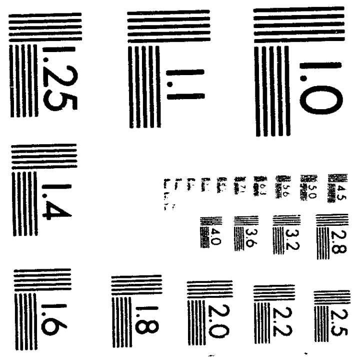

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SENSITIVITY OF CALCULATIONS OF ^{252}Cf-SOURCE-DRIVEN NOISE ANALYSIS MEASUREMENTS TO CROSS-SECTIONS FOR AQUEOUS FISSILE PLUTONIUM SOLUTIONS

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ABSTRACT

Previous experiments have shown large changes in noise-measured parameters for small changes in the measured configuration of fissile material. The sensitivity was investigated by the Monte Carlo code KENO-NR. This code calculates the noise data from the ^{252}Cf-source-driven neutron noise measurement. These calculations have shown that noise-measured quantities are more sensitive to cross sections than the neutron multiplication factor by a factor of as much as ~50 for this fissile plutonium solution. This increased sensitivity means that for verifying calculational methods, a subcritical experiment at a $k \sim 0.9$ by the ^{252}Cf-source-driven noise analysis method is more useful than experiments at $k = 1$.

INTRODUCTION

The ^{252}Cf-source-driven neutron noise analysis method[1] 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 or need for a calibration at a known reactivity condition near delayed critical and dependence on detector efficiency. This method requires measurement of the frequency-dependent CPSD, $G_{23}(\omega)$, between a pair of detectors (detectors 2 and 3) located in or near the fissile material as well as measurements of CPSDs, $G_{12}(\omega)$ and $G_{13}(\omega)$, between these same detectors and a source of neutrons emanating from the ^{252}Cf source ionization chamber (detector 1) positioned in or near the fissile material. The ^{252}Cf source provides neutrons to initiate the fission chain multiplication process. Also required is the auto-power spectral density (APSD), $G_{11}(\omega)$, of the source. A particular ratio of spectral densities $G_{12}^{*} G_{13} / G_{11} G_{23}$ (the asterisk denoting complex conjugation), is independent of detector efficiencies and can be related to the subcritical neutron multiplication factor k. Another useful quantity, the coherence γ_{ij}^2, which is defined as $|G_{ij}|^2/G_{ii}G_{jj}$, is related to higher powers of $\Delta k/k$.

Previous experiments have shown that large changes in noise-measured parameters such as the coherences and ratio of spectral densities result from small changes in the measured configuration of fissile material and from small changes in k. For example, at a $k = 0.9$, the ratio of spectral densities is a factor of 10 more sensitive to changes in k than k itself, and the coherence between detectors is almost a factor of 50 more sensitive. The very high sensitivity of the coherence results from the fact that, for low values of k, the coherence varies as the fourth power of $k/(1 - k)$. Since these noise-measured parameters can be measured to $\pm 1\%$, they can provide a more sensitive test of calculational methods. The ability to calculate these noise-measured quantities that are proportional to some power of $\Delta k/k$ ensures that the calculational method can calculate the margin of subcriticality.

This sensitivity was investigated with KENO-NR[2], which calculates the time sequences of pulses at two detectors near a fissile assembly from the fission chain multiplication process initiated by a ^{252}Cf source in or near the fissile assembly. This code which is a variant of the Monte Carlo neutron transport code KENO-V.a[3] directly calculates the noise analysis data from this measurement method. The time sequences of pulses at the detectors from various initiating ^{252}Cf neutrons are superimposed, consistent with a given source fission rate, and then sampled and Fourier analyzed in a manner identical to the measurement. This Monte Carlo calculation of the measurement is the most valid way of assessing measurement feasibility and sensitivity of noise-measured parameters to cross sections and calculational methods. Direct calculation of the experimental observables by the Monte Carlo method allows the benchmarking of calculational methods and cross sections.

Previous calculations with uranyl (20% ^{235}U) fluoride and uranyl (93.2% ^{235}U) nitrate solutions have shown that these noise-measured parameters at $k = 0.9$ are a more sensitive test of calculation methods and cross sections than critical experiments at $k = 1$ (reference 4). In these previous studies, the sensitivity to cross-section changes was a factor of 10 higher for ratios of spectral densities and a factor of ~40 higher for coherences than for neutron multiplication factor comparisons. Thus, a subcriticality measurement by ^{252}Cf-source-driven noise analysis can measure not only the subcriticality, but it also can be used to provide data for better verification of calculational methods and cross sections than by using comparisons of calculated and measured neutron multiplication factors.

The study reported here explored the sensitivity of the method to changes in cross sections for subcritical plutonium nitrate solutions to investigate the usefulness of these measurements for calculational verification. Three cross-section sets used were 16-group Hansen-Roach[5] with σ_p calculated according to Hansen's method, 27-group ENDF/B-IV,[6] and 27-group ENDF/B-V,[7] as well as variations in the amount of moderator and fissile material in solution. The 27-group cross sections were obtained by using the SCALE system.[8]

AQUEOUS PLUTONIUM FISSILE SOLUTION SYSTEM

The system used in this evaluation is an unreflected right circular cylinders with $k \sim 0.9$. The calculational model included a central source and 5.1-cm-diam ^3He proportional counters on each side

~180° apart as the two detection channels, with the counter axes and the solution cylinder axis parallel. These source detector locations are typical of measurements.

The aqueous plutonium (94% ^{239}Pu) nitrate contains 200 g of plutonium per liter and a solution density of 1.643 g/cm^3, with a molarity of 8. The high molarity was chosen to emphasize the effects of nitrogen. Plutonium isotope content in weight percent is ^{239}Pu = 94.8, ^{240}Pu = 5.42, and ^{242}Pu = 0.58. The hydrogen-to-^{239}Pu ratio is 94.

VARIATIONS IN CROSS SECTIONS

These calculations were performed with Hansen-Roach, ENDF/B-IV, and ENDF/B-V cross sections for KENO. In addition to the variations in the microscopic cross sections, the macroscopic cross sections were also changed by increasing the hydrogen atomic density by 3%, the plutonium by 5%, the oxygen by 10%, and the nitrogen by 10%.

KENO-NR AND KENO-AK CALCULATIONS

The KENO-NR code directly calculates the results of the ^{252}Cf-source-driven noise analysis measurement. This code is an analog version of KENO which directly implements the physics of the neutron interaction process except for the group structure of the problem and the scattering approximations of KENO-V.a. It follows the fission chains to extinction and thus cannot be used for fissile systems at criticality since the fission chain multiplication process does not terminate. No biasing or weighting is used. The average quantities of KENO-V.a such as the average number of neutrons per fission $\bar{\nu}$ are not used, but the known probability distribution of the numbers of neutrons P(ν) are used. Whereas average quantities are used in calculating fluctuating phenomena, the extent of the fluctuating processes is limited, thus reducing the amplitude of frequency-dependent functions.

Since KENO-NR directly calculates the measured quantities, it and the cross-section data set can be verified by measurements. KENO-AK, which uses the same tracking routines and cross sections as KENO-NR, can then be used to obtain the value of the neutron multiplication factor. KENO-AK calculates the fission chain populations resulting from a batch of source fissions distributed with a specified spatial distribution. It follows all the fission chains to extinction. At the completion of this batch, it picks the distribution of the subsequent fissions for the second batch from fission sites from the previous batch. It continues in this fashion for the specified number of batches. At the end of all batches, it sums up all neutron production and divides by the input source to obtain a *k* value as a function of the number of initial batches deleted from the *k* calculation.

RESULTS AND DISCUSSION

The frequency-domain analysis provides a variety of frequency-dependent parameters that can be calculated and compared to measurements. To obtain the correct frequency dependence, the calculation must have the correct neutron energy spectra both in the system and in the leakage because the detectors are external. Frequency-dependent parameters must be compared at specific frequencies. One useful range for comparison is at low frequency, where many of the parameters are constant with frequency. The frequency-dependent functions are APSDs, real and imaginary parts of CPSDs, coherences, phases, ratios of spectral densities, ratios of the products of the frequencies and the real parts of the CPSDs to the imaginary parts that are related to the prompt-neutron decay constant α or the break frequency f_b, where $\alpha = 2\pi f_b$. Some of these quantities depend on the detector reaction rate; but, since the detectors are included in the calculation, they are accounted for explicitly. Other parameters such as the ratios of spectral densities, phases, the break frequencies, and the ratios of the real to imaginary parts of the CPSDs are not dependent on the detector sensitivity.

A. MICROSCOPIC CROSS-SECTION CHANGES

A variety of frequency-dependent functions calculated with Hansen-Roach cross sections can be compared with similar functions obtained with ENDF/B-IV and ENDF/B-V cross sections. Comparisons of the results of calculations with Hansen-Roach and ENDF cross sections are summarized in Table 1. The amplitude of the power spectral densities at low frequency from calculations with ENDF cross sections was more than 30% higher than that with Hansen-Roach cross sections or a factor of 10 more sensitive to cross sections than k calculations. The coherence γ_{12}^2 between detectors and the sources is the fraction of common information in the two signals which arises from the detectors' detecting particles from fission chains initiated by ^{252}Cf fission. The coherence γ_{23}^2 between the two detectors is the fraction of common information in the two detector signals which arises from the two detectors' detecting particles from the same fission chains.

The calculations with ENDF cross sections yield a value of the coherence γ_{12}^2 more than 25% larger at low frequency than that obtained with the Hansen-Roach cross sections. A 4% difference occurs in γ_{12}^2 at low frequency between ENDF/B-IV and ENDF/B-V cross sections. At frequencies above 10^3 Hz, the coherence from calculations with Hansen-Roach cross sections is higher, indicating a larger fraction of higher energy neutrons. The calculation with ENDF cross sections yields a value of the coherence γ_{23}^2 at least 150% larger at low frequency than that from the Hansen-Roach cross sections. An 11% difference occurs in γ_{23}^2 at low frequency between ENDF/B-IV and ENDF/B-V cross-section data sets. The coherence between detectors is a factor of 45 more sensitive to cross sections than the neutron multiplication factor because the coherence at low k is proportional to the fourth power of $(k/\Delta k)$. This high sensitivity of this coherence has been observed in many measurements.

The prompt-neutron decay constant $\alpha = 2\pi f_b$ (where f_b is the break frequency) is related to the ratio of the product of the frequency and the real part of G_{12} to the imaginary part of G_{12}. The value of this ratio for G_{12} changed more than 23% between Hansen-Roach and ENDF cross sections. A 4% difference occurs in α between the ENDF/B-IV and B-V cross-section data sets. The larger value of the prompt-neutron decay constant results partially from a lower k value for the Hansen-Roach set but also from the difference in the prompt-neutron lifetime. The break frequency is the frequency at which G_{12} is 0.70 of its low-frequency limit or 0.5 of its low-frequency limit for G_{23} and can be obtained from the frequency dependence of the calculated G_{12} and G_{23}. This frequency dependence can be calculated and compared with experiment.

The ratio of spectral densities, which has been used in a wide variety of measurements to obtain the neutron multiplication factor with point kinetics interpretation of the experimental data, changed more than 20% between ENDF and Hansen-Roach cross sections. This change was a factor of 6 more sensitive than changes in k and is expected since the ratio of spectral densities is proportional to $\Delta k/k$. The ratio of spectral densities between ENDF/B-IV and ENDF/B-V cross sections differed by 2%.

The phase of the CPSD, G_{12}, is related to the time delay between the correlated signal at the detector and the fission chain initiation by ^{252}Cf source neutrons. The calculated phase as a function of frequency is larger for the ENDF cross sections by 40% at a frequency of 1 kHz (Table 1). The time delay associated with the phase at 1 kHz is 42 μs for Hansen-Roach, 61 μs for ENDF/B-IV, and 63 μs for ENDF/B-V. This difference in phase for a calculation for which the k value differs by less than 3% results from the fact that the k values for the ENDF calculations are higher and the fission chain multiplication process takes longer to develop and die away. Since the detectors in the calculation are symmetrically located with respect to the source and the fissile system (also usually in the measurements), the phase of $G_{23} \approx 0$.

TABLE 1

SENSITIVITY OF CALCULATED FREQUENCY-DEPENDENT PARAMETERS OF A ^{252}Cf-SOURCE-DRIVEN NOISE ANALYSIS MEASUREMENT TO CROSS-SECTION SETS FOR PLUTONIUM NITRATE SOLUTIONS

Parameter	Hansen-Roach	ENDF/B-IV	ENDF/B-V
k	0.863 ± 0.002	0.892 ± 0.002	0.895 ± 0.002
G_{22} (LF)	0.431	0.575	0.603
γ^2_{12} (LF) × 10^{-4}	120 ± 1.0	151 ± 0.6	157.5 ± 0.5
γ^2_{23} (LF) × 10^{-5}	213.2 ± 2.6	531.7 ± 2.9	591.6 ± 4.7
α_p (LF) (sec^{-1})	-18,875 ± 1,500	-14,674 ± 90	-14,057 ± 89
Ratio (LF) × 10^{-4}	2,615 ± 21	2,071 ± 5	2,033 ± 6
Phase @ 1000 Hz (degrees)	-15	-22	-23

B. MACROSCOPIC CROSS-SECTION CHANGES

The macroscopic cross sections were varied by changing the atomic densities of the fissile solution constituents. For these solutions, the hydrogen content was increased 3%; the plutonium content, 5%; and the other constituents, oxygen and nitrogen, 10%. These results for the plutonium nitrate solution are summarized in Table 2. The 5% change in the plutonium atomic density did not change the neutron multiplication factor, whereas the effect of changing hydrogen 3% was a change in k of 0.02. A 10% change in the oxygen and nitrogen content changed k by 0.015 and 0.001 respectively. Since the standard deviation of the calculated k is ±0.002, the statistical significance of the change in k from the change in nitrogen is questionable.

The change in the APSD G_{22} is a factor of 11 more sensitive to a 3% increase in the hydrogen macroscopic cross sections than changes in k, while that for a 10% increase in the oxygen content was a factor of *10* more sensitive than changes in k. The changes in the noise-measured parameters for a 5% increase in the plutonium atomic density were negligible except for the γ_{23}^2, which change 3%. The change in the coherence between detectors on opposite sides of the vessel was a factor of 36 more sensitive to a 3% change in the atomic density of hydrogen than the neutron multiplication factor, and that for the 10% change in oxygen was a factor of 34 more sensitive. These high sensitivities of the noise-measured quantities to atomic densities can be advantageous for validation purposes and for determining which solution constituents need to be measured most accurately by experimenters.

The high sensitivity of the calculations of the noise-measured parameters (which can be measured to an accuracy ±1% and calculated to a precision of ±1%) to cross section sets means that these subcritical noise experiments may be more useful for calculational verification than measurements of the neutron multiplication factor. Differences in the calculations that produce changes in the calculated k value within the statistical uncertainty indicate that ±0.001 (0.1%) can result in statistically significant changes in the noise-measured parameters. This high sensitivity means that noise-measured parameters are more useful for verification of calculations because these parameters are more sensitive to inadequacies in the calculational methods or cross sections than the neutron multiplication factor. At values of k much higher than 0.95, some of the noise-measured parameters are less sensitive to changes; for example, the variation in the coherence is much less than $k/\Delta k$ to the fourth power. This weaker dependence on k begins to manifest itself in this calculation at k of 0.9, where a 1% change in k produces a 19% change in γ_{23}^2, whereas the fourth-power dependence on $k/\Delta k$ would have resulted in an ~40% change.

CONCLUSIONS

These calculations have shown a higher sensitivity of noise-measured quantities to cross sections and calculational methods than the neutron multiplication factor for aqueous plutonium fissile solutions. This increased sensitivity means that for verifying calculational methods, a subcritical experiment at a $k \approx 0.9$ by the ^{252}Cf-source-driven noise analysis method may be more useful than an experiment at $k = 1$. The noise-measured parameters can be obtained from measurements with an accuracy of ±1% or less, and the precision of the Monte Carlo calculation of these quantities can also be ±1% or less. In many cases, changes made that affect k only within the statistical uncertainty of the Monte Carlo calculation produce significant changes in the calculated noise-measured parameters well outside the limits of the uncertainty on these noise-measured parameters (either measured or calculated). Calculation of these noise- measured parameters that are sensitive to the criticality safety margin is a better way of verifying cross-section sets. This calculation and measurement capability allows the validation of calculational methods with in-plant measurements that are less costly and may be more relevant since the variables changed to achieve criticality may make the critical experiment different from the in-plant situation.

TABLE 2

SENSITIVITY OF CALCULATED FREQUENCY-DEPENDENT PARAMETERS OF A ^{252}Cf-SOURCE-DRIVEN NOISE ANALYSIS MEASUREMENT TO CHANGES IN ATOMIC DENSITIES OF PLUTONIUM NITRATE SOLUTION CONSTITUENTS

Parameter	Percentage Change in Parameter for Various Percentage Increases in Atomic Densities of Constituents			
	+5% Pu	+3% H	+10% O	+10% N
k (unperturbed = 0.892)[a]	~0	0.020	0.016	~0
G_{22} (LF)[b]	~0	24 (11)[c]	19 (11)	2
PHASE 12 at 6 kHz	~0	18 (8)	14 (8)	~0
γ^2_{12} (LF)	~0	18 (8)	15 (8)	~0
γ^2_{23} (LF)	3	81 (36)	60 (34)	4
Prompt-neutron decay constant	~0	17 (7)	13 (7)	2
Ratio of spectral densities	~0	13 (6)	10 (6)	2

[a]For the neutron multiplication factor, the values given are Δk.
[b]LF signifies the value of the noise-measured parameter at low frequency.
[c]The numbers in parentheses are the percentage changes in the parameters divided by 100 times Δk.

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