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NONDESTRUCTIVE ASSAY OF SPENT BOILING WATER REACTOR FUEL BY ACTIVE NEUTRON INTERROGATION

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ABSTRACT

Spent boiling water reactor (BWR) fuel from Dresden I was assayed for total fissile mass, using the active neutron interrogation method. The nondestructive assay (NDA) system used has four Sb-Be sources for interrogation of the fuels; the induced fission neutrons from the fuel are counted by four lead-shielded methane-filled proportional counters biased above the energy of the source neutrons.

Spent fuel rods containing 9 kg of heavy metal (~1% fissile) were chopped into 5-cm segments and loaded into three 1-liter cans (~3 kg per can). (Because of mechanical constraints in handling the fuel in the reprocessing test facility, the fuel could not be assayed as entire fuel rods.) The three cans were assayed in seven combinations of one, two, or three cans, enabling an evaluation of the precision and accuracy of the NDA system for different amounts of fissile material. The fissile mass in each combination was determined by comparing the induced-fission-neutron counts with the counts obtained from a known standard comprising chopped segments of unirradiated Dresden fuel. These masses were compared to the masses determined by chemical analyses of the spent fuel.

The results from the nondestructive assays agreed with results from the chemical analyses to within 2-3%. Similar agreement was obtained when two combinations of canned spent fuel were used as standards for the nondestructive assays.

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The assay of BWR spent fuel served as a test of the NDA system which was developed at the Oak Ridge National Laboratory for the assay of spent liquid metal fast breeder reactor (LMFBR) fuel subassemblies at the head-end of a reprocessing plant. Results of previous experiments and calculations reported earlier[1-3] using simulated LMFBR fuel subassemblies indicated that the NDA system can measure the fissile masses of spent fuel subassemblies to within an accuracy of 3%. Results of the assays of spent BWR fuel reported herein support this conclusion.

1. INTRODUCTION

Spent nuclear fuel from a boiling water reactor (BWR) was assayed for total fissile mass using an active neutron interrogation assay system[1-4] developed at the Oak Ridge National Laboratory (ORNL) for the Consolidated Fuel Reprocessing Program (CFRP). The nondestructive assay (NDA) system was developed for the measurement of the total fissile masses in liquid metal fast breeder reactor (LMFBR) fuel and blanket subassemblies at the head-end of a reprocessing plant.

In previous tests of the NDA system, simulated, unirradiated fuel subassemblies containing known amounts of ^{235}U as the only fissile constituent were assayed to determine the achievable count rate, spatial effects due to location of the fuel within the assay cavity, linearity of count rate as a function of enrichment, effects of simulated fission products, and instrument stability.[1-4] As a result, assay accuracies of 2-3% were predicted for the assay of spent LMFBR subassemblies.

The series of measurements reported herein, however, tested the NDA system using actual spent

fuel with unknown amounts of uranium and plutonium fissile isotopes (LMFBR fuel was not available but the use of spent BWR fuel, as discussed in Section 3, provided a stringent test of the system).

Following the nondestructive assays, the spent BWR fuel was dissolved and assayed using chemical techniques to obtain a reference against which to determine the assay accuracy of the NDA system. These measurements were important for two reasons. First, the performance of the NDA system was examined using spent fuel (for which it is primarily intended) rather than unirradiated fuel. Second, because the assays were made of unknown spent fuel and then compared to accurate chemical analyses, the accuracy of the NDA system was determined directly, rather than inferred from other measurements.

2. DESCRIPTION OF NDA SYSTEM

The NDA system used in this investigation has been under development for several years at ORNL and has been reported by Ragan et al.[1-4] The system, similar in design to that of Menlove et al.,[5] though extensively modified, is basically a lead and borated polyethylene structure with a central cavity and channels for sources and detectors (Fig. 1). As a fuel subassembly is passed through the cavity, it is interrogated by 3.7-fJ (23-keV) neutrons produced by four ^{124}Sb-Be sources. A spatially uniform interrogation is ensured by a 3-mm-thick, ^{10}B sleeve which surrounds the cavity and absorbs source neutrons degraded in energy before reaching the assay cavity.

Four methane-filled proton recoil detectors, symmetrically placed 41 cm from the center of the cavity, detect the induced prompt- and delayed-fission neutrons. The detectors are shielded by 30 cm of lead from the gamma backgrounds emitted by the ^{124}Sb sources and the spent fuel. Also, the electronics associated with the detectors are biased to exclude low amplitude pulses [<72 fJ (450 keV)] so that only fission neutrons are counted. This bias also discriminates against pulses resulting from gamma pile-up in the detectors.

Background neutron effects from spontaneous neutron production and (α,n) reactions in spent fuel can be minimized by using intense interrogation sources [up to $\sim 10^{10}$ neutrons/s for LMFBR fuel, requiring ~89 TBq (~2400 Ci) of ^{124}Sb], which ensures production rates of induced fission neutrons equal to or greater than the rates of background neutron emission. However, for low burnup or unirradiated fuels, background radiations are lower, and consequently, less intense ^{124}Sb sources can be used. For the assay of BWR spent fuel described in this report, the intensity of the four ^{124}Sb sources was ~43 TBq (~1160 Ci).

3. DESCRIPTION OF SPENT FUEL

The spent fuel was Dresden I BWR fuel that had an initial ^{235}U enrichment of 2.342% and an average burnup of 2.26 TWs/kg (26,160 MWd/tonne). The fissile content after discharge from the reactor was 1.1%, based on Dresden calculations. The cooling time of the fuel prior to assay was about 4.5 y.

The Dresden fuel was chosen solely because spent fuel rods had been received at ORNL for chemical analysis and were available for this use. Spent LMFBR fuel, had it been available, would have been preferred, since it is the fuel for which the system is primarily designed. However, even though the gamma and neutron backgrounds of the 4.5-y-cooled BWR fuel were lower, the fuel provided as stringent a test of the assay system as would have been provided by 60-d-cooled LMFBR fuel, as explained in the following discussion.

Table 1 shows the estimated fissile isotopic percentage of the Dresden fuel in comparison with typical pressurized water reactor (PWR) and LMFBR fuels. Although Table 1 shows that the gamma emission rate from the 4.5-y-cooled Dresden fuel is between one and two orders of magnitude less than that of the 60-d-cooled fuels, this was not considered a factor that would affect the test of the assay system. This is the case because the gamma background at the detector locations is dominated by the gamma radiation from the ^{124}Sb sources, regardless of the type of fuel being assayed, and the neutron detectors, though sensitive to gamma pile-up effects, are heavily shielded by lead.

Although the neutron emission rate of the 60-d-cooled LMFBR fuel is nearly an order of magnitude greater than that of the 4.5-y-cooled Dresden fuel, the neutron emission rate per unit fissile mass from Table 1 is greater for the Dresden fuel even after 4.5 y of cooling than for the LMFBR fuel after 60 d of cooling. The reason is that the Dresden fuel had received sufficient exposure to produce a proportionately high amount of neutron-emitting curium isotopes in comparison to the amount of fissile material present. Thus, for active neutron NDA measurements, the ratio of the neutron background count rate to the induced count rate from the fissile isotopes is higher for the Dresden fuel than for the LMFBR fuel.

From the preceding arguments it is seen that, because of the relative unimportance of the gamma emission rate of the fuel being assayed and the relatively high ratio of the neutron background to the induced count rates, the Dresden fuel presents as great a challenge to the NDA system as the 60-d-cooled LMFBR fuel.

4. DESCRIPTION OF EXPERIMENT

Because of mechanical constraints in handling the fuel in the reprocessing test facility, the fuel could not be assayed as entire fuel rods. Instead, rods comprising ~9 kg of heavy metal (~1% fissile) were chopped into 5-cm (2-in.) segments and loaded into three 1-liter (10.8 cm diameter, 12.3 cm length) cans (3 kg per can). When loaded in this manner, however,

Table 1. Comparison of Various Spent Fuels

	LMFBR (60 d)	PWR (60 d)	Dresden (60 d)	Dresden (4.5 y)
Initial Enrichment (%)[a]	15	3.3	2.34	2.34
Final Composition (%)[a]				
^{235}U	0.08	0.82	0.58	0.58
^{239}Pu	12.3	0.54	0.41	0.41
^{241}Pu	1.11	0.11	0.12	0.09
Total Fissile (%)	13.5	1.5	1.11	1.08
Burnup (TWs/kg) (MWd/tonne)	8.61 (99,700)	2.85 (33,000)	2.26 (26,200)	2.26 (26,200)
Gamma Background MeV·s^{-1}·kg^{-1}	2.80 E+14	1.3 E+14	∿7.0 E+13	∿3.2 E+12
Neutron Background neutrons·s^{-1}·kg^{-1}, heavy metal	2.19 E+6	6.0 E+5	3.4 E+5	2.6 E+5
neutrons·s^{-1}·kg^{-1}, fissile	1.6 E+7	4.0 E+7	3.1 E+7	2.4 E+7

[a]Weight % relative to the total heavy metal.

the total mass per unit length of one or more cans of spent fuel approximated that of an LMFBR fuel subassembly. Also, about 9 kg of unirradiated Dresden fuel (1.5% ^{235}U enriched) was chopped into segments and similarly loaded into three cans for use as NDA standards (the accuracy of the fissile mass of the unirradiated fuel standard was estimated from fabrication data to be 0.5%).

The approach in this investigation was to assay various combinations of the three cans of spent fuel in the NDA system and to compare these results with those obtained after dissolution by chemical analysis of the fuel. The three cans of spent fuel, designated A, B, and C, were assayed in the following seven combinations of one or more cans: A, B, C, AB, BC, CA, and ABC. Assaying the fuel in these combinations afforded the opportunity to evaluate the accuracy of the NDA system for wide variations in the amount of fissile material (because the cans differed slightly in total fissile mass and isotopic fractions, each of the seven combinations of cans contained a different quantity of fissile material); in particular, neutron multiplication effects could be determined.

Nondestructive assays were accomplished by two methods. By the first method, the fissile masses of combinations of spent fuel cans were determined by comparing the nondestructive assay counts of the spent fuel to those obtained from similar counts from corresponding combinations of cans of known amounts of the unirradiated Dresden fuel. The second method used was similar, except that one can (or two cans where appropriate) of the spent fuel instead of the unirradiated fuel was used as a standard. The second method was possible only after the fissile mass in the can (or cans) adopted as a standard had been determined by chemical analyses.

To assay a combination of one or more cans of fuel, the cans were loaded into an aluminum canister 66 cm (26 in.) long. This canister was inserted into a slightly larger steel container which was placed in the cavity of the assay system. The measurement of the combination consisted of a series of scans in which the cans were translated from below the sensitive volume to above the sensitive volume of the NDA system in 17 discrete vertical positions, 6 cm apart. For each position a 200-s count was taken, and the counts were summed for each scan. Scans were taken for all combinations of sample and sources in place and removed from the system to get the proper background corrections for the gross counting rate. The total counting time was about 1 h per scan, which was required to ensure that statistical errors due to counting were in the 1% range.

Assays were performed for both spent and unirradiated fuel combinations. Following the assays of the seven combinations of the cans of spent fuel, the three cans of spent fuel were chemically analyzed to determine the U and Pu masses in each can. Isotope dilution mass spectrometry was used to determine the fuel isotopics (weight fractions of fissile isotopes).

The fissile mass M_F of an unknown spent fuel combination was determined by the following expression:

$$M_F = \frac{C_{TF}}{C_{TS}} \cdot \frac{M_S}{f} \quad , \qquad (1)$$

where C_{TF} and C_{TS} are the total induced-fission neutron counts for the spent and standard fuels, M_S is the fissile mass of the standard, and f is a correction factor (close to unity) to account for estimated differences between the standard and the assay sample. The correction factor f takes into account differences in the fissile isotopic fractions, approximate enrichments, and total heavy metal masses. Values of f were determined by simulations of the experimental system with the two-dimensional, discrete-ordinates code DOT[6] using information that was independent of the chemical and nondestructive assays. For each assay, in order to determine the correction factor it was necessary to assume a priori an approximation of the spent fuel isotopics, since the assay system exhibits different sensitivities for different fissile isotopes.[1-4] Calculations were made using a correction factor f based, first, on the isotopics calculated from data supplied by Dresden personnel, and, second, on the isotopics of the spent fuel determined by ORNL chemical analyses. The latter method violates our premise that only information independent of the chemical and nondestructive assays should be used to determine the correction factor f, but simulates the situation in which the isotopics of the fuel are measured by an independent NDA method (several such methods are under study at ORNL).

Table 2 shows a comparison of the isotopics as determined from the Dresden data and the ORNL analyses for the three cans of spent fuel. The cans were loaded with chopped fuel from different parts of the fuel rods and the isotopic fractions in the cans show the variation of burnup. Although the isotopic fractions in the cans differ by as much as 50% from the Dresden values, the correspondence between the average values for the three cans and the Dresden values is reasonably good (1% for ^{235}U, 6% for ^{239}Pu, and 25% for ^{241}Pu). The mass of the ^{241}Pu isotope represents about 7% of the total fissile mass.

5. EXPERIMENTAL RESULTS

5.1 NDA of Spent Fuel Using Unirradiated Fuel Standards

Table 3 shows the results of the NDA of the seven combinations of spent fuel cans using the unirradiated fuel standards. For each fuel combination, the ratio of the fissile mass determined by NDA measurement to that determined by chemical analysis is given. Two columns of results are shown; for one column the correction factor f is based on the Dresden spent fuel isotopics (f = 1.023), and for the other column, chemical assay isotopics are assumed. A mean value of the seven ratios (mean ratio) for each of the two sets of data is given along with the standard deviation (σ) of the distribution.

Table 2. Comparison of Isotopic Data for Dresden Fuel

Fissile Isotope	Fraction of Total Fissile Mass: Calculated from Dresden Data[a] (all cans)	Analytical Chemistry Determinations: Average	Can A	Can B	Can C
^{235}U	0.536	0.529	0.575	0.514	0.497
^{239}Pu	0.378	0.402	0.368	0.413	0.426
^{241}Pu	0.086	0.069	0.057	0.074	0.076

[a]Corrected for 4.5-y cooling time.

For Dresden-supplied isotopic data, the mean ratio is 0.989. For isotopic information determined from chemical analyses, the mean ratio is 1.002. In both cases, the standard deviation of the distribution is ±2.2%. These results indicate that higher accuracy is obtained when the chemically determined isotopics rather than the Dresden isotopics are used, because the mean ratio is closer to unity. This outcome was anticipated, since isotopic information supplied by a utility and based on calculational predictions cannot be expected to be as accurate as that determined by direct chemical analyses. However, this indication is inconclusive, since only seven measurements were performed, and because the width of the distribution in each case (σ = ±2.2%) is large compared to the possible bias introduced by using the Dresden isotopics. Although it is not shown conclusively in this experiment, current work at ORNL indicates that valid isotopic data obtained by direct measurements probably will be required to achieve total fissile assay accuracies within 1%.

5.2 NDA of Spent Fuel Using Spent Fuel Standards

The total fissile masses of the spent fuel cans were also determined using spent fuel can combinations B and BC as NDA standards for the one- and two-can combinations, respectively. Results of chemical analyses were used to calibrate standards B and BC. Spent fuel assay results were again based on the two different isotopic determinations. For the first determination in which the Dresden-supplied isotopics were used, the standard and the assayed fuel were assumed identical in total mass, fissile mass, and isotopic content; thus no correction factor was required (f = 1). For the second determination in which the chemical analyses isotopics were used, a correction factor f was determined for each combination of fuel cans assayed, based on slight variations of the isotopics of each can of spent fuel. The results given in Table 4, in a format similar to that of

Table 3. Comparison of NDA Measurements Using Unirradiated Fuel Standards and Dresden and Chemical Isotopics with Chemical Analyses

Spent Fuel Combination	Fissile Mass (kg) (Chemical Analysis)	Fissile Mass (NDA)/ Fissile Mass (Chemical Analysis)	
		Dresden Isotopics (f = 1.023)	Chemical Isotopics (f for each combination)
(1) A	0.0333	0.987	0.997 (f = 1.013)
(2) B	0.0299	1.016	1.031 (f = 1.008)
(3) C	0.0289	0.980	0.996 (f = 1.006)
(4) AB	0.0633	0.991	1.003 (f = 1.011)
(5) BC	0.0589	0.981	0.997 (f = 1.007)
(6) CA	0.0623	1.010	1.023 (f = 1.010)
(7) ABC	0.0922	0.943	0.956 (f = 1.009)
Mean Ratio		0.989	1.002
Standard Deviation of Distribution (σ)		±2.2%	±2.2%

Table 4. Comparison of NDA Measurements Using Spent Fuel Standards and Dresden Isotopics and Chemical Isotopics with Chemical Analyses

Spent Fuel Combination	Fissile Mass (kg) (Chemical Analysis)	Fissile Mass (NDA)/ Fissile Mass (Chemical Analysis)	
		Dresden Isotopics (f = 1)	Chemical Isotopics (f for each combination)
(1) A	0.0333	0.971	0.966 (f = 1.005)
(2) B[a]	0.0299	---	---
(3) C	0.0289	0.964	0.966 (f = 1.008)
(4) AB	0.0633	1.011	1.007 (f = 1.004)
(5) BC[a]	0.0589	---	---
(6) CA	0.0623	1.032	1.029 (f = 1.003)
(7) ABC	0.0922	---	---
Mean Ratio		0.995	0.992
Standard Deviation		±2.8%	±2.8%

[a]Used as standard, calibrated by chemical assay results.

Table 3, show ratios of the nondestructive assays to chemical analyses for different spent fuel can combinations.

The correction factor f is in each case close to 1, since the standards were similar to the unknowns that were assayed. As a result, no noticeable differences are observed when the correction factors are based on chemical analysis isotopic information rather than on Dresden-supplied isotopic information.

6. ERROR ASSESSMENT

Because of the small mass and the low enrichment of the fuel that was assayed, it was not practical to employ counting times long enough to assure negligible counting errors. Overall counting statistics alone (including those from counting the standards) accounted for 1-1.5% measurement errors. Additional errors arise from uncertainties in the fissile masses of the fresh fuel standards (~0.5%), slight gain

fluctuations in the electronics (∿0.4%), and inaccuracies in the chemical assay determinations (∿0.5%). These errors indicate that total errors of 1-3% are to be expected for the assays in this investigation.

7. CONCLUSIONS

(1) The NDA system used in this investigation measured the fissile mass of BWR spent nuclear fuel within accuracies of 2-3%. Results of this investigation support the conclusions from previous experimental work with the ORNL NDA system, which stated that measurement of LMFBR subassemblies within an accuracy of <3% is achievable. Because of the high enrichment of LMFBR fuel, counting rates will be much higher than from Dresden fuel. Thus, counting errors (∿1.5%) resulting from the low count rates encountered in this investigation will be greatly reduced when LMFBR fuel is assayed. Neutron background-to-signal ratios for LMFBR fuels will not be as high as for the Dresden fuel. Also, self-shielding and multiplication effects of the LMFBR assembly will not present measurement difficulties, since earlier work[1-4] has shown the response for dense subassemblies is essentially spatially uniform and linear with fissile mass.

(2) Assay accuracies of 2-3% have been achieved with approximate isotopic data obtained from burnup calculations supplied by a utility (Dresden). If the information used herein is typical of the information that other utilities could provide, then assay accuracies of 2-3% could be achieved routinely without further measurements or calculations of isotopic fractions. For assay accuracies of <1%, more accurate isotopic data will be required, which probably will necessitate direct measurement of the isotopic fractions.

8. REFERENCES

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9. FIGURE CAPTION

(1) Spent fuel nondestructive assay system.

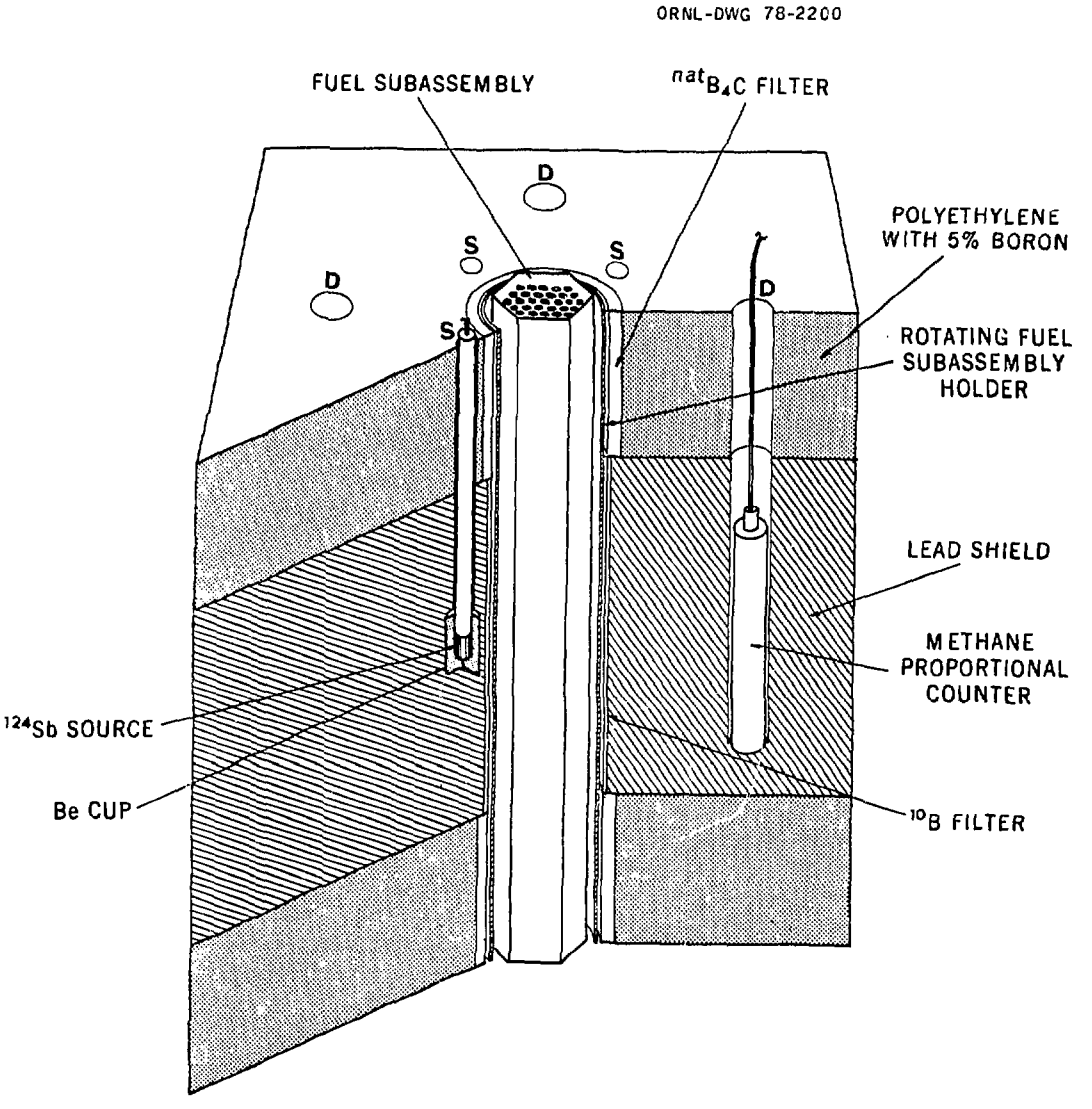

Figure 1. Spent fuel nondestructive assay system.