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ABSTRACT

A nondestructive assay system was developed for determining the total fissile content of spent fuel subassemblies at the head end of a reprocessing plant. The system can perform an assay in 20 min with an uncertainty of <5%. Antimony-beryllium neutrons interrogate the subassemblies, and proton recoil counters detect the resulting fission neutrons. Pulse-height discrimination differentiates between the low-energy interrogation neutrons and the higher-energy fission neutrons.

To optimize the performance of the system, 51-energy-group neutron-transport calculations were made, first with a one-dimensional (1-D) computer model, followed by a 2-D computer model. The performance of the as-built system was calculated using a 2-D model. The cross-sections were from the ENDF/B-IV data file and were processed for use in liquid-metal-cooled fast breeder reactor (LMFBR) calculations.

Calculated and measured results were compared for (1) interrogation-neutron penetrability, (2) fission-neutron detectability, (3) radial variation of assay sensitivity, (4) axial variation of assay sensitivity, and (5) the variation of detector count rate as a function of the number of fuel rods in a special 61-rod, LMFBR-type subassembly.

The calculational procedures were validated by comparison with experimental measurements, thus permitting further exploration of system performance by means of additional calculations. In this manner, the following system characteristics were investigated: the relative assay sensitivities of various fissile nuclides; the applicability of the system to a wide variety of fuels, either fresh or spent; the effect on assay accuracy of uncertainties in the isotopic abundances in the fuel; and the efficacy of possible methods of determining isotopic abundances in the sample.

Keywords: Nondestructive assay; neutron interrogation; spent nuclear fuel; nuclear fuel subassembly; calculations; measurements; nuclear safeguards; material accountability; process control; criticality control; assay accuracy.

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INTRODUCTION

A system was developed (Fig. 1) to perform a nondestructive assay (NDA) of the fissile content, either Pu or U, of a spent fuel subassembly as it enters a fuel reprocessing plant and still retains its identity. The assay results can be applied to materials accountability, process control, criticality control, and safeguards. The design objectives of the system are to assay the total fissile content of a subassembly of spent fuel in 20 min with an uncertainty of <5%.[1]

To achieve uniformity of assay sensitivity over the large cross-sectional area (~120 cm^2) of a subassembly, the subassembly is actively interrogated by 23-keV neutrons from an ^{124}Sb-Be source. A neutron filter (3-mm-thick boron enriched to 92% ^{10}B) surrounding the subassembly attenuates those source neutrons that would not penetrate far into the subassembly because they have lost energy through moderation. To obtain a signal larger than that from the intense neutron background from spent fuel (~10^8 neutrons/s), (a) the total number of fission neutrons is measured instead of only the far fewer delayed neutrons, and (b) four intense interrogating sources are required (each with ~200 Ci ^{124}Sb, producing ~8×10^8 neutrons/s). The high-energy fission neutrons are differentiated from the lower-energy interrogating neutrons by pulse-height discrimination applied to the detectors (methane-filled proton-recoil counters). The counters are shielded against the gamma radiation from the ^{124}Sb sources and the spent fuel by >20-cm-thick lead shields.

The assay system parameters were optimized on the basis of 51-energy-group neutron-transport calculations (P3-S8 discrete ordinates), first with a one-dimensional (1-D) model (ANISN code[2]), followed by a 2-D model (DOT code[3]). Finally, calculations were made, using the 2-D model of the as-built system, to obtain values for comparison with those measured in various experimental situations. Cross sections were from the ENDF/ B-IV data file and were processed for use in LMFBR calculations.[4]

COMPARISON OF CALCULATIONS AND MEASUREMENTS

Comparative calculated and measured results for the variation of assay sensitivity with radius are given in Table I. The test fuel was unirradiated UO_2, enriched to 7.1% in ^{235}U. The detectors were biased to record only those neutrons that imparted an energy of 450-650 keV to the recoil protons. All values are for the indicated ring relative to the first (innermost) ring of six fuel rods surrounding the central rod of a 61-rod subassembly. The four rings contained 6, 12, 18, and 24 rods. For part A (for the plane perpendicular to the axis at its center), *sensitivity* is taken as the product of two components: the interrogating neutron *penetrability* times the *detectability* of the induced fission neutrons. Calculated and measured penetrabilities are defined, respectively, as the calculated local fission density and as the local fission-product gamma activity measured by a NaI detector following several days of activation by the interrogating sources. Calculated and measured detectabilities are defined, respectively, as the calculated local adjoint flux (which gives the local contribution to the detector signal in a detector-based adjoint calculation) and as the measured detector signal from a ^{252}Cf neutron source placed at the given locality in the subassembly.

For part B (for the entire length), sensitivity is defined as the increase in count rate when six normal (7.1% enriched) fuel rods in a given ring were replaced by six higher-enrichment (19.8%) rods. Part B thus involved an axial average over the assay region. The calculations simulated the experimental conditions: each ring of rods constituted a zone of the appropriate composition, with corresponding detector activities determined in forward calculations--no adjoint calculations were needed.

The calculated and measured axial variation of assay sensitivity can be compared by reference to Fig. 2. The penetrability, detectability, and sensitivity for both calculated and measured cases are defined as Part A of Table I. The axial variations of the components (penetrability and detectability) of the sensitivity also showed comparable agreement between calculations and measurements.

The shape of the axial sensitivity curve is not important, because the assay procedure will be to program the scan so that each axial element of the subassembly traverses the entire sensitive region of the assay equipment. These scan results will be compared with those from a nearly identical standard subassembly to determine the total fissile content of a subassembly by integrating the axial scan data.

The variation of calculated and measured count rate with the number of 7.1%-enriched fuel rods can be compared by examing Fig. 3. Incidentally, a slight nonlinearity of both measured and calculated points is evident. Less nonlinearity is observed when enrichment (at constant number of rods) is varied and plotted as abscissa.

The foregoing results present quantitative comparisons of the calculated and measured spatial variation of assay sensitivity, in both radial and axial directions. These results are for ^{235}U-enriched UO_2 fuel, using cross sections from the ENDF/B-IV file processed into a 51-group set for LMFBR calculations.[4] The quantitative aspects of the observed agreement between calculations and measurements indicates the degree to which the 2-D geometric model and the cross-section set used may be considered to be adequate for an expanded calculational program.

This validation of the calculational procedure permits the extension of measured results by additional calculations. Among the characteristics that now can be explored are: the relative sensitivities of various fissile nuclides; the applicability of the NDA system to a variety of fuels, either fresh or spent;[5] the effect on assay accuracy of uncertainties in the isotopic abundances in the fuel; and the efficacy of possible methods of determining isotopic abundances in the sample. The calculations related to these issues are discussed in the next section.

CALCULATIONAL RESULTS

A 2-D calculation was made for a spent LMFBR core subassembly in the normal assay configuration. The isotopic sensitivities (fission neutrons produced per atom of the

given nuclide) relative to ^{239}Pu are given in the second column of Table II. Although ^{233}U is not normally present in this fuel, a trace amount was included to obtain its relative sensitivity. The sensitivity range (1.73) is far better than it would be (∿3.4) if only delayed neutrons had been counted in this same system. We believe that the sensitivity range of 1.73 also is better than that for other systems designed specifically to detect only delayed neutrons.

The 1.73 range of relative isotopic sensitivities means that the results of these experiments, based on ^{235}U-enriched fuel, are qualitatively correct (and not far off quantitatively) for other fuels based on different fissile isotopes. Performance estimates have been given elsewhere for the assay, in this NDA system, of four other fuel subassemblies.[5] The four fuels include three LMFBR fuels (containing spent Pu, fresh Pu, or fresh ^{233}U) and one light-water-cooled reactor (LWR) fuel (requiring an enlarged center cavity in the NDA system).

In the third column of Table II, relative sensitivities are given for our NDA system with the ^{10}B neutron filter removed. Because of the presence of a greater number of low-energy neutrons, the relative sensitivities changed as shown. Not shown, but indicative of the value of the filter, is that the edge-to-center neutron production density increased from the normal-system value of ∿1.15 to ∿1.30 when the ^{10}B filter was removed.

The last column of Table II shows the factor by which the observed signal would increase, for each isotope, if the ^{10}B filter were removed. A measurement of this ratio during an assay would provide information relative to isotopic composition. Different filters (e.g., filters containing appropriate moderating and resonance-absorbing materials) might give better isotopic discrimination, and by making four different measurements one might determine the relative abundances of the four fissile isotopes. More refined calculations with cross sections processed to account for various isotopic dilutions are being made to explore these possibilities.

The estimated accuracy of assay achievable in a 20-min measurement with this system is given in Table III. With careful control of the individual contributing parameters (favorable conditions) a standard deviation of ~0.2% should be realizable. The most important unfavorable condition would be poor knowledge of the relative isotopic composition. The indicated 3% corresponds to the following improbable scenario: the transfer documents state that the fissile content of a given spent LMFBR subassembly is 93% ^{239}Pu and 7% ^{241}Pu, and we use the corresponding assay sensitivity factors. If, instead, the subassembly contains either 0% or 14% ^{241}Pu, the assay error would be +3% or -3%, respectively. If the ^{241}Pu content is known more accurately than this, as it most probably will be, this component of the assay error will be correspondingly less.

CONCLUSIONS

Experiments have shown that the NDA system can meet the design objectives. The calculational model and cross-section set were validated by comparison with experiments. Calculations have been used to extend the experimental results to determine the following: relative sensitivities of various fissile nuclides; the applicability of the system to a variety of fuels; the effect on assay accuracy of uncertainties in the isotopic abundances in the fuel; and the efficacy of possible methods of determining isotopic abundances in the sample.

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Table I. Radial variation of assay sensitivity and of its components (fuel was 7.1% enriched and 450- to 650-keV pulses were detected)

	Values Relative to Ring 1[a]		
	Ring 2	Ring 3	Ring 4
A. In Axial-Center Plane			
Penetrability[b] (P_o)			
Measured	1.005	1.042	1.064
Calculated	1.013	1.033	1.054
Detectability[c] (D_o)			
Measured	1.009	1.027	1.042
Calculated	1.009	1.022	1.038
Sensitivity ($S_o = P_o \times D_o$)			
Measured	1.014	1.070	1.108
Calculated	1.022	1.055	1.094
B. Over Entire Length			
Sensitivity[d] (S)			
Measured	1.031	1.077	1.153
Calculated	1.032	1.074	1.127

[a] In the 61-rod subassembly, one center rod was surrounded by four hexagonal rings of rods.

[b] The fission-neutron density produced in a given ring by the interrogating-neutron source. Standard deviation of the measurements is ∿0.5%.

[c] The detector signal produced by a ^{252}Cf source placed in a given ring. Standard deviation of the measurements is ∿0.4%.

[d] The increase in the count rate when the enrichment was raised from 7.1 to 19.8% in six rods in a given ring. Standard deviation of the measurements is ∿2.3%

Table II. Calculated isotopic sensitivities and effect of ^{10}B filter

Nuclide	Sensitivities Relative To Pu-239		Signal W/O Filter / Signal W/Filter
	With B-10 Filter	Without B-10 Filter	
U-233	1.73	1.51	3.62
U-235	1.12	0.89	3.31
Pu-239	1.00	1.00	4.16
Pu-241	1.45	1.26	3.62

8644

Table III. Accuracy Estimates

	Standard Deviation(%)	
	Favorable Conditions	Unfavorable Conditions
Count Statistics	0.1	0.2
Accuracy of Standard	0.1	0.2
Isotopic Composition	0.1	3.0
Detector Temperature	0.1	0.2
Detector High Voltage	0.1	0.2
Combined	0.2	3.0

8645

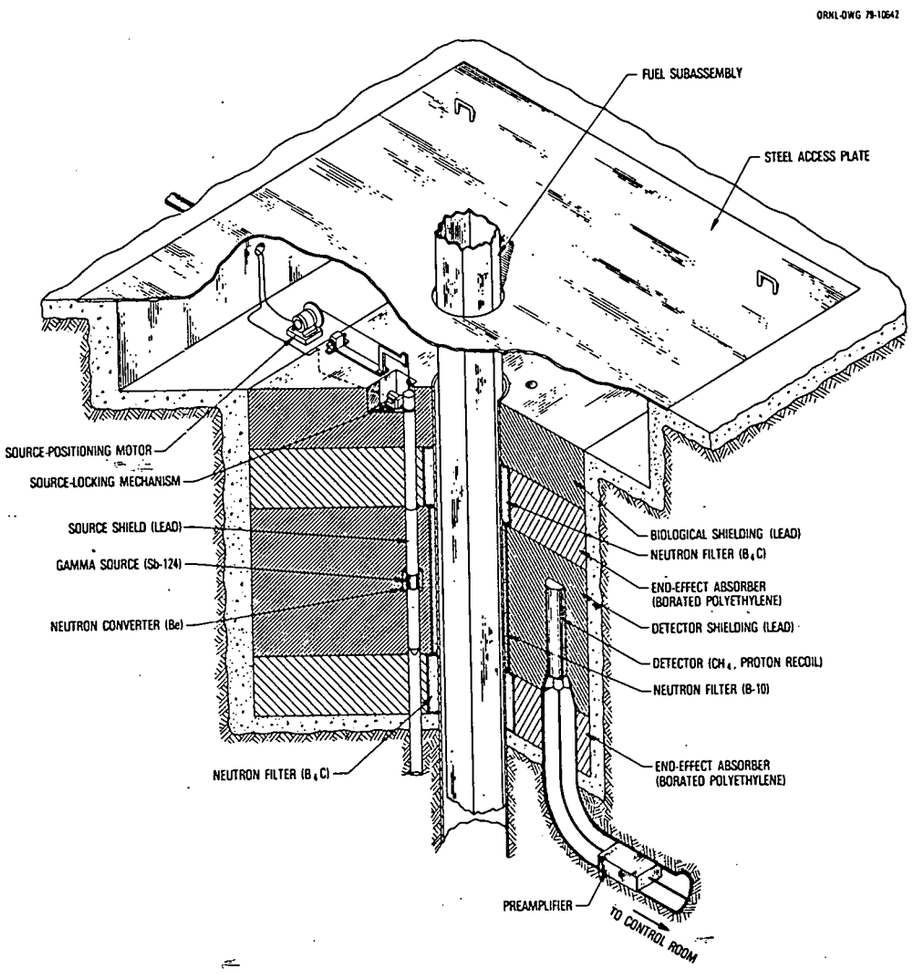

Fig. 1. Experimental system modified for in-the-floor installation. Source and detector locations (four each) alternate every 45°.

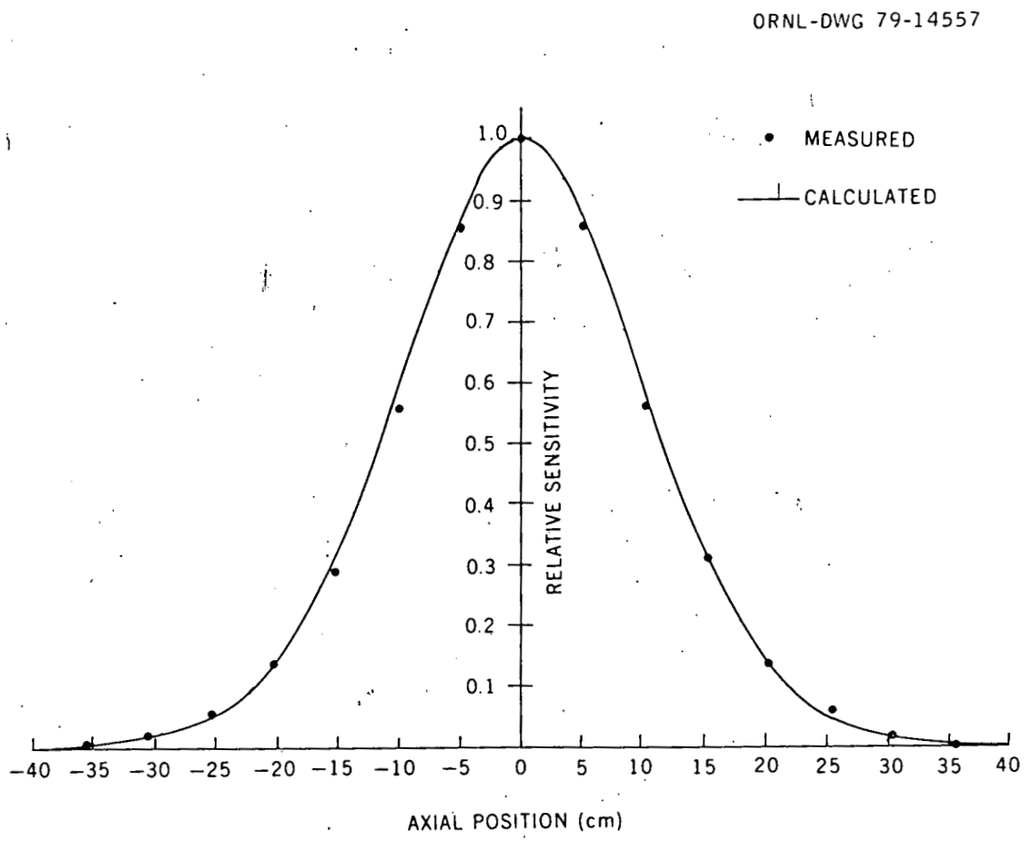

Fig. 2. Comparison of calculated and measured axial sensitivities for 7.1% enriched fuel and 450- to 650-keV pulse detection. Calculated and measured values are normalized to 1.0 at axial position 0.

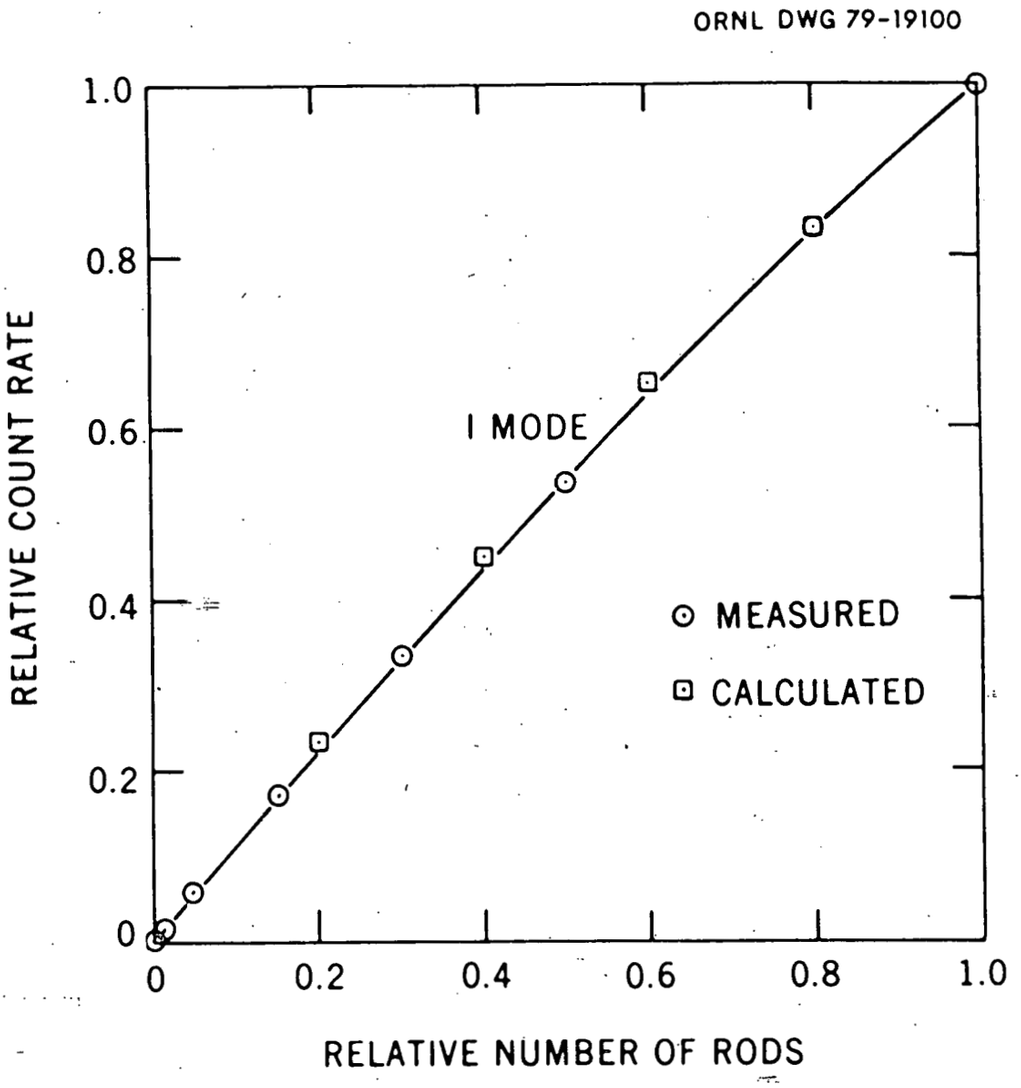

Fig. 3. Variation of count rate with relative number of 7.1%-enriched fuel rods, counting all pulses above 450 keV (I-mode). Calculated and measured count rates are normalized to 1.0 at an abscissa of 1.0.