CONF-871045--7

Consolidated Fuel Reprocessing Program

^{252}Cf-SOURCE-DRIVEN NEUTRON NOISE MEASUREMENTS OF SUBCRITICALITY FOR A SLAB TANK CONTAINING AQUEOUS PU-U NITRATE*

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DE87 013676

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International Seminar on Nuclear Criticality Safety
Tokyo, Japan, October 17-23, 1987

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*Research sponsored by the Office of Facilities, Fuel Cycle, and Test Programs, U.S. Department of Energy, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

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^{252}Cf-SOURCE-DRIVEN NEUTRON NOISE MEASUREMENTS OF SUBCRITICALITY FOR A SLAB TANK CONTAINING AQUEOUS PU-U NITRATE

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ABSTRACT

In order to study nuclear criticality safety related to the development of fast breeder technology, ^{252}Cf-source-driven neutron noise analysis measurements were performed with a Pu-U nitrate solution in a slab tank of various heights and thickness varying 11.43 cm to 19.05 cm. The results and conclusions of these experiments are (1) a capability to measure the subcriticality of a multiplying system of slab geometry to a k_{eff} as low as 0.7 was demonstrated, (2) calculated neutron multiplication factors agreed with those from the experiments within ~0.02, and (3) the applicability of the method for plutonium solution systems was demonstrated. This paper describes measurements in which the height of the slab was varied for a fixed thickness and the thickness varied for a fixed height, which are the first applications of this measurement method to slab geometry.

INTRODUCTION

^{252}Cf-source-driven neutron noise analysis measurements[1,2] were performed with a slab tank at the Critical Mass Laboratory of Battelle Pacific Northwest Laboratories at Richland, Washington, in May 1986. An aqueous Pu-U nitrate solution was pumped into the slab tank to various heights for thicknesses varying from 11.43 to 19.05 cm, and measurements were performed using a measurement system in a mobile laboratory transported from Oak Ridge to Richland. This experiment is part of a program administered by the Consolidated Fuel Reprocessing Program at Oak Ridge National Laboratory and supported by a collaboration between the U.S. Department of Energy and the Power Reactor and Nuclear Fuel Development Corporation of Japan to study nuclear criticality safety related to the development of fast breeder technology[3].

The ^{252}Cf-source-driven neutron noise analysis method for obtaining the subcritical neutron multiplication factor of a configuration of fissile material from cross-power spectral densities (CPSDs) was developed to avoid difficulties inherent in other subcriticality measurement methods such as dependence on detection efficiency and the need for a calibration near delayed criticality. This method requires measurement of the frequency-dependent CPSD, $G_{23}(\omega)$, between a pair of detectors (*Nos.* 2 and 3) located in or near the fissile material and of CPSDs $G_{12}(\omega)$ and $G_{13}(\omega)$ between these same detectors and a source of neutrons emanating from an ionization chamber (No. 1) containing ^{252}Cf, also positioned in or near the fissile material. Also, the auto-power spectral density (APSD), $G_{11}(\omega)$, of the source is required. The particular ratio of spectral densities, $G_{12}^{*}\ G_{13}/G_{11}\ G_{23}$ (* denotes complex conjugation), then formed is independent of detector efficiency and can be related to the subcritical neutron multiplication factor.

Previous measurements performed with a wide variety of systems demonstrated the validity and usefulness of the method.[4-10] In these experiments, the method was used to obtain the subcritical neutron multiplication factor, k_{eff}, for a variety of systems with k_{eff} values that varied from ~0.3 to ~0.99. This paper describes measurements in which the height of the slab was varied for a fixed thickness and the thickness varied for a fixed height, which are the first applications of this measurement method to slab geometry.[11]

DESCRIPTION OF EXPERIMENT

The thickness of the unreflected slab assembly[12] was varied from 11.43 cm to 19.05 cm. The height and width of the tank was 106.7 cm, and for some thicknesses the solution height was varied. The mixed Pu-U nitrate solution, which is a mixture of plutonium and depleted uranium, had a specific gravity of 1.67 g/cm^3; a Pu content of 172.85 g/ℓ; a U content of 261.96 g/ℓ; a H^+ content of 1.23 normal; Pu isotopic constituents were 0.029 wt % for ^{238}Pu, 91.11 wt % for ^{239}Pu, 8.32 wt % for ^{240}Pu, 0.453 wt % for ^{241}Pu, and 0.093 wt % for ^{242}Pu; and the U isotopic constituents were 0.007 wt % for ^{234}U, 0.568 wt % for ^{235}U, 0.024 wt % for ^{236}U, 99.402 wt % for ^{238}U.

The fission rate of the ^{252}Cf source from an average of two independent calibrations was $3.15 \pm 0.10 \times 10^6$ fissions/s for the source for this experiment. Since the ^{252}Cf ionization chamber electronics in the subcriticality measurements counted 1.73×10^6 fissions/s, only 55.0 ± 1.5% of the ^{252}Cf fissions occurring in the source for this experiment were actually detected. The effect of not counting all ^{252}Cf fissions is well understood: previous measurements have been successful even though the percentage of fissions counted was as low as 20%.[1]

Two types of detectors were used in the experiments: ^3He proportional counters and scintillation detectors. The 76.2-cm-long ^3He proportional counters were located vertically at the ends of the slab. Li-glass-plastic scintillators were located on one large face of the slab while the source was adjacent to the other large surface. This positioning minimized the counting of particles directly from the ^{252}Cf source, since particles from ^{252}Cf fission would have to pass through the fuel solution to reach the detectors.

The data-processing system consisted of analog-to-digital converters, a Fourier analyzer, and a control computer. The analog-to-digital sampling system and the array processor (Signal Processing Systems, Inc., Model SPS-1000) used as a Fourier analyzer are capable of sampling and processing three channels of 1024 data points per data block at a throughput rate of 200,000 samples/s for each channel. The array processor and ADC system are controlled by a computer (VAX-750) that intermittently takes the auto-power spectral densities (APSDs), G_{ii}, and cross-power spectral densities, G_{ik}, from the array processor, averages them appropriately, and stores them in data files. The ratio of spectral densities are calculated from these data files.

RESULTS

In these experiments detector-source configurations were varied in order to find detector locations where point kinetics interpretation of the ratio of spectral densities yields meaningful results. In past experiments, several criteria were developed that (in most cases) result in measurements that are interpretable using point kinetics. Some of these criteria are consistent with experience developed previously in pulsed neutron measurements and other kinetic measurements. These criteria are (1) central location of the source, (2) symmetric location of the detectors with respect to the source, (3) detectors located far enough from the source to avoid counting particles directly from the source and to avoid effects from higher spatial modes near the source, (4) use of detectors spanning the full height of the system, and (5) use of the ratio of spectral densities at low frequency only to obtain the neutron multiplication factor. In these experiments, measurements were performed to meet the above criteria as far as practical.

The low-frequency value of the ratio of spectral densities was obtained over the frequency range for which the ratio was found to be constant and statistically significant. This range was selected by visual inspection of the data, but in a practical application of the method this selection would be automated using pattern-recognition techniques. The frequency range chosen for most measurements was such that the cross-power spectral densities with the source G_{ij}, which for fundamental mode falls off at a rate of one decade in amplitude per decade in frequency, could be measured up to a frequency at which it approaches zero. As a result, the CPSDs between the detectors G_{ij} ($i \neq 1$), which for fundamental mode fall off at a rate of 2 decades in amplitude per decade in frequency, are essentially zero for a range of higher frequencies. Thus, over this range of frequencies the ratio of spectral densities had a very large statistical uncertainty.

Table 1 summarizes the measured ratios of spectral densities at low frequency for various slab configurations with the source at the approximate center of the slab face for symmetric source detector locations. Also given in Table 1 are the ratios of spectral densities with the source at the approximate center of the slab face for asymmetric source-detector locations for which a detector was located on each side of the source. These values generally agree with those for symmetric locations of the detectors on each side of the source since the detectors were sampling particles from fission chains propagating over a large fraction of the volume of the slab. For detectors adjacent to each other, the measured ratios were considerably different since these detectors sample fission chains propagating over a small fraction of the slab and are influenced by spatial effects. Interpretation of the experiments

Table 1. Average ratios of spectral densities and neutron multiplication factors for various solution heights and slab thickness with the source located at the approximate center of one large external surface of the slab

Solution height, slab thickness (cm)	Ratio of spectral densities ($\times 10^{-4}$)			Multiplication factor from ratio of spectral densities, k_{eff}[c]
	Symmetric source detector location[a]	Asymmetric source detector location[a]	Average of all measurements[b]	
71.3, 19.05	23.2±0.4	23.5±0.5	23.3±0.3	0.9924±0.0002
61.6, 19.05	50.5±0.9	52.1±1.0	51.3±0.7	0.9830±0.0005
50.7, 19.05	108.5±1.8	110.0±1.7	109.3±1.2	0.963 ±0.001
40.6, 19.05	209.5±3.9	209.7±2.1	209.6±2.0	0.924 ±0.002
71.1, 16.51	232 ± 6	232.0[d]	233.2± 4	0.924 ±0.002
71.1, 13.97	548 ± 6	—	548 ± 6	0.841 ±0.004
71.1, 11.43	1065 ±27	1161 ±100	1097 ±33	0.707 ±0.012

[a]Unweighted average
[b]Uncertainty estimates are one standard deviation of the mean obtained from averaging measurements for the same configuration.
[c]The uncertainties given account for the statistical precision of the ratios of spectral densities and estimates of the uncertainties in the other parameters required to obtain k_{eff}.
[d]Both measurements resulted in a ratio of 0.0232.

with adjacent detectors and also with detectors less than 26 cm from the source will require an interpretation with a spatial kinetics model. Neutron multiplication factors, obtained from the average ratios of spectral densities are also presented in Table 1.

The two other reactivity measurement methods used in these experiments were inverse kinetics rod drop (IKRD)[13,14,15] and break-frequency noise analysis (BNFA).[16] To obtain the subcriticality, the latter method requires a calibration at a known reactivity condition near delayed criticality. This calibration was provided in the experiments by the inverse kinetics method. These well-known methods were used to obtain reactivity estimates with which to compare reactivities obtained from the ^{252}Cf-source-driven neutron noise analysis method.

Independent measurements of subcriticality by the break-frequency noise analysis method, calibrated near delayed criticality by an inverse kinetics rod drop technique, were also performed in order to have an independent measurement to serve as a verification of the Cf-source-driven neutron noise analysis method.

IKRD measurements were made for the 19.05-cm-thick slab with a solution height of 78.1 cm: an acrylic reflector was quickly withdrawn from its position adjacent to the lateral surface of the slab and moved 20 cm away from the slab. The initial and final reactivities from this measurement are given in Table 2. As shown in Table 2, the initial reactivities obtained from all three detectors in two independent measurements varied between 48 and 53 cents subcritical, with average values of 49.6 and 50.6 cents. The final reactivities varied between 165 and 195 cents, with average values of 177.3 and 182.4 cents.

Table 2. IKRD measurements with a 19.05-cm-thick slab with a solution height of 78.1 cm

Counter[a]	Initial reactivity[b] (cents)	Final reactivity[c] (cents)
S-1	50.4±0.4	179.5±5.0
	51.5±1.5	186.5±5.3
S-2	50.3±1.1	187.0±4.0
	53.4±1.0	194.9±4.4
S-3	47.9±1.2	165.4±4.0
	47.7±1.2	165.9±4.3
Average	49.6±0.5	177.3±2.5
	50.8±0.7	182.4±2.7

[a]These counters were located 8 to 17 cm from the slab surface at the ends.
[b]Two independent measurements were performed. The initial reactivity is with the acrylic reflector adjacent to the slab surface. The uncertainty given is one standard deviation.
[c]The final reactivity is with the acrylic reflector withdrawn 20 cm. The uncertainty given is one standard deviation.

For the BFNA measurements, various spectral densities were least-squares fitted to obtain the break frequency, f_b. The reactivity at a given subcritical state is related to the subcritical state break frequency, f_b (or to the prompt-neutron decay constant, $\alpha = 2\pi f_b$), and to the break frequency at delayed criticality, f_{bdc} (or prompt-neutron decay constant, $\alpha_{dc} = \beta/\ell$), as follows:
$(1-k)/k\beta + 1 = (f_b \ell \beta_{dc})/(f_{bdc} \ell_{dc} \beta)$.

The ^{252}Cf source was not present in the break-frequency measurements for the 19.05-cm-thick slab at a solution height of 78.1 cm. The signals from five neutron detection systems were used for noise analysis. The average break frequency was obtained by fitting the five APSDs and the ten CPSDs at the reference reactivity obtained by the IKRD method previously described. The results of this fitting are presented in Table 3 for the reference reactivities with and without the acrylic reflector. The average break frequency for the measurement with the reflector adjacent to the slab surface is 84.5 ± 1.0 Hz, and that with the reflector removed is 155.8 ± 3.0 Hz. These reactivities and break frequencies yield estimates of the break frequency at delayed criticality (f_{bdc}) of 56.2 ± 0.7 and 55.7 ± 1.2 Hz from the measurements with the acrylic reflector inserted and removed respectively. Thus, the average break frequency and α_{dc} at delayed criticality are 55.9 ± 1.0 and 351.5 ± 7.0 s^{-1} respectively. These latter values are used to obtain the reactivity from the BFNA method.

Table 3. Break frequency from noise measurements for a 19.05-cm-thick slab with a solution height of 78 cm

Acrylic reflector position	Reactivity from IKRD (cents)	Break frequency (Hz)	Break frequency at delayed critical (Hz)	Prompt neutron decay constant (s^{-1})
Adjacent to slab surface	49.6±0.5[a]	83.3±0.9	55.7±0.6	349.8±4.0
	50.8±0.7	85.6±1.0	56.8±0.7	356.7±4.5
Removed from slab surface	177.3±2.5	156.5±3.4	56.4±1.3	354.6±8.3
	182.4±2.7	155.1±2.7	54.9±1.1	345.1±6.8

[a]Uncertainties given are one standard deviation.

The APSDs and CPSDs were fitted using least-squares analysis methods to obtain the fundamental mode break frequency for the various experimental configurations of the slab tank. The results, given in Table 4, are the average values from all source-detector configurations for which the data had sufficient statistical precision to be fitted.

The neutron multiplication factors from both experimental methods are compared in Table 5 and agree within the precision of the measurements. The neutron multiplication factors, k_{eff}, were also calculated for a variety of slab configurations using the KENO Monte Carlo method[17] with both Hansen-Roach[18] and ENDF/B-IV cross sections.[19] The geometry of the calcula-

Table 4. Fundamental mode break frequency and neutron multiplication factors from break-frequency noise analysis for various configurations of the slab tank

Slab thickness (cm)	Solution height (cm)	Average fundamental mode break frequency[a] (Hz)	Percentage change in $\frac{\ell}{\ell_{dc}}\frac{\beta_{dc}}{\beta}$	Neutron multiplication factor from BFNA
19.05	71.3	249± 6(3)	0	0.990±0.001
19.05	61.6	507± 3(7)	0.8	0.978±0.001
19.05	50.7	1009±13(13)	1.8	0.953±0.003
19.05	40.6	1732±31(6)	4.7	0.917±0.005
16.51	71.1	1836±14(24)	5.5	0.911±0.005
13.97	71.1	3280±64(34)	13.1	0.835±0.008
11.43	71.0	5690±70(27)	24.5	0.715±0.012

[a]Uncertainties given are one standard deviation. Where the number of measurements is small, the estimates of uncertainties are larger. The number of sets (where a set of experimental data consists of the real and imaginary parts of, for example, three CPSDs, G_{12}, G_{13}, G_{24}, and two APSDs, G_{22}, G_{44}) of the experimental data that were fitted are given in parentheses.

tions includes the egg-crate structure supporting the large flat surfaces of the slab and the room structure, the acrylic hood, and the reflector tank surrounding the slab assembly. The calculated k_{eff} value for the solution height (86.3 cm) that was experimentally determined to be delayed critical was 0.9961. Thus, at least near delayed criticality, the calculations underestimate the k_{eff} value by −0.004. This bias was assumed constant for all slab configurations and hence was added to all calculations for comparison with the measurements. The results of these calculated k_{eff} values for other solution heights are given in Table 5 and agree with the experimental values from the noise measurements to within 0.02.

Table 5. Comparison of calculated neutron multiplication factors with those from the ^{252}Cf source-driven neutron noise and BFNA methods

Slab dimensions (cm)		Neutron multiplication factors			
			Measured from		
			Ratio of spectral densities		
Thickness	Height	Calculated[a]	ORNL	Nagoya-Uni.[b]	BFNA
19.05	71.3	0.992	0.9924±0.0002	0.9917	0.990±0.001
19.05	61.6	0.979	0.9830±0.0005	0.9828	0.978±0.001
19.05	50.7	0.947	0.963 ±0.001	0.9655	0.953±0.003
19.05	40.6	0.925	0.924 ±0.002	———	0.917±0.005
16.50	71.1	0.914	0.924 ±0.002	———	0.911±0.005
13.97	71.1	0.835	0.841 ±0.004	0.8403	0.835±0.008
11.43	71.0	0.720	0.707 ±0.012	———	0.715±0.012

[a]All calculated values have a statistical uncertainty of ±0.005 and include the concrete structure of the cell, the acrylic hood, the steel reflector tank, and the detectors. A bias of 0.004 (obtained by comparing experiment and calculation for a k_{eff} = 1 experimental configuration) was added to all calculated values.
[b]These values were obtained by data processing with a theoretical equation of Nagoya University[20].

CONCLUSIONS

The results and conclusions of these experiments are (1) a capability to measure the subcriticality of a multiplying system of slab geometry to a k_{eff} as low as 0.70 was demonstrated, (2) calculated neutron multiplication factors agreed with those from the experiments within ~0.02, (3) the criteria developed in previous experiments for choosing source-detector-system configurations for which the data could be interpreted using point kinetics were satisfactory for this experiment, (4) measurement times for this geometry were not significantly different from those in cylindrical geometry and were sufficiently short to allow practical measurements, (5) the applicability of the method and understanding of the theory of the measurement method for plutonium solution systems was demonstrated, and (6) the reactivities obtained by independent measurements using the break-frequency noise analysis method agreed with those obtained from the ratio of spectral densities within the experimental uncertainties. Before the use of this method in slab geometry can considered to be fully understood, additional static and dynamic measurements are required for a variety of slab configurations.

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