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NONDESTRUCTIVE DETERMINATION OF BURNUP AND COOLING TIMES OF IRRADIATED FUEL ASSEMBLIES

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Abstract

The applicability of nondestructive gamma-ray and neutron techniques for the characterization of irradiated fuel assemblies has been investigated. Consistency of relative cooling times can be verified using specific isotopic ratios. The axial activity profiles of assemblies have been measured using a variety of detectors. Declared burnup values for MTR, BWR, and PWR assemblies have been correlated with the $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ isotopic ratios.

1. Introduction

The safeguards groups at the Los Alamos Scientific Laboratory (LASL) have been actively investigating nondestructive techniques for the characterization of irradiated fuel assemblies. At present there does not exist an applicable nondestructive technique for directly measuring the fissile content and in particular, the plutonium content in irradiated fuel assemblies. These values can be inferred from the burnup values which can be calculated indirectly from measured gamma-ray and neutron signatures. We have investigated the correlation of these indirect measurements over specific ranges of declared burnup values for Materials Testing Reactor (MTR), Boiling Water Reactor (BWR), and Pressurized Water Reactor (PWR) fuel assemblies. The relationships have been quantified by using established statistical techniques. This report summarizes some of the more important results and conclusions of the spent fuel examination program at LASL.

We have investigated the applicability of nondestructive gamma-ray and neutron methods for the verification of declared irradiation exposure of fuel assemblies. The verification of irradiated fuel assemblies can involve two distinct levels: qualitative verification and quantitative verification.[1] Qualitative verification involves the identification of the material as irradiated fissile material, while the quantitative verification requires the measurement of parameters which can be related to burnup and/or plutonium content. The qualitative verification can be accomplished by establishing that the fuel material contains the appropriate mixture of fission product gamma rays and has a reasonable level of neutron emission. Quantitative verification is more difficult in that the relationships between the measured parameters and burnup must be established and quantified. In this investigation we have attempted to identify the measurable parameters that show the best correlations with declared values.

This investigation is basically divided into three sections. The first section is concerned with checking the relative consistency of the declared cooling times. As both the gamma-ray and neutron signatures are time dependent, the values must be corrected to a specified time before comparisons can be performed. The second section involves the rapid measurement of the relative burnup profile of the fuel assembly. This profile is then used as an integrating function to determine the total burnup of the assembly. The third section discusses the correlations between both the spectral analysis and the neutron results and the declared burnup values.

2. Material and Method

To investigate the applicability of nondestructive gamma-ray and neutron techniques for the characterization of irradiated materials, we have examined three types of fuel assemblies: Materials Testing Reactor (MTR), Boiling Water Reactor (BWR), and Pressurized Water Reactor (PWR). The specific ranges of operator-declared burnup values and declared cooling times for the assemblies examined are listed in Table I.

TABLE I

IRRADIATED FUEL ASSEMBLIES EXAMINED

Type Assembly	Burnup Range	Cooling Time	Fissile Material
MTR	27.4 - 33.5 at.%	438 - 1456 days	93% ^{235}U
BWR	4356 - 18804 MWD/MTU	119 - 1445 days	3.3-4.3% ^{235}U
PWR	15804 - 32195 MWD/MTU	140 - 836 days	2.1-3.1% ^{235}U
BWR	7400 - 11450 MWD/MTU	627 - 630 days	2.56% ^{235}U
BWR	7480 - 12500 MWD/MTU	627 - 630 days	1.52% Pu

The gamma-ray techniques ranged from the simplicity of ion chambers to the complexity of high-resolution gamma-ray spectroscopy, while the neutron technique was restricted to the use of ^{235}U fission chambers. A typical experimental configuration for a reactor spent fuel storage facility is shown in Fig. 1. Complete gamma-ray spectra were collected at specified axial positions on the fuel assemblies and analyzed for comparison to the other profile monitor systems. Also, the gamma-ray data was correlated with the operator-declared burnup values and cooling times to verify the consistency between the declared values and the measured parameters.

The axial gross gamma activity profiles were measured using ion chambers, cadmium telluride, germanium, and Be(γ,n) detectors. The Be(γ,n) detector (Fig. 2) is only sensitive to gamma rays with with energies greater than the 1660 keV threshold for photoneutron production in beryllium.[2] Neutrons produced in the Be are thermalized by the polyethylene and are counted in the fission chamber. This particular detector is relatively insensitive to high gamma-ray fluxes. In a fission product

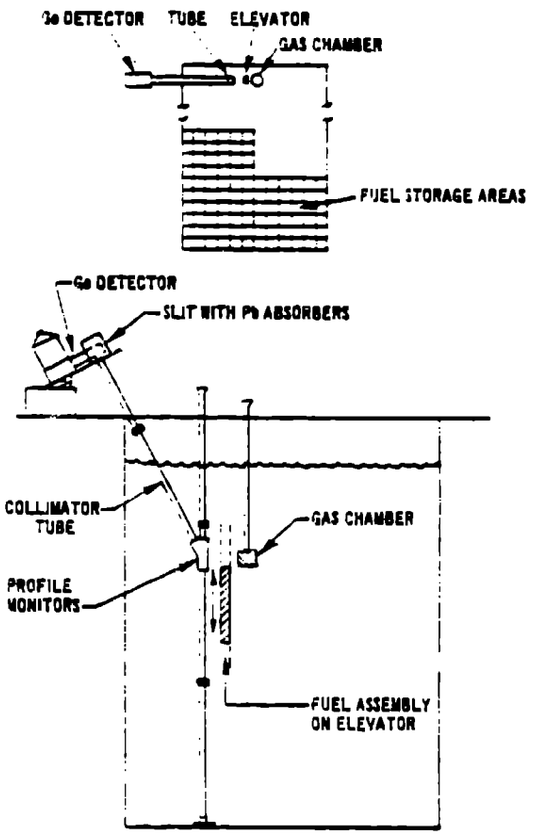

Figure 1.
Schematic of experimental apparatus used to examine the BWR fuel assemblies.

spectra, the principal gamma ray with an energy above the 1660 keV threshold is the 2186 keV gamma ray of ^{144}Pr. The ^{144}Pr isotope has a very short half-life ($t_{1/2}$ = 17.3 min) and is in secular equilibrium with its parent ^{144}Ce (284.5 d). Therefore, the Be(γ,n) profile is essentially the profile of the ^{144}Ce axial distribution.

The relative neutron emission rates of the irradiated assemblies were measured using ^{235}U fission chambers with loadings ranging from 35 mg to 1.5 g. The actual fission chamber used depended upon the neutron source intensity of the fuel assembly as well as the source-to-detector distance.

3. Gamma-Ray Results

Relative Efficiency Corrections. For the BWR and PWR assemblies the relative efficiency was calculated at each axial position to determine if it differed significantly between individual positions on the same assembly and if it differed significantly between individual fuel assemblies. The relative efficiencies were determined using the relative intensities of the ^{134}Cs, ^{106}Rh, and ^{144}Pr gamma-ray

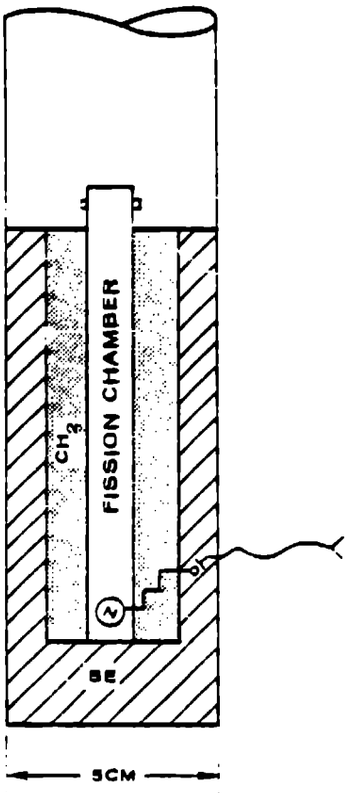

Figure 2.
Be(γ,n) detector for measuring the high-energy (>1660 keV) gamma activity profile.

lines. Statistically the results indicated that the relative efficiencies did not significantly change within each exercise. The variability of the efficiencies between specific axial positions did not vary by more than the uncertainty due to counting statistics from the measured intensities. If longer counting times were used, a difference may have been detected. Therefore, the net areas for all the full-energy peaks were corrected for differences in the relative detection efficiencies using the ^{134}Cs results and normalizing to the 604.6 keV gamma-ray line.

Consistency of Declared Cooling Times. All of the measurable gamma-ray and neutron signatures of irradiated fuel assemblies are functions of time dependent variables. Therefore, the measured parameters must be corrected for the cooling times prior to data interpretation. We have selected the date of discharge from the reactor as the date to which we have corrected all of the gamma-ray results. To evaluate the consistency of declared cooling times, the following nonlinear model relating measured activity ratios to cooling times was used:

$$Tc = \frac{1}{\lambda_j - \lambda_i} \ln \frac{A_i}{A_j} \frac{A_{oj}}{A_{oi}} \qquad (1)$$

where λ_i is the decay constant, A_i, the measured activity at time Tc, and A_{oi} is the

activity at Tc=0 for the ith isotope. Various statistical techniques were used to quantify the ratios that correlated well with the declared cooling times. The isotopic ratios of $^{144}Pr/^{137}Cs$ and $^{106}Rh/^{137}Cs$ appeared to correlate the best. The average percent differences between the declared cooling times and the regression equation were 2.7% and 5.3% respectively. The value of A_{oj}/A_{oi} is assumed to be a constant. This approximation appears to be acceptable for the case of MTR fuel assemblies examined but may not be readily adaptable to LWR fuel assemblies. Figure 3 shows the correlation between the measured $^{144}Pr/^{137}Cs$ activity ratio and the declared cooling times for seven MTR fuel assemblies with cooling times of 662 days to 1456 days. This type of analysis could indicate the presence of a relative measured value lying outside of specified limits. The regression line as well as the 95% confidence bounds are plotted in Fig. 3 to indicate the level of correlation. It should be pointed out that the actual cooling times have not been determined, but rather that all the measured parameters are internally consistent.

Axial Activity Profiles. If a rapid technique for accurately measuring the axial burnup profile of a fuel assembly can be developed, then a detailed measurement can be performed at one location and can be related to the entire assembly by using the profile as an integrating function. We have investigated the applicability of various nondestructive techniques: ion chambers and CdTe detectors operated in the current mode, and CdTe and germanium detectors as gross gamma counters. Activities measured by these techniques were compared with the relative ^{137}Cs isotopic activity determined from the germanium system. The ^{137}Cs profile was assumed to be the best predictor of the burnup profile.

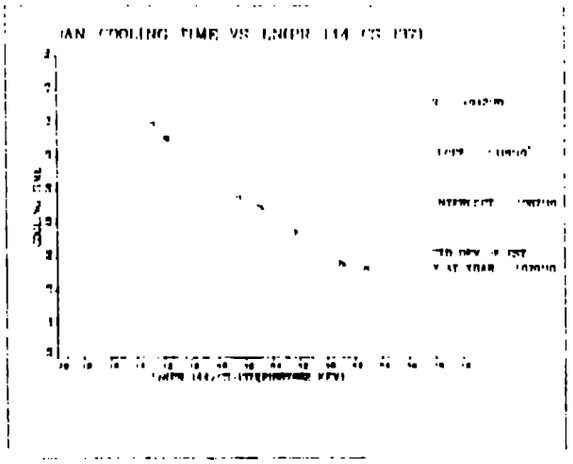

Figure 3.
Comparison of the declared cooling times of MTR fuel assemblies and the calculated regression equation.

The comparisons of the ion chamber results with the ^{137}Cs activity profiles have been previously reported.[3] The correlations between the ^{137}Cs profiles and the Be(γ,n) profiles for the BWR assemblies were very good, with over 91% of the variability in the ^{137}Cs profiles being explained by the Be(γ,n) profile (Fig. 4). A comparison of the gross gamma profiles and the ^{137}Cs profiles showed that only 64% of the variability in the ^{137}Cs profiles can be explained by the gross gamma profile. For these BWR assemblies the maxima of the gross gamma profiles appear to be shifted towards the lower end of the fuel assemblies when compared to the ^{137}Cs and ^{144}Pr profiles.

As discussed previously, the Be(γ,n) detector is primarily responsive to the ^{144}Pr activity which is in secular equilibrium with the ^{144}Ce parent isotope ($t_{1/2}$ = 284.5 d). The Be(γ,n) profile will be influenced more by the recent flux profile, than the ^{137}Cs activity profile ($t_{1/2}$ = 30.12 yr), therefore, the results should be interpreted cautiously when the irradiation period is long relative to the ^{144}Ce half-life.

Relative Burnup Measurements. All the prominent gamma rays of the fission product spectra were correlated directly with the declared burnup values. In addition, selected isotopic ratios were also evaluated as to their linear correlation with the declared burnup values. The accuracy of the declared burnup values was estimated to be approximately 5%. The ^{137}Cs relative activities correlated with the declared burnup values for the MTR, BWR, and PWR fuel assemblies with the percent average differences from the regression equation of 2.5%, 12.3%, and 5.1%, respectively.

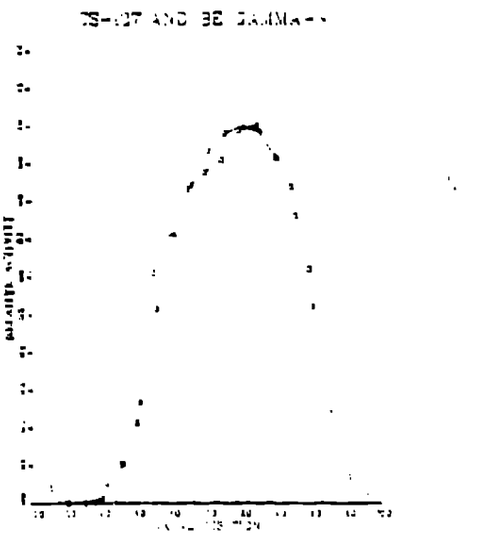

Figure 4.
Axial profile determined from the ^{137}Cs activity (□) and the Be(γ,n) detector (○).

TABLE II

COMPARISON OF OPERATOR DECLARED VALUES AND VALUES CALCULATED FROM REGRESSION EQUATION

Declared Burnup MWD/MTU	Calculated Burnup[1]		
.6606	.6715	.5674	.5313
.7406	.7791	.6252	.6887
.7776	.8028	.7116	.6458
.8279	.8556	.7630	.7298
.8773	.9048	.8330	.8617
.9911	1.0571	1.0085	.9716
.9828	1.0547	1.0113	.9997
1.0066	1.0973	1.0800	1.0458
1.0252	1.0610	1.0972	1.0699
1.0139	1.0535	1.0035	1.0290
1.1050	1.1066	1.0827	1.1751
1.1051	1.1086	1.1796	1.0713
1.2096	1.2178	1.2136	1.2096
1.2195	1.2605	1.1945	1.2088
Average Percent Difference	3.1%	7.1%	5.1%

[1] The regression equations provided the best estimates of the linear relationships between the declared burnup values and the measured parameters.

Each of the gamma-ray variables, single peaks and ratios, were corrected using the operator-declared cooling times to the activities present at the time of discharge. The ^{137}Cs activity correlated very well with the declared burnup values as is indicated in Fig. 5 for the PWR assemblies. Similarly, the linear correlations of the $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ ratios are presented in Figs. 6 and 7, respectively. For each of the plots the 95% confidence bounds are presented. Table II lists the comparison of the declared burnup values and the calculated values from the measured intensities collected at a specified axial position. For the PWR fuel assemblies the axial burnup profiles were relatively uniform through the central region of the assemblies.

The relationship between the isotopic ratios, $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$, and the declared burnup values should not necessarily be linear over this range of burnup values. On selected MTR, BWR, and PWR assemblies, the relationships of the shielded isotopes, ^{134}Cs and ^{154}Eu, with respect to the ^{137}Cs activity were investigated by measuring the relative axial profiles. The following relationship was evaluated,

$$(^{134}Cs \text{ or } ^{154}Eu) = A(^{137}Cs)^B \qquad (2)$$

with particular interest upon the values of B. Results of this analysis are presented in Table III. It is evident that the value of B is significantly less than 2.0 for these ranges of burnup. If the value of B were 2.0, then the relationship between burnup and isotopic ratios $^{134}Cs/^{137}Cs$ or $^{154}Eu/^{137}Cs$, would be linear and the ratios could be directly related to burnup.[5,6] One assumption required for these comparisons is that there was no significant difference in the axial migration properties of these isotopes.

When the ratios are corrected for this nonlinear relationship and correlated with the declared values, the average differences do not significantly improve. However, over a wider range of burnup values this nonlinearity could have a significant affect upon the correlation of the isotopic ratios to the burnup values.

TABLE III

POWER RELATIONSHIP OF ^{134}Cs AND ^{154}Eu WITH ^{137}Cs

Fuel	B-Values ^{134}Cs	^{154}Eu
MTR	1.81 ± 0.11	1.75 ± 0.22
BWR	1.54 ± 0.21	1.53 ± 0.22
PWR	1.53 ± 0.22	1.76 ± 0.36

$^{134}Cs = A(^{137}Cs)^B$

$^{154}Eu = A(^{137}Cs)^B$

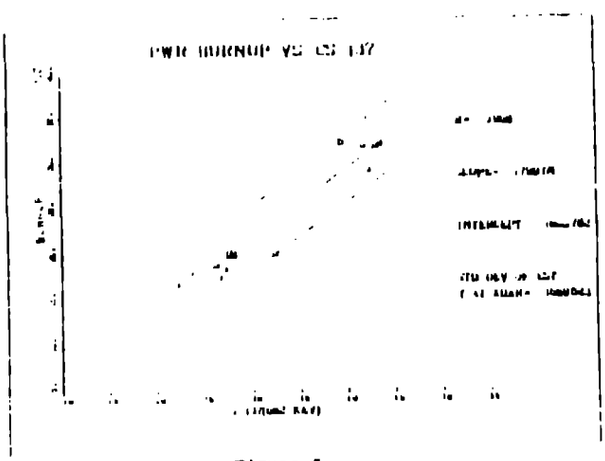

Figure 5.
Correlation of the measured ^{137}Cs activities with the declared burnup values.

Each assembly was oriented in the same manner, that is, all the assemblies were scanned on the same side. To evaluate the effect of the selection of a specific side upon the results, several fuel assemblies were measured on all four sides. For both the BWR and PWR assemblies, the variability in the calculated burnup values at the four sides was approximately the same as the variability between assemblies. These results should be considered as tentative because of the small number of fuel assemblies examined.

4. Neutron Results

Neutrons produced by the spontaneous fissioning of actinide elements and produced via the (α,n) reaction on ^{18}O can provide a signature which can be used to characterize irradiated materials. Calculationally, it has been shown that for a PWR assembly the interior rods of an assembly contribute nearly the same amount to the total neutron emission rate as the exterior rods.[4] The simplicity of the neutron

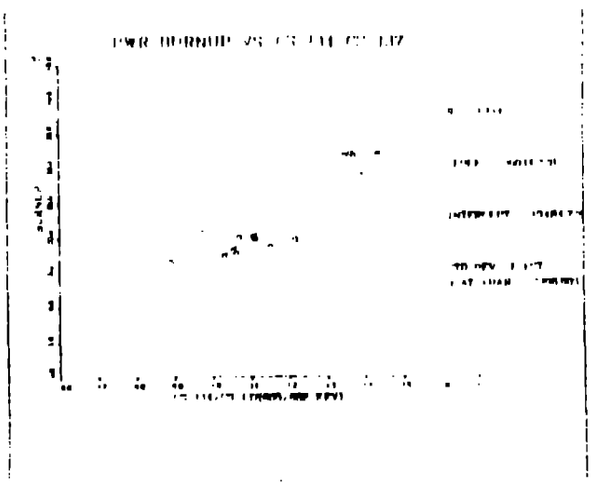

Figure 6.
Correlation of the measured $^{134}Cs/^{137}Cs$ isotopic ratios with the declared burnup values.

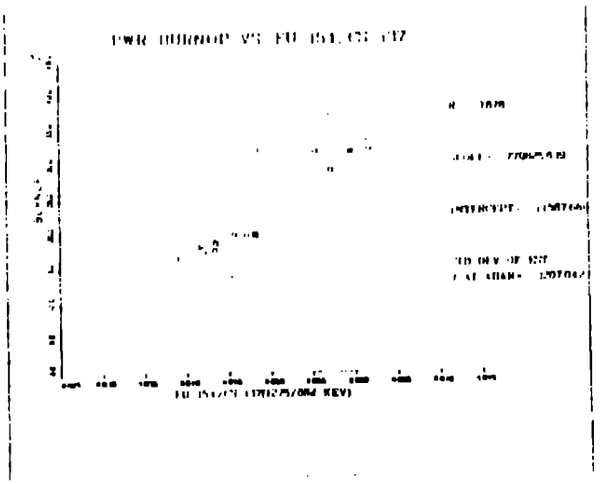

Figure 7.
Correlation of the measured $^{154}Eu/^{137}Cs$ isotopic ratios with the declared burnup values.

measurement technique makes it particularly attractive for the measurement of irradiated fuel assemblies.

Both BWR and PWR assemblies have been examined to investigate the correlation between declared burnup values and the relative neutron emission rates. Only the results of the examination of BWR assemblies are included because this set included a range of burnup values (4356 to 17814 MWD/MTU) with identical cooling times. Table IV lists the measurement results obtained for a set of BWR assemblies with a range of burnup values from 4356 to 17814 MWD/MTU. All of these fuel assemblies had the same cooling times; this eliminated the problem of making a cooling time correction.

A power series relationship would be expected to explain the relationship between neutron count rate and the declared burnup because of the required multiple neutron captures to produce the transuranic isotopes which have been shown to be the major contributors to the neutron emission rate. However, because of the same number of data points a simplified power relationship between the measured count rates over a limited range of burnup values was evaluated at four axial positions. The value of n in the following relationship

$$\text{Count Rate} = k(\text{Burnup})^{n} \qquad (3)$$

was calculated for each axial position (Table IV), with the average value of 4.2 for BWR-2 thru BWR-5. By expanding the range of burnup this assumption fails as is evidenced by a value of 2.6 when the low-burnup assembly, BWR-1, is included in the analysis.

The problem of variability due to the selection of measurement orientation for neutron measurements was investigated for both the BWR and PWR assemblies by measuring the neutron emission rates from the four surfaces. For one BWR assembly the count rate varied by more than the counting statistics depending upon which side was examined. This additional variability may have been the result of varying the source-to-detector distance corresponding to ±1.0 cm. The relative neutron flux decreased by a factor of ten in water as the distance from the centerline of the assembly was increased from 12 to 22 cm. These measured values compared very well with calculated values.[4] The variability measured for four PWR assemblies was approximately the same as the statistical uncertainty. To eliminate the possibility of any dependence, upon which side is measured, the neutron measurements can be obtained from all sides by rotating the assembly or by using an annular detector which measures the neutron rate from a cross section of the assembly. Additional investigations must be completed to allow the complete interpretation of the neutron measurements and how these measurements should be correlated with burnup and/or fissile content of the fuel assemblies.

To understand this relationship better, a ^{235}U irradiated pin from an experimental BWR assembly has been destructively analyzed by mass spectrometry and alpha spectrometry to determine the relative concentrations of actinides as a function of burnup. From these measurements the relative contributions to the total neutron rates for specific isotopes are listed in Table V for burnup values ranging from 7400 to 11450 MWD/MTU for a cooling time of 2.4 years which was the cooling time of the assemblies listed in Table IV. The measurement errors have been estimated to be 0.5% for ^{240}Pu, 1.0% for ^{242}Cm, and 6% for ^{244}Cm. As can be seen from Table V, the neutrons originate primarily from the ^{240}Pu ($t_{1/2}$ = 6570 years) and ^{244}Cm ($t_{1/2}$ = 18.11 years) with the fraction from ^{244}Cm increasing as burnup increases. At the time of discharge the ^{242}Cm isotope is the dominant contributor to the neutron rate, but with a relatively short half-life ($t_{1/2}$ = 163 days) the longer-lived isotopes, ^{240}Pu and ^{244}Cm, become the predominant contributors at cooling times greater than two years. From Table IV, a higher value of n would be expected as the average burnup values increased because of the higher average mass value of the transuranic isotopes.

TABLE IV

RELATIVE NEUTRON COUNT RATES AS A FUNCTION OF AXIAL POSITION

Assembly	Operator Declared Burnup--MWD/MTU	Axial Position A	B	C	D
BWR-1	4356	0.095±0.012	0.105±0.012	0.115±0.013	0.108±0.012
BWR-2	13332	0.953±0.037	1.273±0.043	1.264±0.042	0.800±0.034
BWR-3	15264	3.586±0.072	4.141±0.077	2.656±0.062	0.916±0.036
BWR-4	17122	3.900±0.075	5.017±0.085	3.769±0.073	1.317±0.043
BWR-5	17814	4.215±0.078	5.420±0.088	4.308±0.078	2.015±0.054
Power Relationship:	Count Rate = a(Burnup)^b				
BWR 2 through 5					
	b	4.32	4.86	4.17	2.30

TABLE V

PERCENT CONTRIBUTION TO TOTAL NEUTRON RATE
FOR SPECIFIC ISOTOPES AT A COOLING TIME OF 2.4 YEARS

Declared Burnup (MWD/MTU)	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{242}Pu	^{241}Am	^{242}Cm	^{244}Cm
7400	1.01	0.40	10.26	37.20	1.57	4.10	15.04	30.35
9760	0.50	0.36	5.84	26.55	1.68	3.48	13.60	47.93
9840	0.49	0.41	5.33	25.11	1.57	3.85	15.51	47.66
11450	0.36	0.42	3.80	22.66	1.84	3.42	15.60	51.83

5. Conclusions

A variety of nondestructive techniques have been investigated to evaluate their applicability to the characterization of irradiated fuel assemblies for safeguards. Passive gamma-ray and neutron techniques have been correlated with operator-declared burnup values to identify the variables which best estimate these values. For fuel assemblies having similar irradiation histories, the ^{137}Cs activity, the $^{134}Cs/^{137}Cs$ ratio, and the $^{154}Eu/^{137}Cs$ ratio correlate well with the declared burnup values. For PWR assemblies the average differences between the burnup values provided by the facility operator and the relative burnup values calculated from the regression equation ranged from 5 to 9%.

Various techniques were used to measure the axial burnup profiles of the assemblies. These results were compared to the axial ^{137}Cs activity profile, which was assumed to provide the best estimate of the burnup profile. The relative Be(γ,n) profile compared favorably with the ^{137}Cs profile, while the gross gamma profile compared less favorably. The Be(γ,n) method provides a rapid technique for obtaining an integrating function which can be used in conjunction with the more detailed analyses to obtain the average burnup of the entire assembly.

The analysis of the neutron results indicate that the relative count rates may be related to the operator-declared burnup values for restricted ranges of burnup. Neutron signatures are important in that they can be used to verify the existence of fissionable material whereas gamma-ray techniques can only verify the presence of fission products.

Any safeguards verification system for irradiated fuel assemblies should include both gamma-ray and neutron measurement techniques. The consistency of operator-declared values of irradiation history of a set of irradiated fuel assemblies can be verified using these nondestructive techniques.

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