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OF VARIOUS SPENT OR FRESH FUELS
BY ACTIVE NEUTRON INTERROGATION

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ABSTRACT

Recent studies show that subassemblies containing various spent fuels could be assayed rapidly and accurately by a nondestructive assay system using active neutron interrogation and prompt-neutron detection. Subassembly penetration is achieved by 24-keV (Sb-Be) interrogation neutrons; the spent-fuel neutron background is overridden by using strong interrogating sources and prompt-neutron signals, and background gammas are absorbed by lead. Experiments have demonstrated the potential for assaying, with better than 5% accuracy, three spent plutonium-fueled subassemblies per hour. Calculations, validated by experiments, predict even better performance for fresh or uranium-fueled subassemblies; several performance estimates are given.

INTRODUCTION

A nondestructive assay (NDA) system (Fig. 1) has been developed^{1,2} to assay the total fissile content of complete subassemblies of nuclear fuel rods. The system is required to assay a variety of fuel subassemblies, including those with high burnup and short cooling times. Both core and blanket subassemblies, containing mixed (Pu,U) oxide fuels clad in stainless steel, are included. These subassemblies accentuate two major design problems encountered in assaying various fuel subassemblies: penetration of interrogation and/or signature radiations in the massive subassemblies, and toleration of the high backgrounds of neutrons and gammas from spent-fuel subassemblies. Experiments² indicate that the system will be able to overcome these two problems and meet all other performance requirements, including assay to <5% error.

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Calculations, validated by measurements, indicate that the system is applicable to the assay of subassemblies containing a wide variety of fresh, spiked, or spent fuels--including light-water reactor (LWR) fuels and fuels of alternate fuel cycles.

SYSTEM DESIGN

The method selected is the active NDA method using an external source of interrogating neutrons to induce fissions in the fissile nuclides of the fuel. The neutrons from these induced fissions are detected to provide a measure of the fissile content of the fuel.

Good penetration of the massive subassemblies is achieved by interrogating with 24-keV neutrons from ^{124}Sb -Be isotopic sources. This energy gives good penetration and it is well below the threshold energy for fission of fertile nuclides. The signal count rate is made greater than the background count rate from the spent-fuel neutrons by using intense interrogating sources and by including in the detected signal the far-more-numerous prompt neutrons from induced fissions, rather than limiting the signal to delayed neutrons.

A design concept for the system is illustrated in Fig. 1. Some neutrons from the four ^{124}Sb -Be sources near the fuel interrogate the fuel in the central cavity, after passing through a 3-mm-thick ^{10}B filter which absorbs neutrons of degraded energy. Neutrons from the fissions so induced are detected by four methane-filled proportional counters. These four counters are staggered 45° with respect to the four sources and are farther out in the lead to shield them against the gamma radiation from the sources and from the spent fuel. Electronic pulse-height discrimination is used to reject the smaller proton-recoil pulses produced by the interrogating-source neutrons while accepting the larger pulses produced by the fission neutrons.

The detector-shielding lead is 1 m square and 0.5 m in axial thickness. Above and below it are slabs of borated (5 wt% B) polyethylene, 0.2 m thick, surrounding 12-mm-thick B_4C filters. These end regions serve to define the sensitive assay region and, especially, to reduce the sensitivity of the detectors to the background neutrons from spent fuel outside the assay region. The lead biological shielding serves no operational function in the assay and could be omitted in certain applications.

An axial scan of a subassembly is made by taking a neutron count as the subassembly passes axially through the system. This count is corrected for the fuel-background count which is obtained during a similar scan made without neutron interrogation (^{124}Sb sources removed.) The interrogation-source-background count must also be subtracted. The total-fissile content of an unknown subassembly is obtained by comparison of the net count thus obtained to that for a nearly-identical standard reference subassembly.

SYSTEM EVALUATION

An experimental model with the essential features of Fig. 1 was built and tested.² The fuel subassemblies comprised up to 61 fuel rods containing unirradiated UO_2 pellets of ~12 mm dia. The effective ^{235}U enrichment could be varied and various inhomogeneities could be created by intermixing fuel rods enriched to 1.5% and 7.1%. Six rods of 19.8% fuel were also available for probing spatial sensitivity variation. These test subassemblies simulated those from nuclear power reactors in regard to problems associated with neutron penetration but they lacked the background radiations of spent fuel and were limited to ^{235}U as the fissile nuclide. As shown in the following discussion, these limitations are of no importance to the relevance of the experiments.

Early tests were made using four ^{124}Sb sources of 200 Ci each and a test subassembly of 7.1%-enriched fuel. Neutron count rates and gamma dose rates were measured at three radial (from the system axis) positions of the four detectors. Results are plotted in Fig. 2. Although operation was satisfactory at the ~30 cm (12 in.) detector position, the detectors were near their 2 R/h limit--where gamma pileup interferes with proper operation. Moving the detectors to the ~40 cm (16 in.) position reduced the gamma dose rate by two decades while reducing the count rate by a factor of only 2.5. Our calculations indicate that the gamma dose rate from a spent plutonium-fueled Fast Flux Test Facility (FFTF) subassembly is comparable to that plotted for the 200 Ci sources--about 30% higher at 30 cm and 30% lower at 40 cm. For detector operation below the 2 R/h limit, the 40 cm detector position would be conservative, considering the gamma radiation from spent fuel, and would allow for ^{124}Sb source strengths of up to ~20,000 Ci each.

Our calculations show that four ^{124}Sb sources of 120 Ci each can produce in Be enough neutrons to induce a number of fission neutrons in the fuel equal to the number of background neutrons from a spent plutonium-fueled subassembly. In our tests with 7.1%-enriched fuel, the detectors operated well at the 40 cm position even when four sources of 600 Ci each were used. Under these conditions, there were far higher levels of both gamma dose rate and neutron count rate than would be contributed by spent fuel in the assay of plutonium-fueled subassemblies.

Neutron transport calculations were made for the system of Fig. 1, using a two-dimensional discrete ordinates code with P3-S8 approximation and 51 energy groups. Calculations were made corresponding to several configurations that had been measured experimentally and to some other subassembly assays of interest. Agreement between the measured results and those of the corresponding calculations was satisfactory, thus validating the calculational method. In addition to calculations for the experimental system, calculations were also made corresponding to the assay of a spent plutonium-fueled subassembly and of an unirradiated subassembly containing ^{235}U -enriched UO_2 . In all three cases, penetration of interrogating neutrons and spatial variation of assay sensitivity were satisfactory and similar to those measured in the experimental system.

Relative assay sensitivities for seven common heavy-metal nuclides were determined by isotopic activity calculations for the fuel-zone neutron spectra of these three calculations. The results, expressed as relative total fission-neutron production (relative to ^{239}Pu taken as 100), were essentially independent

of the subassembly fuel and were as follows: ^{232}Th , 0.1; ^{233}U , 170; ^{235}U , 110; ^{238}U , 0.7; ^{239}Pu , 100; ^{240}Pu , 7.7; ^{241}Pu , 140. Thus, the system has comparable sensitivities for the various fissile nuclides and shows good discrimination against fertile nuclides. These results indicate that an approximately 1.7-fold range of fissile-nuclide assay sensitivities is to be expected in assaying various fuels. They also can be used to determine the uncertainty in total-fissile assay results corresponding to a given uncertainty in relative isotopic composition. This calculated 1.7-fold range of fissile-nuclide sensitivities, based on total-neutron detection, is found to be only about half as large as that for a similar system based on delayed-neutron detection--because of the wide variation in delayed-neutron fraction among fissile nuclides. Thus, a system that includes the prompt neutrons in the detected signal tolerates larger uncertainties in knowledge of relative isotopic compositions than does one that detects only delayed neutrons.

Some other results of the experimental program, including those reported earlier², are as follows.

1. For a 61-rod subassembly of 7.1%-enriched fuel, a total signal count (four-detector sum) of 10^6 counts (standard deviation of 0.1%) requires 6 min for a source of 250 Ci ^{124}Sb . Allowing for fuel-background counting and for occasional counting of a reference standard a complete assay might require about 20 min.

2. The differential assay sensitivity (defined as added counts per added gram of ^{235}U) is about 15% higher at the edge than at the center of a subassembly.

3. Compared with that for a homogeneous arrangement (using 30 rods of 1.5% enrichment and 30 rods of 7.1%), the signal is about 3% higher when the 7.1% fuel occupies the outer half of the subassembly and 3% lower when it occupies the inner half.

4. With the 7.1 and 1.5% rods placed on opposite sides of a plane through the subassembly axis, the signal is about 0.5% higher than for the homogeneous arrangement.

5. The maximum achievable eccentricity (subassembly displaced 12 mm from system axis) has no measurable effect (within +0.2%). This statement is true even if the subassembly is not rotated during measurement, but it might not be true without rotation if source strengths and/or detector sensitivities are not azimuthally balanced.

6. The count rate varies linearly with enrichment for mixtures in the range from 0 to 7.1%.

7. The effect of fission products (simulated in our experiments by inserting twelve B_4C poison rods) on the net signal is moderate (~1%) and the effect will be compensated by including real or simulated fission products in the comparison standards used in an actual assay.

8. A measurement precision of ~0.1% (standard deviation for a sequence of 17 successive one-hour measurements) has demonstrated the stability of the instrumentation.

The accuracy attainable in an assay will depend on a number of factors, including especially the following: knowledge of the relative isotopic composition of the fuel and accuracy of the reference standard subassembly (as to absolute fissile content and as to how well it simulates the unknown subassembly in isotopic composition, nuclear poison or fission-product content, and spatial distribution of fissile material). Under the most favorable conditions (e.g., when assaying fresh fuel in a fabrication plant, using well characterized fuel and an essentially identical reference standard whose fissile content is known to 0.1% standard deviation) an assay accuracy of 0.2% (standard deviation) is a reasonable expectation. For less favorable conditions (e.g., when assaying spent fuel with ^{241}Pu known to be $8 \pm 2\%$ of total fissile Pu) an assay accuracy of 2% (standard deviation) is a reasonable expectation.

More study of these considerations is needed to define the accuracy to be expected for various conditions. Because of the importance of the relative isotopic composition of the sample to be assayed, we are investigating methods for the experimental determination of relative isotopic composition of the fuel in a subassembly.

APPLICATIONS

The experiments discussed above and calculational methods validated by comparison with the experiments provide the basis for estimating the characteristics of systems for assaying various types of fuel subassemblies. All such systems follow the basic design portrayed in Fig. 1, but the central cavity is enlarged for larger subassemblies such as those of LWRs.

Some performance characteristics estimated for four types of fuel subassemblies are given in Table 1. Source intensities were chosen (somewhat arbitrarily) to give signal/background ratios ≥ 2 and count rates $\geq 10^6$ counts/min with freshly activated ^{124}Sb sources (60-d half-life). The initial source strengths and the number of detectors can be adjusted to suit the individual requirements and preferences as to signal count rate, signal/background ratio, and time interval for source renewal.

Because of the lower neutron backgrounds from fresh fuels (especially for ^{233}U fuel), the required ^{124}Sb source strengths are lower and the signal/background ratios higher than for spent fuels.

SUMMARY AND CONCLUSIONS

An active NDA system, based on interrogation by ^{124}Sb -Be neutrons and detection of prompt as well as delayed neutrons from the induced fissions, has been developed to determine the total fissile content of spent fuel subassemblies. Experiments² have shown that the system can meet the performance objectives, including assay with $< 5\%$ error. Although tests were made with unirradiated ^{235}U -enriched UO_2 fuel rather than with spent mixed-oxide fuel, calculations have shown that ^{235}U and Pu have comparable assay sensitivities

and that the gamma dose rates and neutron count rates in some of the tests greatly exceeded those that would arise from spent fuel.

A calculational method has been developed and validated by comparison of calculated with experimental results. Calculations have then been used to obtain the relative assay sensitivities of the system for various fissile and fertile nuclides. The assay sensitivities of fertile nuclides were acceptably low and those of fissile nuclides were similar to each other--varying from a low for ^{239}Pu to a high for ^{235}U (1.7 times that of ^{239}Pu). These relative sensitivities are useful for estimating the effect on assay accuracy of uncertainties in the relative isotopic composition of the fuel.

The calculations also have been used to extend the experimental results to estimate the characteristics of such an NDA system for the assay of various types of fuel subassemblies. Four examples are presented in Table 1. The ^{124}Sb source requirements are lower and the signal/background ratios higher for fresh fuels than for spent fuels. Because of its much lower neutron background, ^{235}U -based fuel gives a much higher signal/background ratio than does Pu-based fuel.

Methods for the measurement of isotopic composition are now being explored. Plans for the near future include a demonstration of the system for the assay of spent fuel and a better determination of the limits of precision and accuracy attainable with the system.

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2. G. L. Ragan, C. W. Ricker, M. M. Chiles, and G. C. Guerrant, "Experimental Evaluation of a System for Assay of Spent-Fuel Subassemblies," Trans. Am. Nucl. Soc. 28: 128 (1978).

TABLE 1. ESTIMATED PERFORMANCE FOR THIS
NDA SYSTEM FOR FOUR TYPES OF FUEL SUBASSEMBLY

	<u>FFTF SUBASSEMBLIES</u>			SPENT
	SPENT	FRESH	FRESH	LWR
	<u>Pu</u>	<u>Pu</u>	<u>U²³³</u>	<u>SUBASSEMBLY</u>
I. FUEL NEUTRON BACKGROUND EFFECTS				
Assumed Background, neutrons/s per meter length	1.0E+08	2.4E+06	5.4E+04	8.0E+07
Background Count Rate, ^a counts/min	8.4E+05	5.0E+04	1.1E+03	5.6E+05
II. PERFORMANCE WITH FRESH SOURCES				
Total Sb-124 (four sources), Ci	2000	200	200	2000
Signal Count Rate, ^a counts/min	3.5E+06	9.0E+05	1.1E+06	1.3E+06
Signal/background count rate ratio	4.2	18	1000	2.3

^aFor 12 detectors at R = 30 cm (40 cm for spent fuels).
Proportional counters: 2.5 atm methane; 5 cm
diameter by 25 cm sensitive length; 450 keV pulse
threshold.

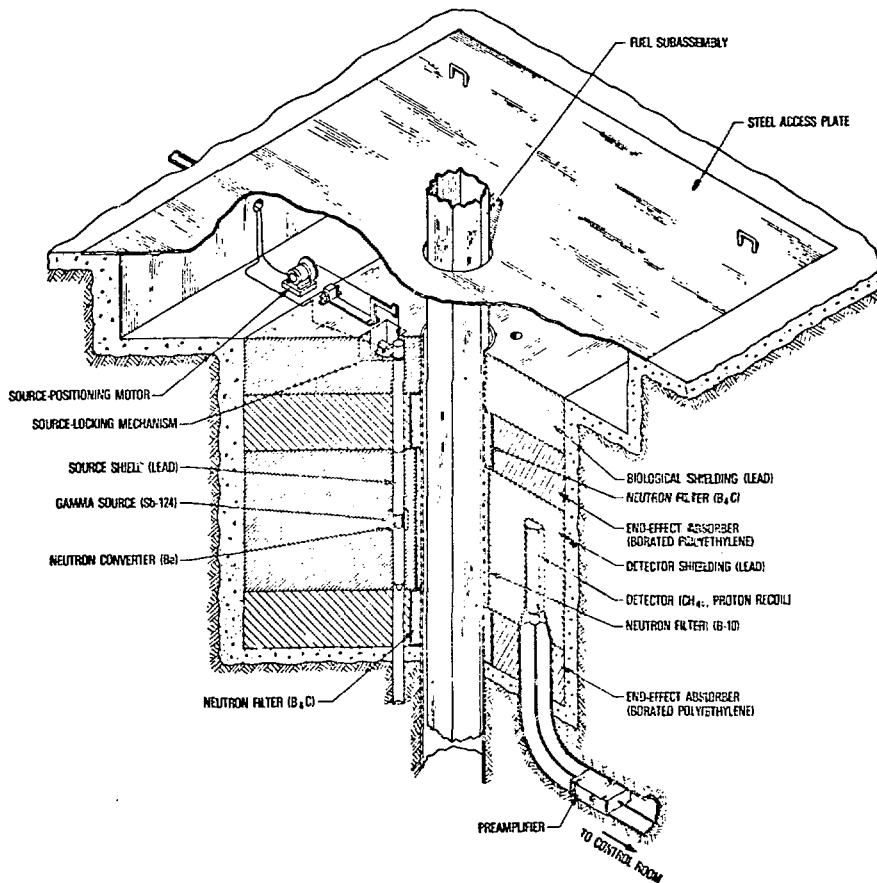


Fig. 1. Experimental system modified for in-the-floor installation. Source and detector locations (four each) alternate every 45°.

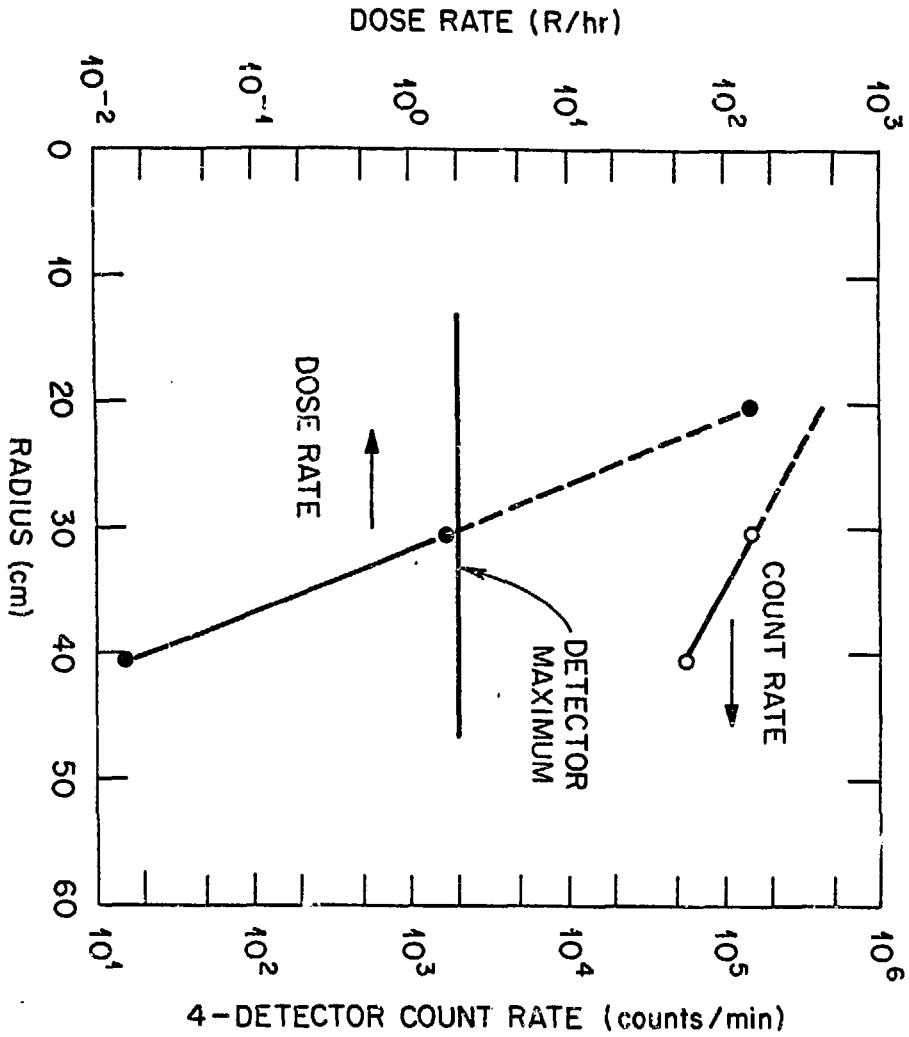


Fig. 2. Variation of gamma dose rate and count rate with radius for 7.1% fuel subassembly and 200 Ci sources.