Source-Driven Noise Analysis Measurements with Neptunium Metal Reflected by High Enriched Uranium

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Subcritical noise analysis measurements have been performed with neptunium (^{237}Np) sphere reflected by highly enriched uranium. These measurements were performed at the Los Alamos Critical Experiment Facility in December 2002 to provide an estimate of the subcriticality of ^{237}Np reflected by various amounts of high-enriched uranium. This paper provides a description of the measurements and presents some preliminary results of the analysis of the measurements. The measured and calculated spectral ratios differ by 15% whereas the "interpreted" and calculated k_{eff} values differ by approximately 1%.

KEYWORDS: Neptunium, highly enriched uranium, subcritical noise measurements

1. Introduction

Subcritical source-driven noise analysis measurements[1] have been performed with a ^{237}Np sphere reflected with high-enriched uranium to provide benchmark data for nuclear criticality safety applications at the Los Alamos Critical Experiments Facility in December 2002. The data from these measurements can be used to determine the subcritical neutron multiplication factor for each configuration. This paper provides a description of the measurements, a discussion of the results of the measurements, and a description of the analysis of the measurements.

2. Description and Results of the Measurements

2.1 Description of the Measurements

The 8.30-cm OD, 237Np sphere had a mass of 6.09 kg and was encased in tungsten (0.259 cm thick) and nickel (0.381 cm thick) casings. The ^{237}Np sphere (98.8 wt% ^{237}Np) was reflected by high-enriched uranium (HEU). The HEU consisted of hemispherical shells of varying thickness. Measurements were performed with 29.3, 34.3, and 39.8 kg of HEU reflector, respectively, by varying the number of shells that surround the ^{237}Np sphere. The Np-HEU spherical assembly was supported on an aluminum stand as shown in Figure 1.

The Np-HEU spherical assembly was placed between two detector arrays that were 180° apart. The detector arrays consisted of two ^{3}He neutron detectors (4 atmosphere pressure) inside a high density polyethylene moderator. The detectors had a 2.54 cm OD and a 12.7 cm active length with a shell comprised of stainless steel. The detectors were configured such that detectors 2 and 3 were located opposite detectors 4 and 5. Detectors 3 and 4 were located nearer to the ^{252}Cf source. The polyethylene moderator was 10.16 cm wide, 12.7 cm thick, and 25.4 cm long. The polyethylene detector moderator blocks were supported on a 1.59-cm thick, 12.7 cm wide, and 15.24-cm long aluminum plate. A ^{252}Cf source ionization chamber was placed on the radial surface of the Np-HEU spherical as shown in Figure 1. The source ionization chamber was placed at the center of the sphere. The source kept in contact with the sphere for all measurements.

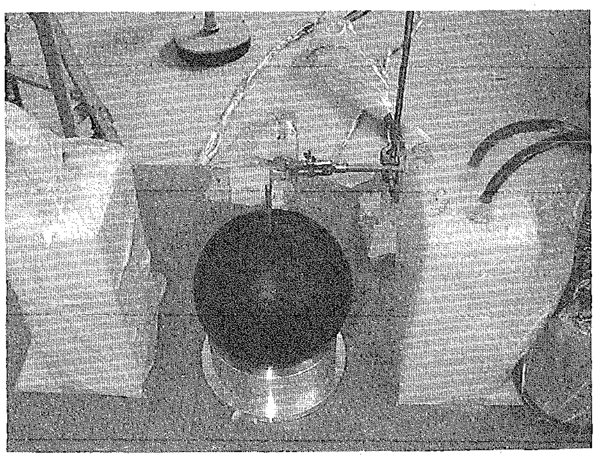

Fig.1 The ^{237}Np sphere reflected by HEU.

An effort was made to center the Np-HEU spherical assembly between the two detector moderators. However, the sphere was not exactly centered for each measurement because the inner surface of the aluminum stand did not make contact with the outer surface of the Np-HEU spherical assembly.

2.2 Results of the Measurements

A certain ratio of frequency spectra is commonly used to determine the subcritical neutron multiplication factor from the measured data. This spectral ratio is independent of detection efficiency and only depends on the efficiency for detecting the

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spontaneous fission in the source ionization chamber. The spectral ratio of interest is defined as

$$R_{ij}(\omega) = \frac{S^{*}_{1i}(\omega)S_{1j}(\omega)}{S_{11}(\omega)S_{ij}(\omega)}. \quad (1)$$

Although the spectral ratio is obtained from the frequency-dependent auto and cross spectra, the spectral ratio is typically constant over the low-frequency range. The spectral ratio values are computed from the measured cross spectra. The source auto spectrum S_{11} is simply a measure of the fission source strength. The detector auto spectra S_{22}, S_{33}, etc. are a measure of the source-induced and inherent fission rate of the system and the background rate and are proportional to the detector count rate. The source-detector cross-spectra S_{12}, S_{13}, S_{14} etc. are a measure of counting events in the detectors correlated with fission events in the source. Consequently, the source-detector cross-spectrum indicates the amount of source-induced fission occurring in the system. The detector cross-spectra S_{23}, S_{24}, S_{25}, S_{34}, etc. are a measure of the events in one detector correlated with events in another detector, so their magnitudes indicate the amount of both source-induced and inherent fission occurring in the system analyzed. A detailed description of this spectral ratio can be found in references 1 and 2. A spectral ratio can be obtained from each pair of detectors. For these measurements, six spectral ratio values are obtained. Due to the location of the source relative to the detectors, the spectral ratio values differ from one detector pair to another. A comparison of the six spectral ratio values for each configuration is provided in Table 1. These spectral ratio values are from repeated measurements performed for each configuration. The uncertainty represents the ability to reconfigure the source and detectors for each measurement and is the standard deviation of the population of the repeat measurements.

Table 1 Average low-frequency value of the measured spectral ratio values as a function of HEU reflector mass

HEU Mass (kg)	R_{23}	R_{24}	R_{25}	R_{34}	R_{35}	R_{45}
39.8	0.0705 ± 0.0028	0.0710 ± 0.0028	0.0640 ± 0.0026	0.0821 ± 0.0033	0.0726 ± 0.0029	0.0645 ± 0.0026
34.3	0.0963 ± 0.0029	0.1062 ± 0.0032	0.0929 ± 0.0028	0.1202 ± 0.0036	0.1068 ± 0.0032	0.0955 ± 0.0029
29.3	0.1145 ± 0.0033	0.1396 ± 0.0040	0.1223 ± 0.0035	0.1609 ± 0.0047	0.1395 ± 0.0040	0.1371 ± 0.0040

The spectral ratio values depend linearly on fissile mass. The spectral ratio values for detectors located on opposite sides of the Np-HEU spherical assembly are less sensitive to the location of the assembly between the detectors whereas the spectral ratio values for detectors that are adjacent would be more sensitive to the location of the assembly. The spectral ratio values for detector combinations 2 and 4 and 3 and 5 should be similar, i.e. R_{24} and R_{35} should be similar in value. As can be seen from the results in Table 1, spectral ratio values R_{24} and R_{35} are very similar as expected. The spectral ratio R_{34} is more affected by direct neutrons from the ^{252}Cf source than the other spectral ratio values whereas the spectral ratio R_{25} is influenced the least from neutrons directly from the ^{252}Cf source.

3. Analysis of the Measurements

3.1 Interpretation of the Measurements

The Monte Carlo code MCNP-DSP[3] was used to interpret the measurements. The frequency dependent spectra, the coherence functions, and the spectral ratio are computed directly using MCNP-DSP. The Monte Carlo codes are also used to interpret the measurement by performing a calculation of the measured quantities and a separate eigenvalue calculation. For example, a comparison of measured and calculated values of the spectral ratio can be used to obtain the "experimental" k_{eff}. If the measured and calculated values of the spectral ratio are in agreement, then the bias in the spectral ratio is zero, and the "experimental" k_{eff} value is equivalent to the calculated value. The bias in the spectral ratio is defined as the difference between measured and calculated values of the spectral ratio (R_m-R_c) where R_m is the measured value and R_c is the calculated value. However, if there is a bias in the calculated spectral ratio, first order perturbation theory can be used to obtain an expression that can be used to determine the "experimental" k_{eff} and the bias in the k_{eff}. The low-frequency value of the spectral ratio has been shown to be linear with k_{eff} over a small range of k_{eff} values. Given the linear dependence of the spectral ratio with a small change in k_{eff}, the bias in the spectral ratio varies linearly as the bias in k_{eff} (k_m-k_c). To determine the "experimental" k_{eff} value and its bias, the Monte Carlo models are slightly perturbed and new values of the spectral ratio (R_p) and k_{eff} (k_p) are obtained. If the linear dependence is valid, then the perturbation calculations can be used to obtain the "experimental" k_{eff} and its bias using the following linear relationship

$$\frac{R_m - R_c}{k_m - k_c} = \frac{R_p - R_c}{k_p - k_c}. \quad (2)$$

This methodology simply uses a linear interpolation or extrapolation between the standard and perturbed values of the spectral ratio and k_{eff} to determine the "experimental" k_{eff}. As previously stated, only small perturbations can be made to ensure that the ratio varies linearly with k_{eff}. Using this relationship, the value of k_m can be determined along with its bias k_m-k_c. Perez et. al.[4] has demonstrated that the relationship given in Eq 2 is valid using first order perturbations of the Boltzmann transport operators for k_{eff} and the spectral ratio. In this paper, Perez summarizes the relationships derived for k_{eff} and the spectral ratio in terms of the transport operator. In this regard, it is demonstrated that using first-order perturbation theory that the relationships between the standard calculated values of the spectral ratio or k_{eff} can be related to the perturbed values. The results of the perturbation analyses directly yield Eq2.

3.2 Results of MCNP-DSP Calculations

MCNP-DSP calculations were performed for all three experimental configurations using the ENDF/B-VI nuclear cross section data. The computed spectral ratio values are provided in Table 2. The calculated k_{eff} values for each configuration are provided in Table 3.

Table 2 Average low-frequency value of the calculated spectral ratio values as a function of HEU reflector mass using ENDF/B-VI cross section data

HEU Mass (kg)	R_{23}	R_{24}	R_{25}	R_{34}	R_{35}	R_{45}
39.8	0.0769 ± 0.0002	0.0831 ± 0.0001	0.0737 ± 0.0001	0.0949 ± 0.0002	0.0844 ± 0.0001	0.0771 ± 0.0000
34.3	0.1081 ± 0.0002	0.1203 ± 0.0003	0.1044 ± 0.0002	0.1379 ± 0.0004	0.1196 ± 0.0002	0.1055 ± 0.0002
29.3	0.1420 ± 0.0003	0.1610 ± 0.0005	0.1414 ± 0.0004	0.1934 ± 0.0003	0.1646 ± 0.0009	0.1467 ± 0.0002

Table 3 Calculated k_{eff} values as a function of HEU reflector mass using ENDF/B-VI nuclear cross section data

HEU Mass (kg)	k_{eff}
39.8	0.88470 ± 0.00005
34.3	0.85255 ± 0.00005
29.3	0.82062 ± 0.00005

The spectral ratio values for detector pairs located on opposite sides of the Np-HEU spherical assembly are consistent with each other and can be used to determine the experimental k_{eff} value for each HEU reflector mass. The calculated spectral ratio values are approximately 15% higher than the measured spectral ratio values for each reflector thickness. The difference between the measured and calculated spectral ratio values is most likely due to inadequacies in the ^{237}Np nuclear cross section data.

A perturbation analysis was performed to determine the subcritical neutron multiplication factor for each configuration. The perturbations included increasing and decreasing the ^{237}Np density, increasing and decreasing the HEU density, and increasing and decreasing both the ^{237}Np density and the HEU density. These various perturbations were made to determine the perturbed spectral ratio values, R_p, and the perturbed k_{eff} values, k_p. These perturbations were then used in the perturbation formula to determine the experimental k_{eff} value for the Np-HEU spherical assemblies.

The spectral ratio values R_{24}, R_{25}, R_{34}, and R_{35} were used to determine the "experimental" subcritical neutron multiplication factor. The spectral ratio values obtained by increasing the Np density 2% are provided in Table 4. The calculated k_{eff} value, k_p, for a 2% increase is 0.88853 ± 0.00005.

Table 4 Average low-frequency value of the calculated spectral ratio values as a function of HEU reflector mass using ENDF/B-VI cross section data for a 2% increase in the Np density

HEU Mass (kg)	R_{23} (R_p)	R_{24} (R_p)	R_{25} (R_p)	R_{34} (R_p)	R_{35} (R_p)	R_{45} (R_p)
39.8	0.0737 ± 0.0001	0.0798 ± 0.0001	0.0717 ± 0.0001	0.0897 ± 0.0001	0.0798 ± 0.0001	0.0741 ± 0.0001
34.3	0.1033 ± 0.0002	0.1148 ± 0.0001	0.1007 ± 0.0002	0.1317 ± 0.0004	0.1165 ± 0.0005	0.1045 ± 0.0003
29.3	0.1380 ± 0.0003	0.1556 ± 0.0003	0.1350 ± 0.0004	0.1828 ± 0.0005	0.1607 ± 0.0005	0.1386 ± 0.0002

The "experimental" k_{eff} value is determined after performing all of the previous mentioned perturbations as the variance weighted k_{eff} for each perturbation and for the applicable spectral ratio values (R_{24}, R_{25}, R_{34}, and R_{35}). The variance weighted k_{eff} values are presented in Table 5. The uncertainty in k_{eff} is the standard deviation of the population of estimates of k_{eff} from each perturbation analysis. This uncertainty does not include the uncertainty associated with the systematic uncertainties in the composition, densities, and geometry of the Np-HEU assembly.

Table 5 "Experimental" k_{eff} values as a function of HEU reflector mass using ENDF/B-VI nuclear cross section data and all perturbations

HEU Mass (kg)	"Experimental" k_{eff}
39.8	0.8952 ± 0.0028
34.3	0.8626 ± 0.0033
29.3	0.8323 ± 0.0032

The spectral ratio values R_{23} and R_{45} are very sensitive to the orientation of the Np-HEU spherical assembly between the two detector arrays. Therefore, these spectral ratio values cannot be used in the analysis of the measurements since the exact position of the Np-HEU spherical assembly between the detector arrays was not controlled.

4. Conclusion

These measurements demonstrate that fairly precise estimates of the subcritical neutron multiplication factor can be obtained from the source-driven noise analysis measurement method using the Monte Carlo code MCNP-DSP. These measurements also demonstrate the high sensitivity of the source-driven noise measurements. The calculated spectral ratio values differ from the measurements by approximately

15%; whereas, the calculated k_{eff} values differed from the interpreted k_{eff} values by only 1%. The results of the measurements also indicate that the nuclear data for Np may be uncertain. However, a complete analysis of the sensitivity of the analysis to the Np nuclear data has not been performed. Furthermore, the measured configurations contain large quantities of highly enriched uranium that make it difficult to ascertain the overall sensitivity of the measurements to the Np nuclear data. The results of these preliminary measurements are encouraging and suggest that additional measurements should be performed with less highly enriched uranium to learn more about the sensitivity of the analysis of the measurements to the Np nuclear data.

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