Sensitivity of Subcritical Measurement Simulations to Neutron Cross-Section Data

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Evaluation of nuclear data typically includes validation of the data through computation of k_{eff} for critical assemblies. The sensitivity of the computed k_{eff} values to the nuclear data is used as an indicator in determining the adequacy of an evaluation. Subcritical measurements offer an alternative to critical experiments as a means to evaluate nuclear data through direct computation of subcritical measurement quantities. In some instances, the subcritical measurement quantities are more sensitive to nuclear data changes than the computed k_{eff} values. Simulations of subcritical source-driven noise measurements were performed for highly enriched uranium metal cylinders and highly enriched uranyl nitrate solutions to demonstrate the sensitivity of the computed subcritical quantities to nuclear data. A particular ratio of spectral quantities from the source-driven noise measurements is of interest because of its independence of detection efficiency and source intensity. These simulations indicate that the spectral ratio is more sensitive to nuclear data evaluations than the computed k_{eff} values for these systems. Direct simulation of subcritical measurements offers additional means to validate nuclear data evaluations.

KEYWORDS: subcritical measurement, evaluation, high enriched uranium, cross sections

I. Introduction

The validation of nuclear data typically involves simulation of critical experiments using Monte Carlo codes and processed nuclear data evaluations. The results of such simulations are typically used to provide information concerning the sensitivity of the calculated neutron multiplication factor, k_{eff}, to slight changes in the nuclear data evaluation or to differences among various nuclear data evaluations. Such sensitivity studies are necessary to provide indicators of inadequacies in nuclear data evaluations. Simulations are typically performed with a wide variety of critical experiments with systems with neutron flux spectrums ranging from thermal to fast in order to properly validate the nuclear data evaluations over broad energy regions.

The direct simulation of subcritical measurements offers an additional means to validate nuclear data evaluations. Conventional Monte Carlo codes have been modified to directly simulate subcritical measurements as they are performed. Simulations of subcritical measurements may be more sensitive to nuclear data evaluations than computations of k_{eff}. Furthermore, subcritical measurements may provide a greater range of neutron spectra than can be achieved with critical experiments because subcritical measurements can be performed with a wider range of materials and geometries than can be achieved with critical experiments. Source-driven subcritical noise measurement has several advantages over other subcritical measurement techniques for benchmarking nuclear data and Monte Carlo codes. One of the primary advantages is that the spectral ratio obtained from the noise measurement does not depend on detection efficiency. This is a significant advantage in that a change in the detection system would not manifest itself in the spectral ratio. Another distinct advantage is that the prompt neutron decay constants can also be obtained from this measurement. Additionally, quantities from the subcritical noise measurement have high sensitivity to small changes in configurations of fissile materials.

II. Subcritical Noise Measurements

Subcritical source-driven noise measurements[1)] are simultaneous Rossi-α and randomly pulsed neutron measurements that provide measured quantities that can be related to k_{eff}. The source-driven noise analysis measurement requires the use of a timed neutron source such as a ^{252}Cf source ionization chamber (detector 1) and two or more neutron detectors (detectors 2 and 3, respectively). The time-dependent responses, f(t), of the source and detectors are correlated in the frequency domain to obtain auto and cross spectra. The source-detector cross spectra are designated as $G_{12}(\omega)$ and $G_{13}(\omega)$ and are the frequency domain equivalent of the randomly pulsed neutron measurement. The detector-detector cross spectrum is designated as $G_{23}(\omega)$ and is the frequency domain equivalent of the two-detector Rossi-α measurement. The auto spectra of the source is designated as $G_{11}(\omega)$ and is related to the spontaneous fission rate of the source. The detector auto spectra are designated as $G_{22}(\omega)$ and $G_{33}(\omega)$ and are the frequency domain equivalent to the single-detector Rossi-α measurement. A certain ratio (R) of the frequency spectra is independent of detection efficiency

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and can be directly computed using Monte Carlo codes. The spectral ratio is defined as:

$$R(\omega) = \frac{G_{12}^{*}(\omega)G_{13}(\omega)}{G_{11}(\omega)G_{23}(\omega)}. \qquad (1)$$

The asterisk in Eq. 1 denotes complex conjugation. If point reactor kinetics models are used to describe the various frequency spectra for the source-driven noise analysis measurement, the spectral ratio is related to reactivity as follows[2]:

$$R(\omega) = \frac{\varepsilon_s \overline{\nu_0}}{\left[\frac{\overline{\nu(\nu-1)}}{|\rho|} + \frac{\overline{\nu_0(\nu_0-1)}}{\overline{\nu_0}}\right]}. \qquad (2)$$

In this expression, ε_s is the efficiency for detecting the spontaneous fission of the source, ν is the number of neutrons from fission in the system, ν_0 is the number of neutrons from the ^{252}Cf source, and ρ is the system reactivity.

The Monte Carlo code MCNP-DSP[3] was developed to simulate a variety of subcritical measurements. In MCNP-DSP, the variance reduction features were disabled to obtain a strictly analog particle tracking to follow the fluctuating processes more accurately. Because biasing techniques are typically employed to reduce the variance of estimates of first moment quantities, they do not preserve the higher moments; therefore, analog Monte Carlo calculations must be performed when analyzing subcritical measurements whose measured quantities are directly related to the higher moments of the neutron production. Because the use of average quantities reduces the statistical fluctuations of the neutron population, average quantities such as the average number of neutrons from fission are not used; instead, appropriate probability distribution functions are sampled. The prompt particle tracking begins with the source event and the subsequent fission chains are followed to extinction. Time series of pulses are obtained at the detectors for each fission chain. These sequences are sampled into blocks of 512 or 1024 data points. The blocks of data are then processed according to the type of measurement being simulated. The auto and cross spectra are computed directly from the Fourier transform of the source and detector time series for each data block. The auto and cross spectra are then averaged over many blocks to obtain the final estimates of the auto and cross spectra. The spectral ratio can then be computed from the auto and cross spectra as is done in the noise analysis measurement.

III. Subcritical Noise Measurements Simulations

Simulations of the source-driven noise analysis measurements were performed for two different experiments. Experiments with highly enriched uranyl nitrate solutions and highly enriched uranium metal cylinders were simulated with MCNP-DSP to examine how the computed spectral ratio varied with nuclear data evaluations.

1. Uranyl Nitrate Solution Tank

Measurements were performed with varying amounts of a highly enriched uranyl nitrate solution in an acrylic tank (25.0825-cm ID, 53.34-cm height) to obtain the spectral ratio as a function of the solution height in the tank.[4] A 0.635-cm stainless steel plate was located at the bottom of the solution tank. A ^{252}Cf source was placed at the axial center of the tank at the mid-height of the solution. ^{3}He neutron detectors (5.08-cm OD, 38.1-cm long) were placed adjacent to the radial surface of the tank and were separated 180° apart. The aqueous uranyl nitrate solution contained 0.29330 gram of uranium per gram of solution. The solution density was 1.64320 g/cm^3 and had a free acid content <0.1 wt% HNO_3. The uranium isotopic content was 93.2 wt% ^{235}U, 5.37 wt% ^{238}U, 1.02 wt% ^{234}U, and 0.41 wt% ^{236}U.

The evaluated spectral ratio[5] values for these measurements are provided in **Table 1**. These evaluated values were obtained from detailed sensitivity study of the various uncertainties associated with the geometry and the materials. Results of MCNP-DSP computations of the spectral ratio values for these experiments are provided in **Table 2**, while the results of the k_{eff} computations are provided in **Table 3**. The results of the simulations indicate that the ENDF/B-V and the ENDF/B-VI.R2 cross sections produce similar results for the computed spectral ratio values and the computed k_{eff} values. The results of the simulations with JENDL-3.2 cross sections are in better agreement with the measured values.

Table 1 Measured spectral ratio values for uranyl nitrate solution tank.

Solution Height (cm)	Evaluated Spectral Ratio (10^{-3})
30.48	98.0 ± 12.0
27.94	139.0 ± 9.7
25.40	187.7 ± 10.6
22.86	248.0 ± 9.7
20.32	303.5 ± 9.7

Table 2 Computed spectral ratio values for uranyl nitrate solution tank.

Sol. Height (cm)	ENDF/B-V (10^{-3})	ENDF/B-VI. R2 (10^{-3})	JENDL-3.2 (10^{-3})
30.48	119.4 ± 0.1	120.0 ± 0.1	102.0 ± 0.1
27.94	163.7 ± 0.1	164.9 ± 0.2	149.1 ± 0.1
25.40	213.3 ± 0.1	218.0 ± 0.1	200.0 ± 0.1
22.86	268.3 ± 0.2	274.4 ± 0.2	256.1 ± 0.3
20.32	334.2 ± 0.4	339.4 ± 0.6	324.8 ± 0.5

Table 3 Computed k_{eff} values for uranyl nitrate solution tank.

Sol. Height (cm)	ENDF/B-V	ENDF/B-VI. R2	JENDL-3.2
30.48	0.9595 ± 0.0003	0.9590 ± 0.0003	0.9666 ± 0.0003
27.94	0.9392 ± 0.0003	0.9385 ± 0.0003	0.9463 ± 0.0003
25.40	0.9140 ± 0.0003	0.9129 ± 0.0003	0.9200 ± 0.0003
22.86	0.8835 ± 0.0003	0.8824 ± 0.0003	0.8900 ± 0.0003
20.32	0.8453 ± 0.0003	0.8437 ± 0.0003	0.8505 ± 0.0003

The differences of the results among the three libraries can be attributed to the differences in the fission, capture, and average number of neutrons per fission obtained using these libraries. The fission rate, capture rate, and average number of neutrons from fission for the three libraries for the 30.48-cm high solution are provided in **Table 4**. The fission rates for the ENDF/B-V and ENDF/B-VI cross sections are very similar, whereas the fission rate for the JENDL-3.2 data is slightly higher than the other libraries. Furthermore, the capture rate obtained using the JENDL-3.2 data is lower than that obtained with either the ENDF/B-V or ENDF/B-VI data. Therefore, the leakage of neutrons from uranyl nitrate solution computed with the JENDL-3.2 data must be greater than that for the ENDF/B-VI data to obtain a k_{eff} value that is only slightly greater than that obtained with the ENDF/B-VI data. However, the computed spectral ratio obtained using the JENDL-3.2 data is significantly different than that obtained using the ENDF/B-VI data. This indicates that the computation of the spectral ratio is much more sensitive to the slowing down of neutrons in the system than the computation of k_{eff}.

Table 4 Computed fission rate, capture rate, and average number of neutrons from fission for 30.48-cm high uranyl nitrate solution.

Quantity	ENDF/B-V	ENDF/B-VI. R2	JENDL-3.2
Fission	0.393	0.393	0.396
Capture	0.130	0.127	0.125
ν	2.426	2.422	2.423

2. Uranium Metal Cylinders

Measurements were also performed with 17.771-cm-OD uranium metal cylinders with different heights.[6)] The ^{252}Cf source was located at the center on the top surface of the cylinders while the ^{6}Li-glass detectors (3.81-cm OD, 2.54-cm long) were placed adjacent to the radial surface of the cylinder and were separated 180° apart. The uranium metal cylinders had a density of 18.76 g/cm^3, and the isotopic content was 93.15 wt% ^{235}U, 5.64 wt% ^{238}U, 0.97 wt% ^{234}U, and 0.24 wt% ^{236}U.

The measured spectral ratio values for these measurements are provided in **Table 5**. A detailed sensitivity study was not performed for these experiments because many of the uncertainties have not been documented. Therefore, the uncertainties in the measured spectral ratio values are only statistical uncertainties and do not represent the true uncertainties. Results of MCNP-DSP computations of the spectra ratio values for these experiments are provided in **Table 6** while the results of the k_{eff} computations are provided in **Table 7**.

The computed spectral ratio values and the computed k_{eff} values obtained from the ENDF/B-V and the ENDF/B-VI.R2 data are similar. The results of the simulations with JENDL-3.2 cross sections are in better agreement with the measured values for the 10.16-cm and 9.14-cm high cylinders. For the 10.16-cm high cylinder, the computed spectral ratio obtained using the JENDL-3.2 data is approximately 4.5% lower than that obtained using the ENDF/B-VI.R2 data whereas the computed k_{eff} values differ by only 0.5%. The increased sensitivity of the computed spectral ratio values is obvious for this example. The results of the simulations for the 9.14-cm high cylinder show a similar trend as the 10.16-cm high cylinder. The computed spectral ratio obtained using all cross section data is statistically the same for the 8.13-cm high cylinder whereas the computed k_{eff} values vary by approximately 0.5%. The computed spectral ratio values for the 7.11-cm high cylinder obtained using the JENDL-3.2 is approximately 0.6% lower than that from the ENDF/B-VI.R2 data and is consistent with the difference in the computed k_{eff} values.

Table 5 Measured spectral ratio values for uranium metal cylinders.

Cylinder Height (cm)	Evaluated Spectral Ratio (10^{-3})
10.16	69.4 ± 0.8
9.14	99.5 ± 1.3
8.13	150.0 ± 3.0
7.11	215.0 ± 6.0

Table 6 Computed spectral ratio values for uranium metal cylinders.

Cyl. Height (cm)	ENDF/B-V (10^{-3})	ENDF/B-VI.R2 (10^{-3})	JENDL-3.2 (10^{-3})
10.16	75.8 ± 0.1	76.3 ± 0.1	72.9 ± 0.1
9.14	119.4 ± 0.2	115.5± 0.2	111.5 ± 0.2
8.13	165.6 ± 0.5	165.2 ± 0.5	166.0 ± 0.3
7.11	230.3 ± 0.8	238.3 ± 0.9	236.7 ± 0.3

The small differences in the computed spectral ratio and k_{eff} values for the 10.16-cm and 9.14-cm high cylinders may be attributed to slowing down of neutrons in the cylinders. The computed fission rates obtained from the three libraries are in close agreement. The computed fission and capture rates and the average number of neutrons from fission for the 10.16-cm high cylinder are presented in **Table 8**. The values for the fission rate and the average number of neutrons from fission are in relatively close agreement; therefore, the differences must be attributed to slight differences in neutron capture and the slowing down of neutrons in the cylinders.

Table 7 Computed k_{eff} values for uranium metal cylinders.

Cyl. Height (cm)	ENDF/B-V	ENDF/B-VI.R2	JENDL-3.2
10.16	0.9226 ± 0.0003	0.9205 ± 0.0003	0.9253 ± 0.0003
9.14	0.8832 ± 0.0003	0.8807 ± 0.0003	0.8856 ± 0.0003
8.13	0.8378 ± 0.0003	0.8350 ± 0.0003	0.8405 ± 0.0003
7.11	0.7854 ± 0.0003	0.7825 ± 0.0003	0.7873 ± 0.0003

Table 8 Computed fission rate, capture rate, and average number of neutrons from fission for 10.16-cm high uranium metal cylinder.

Quantity	ENDF/B-V	ENDF/B-VI. R2	JENDL-3.2
Fission (10^{-3})	2.92	2.93	2.92
Capture (10^{-4})	3.18	3.34	3.32
ν	2.595	2.586	2.597

III. Conclusions

Computations of subcritical measurement quantities offer an alternate method to investigate differences among nuclear data evaluations. The sensitivity of subcritical measurement quantities to nuclear data in some instances is greater than the sensitivity of computed k_{eff} values to nuclear data. The computation of the spectral ratio from the source-driven noise analysis measurement was shown to be more sensitive to nuclear data evaluations than the computation of k_{eff} for uranyl nitrate solution systems and for some cylindrical uranium metal cylinder systems. The computation of the spectral ratio was shown to be more sensitive to the slowing down of neutrons in the system given the fact that the computed fission rates were very similar for each nuclear data set. Furthermore, for this limited set of subcritical measurements, the computed spectral ratio obtained from the JENDL-3.2 data agreed closer with the measured values than the ENDF/B-V or ENDF/B-VI data.

Acknowledgment

Based on work performed at Oak Ridge National Laboratory, operated for the U.S. Department of Energy under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed by UT-Battelle, LLC.

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