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TITLE: ASSAY OF SPENT FUEL BY A SIMPLE REACTIVITY METHOD

AUTHOR(S): D(avid) M. Lee, MP-13
L(oyd) O. Lindquist, Q-2

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ASSAY OF SPENT FUEL BY A SIMPLE REACTIVITY METHOD*

D.M. Lee and L.O. Lindquist
Los Alamos National Laboratory
PO Box 1663
Los Alamos, NM 87545

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.

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.¹ 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.

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 ²⁵²Cf, ¹²⁴Sb-Be, AmLi, etc., or accelerator sources such as sealed-tube neutron generators. Systems based on prompt and delayed neutron counting using either ²⁵²Cf or ¹²⁴Sb-Be sources or neutron generator have been designed and tested.²⁻⁴ These systems require strong sources (5 Ci ²⁵²Cf; 800 Ci ¹²⁴Sb-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. The 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 the self-interrogation technique.

PRINCIPLES AND COMPARISON TO ACTIVE NEUTRON INTERROGATION METHODS

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.

If the fuel assembly is treated as a multiplying system, the passive neutron count rate can be treated as,⁵

$$N_p = M_1 S ; M_1 = \frac{1}{1 - k_1} \quad (1)$$

where S is an unmultiplied source term due to the neutron emission from the plutonium and curium isotopes, M_1 is the multiplication of the fuel assembly and k_1 is the effective multiplication constant. Similarly, the induced rate with the external source can be written as

$$N_1 = M_1 S_0 \quad (2)$$

where the assumption is made that the multiplication M_1 is the same for the external source neutrons. An assay is accomplished by measuring the passive neutron rate, N_p , the active neutron rate, N_A with the external source and forming the quantity

$$N_1 = N_A - N_p = M_1 S_0$$

and correlating N_1 to the fissile content.

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.

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

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

*This work was performed under the auspices of the US Department of Energy, Contract W-7405-ENG-36.

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

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 as⁵

$$k_{eff} = k_{\infty} P_T P_F < 1, \quad (3)$$

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} .

A similar expression to equation (2) can be written for the neutron rate N_R when an absorbing reflector is in place,

$$N_R = M_2 S; \quad M_2 = \frac{1}{1 - k_2}$$

$$k_2 = k_1 - \Delta k. \quad (4)$$

By expressing Eq. (4) 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. (1) and (4) can satisfactorily describe this system.

Combining and rearranging Eqs. (1) and (4) gives the following:

$$N_R - N_P = M_1 M_2 \Delta k S$$

$$= -N_P M_2 \Delta k$$

$$= -\left(\frac{\Delta k}{1 - k_1 + \Delta k}\right) \cdot N_P. \quad (5)$$

An assay is accomplished by correlating the quantity $(N_R - N_P)/N_P$ to the fissile content. The sensitivity of this technique is determined by the magnitude and sign of the change in Δk and the value of k_1 .

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).⁶ 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 tracing 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

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

Table I and geometry is shown in Fig. 3. This fuel assembly is identical to that used for the experimental measurements. 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 and 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 fuel cell.

The calculation involved determining k_{eff} for the fuel assembly with a 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.⁷ 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. (5), 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

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

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.

The experimental configuration is shown in Fig. 5. 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 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-atm³He 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 ²⁵²Cf isotopic source (~3.5 x 10⁴ n/s) into one of the cooling channels and observing the count rates in the thermal detector and epithermal detector with one cadmium sheet inserted. Additional measurements were made with cadmium sheets on more than one side.

A comparison of the results between the measured changes and the calculated changes [Eq. (5) and Fig. 4] is shown in Table II. 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. (5) 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

Fig. 5. Measurement geometry for simulated spent-fuel assembly.

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.

TABLE II

CALCULATED AND EXPERIMENTAL AVERAGE COUNT-RATE CHANGES

Cd	Experimental		Calculated
	Epithermal	Thermal	
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

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 3.14-enriched PWR fuel pins is shown in Fig. 6. By comparing the k_{eff} value for a fresh assembly (0.84) to the data shown in Fig. 6, the combined nonleakage probability can be estimated to be $(P_T P_F) = 0.60$. If we assume that the change in the criticality is due entirely to a change in the combined nonleakage probability, that is,

$$\Delta k_{eff} \approx k_{\infty} \Delta(P_T P_F), \quad (6)$$

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

$$\Delta(P_T P_F) = \frac{0.101}{1.41} = 0.072. \quad (7)$$

Fig. 6. Calculated infinite multiplication constant vs burnup. (Data taken from Ref. 5.)

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. 5. The results are shown in Fig. 7. The slope at 35 000 MWD/tU is 0.0038 Δf /1000 MWD/tU. This implies that a precision of 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 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 [See equation (5)]

$$N_j = N_R - N_P$$

$$\frac{S}{N} = \frac{N_j}{N_P} = \frac{\Delta k}{1 - k_1 + \Delta k} \quad (8)$$

where the dependence on Δk and k 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

$$\frac{S}{N} = \frac{N_1}{N_P} = \frac{S_0}{S} \quad (9)$$

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

Fig. 7. Estimated fractional count-rate change vs burnup.

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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Los Alamos National Laboratory
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Fig. 1. A generic spent-fuel active assay system.

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