SUBCRITICAL MEASUREMENTS AND THEIR APPLICATIONS

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Subcritical measurements have typically been used at critical experiment facilities as a means to monitor the approach to the critical condition. However, subcritical measurement methods have many more applications than simply to serve as indicators for the approach to critical. One of the more fundamental uses involves using them as benchmark measurements. Monte Carlo codes have been developed that can directly simulate a variety of subcritical measurements in the same manner in which the measurements are performed. Therefore, subcritical measurements can be used to validate Monte Carlo codes and nuclear data through direct calculation of subcritical quantities. Subcritical measurement methods can also be used to determine the subcriticality of fissile materials in-situ. This requires measurements to be performed and subsequent analyses of the measurements to determine the subcritical k_{eff} value. Subcritical measurement methods can also be used to monitor the reactivity of a fissile operation. The purpose of reactivity monitoring would not be to provide a highly precise k_{eff} value for the fissile operation but to provide an indication of a change in reactivity and when this change exceeds a specified threshold. This paper addresses the areas of subcritical benchmarking, in-situ subcritical measurements, and subcritical monitoring. Descriptions of the various applications are provided along with the issues and challenges in applying subcritical measurement methods. Finally, recommendations for resolving the issues and challenges for implementation of subcritical measurements are discussed.

Keywords: subcritical, benchmarking, in-situ, monitoring, noise analysis

1. INTRODUCTION

Subcritical measurements methods have many more applications than simply serving as indicators of the approach to critical. Subcritical measurements can serve multiple functions that include using them as benchmark measurements, as a means to determine the subcriticality of fissile materials in-situ, and as a method to monitor the reactivity for fissile material operations. These applications are related to each other but require slightly different approaches to implement the measurements and different levels of knowledge about the system being measured are needed.

The source-driven noise analysis measurement[1] is one subcritical measurement method that has several advantages as compared to other subcritical measurement techniques and has been applied to benchmark measurements and for the determination of the subcriticality of fissile configurations. Measurements have also been performed to investigate the use of this method as a reactivity indicating measurement.

This paper provides a brief description of the subcritical source-driven noise analysis measurement in Section 2. A description of how this method is used to benchmark Monte Carlo codes and nuclear data is provided in Section 3. Application of this measurement for in-situ reactivity measurements is provided in Section 4, while the use of this method as a reactivity monitor is provided in Section 5. Some concluding remarks are provided in Section 6 that include recommendations for extensions of subcritical measurement methods.

2. SOURCE-DRIVEN NOISE ANALYSIS MEASUREMENTS

The subcritical source-driven noise measurement[1] (known as the ^{252}Cf-source-driven noise analysis method) evolved from a simultaneous randomly pulsed neutron measurement and the Rossi-α measurement as an attempt to overcome some of the limitations of other subcritical measurement methods. Other subcritical measurement methods require a calibration measurement at delayed critical or knowledge of the effective delayed neutron

fraction to determine the neutron multiplication factor, k_{eff}. Additionally, many of these methods required some knowledge of the detection efficiency or source intensity to determine k_{eff}. A certain spectral ratio from the source-driven noise measurement can be directly related to the subcritical reactivity without need for a calibration measurement at delayed critical.

The source-driven noise measurement is based on simultaneous performance of the randomly pulsed neutron and Rossi-α measurements. The source-driven noise measurement differs from other subcritical measurements in that the data acquisition is actually performed using Fourier processors. The measurement involves the use of two or more detectors and a timed neutron source. The correlation of the various time dependent signatures is performed in the frequency domain over a time period specified by the data acquisition software. The frequency dependent signatures that are acquired include auto spectra and cross spectra. The simplistic point reactor kinetics models can be used to describe the time-evolution of neutrons in a fissile system and to determine the dependence of the various auto and cross spectra on the properties of the fissile system. The source-detector cross spectra $(S_{SX}(\omega))^2$ can be defined as

$$S_{SX}(s) = \frac{\varepsilon_S \varepsilon_1 \overline{\nu}_0 F_S}{\overline{\nu}\Lambda} \frac{1}{(j\omega + \alpha)}. \tag{1}$$

In this expression, s is the frequency variable, α is the prompt neutron decay constant, F_s is the average fission rate of the source, ε_s is the efficiency for detecting the source event, ε_1 is the detector efficiency in counts per fission, $\overline{\nu_0}$ is the average number of neutrons from the source, $\overline{\nu}$ is the average number of neutrons from fission, and Λ is the neutron generation time. The detector-detector cross spectrum $(S_{xy}(\omega))^2$ is

$$S_{xy}(\omega) = \frac{\varepsilon_1 \varepsilon_2}{(\overline{\nu}\Lambda)^2} \left[\overline{\nu(\nu-1)} \frac{\overline{\nu_0} F_S}{\alpha\Lambda} + \overline{\nu_0(\nu_0 - 1)} F_S + \overline{\nu(\nu-1)} \frac{\overline{\nu_I} F_I}{\alpha\Lambda} + \overline{\nu_I(\nu_I - 1)} F_I \right] \bullet \frac{1}{(\alpha^2 + \omega^2)}. \tag{2}$$

In this expression, ε_1 is the detector efficiency in counts per fission for the first detector, ε_2 is the detector efficiency in counts per fission for the second detector, $\overline{\nu_0}$ is the average number of neutrons from the source, $\overline{\nu}$ is the average number of neutron from fission, $\overline{\nu_I}$ is the average number of neutrons from an inherent source, $\overline{\nu(\nu-1)}$ is the reduced second moment of the neutron emission distribution for fission, $\overline{\nu_0(\nu_0 - 1)}$ is the reduced second moment of the neutron emission distribution for ^{252}Cf spontaneous fission, $\overline{\nu_I(\nu_I - 1)}$ is the reduced second moment of the neutron emission distribution for the inherent source, and F_I is the spontaneous fission rate of the inherent source. The detector-detector cross spectrum defined by Eq. 2 is the frequency domain equivalent of the two-detector Rossi-α measurement. The detector auto spectrum $(S_{xx}(\omega))^2$ is given by

$$S_{xx}(\omega) = \frac{\varepsilon_1 \varepsilon_1}{(\bar{\nu}\Lambda)^2}\left[\overline{\nu(\nu-1)}\frac{\overline{\nu_0}F_s}{\alpha\Lambda} + \overline{\nu_0(\nu_0-1)}F_s + \overline{\nu(\nu-1)}\frac{\overline{\nu_I}F_I}{\alpha\Lambda} + \overline{\nu_I(\nu_I-1)}F_I\right] \bullet \frac{1}{(\alpha^2+\omega^2)} + \varepsilon_1\left[\frac{\overline{\nu_0}F_s}{\alpha\Lambda} + \frac{\overline{\nu_I}F_I}{\alpha\Lambda}\right]. \quad (3)$$

The most significant advantage of the source-driven noise measurement is that a certain ratio of frequency spectra can be obtained that does not depend on detection efficiency. The detection efficiency is stated in terms of counts per fission and is not directly measurable. The spectral ratio of interest is defined as[2]

$$R(\omega) = \frac{S^*_{sx}(\omega)S_{sy}(\omega)}{S_{ss}(\omega)S_{xy}(\omega)}. \quad (4)$$

The term S_{sx} is the source-detector cross spectrum between the source s and the detector x. The asterisk denotes taking the complex conjugate of the source-detector cross spectrum, S_{sx}. The term S_{sy} is the source-detector cross spectrum between the source s and the detector y. The term S_{xy} is the detector-detector cross spectrum between detectors x and y. The term S_{ss} is the source auto spectrum and is simply equal to the source efficiency times the source fission rate ($S_{ss}=\varepsilon_s F_s$). The spectral ratio that includes an inherent source is of particular interest for interpreting measurements for spent nuclear fuel applications or for systems with significant amounts of plutonium. The spectral ratio including an inherent spontaneous fission source is expressed as

$$R(\omega) = \frac{\varepsilon_s \overline{\nu_0}}{\left[\frac{\overline{\nu(\nu-1)}}{|\rho|} + \frac{\overline{\nu_0(\nu_0-1)}}{\overline{\nu_0}} + \frac{\overline{\nu(\nu-1)}}{|\rho|}\frac{F_I}{F_s} + \frac{\overline{\nu_I(\nu_I-1)}}{\overline{\nu_I}}\frac{F_I}{F_s}\right]}. \quad (5)$$

As can be seen from Eq. 5, the spectral ratio that includes the effects of an inherent spontaneous fission source depends on the ratio of the inherent spontaneous fission rate to the ^{252}Cf source fission rate. These expressions are similar to those derived by others except that spatial correction factors and importance functions have been omitted in these equations. This dependence can be included in the appropriately weighted cross sections and neutron emission probabilities. The actual use of such expressions to interpret the measurements is quite limited and a more general approach using the Monte Carlo method has been developed that does not have the limitations of the point reactor kinetics models.[2] The expressions are only provided to provide an indication of how the various frequency spectra relate to the reactivity of a fissile system.

3. BENCHMARKING MONTE CARLO CODES

The use of subcritical measurements as a means to validate Monte Carlo codes and nuclear data substantially increases the reactivity and neutron spectrum regimes over which the codes and data are validated. In fact, subcritical benchmark measurements are the key to determining whether the code bias; i.e. the difference between measured and calculated reactivity indices, is a function of the degree of subcriticality. Subcritical measurements can be used to validate Monte Carlo codes and nuclear data through direct calculation of the measured quantities. Alternatively, the Monte Carlo codes and nuclear data can be validated from the k_{eff} value determined from the measurements. Interpretation of measurements

requires a model to relate what is measured to the neutron multiplication factor. In the past, the subcritical measurements were interpreted using equations developed from point kinetics models for the time-dependent behavior of neutrons in the subcritical configuration. This limited the application of the measurement to situations in which point kinetics was applicable. The most general model to relate the measured quantities to k_{eff} would involve the use of the generalized stochastic model developed by Munoz-Cobos et al.[3] Although an analytical solution of subcritical measurements in terms of the stochastic model is not practical, the Monte Carlo method provides a means to simulate the subcritical measurements and to also calculate the neutron multiplication factor. In fact, the same Monte Carlo code and nuclear data can be used for both calculations.

The Monte Carlo code MCNP-DSP[4] was developed to simulate a variety of subcritical measurements. MCNP-DSP can be used to directly calculate the frequency dependent spectra from the source-driven noise analysis measurement. Furthermore, MCNP-DSP can be used to determine k_{eff} using a perturbation method. The Monte Carlo codes are used to interpret the measurement by performing a calculation of the measured parameters 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. 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. 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 in numerous experiments to be linear with k_{eff} over a wide range of values of k_{eff} with the values of k_{eff} being interpreted using point kinetics models. Given the linear dependence of the spectral ratio with 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:

$$\frac{R_m - R_c}{k_m - k_c} = \frac{R_p - R_c}{k_p - k_c}. \tag{6}$$

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}. Using this relationship, the value of k_m can be determined along with its bias k_m-k_c. Propagation of error is used to obtain the uncertainty in k_m and its bias. Even if the measured spectral ratio value and the calculated value agree, the perturbation analysis is performed. This is required to equate the uncertainty in the measured spectral ratio to the uncertainty in the inferred k_{eff} value.

This methodology has been successfully applied to benchmark source-driven noise measurements with uranyl nitrate solutions. The analysis of these measurements have been documented in the Handbook of the International Criticality Safety Benchmark Evaluation Project.[5] In the analysis of the uranyl nitrate solution experiments, a variety of perturbations were used to interpret k_{eff} and a variety of nuclear data sets used in the interpretations. The results demonstrated that the interpreted k_{eff} was independent of the perturbation method and had a very slight dependence on the nuclear data set. The interpreted (experimental) k_{eff} values for each different cross section data set are presented in Table I. As can be seen in this table, the interpreted k_{eff} values differ slightly depending on which cross section data set was used.

A general trend is observed that the interpreted k_{eff} values obtained using the ENDF/B-V cross section data are consistently higher than the interpreted values obtained from the ENDF/B-VI and JENDL-3.2 cross section data sets. The largest difference between the interpreted k_{eff} values among the results produced using the three different cross section data sets is approximately 0.003 Δk.

Table I. Interpreted experimental k_{eff} values for ENDF/B-V, ENDF/B-VI, and JENDL-3.2 cross section data sets for the high-enriched uranyl nitrate solution experiments.

Solution Height (cm)	ENDF/B-V "Experimental" k_{eff}	ENDF/B-VI "Experimental" k_{eff}	JENDL-3.2 "Experimental" k_{eff}
30.48	0.9631 ± 0.0052	0.9602 ± 0.0052	0.9610 ± 0.0050
27.94	0.9448 ± 0.0045	0.9430 ± 0.0045	0.9432 ± 0.0041
25.40	0.9229 ± 0.0052	0.9215 ± 0.0050	0.9204 ± 0.0056
22.86	0.8889 ± 0.0051	0.8888 ± 0.0048	0.8883 ± 0.0055
20.32	0.8582 ± 0.0055	0.8580 ± 0.0061	0.8571 ± 0.0052
15.24	0.7464 ± 0.0167	0.7511 ± 0.0200	0.7507 ± 0.0234

4. IN-SITU SUBCRITICAL MEASUREMENTS

The second area of applicability of subcritical noise measurements is for in-situ determinations of the subcritical neutron multiplication factor. The two ways that this can be achieved is by post analysis of the measurements using the Monte Carlo method previously described or by using the limited point reactor kinetics models to relate the measured quantity to the subcritical neutron multiplication factor. The latter method should only be considered if an on-line determination of k_{eff} is desired. In such applications, care must be taken to properly select the location of the source and detectors such that the point kinetics models are applicable. Furthermore, calculations of spatial correction factors and importance ratios are also required and must be supplied to the point reactor formulas to determine k_{eff}. The use of point reactor kinetics is discouraged because uncertainties in correction factors contribute directly to the uncertainty in the reactivity estimates. The Monte Carlo method is the preferred way to interpret the measurements because of the limitations of the point reactor models and because correction factors do not have to be computed.

In-situ subcriticality measurements performed at the Oak Ridge Y-12 Plant in Vault 16 demonstrated that these types of measurements are practical[6]. In these measurements, the subcriticality of an array of high-enriched uranium metal castings inside a concrete storage vault was determined using the Monte Carlo method. Measurements were performed with the source and detectors placed inside the array. The source was located in the center of the central casting and the detectors were placed on the periphery of the array. Prior to performing the noise measurements, flux profile measurements were performed to determine an array size that would approximate the results expected if the array was infinite. The detectors were placed on the outside of the essentially infinite array. The noise measurements were performed over many weeks to obtain statistically converged spectra because of the low coherence values. The measurements were interpreted using MCNP-DSP simulations. These simulations were also performed over several weeks. The Monte Carlo models contained as much detail as possible about the storage vault configuration; however, the composition of the concrete storage vault was not known. The analyses were performed assuming that the concrete was dry or wet. The resulting k_{eff} value obtained from the analyses did not depend on the exact knowledge of the concrete composition. Although the concrete composition was not exactly known, the concrete vault was 25 years old and should have been comprised of dry

concrete. Coincidentally, the calculated spectral ratio value for the dry vault agreed within one standard deviation of the measured value. The Monte Carlo models included as much detail as possible concerning the storage vault geometry and composition. The storage trays were modeled in great detail, as were the uranium metal castings. The composition of the storage trays, detectors, and the castings were well known and included in the models. Although the interpreted k_{eff} value for the HEU storage vault does not depend on the composition of the concrete, all other details about the measurement are known. The success of the interpretation of the HEU storage vault measurements occurs because the spectral ratio depends only on the induced fission events from the ^{252}Cf source. A significant inherent neutron source does not exist in this configuration.

The MCNP-DSP computed k_{eff} values and spectral ratio values for these measurements are provided in Table II along with the interpreted k_{eff} values from the perturbation analysis. Perturbations were made to various components of the vault and the subsequent interpreted k_{eff} values did not depend significantly on the perturbation. In fact, the assumption that the concrete was wet is a rather large perturbation as compared to the assumption that the concrete is dry. Even though large perturbations are necessary for the wet concrete model, the interpreted k_{eff} values are statistically equivalent. This application of the perturbation methodology demonstrates the robustness of the model. The success in the interpretation is due in great part to the fact that detailed information is known about the major components of this system with only the concrete composition being the major unknown in the measurements. If the vault system had a significant inherent source, the interpretation would have been further complicated because the inherent source may not permit a unique one-to-one relationship between the spectral ratio and k_{eff}.

Table II. MCNP-DSP calculated spectral ratio and k_{eff} values and interpreted k_{eff} values for HEU storage vault in-situ measurements.

Concrete Condition	Calculated Ratio (R_c)	Calculated k_{eff} (k_c)	Perturbation	Interpreted k_{eff}
Dry	0.3894 ± 0.0027	0.7920 ± 0.0005	Hydrogen content	0.7888 ± 0.0068
			^{235}U enrichment	0.7902 ± 0.0041
			Casting length	0.7901 ± 0.0045
Wet	0.5004 ± 0.0151	0.7354 ± 0.0005	Casting inner radius	0.7798 ± 0.0054
			Reduced concrete density	0.7945 ± 0.0068

[a]Measured spectral ratio (R_m) is 0.395 ± 0.012.

5. SUBCRITICAL MONITORING

Another useful application of subcritical noise measurements is to provide a way to monitor fissile material processing operations. This application would not focus on determining the subcritical neutron multiplication but would use the measured quantities as a way to ascertain normal operation. The enhanced sensitivity of the subcritical noise measurement would allow fissile processing operations to be monitored and controlled using the changes in spectral signatures as indicators to changes in the system. The spectral ratio is an ideal candidate for process monitoring because it is independent of detection efficiency and would not change due to drifts in the electronic system. Measurements and Monte Carlo simulations of a process condition could be performed to provide bounding values for the spectral ratio. The measured spectral ratio values could be equated to k_{eff} values for various operating conditions. Changes in the spectral ratio value could be monitored and used to ensure that the reactivity of the system remains below some desired value. This would enhance safety in operations with fissile material by providing some means estimate the

subcriticality of the fissile process. If the system is simple enough and the geometry is appropriate, then the point reactor kinetics models could possibly be used to provide an on-line estimation of k_{eff}; however, great care would need to be taken to properly estimate the uncertainty on the k_{eff} value from the point reactor kinetics model to ensure that the limits on the fissile operation can be properly constrained.

The greatest challenge in applying subcritical measurements for process monitoring is to measure a quantity that does not depend on changes in the detection system such as detector efficiency or dead time, on changes in any inherent neutrons sources that are capable of inducing fission, and on source intensity. The spectral ratio from the source-driven noise analysis measurement satisfies two of these criteria. However, the spectral ratio as shown in the simplistic models in Eq. 5 is dependent on the inherent sources. If the contribution of the inherent source to the spectral ratio is significant, then care must be taken to provide an indication of the change in the inherent neutron sources in the process. To circumvent this limitation, a modified spectral ratio termed the bi-spectral ratio could be acquired. The bi-spectral ratio depends only on induced fission events caused by the ^{252}Cf source and is independent of the source detection efficiency has been developed that requires the measurement of higher-order correlation functions[7]. These higher-order correlation measurements have yet to be applied to subcriticality measurements, but they have been theoretically shown to possess higher sensitivity to reactivity and are independent of all inherent sources. The bi-spectral ratio is closely related to the standard spectral ratio; however, the bi-spectral ratio involves only events correlated with the timed neutron source. The summary provided in this section follows directly from the developments of Mattingly[7]. The bi-spectral ratio is defined as

$$\Psi_{XY|S} = \frac{S_{SX}(0)S_{SY}(0)}{\bar{s}S_{SXY}(0,0)} = \frac{\overline{\nu_0}}{\dfrac{\overline{\nu_0(\nu_0-1)}}{\bar{\nu}_0} + \dfrac{\overline{\nu(\nu-1)}}{\bar{\nu}|\rho|}}. \tag{7}$$

In this expression $\bar{s}$ is the average source fission rate. The bi-spectral ratio is essentially equivalent to the standard spectral ratio for systems but is independent of the source detection efficiency, the detector efficiency, or an inherent source. This measurement requires pulse neutron measurements to be performed with two detectors. In addition, the bi-spectral ratio requires the correlation of the response of two detectors with the neutron source. This measurement can be thought of as a dual pulse neutron measurement. This measurement is only dependent on fission chains initiated by the neutron source and hence would be more generally applicable to in-situ measurements with plutonium and irradiated nuclear fuels. For systems close to delayed critical, the standard spectral ratio will be dependent on the delayed neutrons that will behave as an inherent neutron source. However, the bi-spectral ratio will not be affected by delayed neutrons and hence would be more useful for systems close to delayed critical. In theory, the bi-spectral ratio can be measured up to prompt critical. An equivalent signature can be acquired in the time domain by performing a three-way coincidence measurement between a timed neutron source and two detectors while at the same time performing two two-way coincidence measurements between the source and each detector. A time dependent signature could be acquired by varying the gate of the coincidence period. A ratio of the two source-detector coincidence measurements to the three-way coincidence measurement times the source detection efficiency would produce a quantity similar to the bi-spectral ratio that should also be independent of inherent neutron sources.

6. SUMMARY

Subcritical measurements can serve multiple functions. The measurements are ideal for use as benchmark measurements because many subcritical measurements such as the Rossi-α, noise analysis, and Feynman variance can be simulated using Monte Carlo codes. Quantities from the source-driven noise analysis measurements have been shown to be more sensitive to nuclear data than direct calculation of k_{eff} thereby making the simulation of the source-driven noise analysis measurements more applicable for testing Monte Carlo methods and nuclear data. Subcritical measurements can also be used to interpret in-situ applications. In this regard, the measurements are applied to determine k_{eff}. A model that relates the measured quantity to k_{eff} is used to interpret the measurements and should be fairly robust. Source-driven noise measurements have been performed to determine the subcritical reactivity of high-enriched uranium metal in storage configurations. These measurements demonstrated that the k_{eff} value could be determined without exact knowledge of all constituents of the storage vault. However, the success of the use of measurements for in-situ verifications is highly dependent on whether or not an inherent neutron source is present. The presence of an inherent source complicates the interpretation of the measurements and may limit the application of the measurements. The use of subcritical measurements as reactivity monitors is also feasible. For process monitoring applications, the subcritical measurement quantities should be evaluated for the limits of the standard operating conditions to equate the expected value of the subcritical measurement quantity to k_{eff}. The spectral ratio from the source-driven noise analysis measurement is an ideal candidate for process monitoring because this quantity does not depend on the source strength or changes in the detection system. However, the spectral ratio is dependent on inherent neutron sources that may change the spectral ratio value although the reactivity state may not have changed. For systems with an inherent neutron source, a signature that is independent of the inherent source such as the bi-spectral ratio or a multi-way coincidence measurement must be performed if the addition of the inherent neutron source is outside of the standard operating conditions. A detailed model such as the Monte Carlo method must also be used to equate the subcritical measured quantity to k_{eff} if k_{eff} is to be the monitoring parameter.

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