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NEUTRON MEASUREMENT TECHNIQUES FOR THE NONDESTRUCTIVE ANALYSIS OF IRRADIATED FUEL ASSEMBLIES

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ABSTRACT

Nondestructive measurement of the passive neutron signatures of irradiated light-water reactor fuel assemblies is a rapid and simple technique for verifying operator-declared exposure values. Fuel assemblies from four different reactor facilities have been measured to establish the functional relationship between the operator-declared exposure values and the experimentally measured neutron emission rates. Experimentally measured neutron emission rates of small fuel rod sections have been shown to agree with the predicted results from our calculational model. Destructive results for the actinide isotopes also agreed very well with our prediction. Neutron emission rates varied by 30 to 40% between opposite corners of the source fuel assembly. Symmetrical neutron detector systems that measure all sides simultaneously were evaluated.

I. INTRODUCTION

Passive neutron measurement of spent-fuel assemblies is a convenient and rapid technique for verifying operator-declared exposure values. This technique is being developed for use by International Atomic Energy Agency (IAEA) safeguards inspectors. To assist the IAEA, the United States is providing direct technical assistance through the International Safeguards Project Office. As part of this assistance program, the Los Alamos National Laboratory was assigned the task of developing a nondestructive assay method using neutron techniques for verifying the fissile content of irradiated light-water reactor (LWR) fuel assemblies at fuel storage facilities. This task was separated into two parts: (1) passive neutron techniques for safeguards inspection of LWR

irradiated fuel assemblies, and (2) investigation of the applicability of active neutron techniques to measurement of the remaining fissile content in irradiated fuel assemblies. This report covers only the first task, passive neutron techniques. We are not working on active neutron techniques at present.

Neutrons emitted from an irradiated fuel assembly can originate primarily from three sources: (1) spontaneous fission of actinide isotopes; (2) interaction of alpha particles from radioactive decay with light materials, ^{18}O in particular; and (3) induced fission in the fissile material from the first two sources. Over short cooling times, photoneutron production from the 150 ppm of ^2H in natural water can be significant. Principal gamma rays with energies greater than the 2.2 MeV photoneutron reaction threshold come from the decay of ^{140}La (40.2-h half-life), which is in equilibrium with its parent isotope ^{140}Ba (12.8-day half-life). Another minor source of photoneutrons comes from a small number of high-energy gamma rays from ^{106}Rh (29.8-s half-life), which is the daughter of ^{106}Ru (366.4-day half-life). For the report we assume that the number of neutrons originating from the photoneutron reaction is insignificant and that source neutrons originate primarily from the isotopes listed in Table I by the (α, n) reaction and spontaneous fission.

The curium isotopes are the dominant sources of neutrons within irradiated fuel assemblies with exposures above 10 GWd/tU (gigawatt days per metric ton of uranium).¹ These two isotopes are produced by different neutron capture chains above ^{241}Pu as shown in Fig. 1. The ^{242}Cm chain goes

whereas the ^{244}Cm chain goes

*Both ^{242}Am and ^{244}Am have metastable states.

TABLE I

PRINCIPAL SOURCES OF NEUTRONS IN IRRADIATED UO₂ MATERIALS

Isotope	Half-Life (yr)	(α, n) ^a Reaction	Spontaneous ^a Fission	Total ^a
²³⁵ U	7.04×10^8	7.11×10^{-4}	2.99×10^{-4}	1.01×10^{-3}
²³⁸ U	4.47×10^9	8.26×10^{-5}	1.36×10^{-2}	1.37×10^{-2}
²³⁸ Pu	8.78×10^1	1.34×10^4	2.59×10^3	1.60×10^4
²³⁹ Pu	2.41×10^4	3.81×10^1	2.18×10^{-2}	3.82×10^1
²⁴⁰ Pu	6.55×10^3	1.41×10^2	9.09×10^2	1.05×10^3
²⁴¹ Pu	1.47×10^1	1.27×10^0		1.27×10^1
²⁴² Pu	3.76×10^5	2.05×10^0	1.72×10^3	1.71×10^3
²⁴¹ Am	4.32×10^2	2.69×10^3	1.18×10^0	2.69×10^3
²⁴² Cm	4.46×10^{-1}	3.76×10^6	2.10×10^7	2.48×10^7
²⁴⁴ Cm	1.81×10^1	7.73×10^4	1.08×10^7	1.09×10^7

^aNeutrons per gram per second.

As shown in Table I, most of the neutrons from ²⁴²Cm and ²⁴⁴Cm come from the spontaneous fission process with $\bar{\nu}$ being 2.51 ± 0.06 for ²⁴²Cm and 2.681 ± 0.011 for ²⁴⁴Cm (Ref. 2). The plutonium isotopes can contribute a significant proportion to the total neutron source for fuel assemblies with relatively low exposures (<10 GWd/tU) or for fuel assemblies with extremely long cooling times (>100 yr). The uranium isotopes are significant neutron sources only in unirradiated fuel materials.

The remainder of the report is divided into five sections. The first section gives calculational results on the buildup and depletion of actinides in a typical United States pressurized-water reactor (PWR) fuel assembly and their influence upon the neutron source rates. The second section presents the experimental results obtained during two field exercises. The third section includes a discussion of how experimental measurements can be combined with calculational results to obtain an estimate of the residual fissile inventory in

Metastable states exist for ^{240}Np , ^{242}Am , and ^{244}Am

Fig. 1.
Formation of actinide isotopes.

irradiated fuel assemblies. The fourth section discusses detector development. The final section summarizes the evaluation of passive neutron measurements as a technique for verifying the fissile content of irradiated LWR fuel assemblies at fuel storage facilities.

II. CALCULATIONAL RESULTS

Passive neutron emission rates have been measured on irradiated fuel assemblies having limited ranges of exposures and cooling times.^{1,3,4,*}

To investigate the applicability of neutron measurement techniques in safeguarding irradiated fuel assemblies, we must understand the neutron emission characteristics for many types of irradiation histories and exposures. Therefore, we have applied calculational techniques to predict the buildup and depletion of the neutron-producing isotopes and to calculate the neutron production rates from these isotopes. Models can also be used to determine calculationally the sensitivity of the neutron emission rates to initial fuel parameters, such as ^{235}U enrichment and fuel density, as well as to different exposure conditions, power levels, irradiation histories, and thermal-epithermal-fast-flux ratios.

The calculational techniques, which complement the limited set of experimental results, can predict trends and parameter sensitivities that should be investigated more thoroughly to understand fuel parameters, irradiation histories, and measured neutron rates.

The production and depletion of actinide isotopes have been calculated for a typical PWR fuel assembly using the detailed power history from the H. B. Robinson-2 (HBR-2) reactor (Westinghouse 700 MWe reactor). These calculations were performed using an improved version of the single-point depletion code EPRI-CINDER⁵ and associated data library.⁶ Flux and shielded cross-section data were derived from the EPRI-CELL code.⁷

The EPRI-CINDER data library, describing 47 actinides, contains all pertinent problem-independent data necessary in nuclide-inventory and aggregate-property calculations for a wide spectrum of LWR fuel irradiations. These data

*This information was furnished by S. Untermyer of the National Nuclear Corporation.

include four-energy-group absorption cross sections and branching fractions for multiple paths in radioactive decay and transmutation of fission products. Also included are similar basic nuclear data for actinide nuclides produced by the transmutation of the initially present heavy-metal nuclides and their absorption/decay products. Four-group unshielded actinide cross sections complete the nuclear data library. With this library, a problem can be completely specified by defining the fuel sample power history, temporal four-group neutron flux spectrum, and initial nuclide inventory.

Four-group unshielded cross sections used in the basic EPRI-CINDER library have been produced from cross-section data in ENDF/B-IV or preliminary ENDF/B-V using a representative weighting spectrum. More accurate four-group cross-section data can be entered as problem-dependent data to override the basic library data. For calculations described in this report, four-group shielded cross sections for principal contributing actinides ($^{234-238}\text{U}$, $^{239-242}\text{Pu}$) were obtained from the EPRI-CELL code for each time step. Effects such as initial enrichment, exposure, poisons, water/metal ratio, rod pitch, and other heterogeneity considerations can be accurately modeled in this manner.

The calculational validity is demonstrated in Table II by comparing the predicted values of a selected subset of the actinide isotopes with destructively measured actinide concentrations for a fuel assembly irradiated in the HBR-2 reactor. The agreement between the two data sets is reasonably good. Isotopes having the larger ($>10\%$) differences have smaller concentrations and are therefore more difficult to measure experimentally.

Using these data, we calculated the production and depletion of the actinide isotopes of interest for a typical PWR fuel assembly. Exposures from 0 to 50 GWd/tU were simulated to represent a larger range of exposures than a safeguards inspector would normally have to measure. Atom densities for each actinide isotope were calculated at 35 time steps. The results for the fissile isotopes (^{235}U , ^{239}Pu , and ^{241}Pu) are presented in Fig. 2 along with the total fissile inventory as a function of exposure.

The principal sources of neutrons in an irradiated fuel assembly change as the irradiation exposure increases. Figure 3 shows the per cent contribution from the major neutron sources to the total neutron source strength for four levels of exposure, and Fig. 4 shows the neutron production per cubic centimeter of material. For the first case, an exposure of 10.9 GWd/tU, ^{242}Cm is source at the time of discharge. Its per cent contribution drops rapidly as a

Fig. 2.

Depletion of ^{235}U and production of ^{239}Pu and ^{241}Pu as a function of irradiation exposure.

function of cooling time because of its 0.445-yr half-life. After 20 months of cooling time, ^{240}Pu becomes the dominant source, followed in importance by ^{244}Cm (18.1-yr half-life), ^{238}Pu , and ^{241}Am . For higher exposures (>20 GWd/tU) the curium isotopes are the dominant sources of neutrons in irradiated fuel assemblies independent of cooling time.

Figure 5 shows the buildup of the two curium isotopes as a function of irradiation exposure. The calculations show ^{244}Cm beginning to build up rapidly for exposures above 20 GWd/tU, while both of the curium isotopes produce neutrons from the (α, n) reaction yield. The spontaneous fission yield of ^{244}Cm is 100 times greater than its (α, n) reaction yield. Therefore at higher exposures (>20 GWd/tU) the source neutrons arise primarily from spontaneous fission.

The calculated total neutron source ratios as a function of exposure for four specified cooling times are plotted in Fig. 6. For exposures above 10 GWd/tU the neutron source rate appears to be correlated with exposure by a power functional relationship

Fig. 3. Percent contribution of individual isotopes to the total neutron source in an irradiated fuel assembly.

Fig. 4.

Relative neutron source rates for each of the five largest contributions to the total neutron rate.

TABLE II

COMPARISON OF MEASURED AND CALCULATED ISOTOPIC RATIOS

<u>Quantity</u>	<u>Measured⁸ Value</u>	<u>Calculated Value</u>	<u>Difference (%)</u>
Burnup (at.%)	3.221	3.221	+0.00
Exposure Gwd/tU	30.92	31.19	+0.89
²³⁴ U/U	0.00014	0.00012	-12.81
²³⁵ U/U	0.00612	0.00588	-3.84
²³⁶ U/U	0.00352	0.00357	+1.52
²³⁸ U/U	0.99022	0.99043	+0.02
²³⁸ Pu/Pu	0.01676	0.01452	-13.35
²³⁹ Pu/Pu	0.54261	0.53966	-0.54
²⁴⁰ Pu/Pu	0.25101	0.24002	-4.38
²⁴¹ Pu/Pu	0.12998	0.13772	+5.95
²⁴² Pu/Pu	0.05964	0.06807	+11.41
²³⁹ Pu/ ²³⁸ U	0.00518	0.00496	-4.28
¹⁴⁸ Nd/ ²³⁸ U	0.00057	0.00059	+3.96

$$\text{Rate} = \alpha \cdot \text{Exposure}^{\beta} \quad , \quad (1)$$

where α is a scaling parameter and β ranges in value from 3.0 to 4.5 depending on the fuel being measured.

The specific values for β for four different cooling times are given in Table III for the four curves plotted in Fig. 6. Influence of ²⁴²Cm is clearly evident at 0 months cooling time, particularly in the region of lower exposures, that is, 5 to 20 Gwd/tU. These calculations are only for the source

Fig. 5.

Buildup of the two principal sources (^{242}Cm and ^{244}Cm) of neutrons as a function of irradiation exposure.

TABLE III
 PARAMETERS FOR POWER FUNCTION RELATIONSHIP

$$\text{Source Rate} = \alpha \cdot \text{Exposure}^\beta$$

Cooling Time (months)	α	β
0	2.93 E-02	3.54
12	2.60 E-02	4.11
36	6.27 E-04	4.44
120	6.31 E-04	4.37

Fig. 6.
Neutron source rate as a function of exposure for four cooling times.

neutrons and do not include any changes in the absorption or multiplication of the source neutron as a function of exposure.

Another significant factor that can influence the neutron source rates is the irradiation history. Figure 7 shows the influence of discrete 1-yr cycles in which the fuel assembly would be in the reactor (U in the legend) or out of the reactor (D in the legend). As can be seen from the data in Fig. 7, the effect of the irradiation history is most significant for short cooling times. After 36 months, all the simulated histories merge to the same relationship between the source neutron rate and exposure. This peculiar behavior is directly related to the buildup and decay of ^{242}Cm within the fuel material. During the downtimes, ^{242}Cm decays, whereas ^{241}Pu , with a half-life of 14.7 yr, β -decays to ^{241}Am , with a half-life of 432.2 yr. While in the reactor, ^{241}Am captures a neutron to produce ^{242}Am that β -decays to ^{242}Cm . The concentration of the ^{241}Am increases during the downtime, thereby providing more source

Fig. 7. Neutron source rates as a function of exposure for different irradiation cycles. Each cycle lasted for 1 yr (U = in reactor, D = out of reactor).

material from which ^{242}Cm is produced. At higher exposures, the per cent contribution of neutrons from ^{242}Cm to the total source neutrons becomes less (see Figs. 3 and 4); therefore, the irradiation history influence is greatest at the beginning of the second cycle.

Figure 7 shows the results of five irradiation histories that can be found in typical power reactors where the fuel assembly is removed for one or two cycles before being reinserted into the reactor to complete its designed exposure. As stated above, the greatest effect is observed when the fuel material is irradiated one cycle and then cooled for one or two cycles before reirradiation. For example, the same source neutron rates (10^3) are obtained for fuel material with a UDDUU history (15 GWd/tU) and a constant UUU history of 30 GWd/tU because of the increased concentration during shutdown of ^{241}Am , the source material for ^{242}Cm production.

The previous discussion shows the correlation between neutron source rate and exposure. However, safeguards inspectors are not concerned with how long a fuel assembly has been in a reactor, but rather with the fissile content of the fuel assembly. Therefore, we have examined the correlation of source neutron rate with total fissile material (Fig. 8), total plutonium (Fig. 9), and total plutonium minus ^{239}Pu (Fig. 10). The total fissile material includes ^{235}U , ^{239}Pu , and ^{241}Pu . All the functional relationships in Figs. 8-10 are single-valued and approximately linear on a semilog scale in the exposure ranges of 10 to 50 GWd/tU. In each of the plots the influence of ^{242}Cm on the neutron source strength is significant only for short cooling times (<2 yr). These correlations can be used to predict successfully the plutonium content in spent-fuel assemblies.³

The isotopic composition of the plutonium changes significantly as a function of exposure. Figure 11 shows the per cent of the total plutonium for each isotope as exposure increases. At 50 GWd/tU exposure, ^{240}Pu would be expected to be about one-half as large as the ^{239}Pu fraction. With curves such as those in Fig. 11 and information on total plutonium content (obtained from Figs. 9 and 10), estimates can be made on the specific inventory of plutonium isotopes in a spent-fuel assembly as a function of exposure.

Fig. 8.

Total neutron source rate as a function of the wt% of the total fissile material remaining in the fuel assembly for four cooling times.

Fig. 9.
Total neutron source rate as a function of the wt% of total plutonium in the fuel assembly for four cooling times.

Fig. 10.

Total neutron source rate as a function of total plutonium minus ²³⁹Pu remaining in the fuel assembly for four cooling times.

Fig. 11.
Isotopic fraction of each plutonium isotope to the total as a function of exposure.

III. EXPERIMENTAL RESULTS

Passive neutron measurement of spent-fuel assemblies is a convenient and rapid technique for verifying operator-declared exposure values to satisfy safeguards requirements. The instrumentation is simple to operate and the acquired data are reasonably easy to analyze. To demonstrate the applicability of this nondestructive technique for satisfying IAEA safeguards requirements, measurements have been made on a variety of spent-fuel assemblies. Thirty-six PWR fuel assemblies have been examined at a reactor fuel storage facility, 17 fuel assemblies from four different reactors were examined at an away-from-reactor (AFR) facility, and sections from a fuel rod were examined before being destructively analyzed.

A. Reactor Spent-Fuel Storage Facility

We measured 36 PWR fuel assemblies using a ring detector containing two ^{235}U fission chambers located on opposite sides.¹ The ring detector (Fig. 12) contained a circular ion chamber for measuring the gamma dose rate and four

Fig. 12.
Ring detector containing four fission chambers.

locations (one in each quadrant) for fission chambers or ^{10}B -lined detectors. In this particular experiment we used two fission chambers located on opposite sides (separated by 180°) and two ^{10}B -lined detectors. The gamma background was too high for the ^{10}B -lined detectors to function correctly; therefore only the results from the fission chambers are reported. Subsequent experiments have shown that small-diameter ^{10}B -lined detectors can operate in high gamma background.⁹⁻¹¹ The typical gamma dose rate at the midplane of the fuel assemblies ranged between 30 000 and 50 000 R/h.

The ring detector was placed above a fuel assembly in the reactor storage rack; then the fuel handling device was attached and the fuel assembly raised until its midplane was encircled by the detector (Fig. 13). Positioning the ring detector over a specified storage location required about 1 to 3 min with the measurement requiring 1 to 2 min to obtain 1 to 2% counting statistics. Thirty-six fuel assemblies were measured within 3 h. The neutron background for the detector with fuel assemblies in all the storage locations below the detector was less than two counts per minute, proving that only the neutron emission from the fuel assembly being measured contributed to neutron signal.

Results for these 36 measurements (Fig. 14) are given in Table IV. The raw data are the actual data recorded, whereas the corrected data were corrected for the decay of the ^{244}Cm isotope (no correction for the decay of ^{242}Cm was needed). Table V gives the parameters of the data fitted using the power function Eq. (1) for both the raw and corrected data. The correction for the ^{244}Cm decay would raise the relative neutron rates of the low-exposure fuel assemblies more than those of the higher exposure fuel assemblies because of the differences in the cooling times.

The neutron emission rate appears to be related to exposure by a power function. A semilogarithmic relationship has also been suggested for a set of PWR measurements.* This functional relationship did not appear to fit our results as well as the power function relationship. (The power function relationship also fitted the results obtained at the AFR storage facility).

The initial ^{235}U enrichment can possibly affect the results, as was discussed in Sec. II. Lower enriched fuel assemblies would be expected to have a higher neutron emission rate than higher enriched fuel assemblies because of

*This information was furnished by S. Untermyer of the National Nuclear Corporation (April 1981).

Fig. 13.

Ring detector on top of fuel storage rack with fuel assembly in measurement position.

TABLE IV

MEASURED AND CORRECTED NEUTRON RATES

Operator-Declared Values					
Fuel Assembly	Initial ²³⁵ U Enrichment (%)	Exposure (MWd/tU)	Cooling Time (Months)	Neutron Rate	
				Raw Data (Counts/min)	Corrected Data (Counts/min)
A37P	2.25	19 174	40	532	603
A38P		18 845		762	864
A39P		18 932		647	734
A49P		18 813		498	565
A59R		19 619	30	781	858
A58R		19 941		1 078	1 185
A57R		19 723		717	788
A45R		19 728		661	726
A46R		19 956		694	761 ^a
A47R		20 010		832	914
A48R		20 223		683	751
B60R	2.80	31 280	22	3 381	3 623
B61R		30 551		2 727	2 922
B62R		30 546		2 648	2 838
B63R		31 080		2 676	2 868
B50R		30 836		2 322	2 488
B51R		30 668		2 876	3 082
B49R		29 875		2 295	2 459
B48R		27 876		2 091	2 241
B47R		30 571		2 612	2 799
B07P				26 852	17
B19P	30 665		3 054	3 222	
B31P	29 699		2 404	2 536	
B43P	30 675		2 487	2 624	
B55P	31 070		2 618	2 762	
C55R	3.30	36 278	10	4 929	5 086
C57R		33 530		3 263	3 367
C58R		33 764		4 133	4 265
C59R		33 192		3 273	3 377
C47R		33 917		3 287	3 392
C48R		35 055		4 130	4 262
C03P				39 025	4
C15P	38 694		8 110	8 213	
C39P	35 086		6 773	6 859	
C51P	38 860		7 427	7 521	
C63P	32 564		5 026	5 090	

TABLE V

FITTED PARAMETERS OF NEUTRON RESULTS USING
 $\text{Count Rate} = \alpha \cdot \text{exposure}^\beta$

Parameter	Uncorrected Data	Corrected Data
All data ^a		
α	6.49×10^{-12}	2.56×10^{-11}
β	3.27	3.14
R^2	0.9526 ^a	0.09523 ^a
Only 10- to 40-month data ^a		
α	9.88×10^{-11}	3.43×10^{-10}
β	3.00	2.88
R^2	0.9679 ^a	0.9664 ^a

^a R^2 is the sum of the squares of the residuals and gives an indication of the goodness of fit. When R^2 is multiplied by 100 it represents the percentage of the total variation about the mean that is explained by the regression.

the increased production of ^{244}Cm . Insufficient data were obtained during this exercise to verify the theory.

Calculations used Monte Carlo techniques to determine the influence of dissolved boron in the water on the measured neutron emission rate.* Using reflecting boundary conditions, a PWR 15 x 15 fuel assembly was simulated by an $n \times n$ array of rods. Each cylinder represents a fuel pin and can contain a homogenous mixture of fuel materials (or their products) and Zircaloy cladding with water between the rods. Neutrons are started within the source material and their paths are traced until they interact with the detector or are captured. The specific uranium and plutonium isotopic concentrations determined from the EPRI-CINDER calculations were used in the source material for each exposure level.

*This information was furnished by P. M. Rinard of the Los Alamos Scientific Laboratory (July 1980).

PWR spent-fuel storage ponds often have approximately 2000 ppm boron. To a first approximation, it was found that the addition of boron to the pond water reduced the neutron yield but did not significantly alter the slope of the power function relating neutron yield to the declared exposure values. The dissolved boron appears to reduce by approximately half the simulated neutron rate at the detector for 15 x 15 PWR fuel assemblies for our particular counting geometry, in which the detector was located 2 cm from the surface of the fuel assembly and was entirely surrounded with borated water (Fig. 15).*

The presence of boron in the water has two effects: (1) it reduces the multiplication within the fuel assembly, and (2) it reduces the counting efficiency of the detector by absorbing neutrons before they reach the detector. The multiplication within the fuel assembly was reduced by approximately 20% over the exposure range of 18 to 39 GWd/tU. The significant shift between the two data sets given in Fig. 15 can be primarily attributed to the absorption by the boron of the neutrons traveling through ~2 cm of borated water before reaching the detector.

B. Spent-Fuel Storage Facility

International safeguards inspectors will also be required to verify the fissile content of spent-fuel assemblies at places other than reactor sites. These additional sites can either be AFR storage facilities or the receiving areas of spent-fuel reprocessing facilities. A spent-fuel measurement system was installed in the receiving pond at the General Electric (GE) Morris Spent-Fuel Storage Facility to evaluate the use of passive gamma-ray and neutron measurement techniques for verifying the declared fissile inventory of spent-fuel assemblies. This measurement program was funded under the Spent-Fuel Storage Facility Project;¹² however, the results obtained apply equally to this project.

Seventeen irradiated fuel assemblies from both boiling-water reactor (BWR) and PWR facilities were examined (Table VI). The specific discharge dates, ²³⁵U enrichments, and declared exposures are given in Table VII. There were four primary objectives of these measurements:

*This information was furnished by P. M. Rinard of the Los Alamos Scientific Laboratory (July 1980).

Fig. 15.

Simulated neutron flux in a detector similar to the one shown in Fig. 13 for two measurement environments: (a) with 2000 ppm boron and (b) without any boron.

TABLE VI

FUEL ASSEMBLIES EXAMINED AT GE MORRIS FACILITY

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

TABLE VII

INDIVIDUAL FUEL ASSEMBLIES EXAMINED

<u>Fuel Assembly</u>	<u>Discharge Date</u>	<u>²³⁵U Enrichment (%)</u>	<u>Declared Exposure (Gwd/tU)</u>
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-B12	4/16/71	3.23	26.3
B-C056	3/04/77	3.397	40.1
B-C032	4/06/74	3.400	23.6
B-C052	3/04/77	3.397	32.8
C-DN201	4/72	2.13	4.7
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	6/02/73	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	6/02/73	3.865	32.4

-
- Reproducibility: How precisely could replicate measurements be made? Also, could the errors covered by positioning and counting statistics be separated? Which of these two error sources is dominant?
 - Angular Effects: Are the neutron and gamma measurements at all four corners of the spent-fuel assembly consistent?

- Fuel-Assembly Geometry: What is the effect on measurements of variations in fuel geometry—PWR fuel assemblies with 14 x 14 and 15 x 15 arrays, and BWR fuel assemblies with 7 x 7 arrays?
- 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, assuming that the ^{242}Cm has decayed?

Another general objective of this exercise was the demonstration to the facility operator that a simple measurement system can be installed in the receiving pond without adversely interfering with the normal operations.

The measurement system consisted of a V-shaped positioning mechanism attached to a tube containing various gamma-ray and neutron detectors. The device was suspended from the wall in the receiving pond at the GE Morris Spent-Fuel Storage Facility (Fig. 16). The fuel assembly was moved to the V-shaped positioning fixture, which automatically positioned the assembly correctly (Fig. 17). This positioning process required less than a minute of the operation time. The detector pipe was located at the corner of the fuel assembly to ensure a reproducible measurement geometry for all fuel assemblies examined. The detector package consisted of three detectors: (1) an ion chamber to measure gross-gamma dose, (2) a ^{235}U fission chamber (~130 mg ^{235}U) to measure the neutron emission rate, and (3) a $\text{Be}(\gamma, n)$ detector to record the high-energy (>1.66 MeV) gamma signature¹ (Fig. 18). The installed system is shown in Fig. 19 with a PWR fuel assembly in the measurement position.

Reproducibilities of 1 to 2% were obtained for the neutron measurements,

Fig. 16.
Positioning mechanism for prototype verification and assay system.

Fig. 17.
Relative position of the fuel assembly and the detector tube.

Fig. 18.
First detector package used in the GE Morris field exercises.

Fig. 19.
PWR fuel assembly located in positioning assembly for measurement.

TABLE VIII

NORMALIZED RESPONSES FOR EACH OF THE SPENT-FUEL ASSEMBLIES^a

<u>Fuel Assembly</u>	<u>Orientation (corner)</u>	<u>Fission Chamber (counts/s)</u>	<u>Ion Chamber</u>
A-A019	A	1.00	1.00
	B	1.02	1.01
	C	1.02	1.03
	D	0.99	1.02
A-C049	A	1.00	1.00
	B	0.63	0.57
	C	0.60	0.51
	D	0.97	0.95
A-A045	A	1.00	1.00
	B	1.03	1.01
	C	1.02	1.04
	D	0.94	0.98
A-B012	A	1.00	1.00
	B	1.04	1.02
	C	0.98	0.97
	D	1.00	0.99
B-C056	A	1.00	1.00
	B	1.03	1.01
	C	0.98	1.01
	D	0.91	0.93
B-C032	A	1.00 (1.00) ^b	1.00 (1.00) ^b
	B	0.72 (0.72)	0.70 (0.70)
	C	0.85 (0.84)	0.83 (0.83)
	D	1.25 (1.22)	1.12 (1.12)
B-C052	A	1.00	1.00
	B	0.95	0.85
	C	1.37	1.22
	D	1.30	1.24
C-DN201	A	1.00	1.00
	B	0.90	0.88
	C	0.82	0.81
	D	0.88	0.86
D-A051	A	1.00	1.00
	B	0.97	0.96
	C	1.07	1.03
	D	1.00	1.01

TABLE VIII (cont)

<u>Fuel Assembly</u>	<u>Orientation (corner)</u>	<u>Fission Chamber (counts/s)</u>	<u>Ion Chamber</u>
D-C004	A	1.00	1.00
	B	0.77	0.83
	C	0.90	0.90
	D	1.21	1.11
D-D001	A	1.00	1.00
	B	1.00	0.98
	C	1.05	1.02
	D	1.03	1.02
D-C001	A	1.00	1.00
	B	1.16	1.09
	C	0.88	0.92
	D	0.76	0.80
D-A036	A	1.00	1.00
	B	0.94	0.98
	C	0.89	0.93
	D	0.95	0.95
D-D020	A	1.00	1.00
	B	0.87	0.94
	C	0.72	0.76
	D	0.85	0.90
D-A009	A	1.00	1.00
	B	0.99	0.99
	C	1.01	1.00
	D	0.96	0.97
D-D046	A	1.00	1.00
	B	0.92	0.95
	C	1.00	0.98
	D	1.05	1.02
D-C020	A	1.00	1.00
	B	1.14	1.08
	C	1.32	1.18
	D	1.18	1.11

^aAll numbers are normalized to the count rate obtained for position A.

^bNumbers in parentheses are for a second set of data taken at the end of the exercise.

with counting statistics contributing about 0.9% to the total variability. Having established the measurement precision, we investigated the variability in the corner measurements of individual fuel assemblies by obtaining repeated (3 to 6) measurements at each corner. The results of these measurements are given in Table VIII. Neutron emission rates varied by 30 to 40% for individual corners. Similar variability was observed in the ion chamber results.

The results from these angular effects measurements are presented in Figs. 20-23 for reactors A,B,C, and D, respectively. The data were corrected for the decay of ^{244}Cm (18.1-yr half-life) because of the differences in cooling times. No correction was included for the ^{242}Cm 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 only 0.003 of the original ^{242}Cm remained.

The maximum variability in the measurements for each of the four corners for the 17 fuel assemblies is given in Table IX. Each of the data sets from the PWR fuel assemblies had a maximum difference of approximately 40% for at

Fig. 20.
Relative neutron emission rates as a function of the four corner positions for fuel assemblies from Reactor A.

PWR - 14 x 14 Assemblies - 450 MWe Rated Power
Exposure Range 23.6-40.1 GWd/tU

Fig. 21.

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

BWR - 7 x 7 Assembly - 715 MWe Rated Power
DN201 - 4.67 GWd/tU

Fig. 22.

Relative neutron emission rates as a function of the four corner positions for a fuel assembly from Reactor C.

PWR - 14 x 14 Assemblies - 430 MWe Rated Power
Exposure Range 26.5-32.4 GWd/tU

Fig. 23.

Relative neutron emission rates as a function of the four corner positions for a fuel assembly from Reactor D.

TABLE IX

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

least one fuel assembly. This high variability does not appear to be related to 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 cruciform control blades are used in BWRs.

The results shown in Fig. 21 exhibited changes ranging from 12 to 42% between the corners of the fuel assemblies. The individual irradiation history of each of the fuel assemblies in the reactor was determined and is listed in Table X. A schematic of the reactor core is given in Fig. 24 with the specific angular orientation of each fuel assembly. In this case, the fuel assemblies cannot be rotated. As Table VIII and Fig. 24 show, the corners with the lowest relative neutron rates were always on the periphery of the reactor core. This effect appears to persist even if the fuel assembly is moved to an interior position, as was the case for B-C052. From these data, it appears that the first cycle of a PWR fuel assembly defines a gradient that persists through the irradiation history.

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. 25 for each of the three PWR data sets. The parameters for the best fits of the power functional relationship are given in Table XI. The number of fuel assemblies

TABLE X
INDIVIDUAL IRRADIATION HISTORIES OF FUEL ASSEMBLIES

<u>Fuel Assembly</u>	<u>Core Position</u>	<u>Average Exposure (GWd/tU)</u>
C032	F-1	10.80
	D-4	12.77
		<u>23.57</u>
C056	K-10	17.13
	C-5	13.34
	C-5	9.60
		<u>40.07</u>
C052	A-8	11.23
	F-2	12.75
	F-2	8.84
		<u>32.82</u>

Fig. 24.
Reactor core in which fuel assemblies B-C032, B-C056, and B-C052 were irradiated.

Fig. 25.
Data from the three PWR data sets with the individual power functions (best fit) and the collective results (dashed line).

TABLE XI

PARAMETER OF 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 = $\alpha \cdot \text{exposure}^\beta$.

^b R^2 is the correlation coefficient and when multiplied by 100 it represents the percentage of the total variation about the mean that is explained by the regression.

measured was limited, as well as the range of exposures, but the data appears to be consistent with the previously obtained measured and calculated results.

We could not determine if there were any significant differences between the PWR fuel assemblies with 15 x 15 and 14 x 14 arrays. Figure 26 shows a comparison of the results from the 15 x 15 fuel assemblies (C056 and C052) and the 14 x 14 fuel assemblies (C020, D046, and D001). The amount of data is insufficient to draw any specific conclusions; however, note that the results are similar for the fuel assemblies with approximately the same exposures.

Two specific conclusions can be based upon these results:

1. Precise neutron measurements (~1%) can be obtained using simple measurement systems.
2. Passive neutron measurements must be made on all four corners or sides of a fuel assembly, otherwise the results may be erroneous.

IV. DESTRUCTIVE ANALYSIS

At the Battelle Columbus Laboratories (BCL) in Columbus, Ohio, fuel rods from spent-fuel assemblies are sectioned for eventual dissolution and isotopic

Fig. 26.

Comparison of results from 15 x 15 (B-C056 and B-C052) fuel assemblies with 14 x 14 (D-C020, D-D046, and D-D001) fuel assemblies.

analysis. A program is in progress to make nondestructive measurements on these rod sections before they are dissolved. For convenience, a portable instrument, the High-Level Neutron Coincidence Counter (HLNCC),¹³ was used for these measurements. This detector not only measures the signal from all neutrons emitted by the sample but also determines coincidence counts from neutrons emitted in spontaneous fission events. The nondestructive measurements at BCL were valuable because direct correlations can be made between the neutron count rate data and the isotopic composition determined by destructive means. The destructive data, which include analysis for most actinide isotopes from uranium through curium, are also valuable for comparison with calculationally estimated quantities.

The rod sectioning, isotopic analysis program is being performed at BCL as part of a joint Baltimore Gas and Electric Company, Combustion Engineering Company, and Electric Power Research Institute Demonstration Fuel Assembly Program. The particular fuel assembly being analyzed was a specially adapted assembly from the Calvert Cliffs reactor facility. The demonstration program specified that this assembly be in the core through four cycles and that specified rods be removed from the assembly for various types of examination after

each cycle. The assembly was designed to be reconstituted with new rods before being returned to the core. Rods remaining in the assembly through the four cycles would obtain exposures between 40 and 50 GWd/tU.

Unfortunately, Los Alamos involvement in this program did not begin until after rods were removed at the end of three cycles. No neutron measurements were made on rods removed after the first two cycles. For the three-cycle rods, exposures ranged from 34 to 37 GWd/tU. These rods had initial ^{235}U enrichments of 2.33 or 2.45% and cooling times of 17 months. From Fig. 6, which is for a 2.56% initial ^{235}U enrichment, one can estimate the neutron source rate for samples with 36 GWd/tU exposures and 17 month cooling times to be approximately $7000 \text{ n}/(\text{s}\cdot\text{cm}^3)$. From the experimental measurements, the neutron source rate is approximately $6700 \text{ n}/(\text{s}\cdot\text{cm}^3)$. This experimental value was determined assuming a 12% detector efficiency and a mass density of $10.3 \text{ g}/\text{cm}^3$ for the fuel.

The HBR-2 model used for the calculational results shown in Fig. 6 is for a 15×15 array, PWR assembly and not for a 14×14 array like the Calvert Cliffs fuel. Also the calculational source rate terms do not include any transport effects. Self-shielding in the actual samples probably would have lowered the experimental count rates. The reasonably close agreement between the estimated calculational and experimental values gives some confidence that the calculational model is correctly predicting neutron source rates. Data from the four-cycle analysis are needed to confirm this conclusion.

In Fig. 27 we compare the calculated data and the destructive data for the Calvert Cliffs rod sections. The area indicated by the box in Fig. 27 is enlarged and shown as Fig. 28. In these figures the $^{244}\text{Cm}/^{239}\text{Pu}$ ratio is shown as a function of exposure. This ratio is used because the destructive data for the curium isotopes were reported in this fashion. As has been stated previously, ^{244}Cm is the dominant neutron source in fuels with high exposures and long cooling times. The ability to relate neutron count rates to ^{244}Cm content and then correlate the ^{244}Cm to parameters such as total fissile, total plutonium, or total plutonium minus ^{239}Pu , as in Figs. 8, 9, and 10, depends on calculational models. If these models are proved correct, then they can be used in conjunction with experimental measurements to estimate the fissile or plutonium content of spent-fuel assemblies.

Information in Figs. 27 and 28 for the limited data currently available tends to confirm calculational predictions of ^{244}Cm concentrations at least

Fig. 27.
Comparison of calculated and experimentally measured results.

Fig. 28.
Expanded curve showing the detailed comparison of experimental and calculational results.

for exposures up to 37 GWd/tU. As shown in Fig. 28 most of the destructive data are within the indicated error limits of the calculational curves. When the four-cycle data are available, they will be very important in this analysis. As seen in Fig. 27, the $^{244}\text{Cm}/^{239}\text{Pu}$ ratio increases dramatically for exposures between 35 and 50 GWd/tU. If the calculational models are incorrect, a larger error between experimental and calculational results would probably occur for the high exposures of the four-cycle rods.

A third comparison between calculated data and destructive analysis data is shown in Table XII. In this table, plutonium isotopic data are compared. The calculated data were taken from the curves shown in Fig. 11. Comparisons in Table XII are reasonably good. Certainly the calculated data have the proper trends. Discrepancies, particularly in the ^{240}Pu and ^{241}Pu comparisons, may be related to using the HBR-2 calculational model. Further calculations will be performed using specific Calvert Cliffs models to see if there is better agreement on these plutonium isotopic data.

These preliminary results indicate that calculational techniques are effective in predicting spent-fuel parameters. In some cases, for example, the $^{244}\text{Cm}/^{239}\text{Pu}$ ratio, the value may be insensitive to specific reactor parameters. In this situation, modeling of a generic type of reactor may be sufficient to generate calculated quantities. For data such as the plutonium isotopics,

TABLE XII

PLUTONIUM ISOTOPIC COMPARISONS FOR ROD SECTIONS
WITH EXPOSURES OF APPROXIMATELY 36 GWd/tU

Isotope	Per Cent of the Total	
	Calculated Value	Measured Value
^{238}Pu	1.7	2.277
^{239}Pu	49.9	48.533
^{240}Pu	24.3	28.216
^{241}Pu	15.4	12.553
^{242}Pu	8.7	8.421

which may be more dependent on specific reactor parameters, precise modeling might be necessary to achieve very accurate data. Further investigations will be made to determine the applicable ranges for the calculated data.

V. DETECTOR DEVELOPMENT

The measurement of the neutron yield from spent-fuel assemblies is complicated by the very high gamma dose (50 000 to 100 000 R/h) originating from fission products. Thus any detector used to count neutrons should be insensitive to gamma-ray pile-up problems. High efficiency for counting neutrons is desirable to reduce measurement times, but the neutron emission rate from spent-fuel assemblies is large enough (10^7 to 10^8 n/s) that the efficiency requirement can be relaxed.

Several different types of thermal-neutron detectors have been considered for the application to the measurement of spent-fuel assemblies. Fission chambers have received most attention because of their excellent gamma-ray rejection capabilities. Fission chambers with diameters up to 25 mm can be operated in gamma fields of over 10^5 R/h.

A. ^{10}B Detector

Fission chambers containing ^{235}U can be a transportation problem because of limitations placed on the transport of special nuclear material across national boundaries. Applicability of neutron detectors other than ^{235}U fission chambers has been evaluated by various experimenters.⁹⁻¹¹ Other detectors such as $^{10}\text{BF}_3$ gases and ^{10}B -lined detectors have significantly better neutron detection efficiencies. Table XIII lists some of the detectors that were considered for the present application.

The sensitivity of neutron detectors to gamma-ray pile-up problems is also a function of the detector size. In general, the larger the diameter and length of the detector, the greater is the gamma-ray pile-up problem. Small ^{10}B detectors (5 to 13 mm in diameter and a few inches long) can operate in gamma doses of over 10^4 R/h.^{9,10} One or two centimeters of lead shield can extend this range of operation up to 10^5 R/h. Measurements* on spent-fuel

*This information was furnished by G. W. Eccleston and H. O. Menlove of Los Alamos Scientific Laboratory (December 1980).

TABLE XIII

THERMAL-NEUTRON DETECTOR COMPARISON

Detector Type	Diam x Length (mm)	Fill Pressure (atm)	Operating Voltage (V)	Efficiency (cps/nv/cm)	Max γ^a Dose (R/h)
^{235}U fission chamber	25 x 152	1	400	0.01	10^5
^3He detector	25 x 152	4	1500	1.1	2
BF_3 detector	25 x 152	1	1800	0.32	10
^{10}B detector	25 x 152	<1	900	0.10	10^3
^{10}B detector	12 x 152	<1	700	0.05	10^4
^{10}B detector	6 x 152	<1	700	0.02	2×10^4

^aMaximum γ dose corresponds to unshielded condition. The addition of 1 or 2 cm of lead will increase this value 10 times.

assemblies have shown that the dose level is reduced more than 300 times using 5 cm of lead. The lead shielding has little or no effect on the neutron counting efficiency.

Because the ^{10}B detectors contain no radioactive material and they have higher efficiency than fission detectors, they might be used as a substitute for fission detectors in spent-fuel applications.

B. Neutron Ring Detector

The problem of variability in the neutron emission rates from the individual sides or corners can be partially resolved by using a detector that measures all four sides or corners at the same time. To attain this goal, we have developed a square detector (Figs. 29 and 30) in which four ^{235}U (130 mg) fission chambers and four ion chambers are placed parallel to the sides of the fuel assembly. This detector was designed to measure PWR fuel assemblies. Similar detectors can be made for measuring BWR and other types of fuel assemblies. This particular detector will be demonstrated to IAEA personnel as part of the equipment for measurement with minimum movement of spent-fuel

Fig. 29.
Square detector system to measure all four sides of a fuel
assembly simultaneously.

Fig. 30.
Ring detector placed around a 15 x 15
PWR fuel assembly.

assemblies. This detector is placed on top of the spent-fuel storage rack; the fuel assembly is then raised by the operator (see Fig. 13).

Some storage facilities for spent-fuel assemblies have boron dissolved in the water to reduce the reactivity of the stored fuel. Boron reduces the neutron counting rate (see Sec. III). The effect of the boron on the detector counting efficiency can be largely eliminated by counting only epithermal neutrons from the assembly. This can be accomplished by surrounding the neutron detector (fission chamber) with a few centimeters of polyethylene moderator covered with a cadmium liner. The cadmium negates the influence of the boron in the water by absorbing the thermal neutrons before they reach the detector. The polyethylene slows down the epithermal neutrons near the detector by scattering reactions and thus increases the detector counting efficiency. The boron in the water still has the effect of decreasing the neutron multiplication in the fuel assembly, but this effect is much smaller than the detector efficiency change, which can be eliminated with the cadmium-lined detector.

In some cases it might be desirable to measure the boron concentration in the water to correct the neutron yield data. This can be accomplished by having two adjacent thermal neutron detectors in the probe. One detector is bare, using the pool water as its moderator, and the other detector is surrounded by polyethylene and cadmium as described above. When the fuel assembly is positioned next to the probe, the ratio of the counting rates in the two detectors will give a measure of the boron concentration in the pool water.

VI. CONCLUSIONS

Passive neutron measurements can verify the exposure and fissile contents of irradiated fuel assemblies. Instrumentation and detectors are uncomplicated and easy to operate. Measurement times of 1 to 5 min are sufficient to obtain 1 to 2% relative precision. Detector systems can be portable (square ring detector) to measure fuel assemblies in the storage racks at reactors or can be in-plant systems to measure fuel assemblies as they are received at a storage or reprocessing facility.

Neutron emission rates from irradiated fuel assemblies can vary significantly (from 30 to 40%) at a specific axial location depending upon the corner or side being measured. Therefore a single position neutron measurement may

not be representative of the true "signature." This variability with respect to angular orientation has been measured on fuel assemblies from one BWR and three PWR facilities. Therefore neutron emission rates must be measured at more than one angular orientation to obtain a representative result.

Calculational techniques should be used to complement experimental measurements because of the limited exposure ranges for fuel assemblies from a single reactor. Calculational techniques can also be used to determine the sensitivity of the neutron source rates to various initial fuel parameters (per cent initial enrichment) and irradiation histories. Simulation of specific detector designs using Monte Carlo techniques can significantly improve the design of the measurement system.

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