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Self-Interrogation of Spent Fuel

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SELF-INTERROGATION OF SPENT FUEL

by

D. M. Lee and L. O. Lindquist

ABSTRACT

A new method for the assay of spent-fuel assemblies has been developed that eliminates the need for external isotopic neutron sources, yet retains the advantages of an active interrogation system. The assay is accomplished by changing the reactivity of the system and correlating the measurements to burnup.

I. INTRODUCTION

The assay of spent fuel is important for nuclear material safeguards. It provides information for detecting diversion of special nuclear material and for ensuring criticality control of spent-fuel storage pools. The information is also useful for process control in reprocessing plants.

Measurement techniques for the assay of spent fuel include passive gamma-ray and neutron methods and active neutron methods. Passive gamma-ray and neutron measurements can be correlated with burnup to verify the fissile material inventory of spent fuel. However, passive gamma-ray assay is not sensitive to the interior of the spent-fuel assembly and therefore cannot verify the integrity of the interior fuel rods.

Passive neutron assay relies on correlations between the neutron emission rate and declared burnup to determine the fissile content of the spent-fuel assembly. The neutron emission rate depends on the multiplication of the assembly and the spontaneous fission rate of the isotopes of plutonium and curium. Although a substantial fraction of the neutron emission rate is due to the curium isotopes, good correlation between the total uranium and plutonium content and the neutron rate has been observed for pressurized-water reactor (PWR) fuel assemblies, primarily because the curium production rate is a predictable function of burnup.¹

Recently, the relationship between the neutron emission rate and burnup for boiling-water reactor (BWR) fuel assemblies has proved to be double valued.² The transuranic production chain depends on the thermal-to-epithermal neutron flux ratio and in BWR reactors this ratio depends on the void fraction. For the same burnup the upper portion of the fuel assembly has more transuranium nuclides than the lower portion; the increased void fraction in the upper portion causes the neutron energy spectrum to be harder in that portion.

Active assay has traditionally been considered to be the best assay method for spent fuel because it is the only technique that measures the fissile isotopes directly. In this technique an external neutron source induces fission in the uranium and plutonium isotopes. The fission neutrons are detected and correlated either to burnup or fissile content. The interrogating sources can be isotopic sources such as ^{252}Cf , $^{124}\text{Sb-Be}$, AmLi , etc., or accelerator sources such as sealed-tube neutron generators. Systems based on prompt and delayed neutron counting using either ^{252}Cf or $^{124}\text{Sb-Be}$ sources or neutron generators have been designed and tested.³⁻⁶ These systems require strong sources (3 Ci ^{252}Cf ; 800 Ci $^{124}\text{Sb-Be}$) so that the induced signals can be measured in the presence of a strong passive neutron emission rate. For high-burnup, freshly discharged reactor fuel, the strength of the interrogating source can at times be prohibitively large.

A new assay technique has been developed for spent-fuel assemblies that eliminates the need for external isotopic sources, yet retains the advantage of an active interrogation system. This self-interrogation technique is potentially capable of assaying any spent-fuel assembly, even a freshly discharged spent-fuel assembly. This report documents the experimental and calculational results of a preliminary investigation of this self-interrogation technique.

II. PRINCIPLES

This section compares the principles of the self-interrogation method with those of the active source interrogation method.

A. Active Neutron Interrogation Method (External Sources)

A typical active interrogation system is shown in Fig. 1. The neutron source of strength S_0 neutrons/second emits neutrons that interact with the fissile material of the fuel assembly and cause fissions. The neutron detector detects the neutrons from the induced fissions as well as the passive neutron background from spontaneous fission of the transuranic isotopes. The neutrons from the induced fissions are proportional to the amount of fissile material present in the fuel assembly.

Mathematically, the detected neutron count rate with active interrogation, N_A , is equal to the passive neutron count rate N_p plus the induced neutron count rate N_i , so that

$$N_A = N_p + N_i . \quad (1)$$

The induced count rate N_i depends on the neutron source strength and the amount of fissile material as well as geometrical factors and cross sections. The induced rate N_i can be written as

$$N_i = g(x, y, z, U, Pu, \dots) S_0 , \quad (2)$$

Fig. 1.
A generic spent-fuel
active assay system.

where the dependence on the source strength is explicitly shown. The undefined function $g(\dots)$ depends on all other variables of the system, including fissile content. Combining and rearranging Eqs. (1) and (2), the induced rate can be written as

$$N_i = N_A - N_p = g(x, y, z, U, Pu, \dots) S_0 . \quad (3)$$

An assay is accomplished by correlating the function $g(\dots)$ with the fissile content.

B. Self-Interrogation Method

A generic self-interrogation system is shown in Fig. 2. The self-interrogation method is based on changing the neutron absorption in the vicinity of the fuel assembly and measuring the subsequent change in neutron signal. This is accomplished by surrounding the assembly with a material that has a high neutron-absorption cross section. Neutron absorption eliminates the induced fissions in the fissile material resulting from those neutrons that normally would have reflected back into the fuel assembly.

Fig. 2.
A generic spent-fuel self-interrogation assay system.

Mathematically, the neutron count rate N_R with the absorber in place can be written in a manner similar to Eq. (1).

$$N_R = N_p - N_j \quad , \quad (4)$$

where N_p is the passive neutron count rate previously defined and N_j is the count rate reduction due to the absorber. N_j depends on the original passive rate N_p , the fissile content, and geometrical factors. By analogy to Eq. (2), N_j can be expressed as

$$N_j = f(x, y, z, U, Pu, \dots) N_p \quad , \quad (5)$$

where the dependence on the passive neutron rate is explicitly shown. The undetermined function $f(\dots)$ contains all other variables. Combining and rearranging Eqs. (4) and (5), the rate N_j can be expressed as

$$-N_j = N_R - N_p = - f(x, y, z, U, Pu, \dots) N_p \quad . \quad (6)$$

A comparison of Eqs. (3) and (6) reveals that one of the differences between the active interrogation technique and the self-interrogation technique is that the thermal neutron source in Eq. (3) is replaced by thermal neutrons in Eq. (6). The unspecified functions f and g , which are very complex, contain all the parameters that describe the fuel assembly, including the geometry, reflector, moderator, and fissile content. Because, in general, the fissile content is the only variable, a correlation to f or g provides a means of assaying the fuel assembly.

In the active interrogation technique, the assay is accomplished by producing more fissions in the fuel assembly system and by keeping all parameters of the system fixed. In the self-interrogation method, the reduction of fissions is caused by a change in the reflector. Because the fuel assembly and

reflector comprise a subcritical assembly with an effective multiplication constant, k_{eff} , the change in the reflector material can be viewed as a change in the reactivity of the system.

The criticality equation for a subcritical finite assembly can be written as⁷

$$k_{eff} = k_{\infty} P_T P_F < 1 \quad , \quad (7)$$

where k_{∞} is the multiplication constant for an infinite system, P_T is the thermal nonleakage probability, and P_F is the fast nonleakage probability. The nonleakage probabilities are less than 1 for finite systems, which means that some neutrons are lost to the system and cannot contribute to the continuation of a chain reaction. However, neutrons that penetrate into the reflector material surrounding the fuel assembly and are reflected back into the fuel assembly are not considered lost from the system because these neutrons can still cause fissions. If the reflector material is replaced by an absorbing material, the neutrons are absorbed. The nonleakage probabilities are decreased; that is, more neutrons are lost. The decrease in the nonleakage probabilities creates a decrease in the multiplication constant k_{eff} .

By treating the fuel assembly as a multiplying system, the passive neutron rate N_p can be written as⁷

$$N_p \approx M_1 S; M_1 \approx \frac{1}{1-k_1} \quad , \quad (8)$$

where S is an unspecified source term and M_1 is the multiplication of the fuel assembly. A similar expression can be written for the neutron rate N_R when an absorbing reflector is in place.

$$N_R \approx M_2 S; M_2 \approx \frac{1}{1-k_2} \quad (9)$$

$$k_2 = k_1 - \Delta k \quad .$$

By expressing Eq. (9) in this manner, we imply that the changes observed can be expressed entirely by a change in the multiplication of the system. In addition, we assume that Eqs. (8) and (9) can satisfactorily describe this system.

Combining and rearranging Eqs. (8) and (9) gives the following:

$$\begin{aligned} N_R - N_P &= -M_1 M_2 \Delta k S \\ &= -N_P M_2 \Delta k \\ &= -\left(\frac{\Delta k}{1-k_1 + \Delta k}\right) \cdot N_P \quad . \end{aligned} \quad (10)$$

If Eqs. (6) and (10) are compared, the function f can be written as

$$f = \frac{\Delta k}{1-k_1 + \Delta k} \quad , \quad (11)$$

where the relationship to the multiplication constant of the fuel assembly is shown. The sensitivity of this technique is determined by the magnitude and sign of the change in Δk and the value of k .

III. CALCULATION OF k AND Δk

To determine the magnitude of the expected change in the multiplication constant caused by changes in the reflector material, we used the Los Alamos National Laboratory Monte Carlo neutron transport code (MCNP).⁸ This code permits detailed modeling of the geometry of the measurement system and includes the capability of calculating the eigen values (k_{eff}) for multiplying

systems. The neutron cross sections were derived from the ENDF/B-IV library, where linear interpolation was used between energy points. A few hundred to a few thousand tabulated energy points were available, depending on the isotope. For thermal collisions, scattering was determined by either a free-gas model or a thermal scattering model. In the free-gas model, neutrons are assumed to be transported in a monatomic gas that has an isotropic Maxwellian distribution of velocities. In the thermal scattering model, chemical binding and crystalline effects are considered. The thermal scattering model was generally used for neutron energies below 2 eV.

The criticality calculation is based on tracking groups of source neutrons until termination. Neutron termination results from either a fission or a loss from the system by absorption or leakage. After all source neutrons are processed, k_{eff} is calculated by dividing the total number of fission neutrons produced by the source size. This procedure can be repeated for any number of cycles, and the calculated k_{eff} can be an average of the cycles.

A 15-by-15-rod PWR fuel assembly of 3.2% enrichment was modeled for the calculation of k and Δk . Characteristics of the fuel assembly are shown in Table I and the geometry is shown in Fig. 3. This fuel assembly is identical to that used for the experimental measurements described in Sec. IV. The fuel assembly was divided into 225-unit cells, each 1.42 cm square by 120 cm long. The open cells (water) correspond to cooling channels; the fuel cells were a homogeneous mixture of fuel, cladding, and water. The speed and efficiency of the MCNP code were improved by homogenizing the unit fuel cell.

The calculation involved determining k_{eff} for the fuel assembly with the water reflector only and then determining k_{eff} for conditions where a 0.038-cm-thick cadmium sheet was placed next to one or more sides, 1.27 cm from the fuel assembly. The results of the k_{eff} calculation are shown in Fig. 4. The k_{eff} for a totally water-reflected assembly was determined to be 0.84. This result appears reasonable when compared to the k_{eff} (0.89) for a 17-by-17-rod, 3.5%-enriched PWR fuel assembly.⁹ In Fig. 4 the effective multiplication constant is plotted as a function of the number of sides covered with cadmium. As expected, the effective multiplication constant decreases with increasing cadmium coverage and a 10 to 12% change can be achieved. This implies, by substitution into Eq. (10), that as much as a ~40% change in neutron rate is possible.

TABLE I

PWR ASSEMBLY: FUEL ROD CHARACTERISTICS

Fuel Rods

Number	204
Diameter	1.08 cm
Length	129 cm (122 cm of UO_2)
Cladding thickness	0.089 cm

Fuel Rod Material

Mass	1504 g
Matrix	UO_2
Enrichment	3.2%
^{235}U mass	31.0 g
Density	10.5 g/cm ³

Fuel Assembly

Fuel rod locations	204
Open channels	21
Rod spacing	1.42 cm

PWR SPENT FUEL GEOMETRY

Fig. 3.
Calculational model of a spent-fuel assembly.

A second series of calculations examined the sensitivity of the change in k_{eff} using different spacing between the absorber and the fuel assembly. Figure 5 shows the results when two adjacent sides were covered with cadmium. The change in k_{eff} is maximum when the cadmium is placed close to the detector; the change decreases as the separation increases.

IV. MEASUREMENT OF A SIMULATED SPENT-FUEL ASSEMBLY

The self-interrogation technique relies on the capability of changing the passive neutron emission rate by changing the multiplication of the fuel assembly. We simulated a spent-fuel assembly by inserting a fission source into an unirradiated PWR fuel assembly. This arrangement allowed easy access and greatly simplified our measurements. The purpose was to verify experimentally the results of our calculations and to understand the effect of reflector changes for different source locations in the fuel assembly.

The experimental configuration is shown in Fig. 6. The PWR assembly, identical to the assembly described in Sec. III (Table I), was partly immersed in a 200-l container of distilled water. The removable cadmium sheets were held

Fig. 4.
Calculated effective multiplication constant vs the number of sides covered with cadmium.

Fig. 5.
Variation of k_{eff} with different spacing between the fuel assembly and cadmium absorber.

Fig. 6.
Measurement geometry
for simulated spent-
fuel assembly.

in a supporting frame 1.27 cm from the fuel assembly. The cadmium sheets (0.038 cm thick by 21.6 cm wide by 76 cm long) were supported by a 0.156-cm-thick fiber glass sheet.

The thermal neutron detector and the epithermal neutron detector were 4-atm ^3He gas proportional counters, 122 cm long by 2.54 cm in diameter. The epithermal neutron detector was surrounded by a 5.08-cm-diam aluminum cylinder wrapped with a 0.038-cm-thick cadmium sheet; the moderating material between the detector and the aluminum cylinder was water. Thermal neutrons (<1 eV) are absorbed in the cadmium sheet; epithermal neutrons pass through the cadmium, are thermalized in the water moderator, and then detected by the ^3He detector.

The first series of measurements consisted of inserting a ^{252}Cf isotopic source ($\sim 3.5 \times 10^4$ n/s) into one of the cooling channels and observing the count rates in the thermal detector with one cadmium sheet inserted. The results are shown in Fig. 7. The numbers represent the ratios of the count rates observed after the cadmium sheet was inserted to the count rates observed before insertion of the cadmium. The net change in count rate with the cadmium sheet inserted depends on the relative magnitude of the sensitivity

Fig. 7.
The numbers represent the ratios of the count rates observed after the cadmium sheet was inserted to the count rates observed before insertion of the cadmium.

to source neutrons and neutrons that exit the side of the fuel assembly. The source locations closer to the cadmium give a higher probability of neutron leakage and produce the greatest changes in count rate. It is desirable to maximize the count-rate change for each location by placing cadmium on more than one side. This is equivalent to maximizing the change in the multiplication constant of the system.

Figure 8 shows the results for the thermal detector with cadmium sheets on three sides. The measurements were made by removing one fuel rod at a time and inserting the ^{252}Cf source. Although the rod removal produces a small perturbation from the original configuration, the general results should be very similar. The fractional count-rate changes range from 0.10 to 0.40 with a simple average of 0.265. The smallest changes occur for locations closest to the neutron detector, analogous to the situation in Fig. 7.

The results for the epithermal neutron detector are shown in Figs. 9 and 10 for different configurations of cadmium sheets. For coverage on three sides, the fractional count-rate changes range from 0.18 to 0.39 with a simple average of 0.34; for four sides, the changes range from 0.19 to 0.48 with a

Fig. 8.
Fractional count-rate changes for the thermal detector with the ^{252}Cf source in different locations in the fuel assembly. Three sides were covered.

Fig. 9.
Fractional count-rate changes for the epithermal detector with the ^{252}Cf source in different locations in the fuel assembly. Three sides were covered.

Fig. 10.
Fractional count-rate changes for the epithermal detector with the ^{252}Cf source in different locations in the fuel assembly. Four sides were covered.

simple average of 0.375. The increased sensitivity might result from the increased mean-free path for the epithermal neutrons when compared to thermal neutrons.

An interesting comparison (Table II) can be made between the measured changes and the calculated changes [Eq. (11) and Fig. 4]. For the epithermal detection system, the calculated and experimental values show surprisingly good agreement. Evidently the average values measured and the simplistic derivation of Eq. (11) are reasonable approximations and adequately describe the behavior of the system. The values in parentheses in Table II are the results obtained for a different set of measurements (the various sides of the assembly were covered in different order). Some reconfiguration of the detectors occurred between the two measurements. Reasonably consistent values were obtained by the epithermal detector system, irrespective of the order in which the sides were covered. The measured results for the thermal detector system generally show smaller changes than the calculated results and display large differences depending on which side is covered. In the thermal system, adjacent-side coverage produces a greater change than opposite-side coverage. Smaller thermal mean-free paths can account for this difference.

The third series of measurements investigated whether removal of fuel rods could be detected. The geometry for these measurements is shown in Fig. 11. The ^{252}Cf source was separately placed in four locations surrounding the center of the fuel assembly and the sum of the responses for the four locations was used for the analyses. Measurements for the full fuel assembly were first taken with no cadmium sheets and then with various cadmium sheets in place on

TABLE II
CALCULATED AND EXPERIMENTAL AVERAGE COUNT-RATE CHANGES

<u>Cd</u>	<u>Experimental</u>		<u>Calculated</u>
	<u>Epithermal</u>	<u>Thermal</u>	
1	0.19 (0.17)	0.08 (0.17)	0.17
2	0.26 (0.24)	0.17 (0.25)	0.28
3	0.34 (0.36)	0.27 (0.28)	0.35
4	0.38		0.38

Fig. 11.
Assembly geometry for measurement of sensitivity to rod removal.

different sides. (For the thermal system, a sheet was not used in front of the detector.) Then the eight center fuel rods (shown as Xs in Fig. 11) were removed and the measurements were repeated. The results are tabulated in Table III. The errors are due to counting statistics alone. Similar to the response observed previously, the epithermal detector system showed a greater sensitivity than the thermal system. The removal of eight fuel rods can be detected readily with the epithermal detector but poorly with the thermal system.

TABLE III
MEASURED COUNT-RATE CHANGES FOR ROD REMOVAL

<u>Geometry</u>	<u>Epithermal</u>		<u>Thermal (3 sides)</u>
	<u>3 Sides</u>	<u>4 Sides</u>	
Full	0.337 ± 0.002	0.359 ± 0.002	0.248 ± 0.001
Eight rods missing	0.318 ± 0.002	0.344 ± 0.002	0.245 ± 0.001

V. DISCUSSION

Extending the present results to spent-fuel assemblies of different burnup can be accomplished by relating the burnup dependence on criticality to our results. The criticality as a function of burnup for an infinite lattice (k_{∞}) of 3.1%-enriched PWR fuel pins is shown in Fig. 12. By comparing the k_{eff} value for a fresh assembly (0.84) to the data shown in Fig. 12, the combined nonleakage probability can be estimated to be $(P_F P_T) = 0.60$. If we assume that the change in the criticality is due entirely to a change in the combined nonleakage probability, that is,

$$\Delta k_{eff} \approx k_{\infty} \Delta(P_T P_F) \quad ,$$

then, from our results that $\Delta k_{eff} = 0.101$ for fresh fuel, the quantity $\Delta(P_T P_F)$ is approximately

$$\Delta(P_T P_F) = \frac{0.101}{1.41} = 0.072 \quad .$$

Using the combined nonleakage probability (0.60) and $\Delta(P_T P_F) = 0.072$ as the same values for PWR spent-fuel assemblies of different burnup, an estimate for the fractional change in count rate can be determined from Eq. 11. The results are shown in Fig. 13. The slope at 35 000 MWd/tU is 0.0038 $\Delta f/1000$ MWd/tU.

Fig. 12.
Calculated infinite multiplication constant
vs burnup. (Data taken from Ref. 5.)

Fig. 13.
Estimated fractional count-rate change
vs burnup.

This implies that a precision of $\sim 2.4\%$ on f is required for a ~ 1000 MWd/tU precision on burnup. Repeat measurements have shown that this level of precision is possible.

The detection of rod removal for higher burnup fuel will be more difficult because the initial multiplication constant will be smaller and consequently the change in multiplication constant will be smaller. Detailed calculations must be performed before any definitive statement can be made as to the level of sensitivity to rod removal for higher burnup fuel.

The self-interrogation technique can assay any spent-fuel assembly regardless of how strong the passive neutron rate; the signal-to-noise ratio depends on the reactivity changes and not on the passive neutron rate. The signal-to-noise ratio can be expressed as

$$\frac{S}{N} = \frac{N_j}{N_p} = \frac{f(x, y, z, Pu, U, \dots) N_p}{N_p} = f(\dots) ,$$

where the dependence on $f(\dots)$ is explicitly shown. The significant fact is that the signal-to-noise ratio in this approximation does not depend on N_p , the passive rate.

A similar expression for the active technique is

$$\frac{S}{N} = \frac{N_i}{N_p} = g(x, y, z, Pu, U, \dots) \frac{S_0}{N} .$$

Here the signal-to-noise ratio depends directly on the ratio of the source strength to the passive neutron rate. If, as in high-burnup, short cooling-time spent fuel, the passive rate is very high, then the source also must be very strong.

VI. CONCLUSION

The possibility of using the self-interrogation, or reactivity, method for assaying spent-fuel assemblies has been demonstrated. Although the mathematical relations have been simplistic, agreement between the data and the calculations suggest that the approximate treatment is correct and can be used to estimate the effects of reflector changes. Computing time and measurement time limited the scope of this initial work to the investigation of cadmium as the absorber. Future investigation will include additional work on other absorbers in conjunction with more efficient reflectors, as well as tests of the effects of rod replacement.

This self-interrogation method is applicable to the assay of any fissionable material that emits neutrons. In particular, the assay of plutonium-based materials is possible.

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