	24.57	24.66	+0.37	30.92	31.19	+0.89
234U/U	0.00016	0.00014	-14.04	0.00014	0.00012	-12.81
235U/U	0.00816	0.00928	+1.53	0.00612	0.00588	-3.84
236U/U	0.00326	0.00322	-1.08	0.00353	0.00357	+1.52
238U/U	0.98842	0.98835	-0.01	0.99022	0.99043	+0.02
238Pu/Pu	0.01143	0.00979	-14.35	0.01676	0.01452	-13.35
239Pu/Pu	0.59557	0.59365	-0.32	0.54261	0.53966	-0.54
240Pu/Pu	0.23290	0.22766	-2.25	0.25101	0.24002	-4.38
241Pu/Pu	0.11842	0.12385	+4.58	0.12998	0.13772	+5.95
242Pu/Pu	0.04168	0.04506	+8.10	0.05964	0.06807	+11.41
239Pu/238U	0.00494	0.00486	-1.61	0.00518	0.00496	-4.28
148Nd/238U	0.00045	0.00047	+3.74	0.00057	0.00059	+3.96

^aMeasured isotopic ratios reported in Ref. 4, p. 16. Calculated values from 3/81 tandem CELL/CINDER-2 calculations using a detailed power history and 506.75-day cooling.

TABLE D-II
SOURCES OF NEUTRONS IN IRRADIATED UO₂ MATERIALS^a

Isotope	Half Life (yr)	(α, n) Reaction	Spontaneous Fission ^b	Total
235U	$7.038 \pm 0.005 \times 10^8$	$7.11 \pm 0.71 \times 10^{-4}$	$2.99 \pm 0.99 \times 10^{-4}$	$1.01 \pm 0.12 \times 10^{-3}$
238U	$4.468 \pm 0.002 \times 10^9$	$8.26 \pm 0.83 \times 10^{-5}$	$1.36 \pm 0.02 \times 10^{-2}$	$1.37 \pm 0.02 \times 10^{-2}$
238Pu	$8.771 \pm 0.003 \times 10^1$	$1.34 \pm 0.14 \times 10^4$	$2.59 \pm 0.11 \times 10^3$	$1.61 \pm 0.16 \times 10^4$
239Pu	$2.413 \pm 0.002 \times 10^4$	$3.81 \pm 0.38 \times 10^1$		$3.82 \pm 0.43 \times 10^1$
240Pu	$6.570 \pm 0.006 \times 10^3$	$1.41 \pm 0.14 \times 10^2$	$9.09 \pm 0.10 \times 10^2$	$1.05 \pm 0.19 \times 10^3$
242Pu	$3.763 \pm 0.009 \times 10^5$	2.05 ± 0.20	$1.72 \pm 0.02 \times 10^3$	$1.71 \pm 0.02 \times 10^3$
241Am	$4.320 \pm 0.002 \times 10^2$	$2.69 \pm 0.27 \times 10^3$		$2.69 \pm 0.32 \times 10^3$
242Cm	$4.456 \pm 0.001 \times 10^{-1}$	$3.76 \pm 0.38 \times 10^6$	$2.10 \pm 0.08 \times 10^7$	$2.48 \pm 0.09 \times 10^7$
244Cm	$1.810 \pm 0.002 \times 10^1$	$7.73 \pm 0.77 \times 10^4$	$1.08 \pm 0.01 \times 10^7$	$1.09 \pm 0.01 \times 10^7$

^aSpecific neutron emission rates in neutrons per gram-second.

^bThe reference value for spontaneous fission is

$$\bar{\nu}_{sp} = 3.756 \text{ for } {}^{252}\text{Cf}.$$

Fig. D-1. Light-water reactor production chains for the transuranic isotopes from ^{235}U and ^{238}U feed material.

The fissile isotopes of uranium and plutonium are shown in Figs. D-2 through D-5. In Fig. D-2, the ^{235}U concentration is shown. Concentration of this isotope depends primarily on the initial enrichment. Other parameters examined have very little effect. Plutonium-239, shown in Fig. D-3, is affected by initial ^{235}U enrichment and UO_2 mass density, but is relatively unaffected by power level and irradiation history. Similarly, ^{241}Pu , shown in Fig. D-4, is affected by initial enrichment and UO_2 density. Power levels have little effect on the production of ^{241}Pu . Because of the relatively short half-life for ^{241}Pu (14.7 yr), long shutdown periods affect the ^{241}Pu concentration. This is shown in the final plot of Fig. D-4. Figure D-5 shows that total fissile content ($^{235}\text{U} + ^{239}\text{Pu} + ^{241}\text{Pu}$) is affected primarily by initial enrichment.

The principal neutron-producing plutonium isotopes (^{238}Pu , ^{240}Pu , and ^{242}Pu) are shown in Figs. D-6 through D-8. Americium-241, which results from the decay of ^{241}Pu , is shown in Fig. D-9. All

of these isotopes have some sensitivity to initial ^{235}U enrichment. For isotopes with short half-lives or for those formed from other isotopes with short half-lives, there is also some dependence on irradiation history.

The curium isotopes ^{242}Cm and ^{244}Cm are shown in Figs. D-10 and D-11. Curium-242 is very sensitive to all parameters, whereas ^{244}Cm is basically sensitive only to initial ^{235}U enrichment. Because of the short half-life for ^{242}Cm ($t_{1/2} = 163$ days), much of this isotope decays away during extended shutdowns or after discharge.

II. NEUTRON SIGNALS

Production and depletion data for transuranic isotopes described in the previous section have been used to obtain relative neutron source strengths as a function of exposure and cooling time. The neutron source terms relate only to the production of neutrons within the fuel material from spontaneous fissioning and (α, n) reactions

Fig. D-2.
Sensitivity of ^{235}U to various reactor parameters as a function of exposure.

Fig. D-3.
Sensitivity of ^{239}Pu to various reactor parameters as a function of exposure.

Fig. D-4.
Sensitivity of ^{241}Pu to various reactor parameters as a function of exposure.

Fig. D-5.
Sensitivity of total fissile material to various reactor parameters as a function of exposure.

Fig. D-6.
Sensitivity of ^{238}Pu to various reactor parameters as a function of exposure.

Fig. D-7.
Sensitivity of ^{240}Pu to various reactor parameters as a function of exposure.

Fig. D-8.
Sensitivity of ^{242}Pu to various reactor parameters as a function of exposure.

Fig. D-9.
Sensitivity of ^{241}Am to various reactor parameters as a function of exposure.

Fig. D-10.
Sensitivity of ^{242}Cm to various reactor parameters as a function of exposure.

Fig. D-11.
Sensitivity of ^{244}Cm to various reactor parameters as a function of exposure.

with ^{18}O and do not include any multiplication or absorption within the fuel. Transport of these source neutrons from their birth in the fuel assembly to a detector outside the assembly is currently being investigated. Results of neutron yield calculations are shown in Figs. D-12 through D-14 for exposures of 10.86, 19.72, and 51.20 Gwd/tU, respectively. For all these exposures, the curium isotopes are the primary neutron producers. At low exposure (<10 Gwd/tU), the plutonium-amer-
 cium isotopes contribute significantly to the neutron rate because the curium is only beginning to be produced. For higher exposures, the neutron rate is almost totally dominated by the curium isotopes. In the 10-35 Gwd/tU exposure range, both ^{242}Cm and ^{244}Cm contribute to the neutron source until the ^{242}Cm decays away. For exposures above 35 Gwd/tU, the ^{244}Cm concentration becomes so large compared to ^{242}Cm that most of the neutrons come from ^{244}Cm regardless of the cooling time. For reprocessed fuels with the curium isotopes removed, the principal neutron sources would be the plutonium isotopes and ^{241}Am . The ^{241}Am contribution increases with cooling time as the ^{241}Pu decays to ^{241}Am with a 14.3-yr half-life.

Fig. D-12.
 Neutron source rates for the five most prominent neutron-emitting isotopes and an exposure of 10.860 Gwd/tU.

Fig. D-13.
 Neutron source rates for the five most prominent neutron-emitting isotopes and an exposure of 19.717 Gwd/tU.

Fig. D-14.
 Neutron source rates for the five most prominent neutron-emitting isotopes and an exposure of 51.201 Gwd/tU.

Figure D-15 compares the relative neutron rates for the detailed power history and the constant power case (200 W/cm) of the H. B. Robinson-2 data. As can be readily seen, results for the constant power case are similar to those for the detailed power history. In all subsequent comparisons, the constant power case is used as the basis for comparison.

Fig. D-15.

Comparison of neutron source rates at discharge as a function of exposure for a detailed power history and a continuous 200 W/cm linear power history.

The effect of varying the power level on the neutron rate at the time of discharge is shown in Fig. D-16. Differences among the three power levels decrease as the cooling time increases, with the three curves merging to one curve for very long cooling time, as shown in Fig. D-17. For a long cooling time, the neutron rate depends on the amount of ^{244}Cm , which is essentially independent of power level. For a short cooling time, neutrons from ^{242}Cm account for the dependence on power level shown in Fig. D-16.

Changes in the initial mass density of the fuel material do not influence significantly the neutron source strength (Fig. D-18). Differences in the initial ^{235}U enrichment of the fuel material can significantly affect the neutron production rate, as is illustrated in Fig. D-19. In these cases, the power levels were maintained at 200 W/cm; therefore, the lower enrichment fuel was necessarily exposed to a higher flux as well as having slightly more ^{238}U source material available for production of the transuranic isotopes, particularly the curium isotopes. Production of more curium results in higher neutron source rates for lower enrichments.

Irradiation history is one of the most important parameters affecting neutron source terms,

Fig. D-16.

Comparison of neutron source rates at discharge as a function of exposure for continuous power histories of 160 W/cm, 200 W/cm, and 240 W/cm.

Fig. D-17.

Comparison of neutron source rates at a cooling time of 36 months as a function of exposure for continuous power histories of 160 W/cm, 200 W/cm, and 240 W/cm.

particularly for cooling times < 3 yr. Figure D-20 shows the effects of several irradiation scenarios. In these plots, a U implies a 1-yr cycle at full power and a D implies a 1-yr downtime. The relative neutron rates are dramatically altered by the different irradiation histories, even though the total exposures are identical. The principal cause of these perturbations is the buildup of the ^{242}Cm isotope. Plutonium-241, with a half-life of 14.35 yr, is a precursor of

0 month Cooling Time

24 month Cooling Time

60 month Cooling Time

120 month Cooling Time

Fig. D-18.
 The effect of initial UO_2 mass density on the neutron source rate as a function of exposure and cooling time.

0 month Cooling Time

24 month Cooling Time

60 month Cooling Time

120 month Cooling Time

Fig. D-19.
Effect of initial ²³⁵U enrichment on the neutron source rate
as a function of exposure and cooling time.

Fig. D-20.
Effect of irradiation history on the neutron source rate as a function of exposure and cooling time.

^{241}Am ($t_{1/2} = 432$ yr), which captures a neutron and produces ^{242}Am , which decays with a 16.01-h half-life to ^{242}Cm . For a significant downtime, the ^{241}Am inventory increases, thereby producing more source material for the production of ^{242}Cm . The resulting influence upon the relative neutron rates becomes less important as the cooling time increases, with all cases merging after 3 yr.

The above data illustrate the important influence of various fuel parameters and irradiation histories upon the neutron sources in irradiated fuel assemblies. To interpret correctly passive neutron measurements of fuel assemblies, some information about the initial fuel material is necessary, and for short cooling times (<36 months), additional information is required about the irradiation histories.

For irradiated fuel assemblies that have cooled sufficiently to allow the decay of ^{242}Cm to an insignificant level, the neutron emission rate and subsequent detector count rate are directly proportional to the ^{244}Cm density. As shown in Figs. D-21 and D-22, ^{244}Cm density can be correlated with plutonium content. In Fig. D-21, ^{244}Cm is correlated with $\Delta\text{Pu} = \text{Total Pu} - ^{239}\text{Pu}$. In Fig. D-22, the correlation is between ^{244}Cm and total plutonium. At Karlsruhe,⁵ these correlations were used successfully to determine plutonium content in irradiated fuel assemblies. As shown in Fig. D-21, ΔPu vs ^{244}Cm is an excellent correlation because there is little sensitivity to parameters such as initial ^{235}U enrichment, UO_2 density, power level, and irradiation history. The total plutonium vs ^{244}Cm correlation shown in Fig. D-22 is basically insensitive to all parameters except initial ^{235}U enrichment. Thus, if corrections are made for initial ^{235}U enrichment, these two correlations could be used for determining plutonium content from passive neutron measurements. Other correlations are possible and may be developed to relate neutron rates to various irradiated fuel parameters.

III. GAMMA-RAY SIGNALS

Measurable spectral gamma-ray signatures from irradiated boiling-water-reactor (BWR) and pressurized-water-reactor (PWR) fuels come primarily from fission products. These signatures can be used to verify operator-declared values of exposure and cooling time.⁶ Various gamma-ray signatures and isotopic ratios have been investigated calculationally. Some results of these calculations are shown in Figs. D-23 through D-25. In Fig. D-23, ^{137}Cs is relatively independent of the parameters investigated. Because of these characteristics and a relatively long half-life (30.1 yr), measurement of ^{137}Cs concentration is one means for determining exposure.⁶ In Fig. D-24, ^{134}Cs has a very short half-life (2.06 yr) and is therefore very sensitive to shutdown periods. Cesium-134 is also more sensitive than ^{137}Cs to initial ^{235}U enrichment. Europium-154, shown in Fig. D-25, has a half-life of 8.6 yr and behaves similarly to ^{134}Cs . Both ^{134}Cs and ^{154}Eu are shielded isotopes in that they are not formed directly from fission. (Only an insignificant fraction of these two isotopes are formed directly from fission.) Instead, these isotopes are produced from chains of neutron capture events. For ^{134}Cs , the chain is very simple, resulting from capture in ^{133}Cs that comes from ^{133}I through ^{133}Xe . The ^{154}Eu chain is much more complicated, starting from ^{145}Pr , ^{150}Nd , and ^{148}Nd . A complete description of the ^{154}Eu chain is included in Ref. 6.

The isotopic ratios $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ are shown in Figs. D-26 and D-27, respectively. Both ratios depend on initial ^{235}U enrichment. Because of the short half-life of ^{134}Cs , the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio is very dependent on the irradiation history of the fuel assembly. Both of these ratios have been used experimentally to verify exposure and cooling times.⁵

In Figs. D-28 and D-29, these two isotope ratios are plotted as a function of ^{239}Pu content for two exposure histories. The detailed irradiation history for H. B. Robinson includes a

Fig. D-21.
Dependence of correlation between ^{244}Cm and $\Delta\text{Pu} = \text{total plutonium} - ^{239}\text{Pu}$ on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

Fig. D-22.
Dependence of correlation between ^{244}Cm and total plutonium on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

Fig. D-23.
Sensitivity of ^{137}Cs to various reactor parameters as a function of exposure.

Fig. D-24.
Sensitivity of ^{134}Cs to various reactor parameters as a function of exposure.

Fig. D-25.
Sensitivity of ^{154}Eu to various reactor parameters as a function of exposure.

Fig. D-26.
Sensitivity of $^{134}\text{Cs}/^{137}\text{Cs}$ to various reactor parameters as a function of exposure.

Fig. D-27.
Sensitivity of $^{154}\text{Eu}/^{137}\text{Cs}$ to various reactor parameters as a function of exposure.

Fig. D-28.
Dependence of correlation between $^{134}\text{Cs}/^{137}\text{Cs}$ ratio and ^{239}Pu on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

Fig. D-29.
Dependence of correlation between $^{154}\text{Eu}/^{137}\text{Cs}$ ratio and ^{239}Pu on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

61-day downtime between cycles (~ 20 GWd/EU). Once the value of the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio exceeds 0.065, the corresponding quantity of ^{239}Pu does not change by more than 5%. This value of the ratio corresponds to a burnup range from 20 to 50 GWd/tU. Similar results are found for the $^{154}\text{Eu}/^{137}\text{Cs}$ isotope ratio, except that the threshold value of this ratio is ~ 0.022 .

Figures D-30 and D-31 show the results of plotting these two isotope ratios as a function of the remaining fissile inventory ($^{235}\text{U} + ^{239}\text{Pu} + ^{241}\text{Pu}$) in a typical PWR fuel assembly. The functional relationship is approximately linear in both cases. The $^{134}\text{Cs}/^{137}\text{Cs}$ ratio is affected by the 61-day downtime between cycles more than the $^{154}\text{Eu}/^{137}\text{Cs}$ ratio because the ^{134}Cs half-life is only 2.062 yr compared to 8.6 yr for ^{154}Eu . These calculations indicate that information can be obtained from the isotope ratios $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ in addition to estimates of exposure and cooling time. They also may correlate with the fissile inventory of spent-fuel assemblies.

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Fig. D-30.
Dependence of correlation between $^{134}\text{Cs}/^{137}\text{Cs}$ ratio and the concentration of total fissile material on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

Fig. D-31.
Dependence of correlation between $^{154}\text{Eu}/^{137}\text{Cs}$ ratio and the concentration of total fissile material on initial ^{235}U enrichment, initial UO_2 mass density, power level, and irradiation history.

APPENDIX E

PULSED-NEUTRON INTERROGATION TECHNIQUE FOR THE DETERMINATION OF THE FISSILE CONTENT OF SPENT-FUEL ASSEMBLIES

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I. INTRODUCTION

A technique that uses pulsed-neutron sources to verify directly the uranium and plutonium fissile contents of spent light water reactor (LWR) fuel assemblies is under investigation. Because spent fuel is normally stored underwater in storage pools, the initial studies are for fuel assemblies under several meters of water. This constraint has the advantage of supplying the shielding for the neutron interrogation source as well as the intense gamma-ray dose from the fuel assembly.

The fuel assemblies of most interest for the present study are irradiated pressurized-water-reactor (PWR) assemblies that contain both uranium and plutonium fissile components. Boiling-water-reactor (BWR) fuel assemblies present a more complex problem that will be addressed in future work.

Pulsed-neutron techniques^{1,2} have been used for several years in the uranium well-logging industry to assay the quantity of uranium underground in exploration boreholes. Most of the well-logging systems have used sealed-tube neutron generators to supply the interrogation neutrons. Two basic approaches have been used: one is to pulse the neutron source on and off and to count the induced delayed fission neutrons when the interrogation source is off; the other is to count the induced prompt fission neutrons shortly after each interrogation pulse. Both of these approaches are used to assay the ^{235}U content of the ore. This logging procedure led to the development of compact and rugged neutron generators that can be used in a relatively small borehole under several thousand feet of water. The environment in a spent-fuel storage pool is much more favorable than this.

Although the physical constraints for spent-fuel measurements are not as severe as for well-logging, the radiation backgrounds are much higher. The gamma-ray doses could be up to 50 000 R/h adjacent to fuel assemblies. There is also a neutron background that comes primarily from spontaneous fission and (α, n) reactions occurring within the fuel material. The spontaneous fission sources are the curium isotopes, ^{242}Cm and ^{244}Cm , and the even-numbered plutonium isotopes. The (α, n) sources are primarily ^{242}Cm and ^{241}Am . The intensity of this neutron background depends on the cooling time and level of exposure in the fuel assembly, as shown in Fig. E-1. The source strength of background neutrons could be up to 10^8 n/s for the entire fuel assembly. The measured neutron rates (Fig. E-1) are only for a small portion of the assembly.

Pulsed-neutron generators together with the prompt neutron signal have been used by Sandia National Laboratories, Albuquerque,³ for the measurement of fissile material in a portal monitor and by the Detection and Verification Group Q-2⁴ at Los Alamos for the assay of small quantities of ^{235}U in large waste containers. Both of these systems require large quantities of graphite and CH_2 moderators to extend the lifetimes of the thermal neutrons in the system, thereby increasing the sensitivity for small quantities of fissile material.

A modulated (or shuffler type) ^{252}Cf neutron source was developed at Los Alamos for the measurement of ^{235}U content in bulk samples.⁵ More recently, this method was applied to spent-fuel elements and reprocessing waste canisters for

MEASURED NEUTRON RATE

Fig. E-1.
Measured neutron-emission rates vs
declared burnup for 36 spent PWR
assemblies at the Zion Generating
Station.

the Fluorinel and Storage (FAST)⁶ Facility neutron interrogator. In both cases, only the induced delayed fission neutrons are measured to determine the ^{235}U content of high-enrichment uranium fuel.

II. PRINCIPLE OF THE TECHNIQUE

Spent LWR fuel assemblies contain ~0.8-1.0% each by weight of fissile uranium and plutonium. The straightforward assay approach of neutron interrogation and counting of the induced prompt or delayed fission neutrons is not sufficient to determine the plutonium content. During exposure in the reactor, the ^{235}U decreases, the plutonium increases, and the sum of the fissile components changes as illustrated in Fig. E-2. Thus, the measurement of the sum of the two components does not give the plutonium content.

One approach we are presently investigating to assay the plutonium content is to measure both the prompt- and delayed-neutron responses and then to use the prompt/delayed-neutron ratio to separate the two components. This is possible because the delayed neutron fraction is roughly a factor of 2.6 higher for ^{235}U than ^{239}Pu . This same approach for separating ^{235}U and ^{239}Pu in mixed-oxide fuel was investigated by us⁷ previously for fresh BWR fuel rods and by Y. Matsuda et al.⁸ for irradiated fuel pellets. For high burnup

fuel, the ^{241}Pu contribution may interfere with the ability to separate the ^{235}U and ^{239}Pu components.

A. Pulsed 14-MeV Neutron Generator

Czubek¹ has shown that with a pulsed-neutron source, epithermal flux die-away depends on the presence of fissile material, that is, epithermal flux dies away much faster in the absence of fissile material. Thermal flux die-away with and without fissile material does not change as much as the epithermal die-away. In addition, delayed neutrons from fission are present for times that are much longer than either the epithermal or the thermal die-away times. Although Czubek's results were for well-logging applications, similar effects are expected for pulsed-neutron interrogation of spent-fuel assemblies. By selectively choosing measurement times after a pulse, epithermal die-away could be used to verify fissile material presence and the fissile composition could be determined from delayed neutrons. A possible drawback to such a system is the high background of spontaneous fission and (α, n) neutrons in spent-fuel assemblies.

Time-dependent calculational studies are being performed to determine the feasibility and applicability of an active spent-fuel assay system. These calculations use the Los Alamos Monte Carlo code MCNP.⁹ In the current model, neutrons from

CALCULATIONALLY ESTIMATED QUANTITIES **PWR SENSITIVITY SERIES -- CASE A-2**

Fig. E-2.
 Calculation of weight per cent of ²³⁵U, ²³⁹Pu, and ²⁴¹Pu as a function of burnup.

a 14-MeV (D,T) source pulsed for 15 μ s are introduced into a typical 15 by 15 PWR spent-fuel assembly. A fission chamber surrounded by $\sim$ 5 cm of polyethylene and a thin layer of cadmium forms the epithermal neutron detector. The cadmium reduces the detector response to low-energy neutrons coming from the assembly and the (D,T) source. The entire system is surrounded by water to simulate measurement procedures in spent-fuel storage ponds. Fuel-water heterogeneity effects are accounted for by actually modeling fuel pins in the assembly.

A diagram showing the initial geometric setup for the Monte Carlo calculations is shown in Fig. E-3. A version of MCNP, which was developed for tracking delayed neutrons, is being adapted for this study. Previously only the delayed neutrons arising from fissions produced by fast incident neutrons were considered. The modified version of the code considers delayed fission neutrons arising from either fast or thermal neutrons. This is important because most of the source neutrons will be thermalized in the water environment before causing fissions in the spent fuel.

Time discrimination between the 14-MeV source neutrons, the prompt fission neutrons, and the delayed fission neutrons is possible because the

Fig. E-3.
 Geometric configuration of the active neutron interrogation source, the PWR assembly, and the detector for counting prompt and delayed neutrons.

slowing down time of neutrons in water from 14 MeV to epicalcium energy (~ 0.3 eV) takes only a few microseconds, whereas the thermal neutrons have a lifetime of ~ 200 μ s in water. Figure E-4 shows the time sequence for a repetitively pulsed system. During the time period from a few tens of microseconds to a few hundred microseconds, the prompt fission neutrons are counted. The 14-MeV neutrons produced by the generator are thermalized and induce fissions in the fissile material. For the detector shown in Fig. E-3, the cadmium liner prevents most of the thermalized source neutrons from reaching the detector. The induced fission reactions are closer to the detector, and a significant fraction of the prompt fission neutrons retain sufficient energy to pass through the cadmium barrier and reach the detector. The delayed-neutron time gate is opened ~ 1500 μ s after the generator pulse. The neutrons in this time interval are from induced fissions corresponding to many prior pulses. The intensity of the delayed-neutron signal slowly builds up and reaches equilibrium after a few minutes because the longest-lived delayed-neutron precursor has a half-life of ~ 55 s.

B. Modulated ^{252}Cf Neutron Source

As an alternative to the 14-MeV neutron generator, a modulated ^{252}Cf neutron source can be used for the same application. The induced prompt fission neutrons are counted while the ^{252}Cf is in the near vicinity of the fuel assembly. The ^{252}Cf source then is moved rapidly away from the fuel assembly and detector and the delayed neutrons

are counted. To obtain sufficient counting statistics, the source is repetitively transferred back and forth between the sample and its shielded position, as is done in the ^{252}Cf shuffler system⁷ described previously. Figure E-5 shows a schematic diagram of the irradiation and counting cycle. The irradiation time of ~ 10 s is considerably longer than the delayed-neutron counting time (~ 3 s) to improve the signal-to-background ratio. Again, a high neutron background is expected from the spent fuel.

In addition to the background neutrons from the spent fuel, there are background neutrons from the ^{252}Cf source during the prompt-neutron counting interval, as shown in Fig. E-5. The number of these source neutrons reaching the detector can be reduced by absorption in a cadmium sleeve around the detector, as shown in Fig. E-2. Similar to the pulsed-neutron generator approach, the induced fission neutrons are closer to the detector than the californium source, and thus a higher fraction of these signal neutrons are above the cadmium energy cutoff and reach the detector.

C. Separations of Fissile Uranium and Plutonium Contents

The delayed-neutron rate per source neutron is roughly the same for both the pulsed 14-MeV neutron source and the modulated ^{252}Cf source. However, a source intensity of 10^9 - 10^{10} n/s can be obtained more readily with a ^{252}Cf source (~ 1 mg) than with currently available sealed-tube neutron generators.

Fig. E-4.
Active neutron irradiation and counting cycle for a pulsed-neutron generator using both prompt- and delayed-neutron counting.

Fig. E-5.

Active neutron irradiation and counting cycle for a modulated ^{252}Cf neutron source using both prompt- and delayed-neutron counting.

Table E-I shows the delayed fraction and discrimination ratio¹⁰ for the principal fissile components. The ^{235}U has considerably more delayed neutrons per fission resulting in a discrimination ratio of 2.6 relative to ^{239}Pu for a two-component linear system. Because the ^{241}Pu has a delayed-neutron fraction that is very similar to ^{235}U , it will tend to wash out the discrimination factor. We propose to circumvent this problem through iterative techniques, using calculated isotopic correlations. That is, in the first estimate of the ^{235}U and ^{239}Pu components, the ^{241}Pu contribution would be neglected. The estimated ^{235}U and ^{239}Pu ratios would then be used to predict the ^{241}Pu fraction using isotope correlations. The initial data would then be corrected for the ^{241}Pu contribution and the $^{235}\text{U}/^{239}\text{Pu}$ ratio would be re-estimated. These more accurate values would then be used to predict a better value for the ^{241}Pu fraction to correct the initial values, and so forth until convergence. Because the ^{241}Pu is a relatively small fraction of the measured response, we expect that the error introduced by this correction procedure will be small.

Modifications have been made in the Monte Carlo code MCNP so that calculations can be performed to determine signal-to-background ratios for the delayed neutrons, optimal source-detector

TABLE E-I

DELAYED NEUTRON FRACTIONS

Isotope	Fission Cross Section (b)	Discrimination Ratio ^a	Delayed-Neutron Fraction ^b
^{235}U	584	-	0.0168
^{239}Pu	742	2.6	0.0061
^{241}Pu	1010	-	0.0154

^aThe discrimination ratio is defined in Ref. 10 for a two-component system where ^{235}U is one of the components. For ^{241}Pu , the discrimination ratio is not applicable.

^bThe delayed-neutron fraction is the yield of delayed neutrons per fission event.

configurations, sensitivity to changes in the plutonium and uranium isotopic compositions, and sensitivity to the removal of individual fuel pins. These calculations will be performed as part of our program to evaluate active neutron techniques for the verification of spent-fuel assemblies.

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APPENDIX F

EXPERIMENTAL PROGRAM AT LOS ALAMOS

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Design of the active and passive neutron portions of the spent-fuel verification and assay system is being accomplished using both experiments and calculations. This appendix describes the experimental facilities, the available materials, and the experiments in progress at Los Alamos to complete the research necessary to achieve the specified measurement goals. The experimental program at Los Alamos is limited to handling unirradiated fuel assemblies. Gamma-ray and ion-chamber measurements, therefore, have to be obtained with field experiments and are discussed in App. B. Passive and active neutron measurements can be performed on unirradiated assemblies by using neutron sources to induce fissions in the fuel. The primary purpose of the experimental work is to develop a design permitting neutron measurements on light-water-reactor (LWR) fuel assemblies. The neutron measurements will be used to determine the burnup and the total effective fissile content in irradiated fuel assemblies.

I. EXPERIMENTAL FACILITIES AND EQUIPMENT

The experimental measurements are being performed at the Safeguards Hot Cell Facility at Los Alamos. An exterior view of the facility is shown in Fig. F-1. The hot cell contains three shielded viewports with remote manipulators at two of the windows. Additional remote viewing of the cell interior is provided with two television units that have pan, tilt, zoom, and focus controls. Personnel entry to the cell is through an intermediate room and then past an access port that can be sealed with a 1.22-m-thick concrete door. Contaminated materials are not processed in this hot cell, and its major advantages are the capability for handling radiation sources and providing a shield

from other experiments outside the cell. Intense radioactive sources stored in large shield containers are moved into the cell through an overhead access port. This port, sealed by a 1.22-m-diam by 1.22-m-thick concrete plug, can be opened by lifting the plug with a 50-ton-capacity crane.

The interior of the hot cell, shown in Fig. F-2, is 4 m deep, 15 m wide, and 5 m tall. The crane in the cell permits an overhead working clearance of 2.7 m and has a lifting capacity of 5 tons. Crane operations can be controlled from inside and outside the cell. Access ports at each shield window permit instrument cabling between the cell and exterior equipment. Isolated power routed inside and outside the cell limits electrical interference from outside sources and is shielded to reduce ground loop noise. The size and features of the hot cell enable a variety of experiments to be carried out simultaneously using the three shield windows.

Two experimental stations (Fig. F-1) are located at the two shield windows with the remote manipulators. One station has a Canberra Series 80 Single Parameter Multichannel Analyzer (MCA), and the other has a Canberra Multiparameter MCA. Both analyzers are interfaced to Digital Equipment Corporation (DEC) LSI-11 microcomputers. Analyzer data can be collected manually using the Canberra keyboard or remotely with software executed by the computers. Mass storage for computer programs and data is provided by 5-megabyte hard disks. Dual-density floppy diskettes are used for archival storage and to transfer information between computers. Detector signals collected from experiments in the hot cell are routed to nuclear instrument modules (NIM) at the measurement stations outside the cell. A complete set of NIM is available to process detector signals. These signals can be

Fig. F-1. Hot cell exterior view.

Fig. F-2. Hot cell interior view.

routed to the analyzers or directly to the computers using a scaler interface board that resides in the computer backplane. This scaler consists of 6 channels, each with a 24-bit count capacity. Additional interfaces can be added to the computer to provide increased control and monitoring of experiments.

Design constraints for spent-fuel measurements require that data be collected underwater. To enable underwater measurements at the hot-cell

facilities, two tanks were constructed to permit fuel assemblies to be immersed in water. One tank, a cylindrical barrel 140 cm high and 50 cm in diam, enables a large volume of water to surround a fuel assembly. The other tank, also 140 cm high, is square with an inside dimension of 22.2 cm, providing a tight fit for pressurized-water-reactor (PWR) fuel assemblies. This tank is surrounded by a table permitting a variety of materials and detectors to be easily located around the fuel for measurements.

A variety of detectors and neutron sources are available for experimental measurements, including ^3He , ^{10}B , and fission chamber detectors. Water-tight polyethylene cylinders have been designed to house the detectors for underwater measurements. Isotopic neutron sources available for measurements include Am-Li , Pu-Be , Ra-Be , and ^{252}Cf , in addition to $^{124}\text{Sb-Be}$ photoneutron sources. The californium sources are small in size and have a convenient half-life and a large range of neutron yields. Californium sources currently available for measurements have yields ranging from 10^4 to 10^8 n/s and may be obtained with yields to 10^{10} n/s. These sources have been calibrated by the National Bureau of Standards and thus are useful for normalizing experimental data to counts per source neutron for comparison with Monte Carlo calculations.

II. INITIAL CODE VALIDATION EXPERIMENTS

Neutron measurements on spent-fuel assemblies require counters capable of operating in large neutron fields (10^{10} n/s) while subject to intense gamma dose rates (50 000 R/h). A few neutron detectors are capable of meeting these requirements, and among them fission chambers offer some distinct advantages. Fission chambers require little or no gamma-ray shielding and are capable of detecting individual neutrons in the presence of an incident gamma-ray flux of 10^6 R/h. They have low detection efficiency enabling neutron counting to be performed with minimal deadtime effects when exposed to large neutron fields. Research and development experiments pertaining to spent-fuel measurements incorporate fission chambers for both passive and active neutron measurements.

The fission chambers are supplied according to the specifications listed in Table F-I. Chambers with varying dimensions may be ordered. Uranium coatings on the tube wall may have thicknesses up to 2 mg/cm^2 . The detectors currently in use contain 143 mg of UO_2 enriched to 93.2 wt% ^{235}U , corresponding to a 1 mg/cm^2 wall coating.

TABLE F-I

SPECIFICATIONS FOR MATCHED 2.54-CM-DIAM ^{235}U PROPORTIONAL COUNTERS

- 12.7-cm active length, 2.54-cm diam; overall length not to exceed 36.5 cm.
- Optimum operating voltage must be less than 1000 V.
- Uranium coating on tube wall must be between 1 and 2 mg/cm^2 . Total quantity of U-235 shall be $143 \text{ mg} \pm 10\%$. Uranium shall contain 93% or more ^{235}U by isotope weight. All detectors shall be matched for total uranium content within 5%.
- Tube walls must be made of 1100 Al.
- All detectors must be equipped with standard female HN connectors mounted axially at one end of the detector in a manner such that the detector and connector form a mechanically and electrically sound unit.
- Detectors must be capable of detecting individual neutrons (counting mode) in the presence of an incident gamma flux of 10^6 R/h.
- Detectors shall be able to operate at 200°C and exhibit no more than a 10% decrease in sensitivity for a 3×10^{20} thermal neutron fluence.
- The maximum in the pulse-height distribution for all detectors must lie within 5% of the average of the peaks for all detectors operated at the same optimum high voltage.
- To meet the above specifications, the vendor is expected to perform the necessary calculations and tests to arrive at the gas mixture, wall coating to give sufficient multiplication, stopping power, and minimum rise time.

Design of a spent-fuel assay system depends on understanding the neutronics of background neutrons (primarily spontaneous fission from ^{242}Cm and ^{244}Cm) and neutrons from fission produced by an external neutron-interrogation source. Measurements that enable the neutronics to be studied are difficult to obtain in a laboratory environment where the actual spent-fuel assemblies cannot be handled. Computer calculations using Monte Carlo and Discrete Ordinates codes enable parametric data to be obtained to enhance the neutronics studies using laboratory measurements. Experiments are possible on unirradiated fuel assemblies that

represent various stages of burnup. These experiments are conducted using appropriate combinations of cold fuel rods containing either plutonium or uranium to simulate various burnups. Calculations that simulate the experimental geometries are compared with the measured data to check the validity of the computer models. In addition, the calculations, once validated, provide information on the suitability of the experimental mock-up assemblies for providing design data that correspond to actual spent-fuel assemblies.

To simulate the underwater environment for experiments, a large water-tight aluminum barrel, 50 cm in diam by 140 cm (4.5 ft) high, was constructed. Experiments using this barrel were completed to provide initial validation of Monte Carlo calculations. The experiments consisted of filling the barrel with water and positioning a ^{252}Cf source and fission chamber at the midplane of the barrel. Fission chamber count rates were obtained for a series of source-detector separations. The ^{252}Cf source produced 4.51×10^5 n/s ($\pm 1.22\%$ on 12/1/80) at the time of the experiment. The Monte Carlo code tallies events on a per-source-neutron basis, so that the measured count rates had to be converted to counts per source neutron for direct comparison with calculations.

Monte Carlo calculations were performed using a model geometry that described the experimental source-detector configuration. The fission chamber was simplified in the calculations by modeling the UO_2 wall coating as a gas uniformly distributed throughout the detector. The UO_2 gas density in the fission chamber was computed to be 2.22×10^{-3} g/cm³, corresponding to 5.0×10^{18} UO_2 molecules/cm³ or 1.5×10^{19} atoms/cm³.

A series of calculations was performed for different detector-source separations corresponding to the experimental measurements. The neutron source in the computer runs was modeled as a point, isotropic neutron emitter with a fission-energy-spectrum distribution. Continuous energy cross sections supplied with the Monte Carlo code were used for all materials. Interactions of neutrons with water were treated using the thermal scattering model. This model is generally referred to as $S(\alpha, \beta)$ and provides a complete representation

of thermal neutron scattering by molecules and crystalline solids. Fission events in the detector were determined as fissions per source neutron. The calculational data are plotted as the upper curve of Fig. F-3.

The experimental and calculational data of Fig. F-3 are listed in Table F-II. The data are independent and no normalization factor has been applied between the experiments and calculations. The experimental measurements (counts/neutron) are systematically smaller by 7-50% compared to the calculations. The code calculates fissions per source neutron and not counts. The measured count rate for a fission chamber with a 1 mg/cm^2 UO_2 coating may require a correctional increase of up to 25% to obtain the fission rate. This correction is required with detectors that have a thick wall coating. A certain percentage of the heavy mass fragments produced in fission will lose enough energy escaping the UO_2 to be below the alpha threshold, thus preventing the register of a fission count.

The percentage difference between the calculational and experimental data increases as the distance between the source and detector decreases. The calculated thermal and epithermal neutron flux in water from a fission spectrum as a function of distance from the source is shown in Fig. F-4. The energy spectrum from a fission source in water reaching a detector is essentially all epithermal (>0.25 eV) for distances of 5 cm or less between the source and detector. Resonance self-shielding effects that depend on neutron energy and approximations in modeling the detector as filled with UO_2 gas may account, at least in part, for the increasing difference observed between experiment and calculation for small source-detector separation.

Active and passive neutron measurements on spent-fuel assemblies may measure either total or epithermal counts. Epithermal events are obtained by wrapping the counters in cadmium. Generally, to increase detection efficiency for epithermal events, the counters are encased in a polyethylene sleeve that is wrapped in cadmium. Determination of the effect of cadmium on a fission source and a

Fig. F-3.
Fission chamber response to a
 ^{252}Cf source in water.

TABLE F-II

COMPARISON BETWEEN EXPERIMENTAL AND CALCULATIONAL
RESPONSES TO A ^{252}Cf SOURCE FOR A FISSION
CHAMBER IN WATER

Source-Detector Separation (cm)	Experiment (counts/ source neutron)	Calculation (fissions/ source neutron)	Difference (%)
0	-	$1.142\text{-}3 \pm 7.526\text{-}5^a$	-
1.5	$8.34\text{-}4 \pm 2.43\text{-}5$	$1.112\text{-}3 \pm 6.272\text{-}5$	33.3
2.0	$8.37\text{-}4 \pm 1.05\text{-}5$	$1.261\text{-}3 \pm 7.780\text{-}5$	50.7
3.0	$8.23\text{-}4 \pm 1.03\text{-}5$	$1.158\text{-}3 \pm 7.133\text{-}5$	40.7
4.0	$7.61\text{-}4 \pm 9.58\text{-}6$	$1.147\text{-}3 \pm 6.148\text{-}5$	50.7
5.0	$6.77\text{-}4 \pm 8.77\text{-}6$	$9.268\text{-}4 \pm 3.679\text{-}5$	36.9
7.5	$4.40\text{-}4 \pm 6.10\text{-}6$	$6.020\text{-}4 \pm 2.703\text{-}5$	36.8
10.0	$2.79\text{-}4 \pm 4.53\text{-}6$	$3.322\text{-}4 \pm 2.100\text{-}5$	19.1
12.5	$1.57\text{-}4 \pm 2.27\text{-}6$	-	-
15.0	$8.41\text{-}5 \pm 1.41\text{-}6$	$9.738\text{-}5 \pm 6.583\text{-}6$	15.8
17.5	$4.86\text{-}5 \pm 8.52\text{-}7$	-	-
20.0	$2.73\text{-}5 \pm 6.77\text{-}7$	$3.098\text{-}5 \pm 2.500\text{-}6$	13.4
22.5	$1.37\text{-}5 \pm 3.47\text{-}7$	-	-
25.0	$7.72\text{-}6 \pm 1.91\text{-}7$	$1.062\text{-}5 \pm 1.076\text{-}6$	37.6
27.5	$4.35\text{-}6 \pm 1.05\text{-}7$	-	-
30.0	$2.62\text{-}6 \pm 1.19\text{-}7$	$2.735\text{-}6 \pm 3.320\text{-}7$	7.2

^a $1.142\text{-}3 = 1.142 \times 10^{-3}$.

Fig. F-4.
Thermal and epithermal neutron flux
at various distances in water from
a ^{252}Cf source.

fission chamber in water was accomplished experimentally. Counts were collected from a bare and a cadmium-wrapped fission chamber immersed in water for various californium source-detector separations. The ratio of cadmium-to-bare-detector counts is plotted in Fig. F-5. Increasing the separation between source and detector provides more thermalization of neutrons and produces a lower ratio of epicadmium-to-total counts. Typically, the detector will be separated from an actual spent-fuel assembly by about 5 cm or greater. The response of a cadmium-wrapped fission chamber will be between 1 and 2% of the bare detector response to a fission spectrum for source-detector separations of 5-15 cm.

III. FUEL MATERIAL INVENTORY

Fuel materials are available to simulate experimentally both PWR and boiling-water-reactor (BWR) fuel assemblies. PWR assemblies have a uniform fuel loading, are larger than BWR assemblies, and their isotope inventories can be calculated as a function of exposure using the CINDER code (App. D). The smaller size of BWR assemblies, compared with PWR, should enable a more uniform interrogation of their matrix. However, the non-uniform fissile loading and the addition of burnable poison rods in the BWR fuel make this measurement more difficult and less accurate compared

to PWR measurements. The present lack of a suitable method to determine BWR isotope inventory as a function of exposure poses a problem in attempting to construct experimental configurations that are representative of various exposure levels. Therefore, experimental work is focused currently on the PWR measurement problem.

The materials currently available for constructing PWR measurement geometries are listed in Table F-III. A schematic drawing of the 15 by 15 PWR fuel assembly skeleton is shown in Fig. F-6, and a picture of the assembly, loaded with fuel and being lowered into the large cylindrical tank, is shown in Fig. F-7. Fuel cladding tubes for BWR and PWR fuel are available to permit experimental rods to be filled with various materials, so that a variety of test configurations can be constructed. Both depleted and 3.3% enriched UO_2 fuel pellets are available to construct fuel rods. A californium source can be inserted at the center of two special rods built using depleted and enriched pellets. These source-fuel rods allow detector response measurements to be obtained corresponding to neutron emission from fuel at various rod positions in the assembly matrix. Matrix sensitivity effects for passive and active neutron measurements are determined using these rods.

The inventory of available BWR fuel components is listed in Table F-IV. An additional 100 mixed-oxide BWR rods will be available soon. The BWR

Fig. F-5.
Fission chamber response ratio (cadmium-covered/bare) for a ^{252}Cf source in a water environment.

Fig. F-6.
15 by 15 PWR assembly available for experiments.

rods currently available for measurement are characteristic of assemblies used in older generation reactors. Experimental work using the current BWR inventory will begin following completion of measurements with the PWR components.

IV. PWR FUEL ASSEMBLY MEASUREMENTS

Approach-to-critical measurements are required to ensure that the 15 by 15 PWR-type fuel assembly can be safely immersed in water for specified geometries and fissile material loadings. Two experimental geometries were constructed, the approach-to-critical measurements were completed,

and approval was granted by the criticality safety committee to proceed with experiments. The first geometry approved for experiments is the large cylindrical tank shown in Fig. F-7 using the 15 by 15 PWR assembly. The second approved geometry, also shown in Fig. F-7, is a rectangular tank providing a close fit to the 15 by 15 PWR assembly. This rectangular tank is surrounded by a table permitting various materials to be stacked around the assembly.

The approach-to-critical procedure consists of securing the PWR assembly, with no fuel rods, in the measurement geometry of interest and then filling with water. A ^{252}Cf neutron source is

TABLE F-III

PWR FUEL MATERIALS AND COMPONENTS

Fuel Assemblies

Array size	15 by 15
Fuel positions	204
Guide tube positions	21
Outside dimension	21.59 cm
Assembly height	127.00 cm
Rod pitch (center-center)	1.42 cm
Skeleton #1	Stainless steel guide tubes
Skeleton #2	Zircaloy guide tubes

Fuel Rod InventoryUO₂ Ceramic Fuel

14 aluminum clad 3.2 wt% ²³⁵ U
189 Zircaloy clad 3.19 wt% ²³⁵ U
15 Zircaloy clad 3.3 wt% ²³⁵ U
10 Zircaloy clad 0.7 wt% ²³⁵ U (natural)
204 Zircaloy clad 0.2 wt% ²³⁵ U (depleted)

Mixed Oxide Fuel

530 Zircaloy clad rods containing PuO₂ (~5%) mixed with UO₂

Plutonium Isotopic Composition (10/1/80)

Isotope	Range (wt%)
²³⁸ Pu	0.478- 1.03
²³⁹ Pu	73.193-76.512
²⁴⁰ Pu	15.046-18.668
²⁴¹ Pu	4.111- 6.241
²⁴² Pu	1.452- 1.975

located in one of the fuel-assembly guide tubes, and two neutron detectors are located in the water region outside the assembly. The measurement procedure consists of collecting an initial count, C_0 , with no fuel, followed by additional counts, C_1 , as fuel rods are added to the assembly. The ratio of counts, C_0/C_1 , is plotted vs fuel rod loading and used to determine the number of rods that can be added to the assembly at each step of the approach-to-critical measurement. When the array is full, the ratio C_0/C_1 provides an experimental value for fuel-assembly multiplication. A plot of the approach-to-critical data collected for the small rectangular tank, with lead and polyethylene surrounding the fuel, is

Fig. F-7.

Rectangular and cylindrical fuel assembly measurement tanks. A 15 by 15 PWR fuel assembly is being lowered into the cylindrical tank.

shown in Fig. F-8. The multiplication for this geometry was determined to be between 8 and 10. The curve in Fig. F-8 becomes less steep as more fuel rods are added, indicating a much larger quantity of fuel would be required to reach a critical (unsafe) state.

Sensitivity of neutron measurements to changes in the ²³⁵U loading of one fuel pin placed at various locations in the 15 by 15 PWR fuel assembly immersed in water was determined experimentally. A ²⁵²Cf neutron source, centered 4 cm from the assembly edge and positioned at the fuel midplane, induced fissions in the fuel. A 4-atm ³He counter, centered 3 cm from the assembly on the side opposite the source, detected neutrons produced from the source and from fissions in the fuel. The sensitivity study consisted of comparing count rates from the assembly before and after one of the 204 3.2% enriched fuel rods was replaced with a natural (0.7% enriched) rod. The matrix positions corresponding to rod locations are listed in

TABLE F-IV

BWR FUEL MATERIALS AND COMPONENTS

Fuel Assembly

Array size	6 by 6
Fuel rod positions	36
Outside dimension	16.51 cm
Assembly height	152.40 cm
Rod pitch (center-center)	2.06 cm

Fuel Rod InventoryUO₂ Ceramic Fuel

36 Zircaloy clad 2.34 wt% ²³⁵ U
6 Zircaloy clad 1.77 wt% ²³⁵ U

Mixed Oxide Fuel

36 Zircaloy clad 2-3 wt% PuO ₂

Plutonium Isotopic Composition

Isotope	wt%
²³⁸ Pu	0.28
²³⁹ Pu	75.52
²⁴⁰ Pu	18.12
²⁴¹ Pu	5.00
²⁴² Pu	1.09

Fig. F-6. Column and row positions are along the top and side of the assembly, respectively, and range from 1-15. The count-rate difference for rod substitution along column 7 is shown in Fig. F-9. The ²⁵²Cf source was adjacent to pin 8 in row 15. The sensitivity measurements indicate a decrease in the ²³⁵U loading of one rod from 3.2-0.7% can be reliably detected. The average count-rate difference along column 7 is $0.93 \pm 0.17\%$. Nearly all rods along the column produced nearly the same count-rate difference, indicating that the sensitivity to change in ²³⁵U loading is independent of the matrix location. The exception to this observation occurs for fuel rods adjacent to the neutron source. These rods produce a significantly larger count-rate difference than the average, indicating an increased sensitivity to changes in ²³⁵U loading. This same trend was observed for columns 1-6; however, the average count-rate difference is smaller, but

Fig. F-8.

Approach-to-critical data for the rectangular tank.

still observable, for fuel rod columns farther from the source and detector.

V. EXPERIMENTAL GOALS AND MEASUREMENT OBJECTIVES

Future measurements are planned with fission chambers encased in water-tight polyethylene sleeves. Experiments will first concentrate on reducing the sensitivity of detector responses to the assembly matrix. Data will be collected for various neutron source positions with different numbers of detectors at various locations around the assembly. Figure F-10 is a picture of the fission chamber and ²⁵²Cf source-positioning jig to be used for these studies. The jig is shown positioned on the top of the 15 by 15 PWR-type fuel assembly with the polyethylene detector sleeve in a horizontal position. This jig and fuel assembly will be firmly secured in the large cylindrical water tank for measurements and is shown here free-standing for visual clarity. Results of the sensitivity studies will be used to guide design of a neutron tailoring and distribution region for a ²⁵²Cf source and to specify the number of detectors and their positions around the assembly.

The primary objective of the experimental program is to develop and test the capacity of various measurement geometries to provide adequate

Fig. F-9.
Sensitivity of active neutron measurements
to fuel rod substitution.

spent-fuel measurements over a representative range of exposures and cooling times. Preliminary measurements to validate the Monte Carlo code have been completed. Initial sensitivity measurements indicate that the measurement goals may be achieved with proper design. Additional detailed sensitivity measurements will be completed to determine a design that is insensitive to the assembly matrix.

Experiments are planned on PWR and BWR assemblies using combinations of UO_2 and PuO_2 materials to simulate fuel burnups up to 50 GWd/tU. Measurements over this burnup range will be collected and evaluated in terms of developing calibration curves relating neutron measurements to effective fissile content and fuel assembly exposure. Contributions to the steady-state neutron count rate from plutonium and uranium fissions will be studied in an attempt to determine the plutonium/uranium ratio. Field measurements to test the active neutron method will follow once a measurement system has been developed, constructed, and tested at Los Alamos.

Fig. F-10.
Positioning jig containing a polyethylene-encased
fission chamber and a ^{252}Cf holding rod located
on top of a 15 by 15 PWR-type fuel assembly.