

Instrumentation and Controls Division
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FEASIBILITY OF SUBCRITICALITY AND NDA MEASUREMENTS FOR SPENT
FUEL BY FREQUENCY ANALYSIS TECHNIQUES WITH ^{252}Cf

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ABSTRACT

The ^{252}Cf -source-driven frequency analysis method can be used for measuring the subcritical neutron multiplication factor of arrays of LWR fuel and as little as a single PWR fuel assembly. These measurements can be used to verify the criticality safety margins of spent LWR fuel configurations and thus could be a means of obtaining the information to justify burnup credit for spent LWR transportation/storage casks. In addition, the data provided by such a measurement can be used to validate calculational methods for criticality safety. These measurements provide parameters that have a higher sensitivity to changes in fissile mass than neutron multiplication factor and thus serve as a better test of calculational methods.

The analyses have also shown that measurement of the cross power spectral density (CPSD) between detectors on one side of a single fuel assembly and an internal or external ^{252}Cf source driving the fission chain multiplication process can be used for nondestructive assay of fissile mass along the length of the assembly. This CPSD is a smooth function of fissile mass and does not depend on the varying inherent source in the fuel assembly and thus is ideal for fissile mass assay.

1. INTRODUCTION

As nuclear spent fuel storage pools at operating nuclear reactors reach their storage limitations, a subcriticality measurement for more compact storage that takes credit for burnup becomes more desirable both for assessing by measurement the criticality safety of proposed storage configurations and for providing data that can be used for validation of calculational methods for criticality safety of spent fuel storage. This type of measurement¹ is also useful for the design and qualification of shipping and storage casks for spent fuel. The usefulness of such a measurement in the past has been limited by the theoretical models used to interpret the experimental data. These limited point kinetics models for interpretation of measurements have been replaced by more general Monte Carlo models.² These more general models can be used to obtain two quantities from the measured data: (1) the subcritical neutron multiplication factor, k , and (2) the bias in the calculation methods and cross section data used for criticality safety. The latter is also of keen interest to the criticality safety specialist in assessing the

accuracy of his analysis methods. It is especially useful that the codes used to interpret the measured data can also be used for criticality safety assessments. As a result, these measurements can be used to evaluate the adequacy of calculational methods for criticality safety. Recent work has shown how this measurement method and its interpretation can be applied successfully to two types of subcritical measurement: the first where accurate, detailed knowledge of the subcritical configuration is known (i.e., geometry, isotopic composition of fuel, atomic densities of all constituents, etc.) and the second where the configuration is not accurately known. In the latter case, some assumptions have to be made about the system. It has been shown by interpretation of measurements that these types of assumptions may not affect the subcritical neutron multiplication factor (only its uncertainty) obtained from interpretation of the measured data but do affect the bias obtained by this method.³ However, if the criticality safety specialist used the same assumption in his criticality safety assessment, he will reach the same conclusions in the assessment. Recent interpretation of measurements with a mockup of a 17×17 fuel pin pressurized water reactor (PWR) fuel element has shown that a single fuel assembly can be used to provide data to validate calculational methods.

This paper reviews previous work in the application of this method to spent fuel; presents the results of application of this general model for interpretation of previous fresh fuel measurements; provides assessments of the practicality of measurements with spent fuel for both subcriticality for burnup credit and for nondestructive assay (NDA) of fissile mass as a function of fuel assembly length; and addresses the availability of the hardware for such measurements.

2. MEASUREMENT METHOD

2.1 Measurement Concept

The ^{252}Cf -source-driven frequency analysis method for obtaining the subcritical neutron multiplication factor of a configuration of fissile material was developed to avoid difficulties inherent in other subcriticality measurement methods such as need for a calibration at a known reactivity condition near delayed critical and an interpretation that depends on detector efficiency. Although the interpretation of the measured data does not require knowledge of detection efficiency, the time to complete a measurement does depend

on the detection efficiency. This measurement concept has been under development since 1968, and frequency analysis measurements with reactors were first made in 1959.⁴ This method requires measurement of the frequency-dependent cross power spectral density (CPSD), $G_{23}(\omega)$, between a pair of detectors (detectors 2 and 3) located in or near the fissile assembly as well as measurements of CPSDs, $G_{12}(\omega)$ and $G_{13}(\omega)$, between these same detectors and a source of neutrons emanating from a ^{252}Cf source ionization chamber (detector 1) positioned in or near the fissile assembly. The ^{252}Cf source emits neutrons to initiate the fission chain multiplication process in the spent fuel array. In addition, fission chains are initiated by the inherent neutron sources in the spent fuel. Also required is the autopower 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 denotes complex conjugation), is independent of detector efficiencies and can be related to the subcritical neutron multiplication factor. Another useful quantity, the coherence, γ_{ij}^2 , is defined as $|G_{ij}|^2 / G_{ii} G_{jj}$ and is the fraction of common information in two signals and varies between 0 and 1. The common information in the signals arises from the fission chain multiplication process in the fissile material of the spent fuel.

2.2 Advantages of the Method

One advantage of the method is the higher sensitivity of the frequency measured parameters to fissile mass than other methods, and this has been observed experimentally and demonstrated theoretically⁵ for a variety of configurations of fissile materials and can be the basis of the use of this method for process control.⁶ This sensitivity for the spent fuel application has been reported.⁷ Another advantage of the method is that since the ratio of spectral densities $G_{12}^* G_{13} / G_{11} G_{23}$ is independent of detection efficiency, it depends more directly on $\Delta k/k$, where k is the neutron multiplication factor.⁸ This method measures some quantities G_{12} and G_{13} that are only sensitive to detected particles from induced fission by ^{252}Cf neutrons, some that depend on all fission neutron sources, G_{23} , and some that depend on all neutron sources, G_{22} and G_{33} . Another advantage recently demonstrated is the ability to measure the subcriticality without very accurate knowledge of the configuration of fissile materials.

2.3 Previous Uses

The ^{252}Cf -source-driven noise analysis method has been used in measurements for subcritical configurations of fissile systems for a variety of applications. Measurements on over 30 fissile systems have been performed with a wide variety of materials and configurations. This method has been applied to measurements for (1) initial fuel loading of reactors,⁹ (2) quality assurance of reactor fuel elements,¹⁰ (3) fuel preparation facilities,¹¹ (4) fuel processing facilities,¹² (5) fuel storage facilities,¹³ (6) zero-power testing of reactors,¹⁴ and (7) verification of calculational methods for assemblies with $k < 1$.¹⁵ These previous measurements, performed with a wide variety of multiplying systems, demonstrated the usefulness of the method. In these measurements, the ^{252}Cf -

source-driven frequency analysis method was used to obtain the subcritical neutron multiplication factor for a variety of static systems with values that varied from ~ 0.35 to ~ 0.99 . Four of these measurements were supported by the Power Reactor and Nuclear Fuel Development Corporation of Japan (PNC) and addressed the application of this method to fuel reprocessing for breeder reactors. In one measurement, the k value was measured every 6 s as a tank of uranyl nitrate solution was drained from a k of 0.96 to 0.35 at a rate of 0.01 $\Delta k/s$.¹⁶ This dynamic measurement, which was part of the PNC program, was the first step in applying this method to dynamically monitor fuel in a future reprocessing plant. These measurements have shown that this method has potential to monitor many dynamic situations in processing plants such as in a continuous dissolver or batch dissolvers either to obtain the neutron multiplication factor k or to be used as a signature to verify that various processes are occurring in a repeatable or bounded way. In addition to these applications, the method has been used for nuclear weapons identification^{17,18} and can be used for nondestructive assay for nuclear material control and accountability.¹⁹

3. INTERPRETATION OF DATA

3.1 MCNP-DSP

MCNP-DSP is an analog Monte Carlo code developed from MCNP that calculates the time and frequency analysis parameters of the ^{252}Cf -source-driven frequency analysis measurement and the neutron multiplication factor of fissile assemblies. In MCNP-DSP, average quantities like the average number of neutrons from fission, $\bar{\nu}$, have been removed and replaced with the appropriate probability distributions because average quantities reduce the statistical fluctuation in the fission chain populations. These distributions are used in the calculation of the frequency analysis measurement parameters and also in the calculation of the neutron multiplication factor.

For the frequency analysis calculations, the particle tracking begins with the source fission. The fission chains are then followed to extinction. The time series of pulses are obtained at each detector for each fission chain. The time series of detector pulses from all contributing fission chains are superimposed consistent with a source fission rate to obtain the proper time-ordered sequences of pulses at the detectors. The sequences are sampled into blocks of 512 or 1024 data points. These blocks of data are then Fourier transformed using standard algorithms. The Fourier transformed blocks are complex multiplied and averaged to obtain estimates of the APSDs and CPSDs. This process is repeated until the desired statistical uncertainty is obtained. The calculation of the neutron multiplication factor proceeds in the same manner as the standard MCNP eigenvalue calculation.

3.2 Neutron Multiplication Factor and Bias in Calculation Methods

The MCNP-DSP code is used to obtain the neutron multiplication factor from the measurement in the following

manner: (1) A measurement is performed to obtain the ratio of spectral densities, R_m . (2) A detailed Monte Carlo model of the measurement is used to obtain the ratio of spectral densities, R_c , and the neutron multiplication factor, k_c . (3) If the measured and calculated values of the ratio of spectral densities agree, then the measured neutron multiplication factor, k , is the neutron multiplication factor, k_c , obtained with the Monte Carlo model. (4) If the measured and calculated values of the ratio of spectral densities do not agree, then the Monte Carlo model is perturbed in a manner such that the ratio of spectral densities from the perturbed model, R_p , agrees with the measured value of the ratio of spectral densities, R_m . (5) Then the neutron multiplication factor inferred from the measurement is that for the perturbed model, k_p . (6) The bias in the neutron multiplication factor is the difference between k_p and k_c . To verify that the neutron multiplication factor obtained does not depend on the method of perturbation, the method of perturbation is varied. Ideally, the k obtained from the measurement should not depend on how the system is perturbed. With this approach to interpretation of the measured data, this more general Monte Carlo model replaces the limited point kinetics model to interpret the measured data. In the cases examined so far, the multiplication factor obtained does not depend on the method of perturbation nor require an accurate knowledge of the system, but, of course, the bias and its uncertainty and the uncertainties in k depend on the accuracy of the knowledge of the system. Steps 1 and 2 allow a direct calculation of the measured data and thus can be used to validate the calculation methods and cross sections. Steps 1 and 2 determine the calculational bias in the frequency analysis parameters. The calculational bias in k is obtained by use of the sensitivity coefficient $\Delta R/\Delta k$ obtained from the Monte Carlo model. Other measured frequency analysis parameters could be used as well since all the measured frequency analysis parameters are relevant for criticality safety because they depend on powers of $\Delta k/k$ up to fourth order.

3.3 Measurements with Fresh Fuel Assemblies

An example of the interpretation of the data from this type of measurement for PWR fuel pin arrays using this general Monte Carlo model MCNP-DSP is given in this section; it illustrates how the method can be used for subcritical configurations of light water reactor (LWR) fuel. Extensive measurements with subcritical arrays of fresh PWR fuel pins were performed in 1983 at the Babcox and Wilcox Company Critical Experiments Facility in Lynchburg, Virginia, using the ^{252}Cf -source-driven frequency analysis method.²⁰ These experiments have not been extensively interpreted and reported because of the limitations of the point kinetics models used to interpret the measured data. This section presents some of the measured data and the results of this Monte Carlo method of interpretation of these ^{252}Cf source-driven frequency analysis measurements as applied to subcritical arrays of PWR fuel pins. Measurements for arrays of PWR fuel pins, even as small as a 17×17 fuel pin array, and 18-element arrays that

were highly poisoned with boron (up to 4303 ppm) in the water are presented. The Monte Carlo model MCNP-DSP was used to interpret the measured data to obtain the subcritical neutron multiplication factor. This type of measurement can be used in two ways: to determine by measurement the subcriticality of arrays of spent fuel or, with extensive knowledge of the fuel element in the array, to serve as a benchmark of calculational methods.

The experimental vessel for these measurements was a 152.4-cm-diam tank with grid plates that supported 153.3-cm-long aluminum-clad UO_2 fuel pins with a square pitch of 1.637 cm. The fuel pin diameter was 1.206 cm, and the UO_2 pellet diameter was 1.044 cm. The uranium was enriched to 2.46 wt % ^{235}U . For some of these measurements, the boron concentration of the water reflector and moderator was 1510 ppm natural boron. The configurations of fuel pins were varied for a fixed boron concentration of 1510 ppm. The cylindrical pin configurations contained 4961, 3713, 2533, 1281, 749, and 333 pins with effective radii of 66.05, 56.26, 46.65, 33.06, 25.29, and 16.89 cm respectively. A 17×17 square array of fuel pins was also assembled to simulate a single PWR fuel element. For the full array of 4961 fuel pins, the boron concentration was varied in several steps up to 4303 ppm. The characteristics of these experimental configurations are very precisely known, and thus these measurements can validate calculational methods.

This section presents an analysis for a small portion of these measurements, that is, the 4961 fuel pin configuration with boron concentrations of 2300 and 4303 ppm, and a 17×17 fuel pin array simulating a single PWR fuel element with 1510 ppm boron in the water. These subcritical configurations represent significant changes in either the neutron spectrum or the neutron leakage from the delayed critical configuration. The calculational bias for the subcritical configurations can be compared with that from the delayed critical configuration. In the subcritical measurements analyzed in this paper, the californium source was in the center of the arrays both vertically and axially, and the ^3He proportional counters were located as shown in Fig. 1 for the 17×17 fuel pin array. The signals from three counters on each side were summed to serve as one detection channel.

MCNP-DSP was used to obtain the subcritical neutron multiplication factor from the measured data and the calculational bias. For this analysis, both the boron concentration and the enrichment were varied for the most subcritical configuration, and some of the results are given in Table 1, using ENDF/B-V cross section data for these configurations. The negative bias from the subcritical experiments (0.01 to 0.024) is larger in magnitude than that from the comparison of the calculation with measurement for the delayed critical configuration (0.0013). Also, the neutron multiplication factor does not depend on the method of perturbation used in the analysis, which was varied for the 17×17 fuel pin array. The uncertainties of the neutron multiplication factor obtained from the subcritical experiments

varied from ~ 0.004 to ~ 0.007 . From these experiments the average neutron multiplication factor for a single fuel element (17×17) is 0.671 ± 0.0044 .

This generalized interpretation of these experiments has shown that this method can be used for LWR fuel with array configurations as small as one fuel element and that this measurement method and its interpretation are useful for spent fuel subcriticality measurements that can be used as a basis for burnup credit and also for validation of calculational methods for criticality safety.

4. SPENT FUEL APPLICATIONS

4.1 Subcriticality for Burnup Credit

The californium-source-driven noise analysis method is considered for this application because (1) measurements had been performed in 1983 at the Babcock and Wilcox (B&W) Critical Experiments Facility with mockups of arrays of PWR fresh fuel pins of the size typical of large cask configurations, (2) neutron detectors are available that had been designed for use with spent fuel arrays, and (3) the technology necessary to construct ionization chambers containing ^{252}Cf of adequate intensity for these measurements has been demonstrated. The practicality of a measurement depends on the ability to install the hardware that is required to perform the measurement. Oak Ridge National Laboratory has a long history of installing detectors in reactor pools for noise analysis and other measurements. Mechanical installation and operation under water will require dry tubes or other waterproofing of the detectors and source chambers. The major requirements that must be met are (1) sufficient neutron source intensity to produce a discernible signal above that produced by the inherent source of the fuel (estimated from past experience to be at least $\sim 5\%$ of the inherent source) and (2) sufficient detection efficiency to perform the measurements in a reasonable time. A high-sensitivity fission detector has been designed by Reuter Stokes, Inc., under the Small Business Innovative Research Program of the Department of Energy for measurements with spent fuel.²¹ Source chambers containing the ^{252}Cf of the required source intensity for this application have been constructed and have operated successfully for ~ 10 years. Sources small enough to fit in control rod guide tubes have been fabricated.²²

An analysis²³ had been performed to estimate measurement times for a variety of spent fuel configurations using two-dimensional transport theory calculations for a variety of configurations of spent PWR fuel to obtain estimates of the detection efficiency. Both types of calculations used 27-group cross sections obtained from ENDF/B-IV.²⁴ These calculations included (1) a Castor V cask with borated and unborated water; (2) pool storage racks at Virginia Power's Surry plant (both with 2.57% ^{235}U initial enrichment, 23-GWd/MTU burnup, and 14-year cooling time); (3) regions 1 and 2 pool storage racks at Duke Power's McGuire plant; and (4) a special, tightly packed 3×3 array with 6.3 mm of stainless steel between fuel elements, 9.4-mm-thick outer walls, and 5.1 mm of water on all sides of the fuel element in

borated and unborated water (both with 3.05% ^{235}U initial enrichment, 26-GWd/MTU burnup, and 10-year cooling time). In addition, estimates of the detection efficiencies were made for the typical burnup credit rail cask with borated and unborated water and the 2.57% ^{235}U initial enrichment fuel.

The calculated values of k and the detection efficiencies for a single fission counter (51 mm OD, 2.44 m long) and a single ^3He ionization chamber for the various configurations are plotted in Fig. 2. From the theory of the ^{252}Cf noise analysis method, it is possible to estimate the coherences (which depend on the detection efficiencies) and thus the measurement times required to achieve a specified statistical precision. Estimates of the measurement times (within a factor of 2) for various coherences are also indicated on Fig. 2 for an uncertainty of 5% in the ratio of spectral densities or an uncertainty of $\sim 5\%$ in $\Delta k/k$. Obviously, longer measurement times will reduce the statistical uncertainty. The use of ^3He ionization chambers requires gamma ray shielding and special additions to the chamber gas. The ^3He chamber for the calculations had a 51-mm OD and 2.44-m length, surrounded by 25-mm-thick polyethylene and 76 mm of lead shielding between the chamber and the fuel.

The practicality of a spent fuel measurement depends on the use of detectors that meet the detection efficiency requirements. From the data of Fig. 2, the existing fission counters would allow measurements for a special 3×3 spent fuel array ($k = 0.90$) and a typical burnup credit rail cask with spent fuel in unborated water ($k = 0.92$). Adding a moderator around the fission counters would allow measurements with the typical burnup credit rail cask with borated water ($k = 0.76$) and the special 3×3 array with borated water ($k = 0.75$) but using a single detector would be marginal for the McGuire pool racks ($k = 0.68$). For the special 3×3 array and rail cask in unborated water, the measurement time estimates are ~ 1 min, while for these arrays in borated water the measurement time estimates are ~ 40 min with fission counters. A ^3He ionization chamber, which would have to be designed and fabricated, would certainly be adequate for the above configurations and would also reduce the measurement time considerably for the McGuire region 2 racks and for configurations with k values as low as 0.6. Using multiple detectors on each side of the array would decrease the measurement time considerably; for example, using three counters in each detection channel increases the coherence between detector channels and thus would reduce the measurement time a factor of ~ 10 , making measurements for all arrays practical. These calculations show that the measurements are feasible. Measurement in unborated water are more relevant for the storage/transportation cask application and in general require shorter measurement time because of the higher k values for the arrays

4.2 Validation for Criticality Safety Calculations

The method described in Sects. 3.1 and 3.2 as applied to the measurements of Sect. 3.3 have shown that the data provided by this type of measurement can provide the neutron

multiplication factor and the bias in calculational methods for as little fuel as a single fuel assembly. The direct comparison of the calculated frequency analysis parameter with the measured can be used to validate calculational methods and cross section data if the fuel assemblies can be accurately described. These validations are especially useful since all frequency analysis parameters depend on $\Delta k/k$ to some power up to fourth order.

4.3 Fissile Assay of Spent Fuel

Since the method has been shown to be able to measure the subcriticality of as little as a single PWR fuel element and since both previous experiments and Monte Carlo calculations have shown that the frequency analysis parameters have a high sensitivity to fissile mass, the obvious question was, "Can this method be used for fissile mass nondestructive assay of spent LWR fuel?" The feasibility of fissile mass assay of spent fuel using the ^{252}Cf -source-driven frequency analysis method was investigated in a series of calculations using MCNP-DSP. This analysis used a central source in a control rod guide tube and two arrays of three 1-ft-long ^3He counters in polyethylene, one on each side of a 17×17 pin PWR fuel assembly. Each array of three detectors was a detection channel. This was done to increase the statistical accuracy of the calculations. In an actual measurement an external source adjacent to the fuel assembly and fission counters that can operate in the high-gamma-ray radiation field around spent fuel would be used. These source detector assumptions should not significantly affect any conclusions drawn from these analyses. The calculational model is given in Fig. 3 with the division of the fuel assembly into seven axial zones with varying burnups. Three average fuel assembly burnups were used: 10, 21, and 32 GWd/MTU.

The frequency analyses parameters that were calculated were those defined in Sect. 2.1. The APSDs depend on all neutron sources. The CPSDs G_{12} and G_{13} depend only on induced fission in the fuel element or fissile mass, and G_{23} depends on all fission sources, including spontaneous fission. Since the CPSDs G_{12} and G_{13} are the only spectra that depend only on fissile mass and not on inherent sources, these quantities are the most favorable for fissile mass assay. All calculations involved only 3.2×10^6 californium fissions, which is a small number, since a $6\text{-}\mu\text{g}$ ^{252}Cf ionization chamber has this fission rate per second.

The magnitude of G_{12} vs frequency is given in Fig. 4 for position 3 in a fuel assembly with 32-GWd/MTU burnup. This function is constant with frequency up to ~ 700 Hz and then falls off with frequency. The average value at low frequency can be obtained from these data and can be used for a measurement of fissile mass. A plot of $|G_{12}|$ at low frequency vs ^{235}U content is given in Fig. 5. The values for the end positions fall on one curve, and those for positions 2 \rightarrow 6 fall on another curve. A similar plot vs fissile mass including ^{239}Pu is given in Fig. 6 and shows similar behavior. All other frequency analysis parameters at low frequency vs fissile mass can also be separated into two curves. Thus, with two

calibration curves, one for the ends and one for the rest of the fuel assembly, fissile mass assay could be obtained from a measurement of the CPSD between a detection channel and the source. If three ^3He counters are used, a measurement to $\pm 1\%$ could be made in less than 60 s, and using three fission counters and an external source, a measurement could be made to $\pm 3\%$ in less than 60 s. The preferred arrangement for fissile mass assay should be a source adjacent to one side of a fuel assembly with a group of three 1-ft-long chambers in polyethylene on the other side.

The hardware for this measurement exists, with the fission counters, detection electronics, and the Fourier processor commercially available. Californium source ionization chambers with $5\text{ }\mu\text{g}$ would be adequate and have been built and satisfactorily operated for over 10 years.

5. CONCLUSIONS

The ^{252}Cf -source-driven frequency analysis method can be used for measuring the subcritical neutron multiplication factor of arrays of LWR fuel and as little as a single PWR fuel assembly. These measurements can be used to verify the criticality safety margins of spent LWR fuel configurations and thus could be a means of obtaining the information to justify burnup credit for spent LWR transportation/storage casks. In addition, the data provided by such a measurement can be used to validate calculational methods for criticality safety. Calculations for a special array of spent fuel in unborated water show that these measurements can provide data for a more sensitive test of calculational capability than comparisons between calculated and measured neutron multiplication factors since the sensitivity to changes in fission product cross sections was a factor of ~ 10 higher for ratios of spectral densities and a factor of ~ 40 higher for coherences than that for neutron multiplication factor comparisons.⁷

The analysis has also shown that measurement of the cross power spectral density between detectors on one side of the fuel assembly and an internal or external ^{252}Cf source driving the fission chain multiplication process can be used for nondestructive assay of fissile mass. One of the frequency analysis parameters is a smooth function of fissile mass and does not depend on the varying inherent source in the fuel assembly. In addition, fissile mass assay as a function of length along a fuel assembly can be performed by this measurement method for single fuel assemblies.

The practicality of a subcriticality measurement depends on the ability to install the hardware required to perform the measurement. Source chambers containing the ^{252}Cf at the required source intensity for this application have been constructed and have operated successfully for >10 years. A 9.3-mm-OD ^{252}Cf ionization chamber that fits into the control rod guide tubes of PWR fuel elements (if necessary) has been designed and operated successfully. Fission counters developed by Reuter Stokes, Inc., for spent fuel measurements allow subcriticality measurements for a variety of spent fuel cask configurations. For other fuel casks with lower neutron multiplication factors, shielded ^3He counters with special gas

additives to reduce the effect of gamma rays can be used or multiple detectors on each side of the array. For the fissile mass assay application, the same sources used for subcriticality measurement can be used, and commercial 31-cm-long 5.1-cm-diam fission chambers would be adequate. For both types of measurements, commercially available detection electronics and Fourier processors are adequate.

REFERENCES

1. J. T. Mihalcz, V. K. Paré, G. L. Ragan, M. V. Mathis, and G. C. Tillet, "Determination of Reactivity from Power Spectral Density Measurements with Californium-252," *Nucl. Sci. Eng.*, **66**, 29 (1978).
2. T. E. Valentine and J. T. Mihalcz, "MCNP-DSP: A Neutron and Gamma Ray Monte Carlo Calculation of Source-Driven Noise-Measured Parameters," submitted to *Annals of Nuclear Energy*, 1996.
3. T. E. Valentine and J. T. Mihalcz, "MCNP-DSP Calculations of the ^{252}Cf -Source-Driven Noise Analysis Measurements of High-Enriched Uranium Metal Cylinders," ICNC '95, The Fifth International Conference on Nuclear Criticality Safety, September 17–22, 1995, Albuquerque, New Mexico.
4. J. A. Thie, *Reactor Noise*, Rowman and Littlefield, Inc., New York, 1963.
5. J. T. Mihalcz and T. E. Valentine, "Calculational Verification and Process Control Applications Utilizing the High Sensitivity of Noise Measurement Parameters to Fissile System Configuration," *Nucl. Sci. Eng.*, **121**, 286–300 (1995).
6. J. T. Mihalcz, "Use of Noise Analysis Methods in Process Monitoring of Future Fuel Cycles," *Proc. Global '93 Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options*, Seattle, Washington, September 12–17, 1993, Vol. 1, p. 510, American Nuclear Society, 1993.
7. A. W. Krass, T. E. Valentine, and J. T. Mihalcz, "Sensitivity of the ^{252}Cf -Source-Driven Noise Analysis Method to Fission Product Content of Spent LWR Fuel," *Trans. Am. Nucl. Soc.*, **65**, 253 (1992).
8. J. T. Mihalcz and E. D. Blakeman, "Safer Fuel Loading and Initial Reactor Startup Using the ^{252}Cf Source Driven Noise Analysis Method," *Trans. Am. Nucl. Soc.*, **61**, 244 (1990).
9. W. T. King, J. T. Mihalcz, and E. D. Blakeman, "Preliminary Investigation of the ^{252}Cf -Source-Driven Noise Analysis Method of Subcriticality Measurement in LWR Fuel Storage and Initial Loading Applications," *Trans. Am. Nucl. Soc.*, **47**, 239 (1984).
10. J. T. Mihalcz and W. T. King, "Quality Assurance Verification of High-Flux Isotope Reactor Fuel Elements by the ^{252}Cf -Source-Driven Noise Analysis Method," *Nucl. Technol.*, **84**, 205 (1989).
11. J. T. Mihalcz, W. T. King, and E. D. Blakeman, " ^{252}Cf -Source-Driven Neutron Noise Analysis Measurements for Coupled Uranium Metal Cylinders," *Trans. Am. Nucl. Soc.*, **49**, 241 (1985).
12. J. T. Mihalcz, E. D. Blakeman, and W. T. King, "Subcriticality Measurements for Two Coupled Uranium Nitrate Solution Tanks Using ^{252}Cf -Source-Driven Neutron Noise Analysis Methods," *Trans. Am. Nucl. Soc.*, **52**, 640 (1986).
13. J. T. Mihalcz, W. T. King, and E. D. Blakeman, "Decoupling of Uranium Metal with Borated Plaster Using ^{252}Cf Noise Analysis Methods," *Trans. Am. Nucl. Soc.*, **50**, 307 (1985).
14. W. T. King and J. T. Mihalcz, "Power Spectral Density Measurements with ^{252}Cf for a Light-Water-Moderated Research Reactor," *Trans. Am. Nucl. Soc.*, **33**, 796 (1979).
15. J. T. Mihalcz, W. T. King, E. B. Johnson, and E. D. Blakeman, "Subcriticality Measurements for a Fuel Solution Tank with Changing Fuel Concentration Using ^{252}Cf -Source-Driven Neutron Noise Analysis," *Trans. Am. Nucl. Soc.*, **45**, 337 (1983).
16. J. T. Mihalcz, E. D. Blakeman, G. E. Ragan, E. B. Johnson, and Y. Hachiya, "Dynamic Subcriticality Measurements Using the ^{252}Cf -Source-Driven Noise Analysis Method," *Nucl. Sci. Eng.*, **104**, 314 (1990).
17. J. T. Mihalcz and V. K. Paré, "Nuclear Weapons Identification System (NWIS)," p. 24 in *Arms Control and Nonproliferation Technologies*, Report on Nuclear Warhead Dismantlement, DOE/AN/ANCI-94C, Lawrence Livermore National Laboratory, Third Quarter 1994.
18. J. T. Mihalcz and V. K. Paré, "NWIS Signatures for Confirmatory Measurements with B33 Trainers," *Proceedings of the 36th Annual Meeting of the Institute of Nuclear Materials Management*, Palm Desert, California, July 1995.
19. T. E. Valentine, J. T. Mihalcz, and P. E. Koehler, "Calculated NWIS Signatures for Enriched Uranium Metal," *Proceedings of the 36th Annual Meeting of the Institute of Nuclear Materials Management*, Palm Desert, California, July 1995.
20. W. T. King, J. T. Mihalcz, and E. D. Blakeman, "Preliminary Investigation of the ^{252}Cf -Source Driven Noise Analysis Method of Subcriticality Measurement in LWR Fuel Storage and Initial Loading Applications," *Trans. Am. Nucl. Soc.*, **47**, 239–40 (1984).
21. J. A. Williams, J. T. Mihalcz, C. W. Ricker, F. L. Glesius, and T. A. Kniss, "A High-Sensitivity, Position-Sensitive Fission Counter for Subcriticality Measurements of Spent Fuel," *Nucl. Instrum. Meth. Phys. Res.*, **A299**, 187–90 (1990).
22. M. M. Chiles, J. T. Mihalcz, and C. E. Fowler, "Small, Annular, Double-Contained ^{252}Cf Fission Chamber for

Source-Driven Subcriticality Measurements," Institute of Electrical and Electronics Engineers Nuclear Science Symposium, Orlando, Florida, October 27-31, 1992, *IEEE Trans. Nucl. Sci.*, **40**, 816-18 (1993).

23. J. T. Mihalczo and B. L. Broadhead, "Feasibility of Spent LWR Fuel Subcriticality Measurements by the Cf Noise

Method," *Trans. Am. Nucl. Soc.*, **62**, 322 (1990).

24. G. D. Garber, *ENDF-201, ENDF/B Summary Documentation*, Summary Documentation for ENDF/B-IV, BNL 1754, Brookhaven National Laboratory, October 1975.

Table 1. Neutron Multiplication Factors and Bias in Calculations with ENDF/B-V Data from Subcritical Measurements with PWR Fuel

Configuration	Ratio of Spectral Densities		Perturbation	k_{eff}	Negative Bias
	Measured, R_m	Calculated, R_c			
Critical, 4961 fuel pins, 1511 ppm B	NA	NA	NA	0.9987 ± 0.0006	0.0013 ± 0.0006
4961 fuel pins, 2386 ppm B	0.2483 ± 0.0041	0.2648 ± 0.0024	Boron content.	0.9142 ± 0.0069	0.0243 ± 0.0070
4961 fuel pins, 4303 ppm B	0.4301 ± 0.0067	0.4696 ± 0.0110	Boron content.	0.7421 ± 0.0075	0.0124 ± 0.0075
289 fuel pins, 1510 ppm B	0.5460 ± 0.0069	0.5643 ± 0.003	Boron content.	0.6691 ± 0.0042	0.0101 ± 0.0043
			Enrichment	0.6784 ± 0.0074	0.0195 ± 0.0074

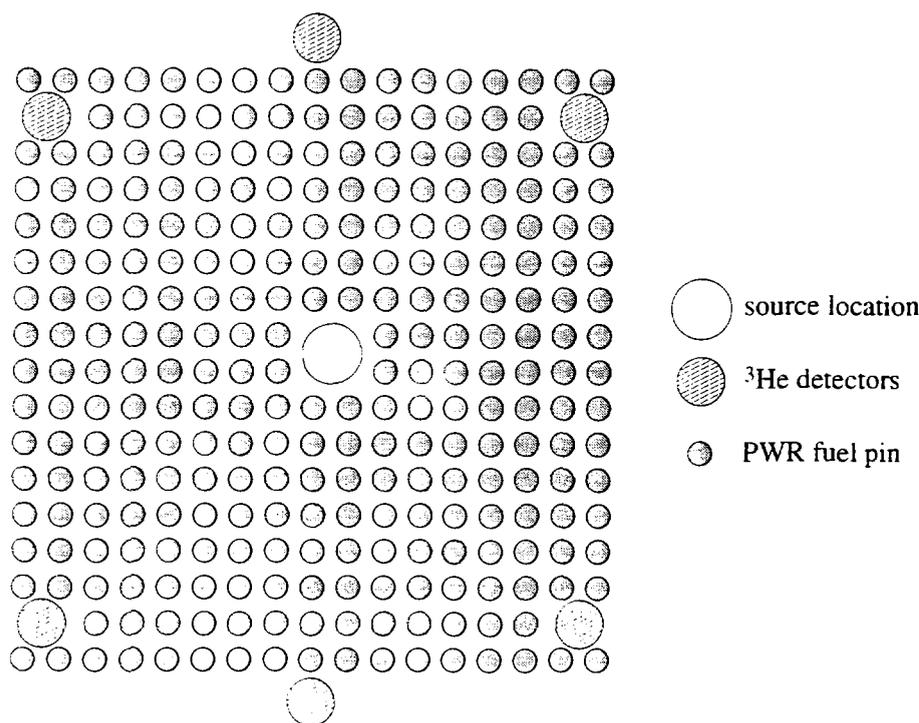


Fig. 1. Source-detector-system configuration for ^{252}Cf source driven frequency analysis measurements for a mockup of a 17×17 fuel pin PWR fuel assembly.

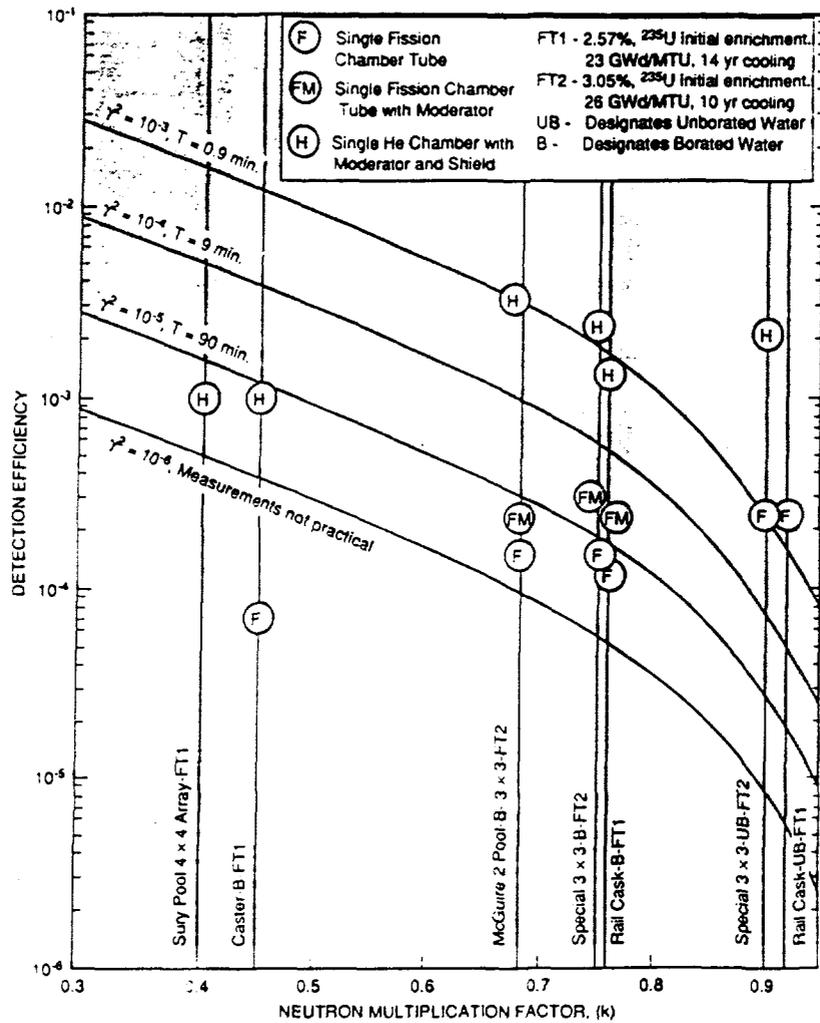


Fig. 2. Detection efficiency for various detectors vs neutron multiplication factor for a variety of spent LWR fuel arrays.

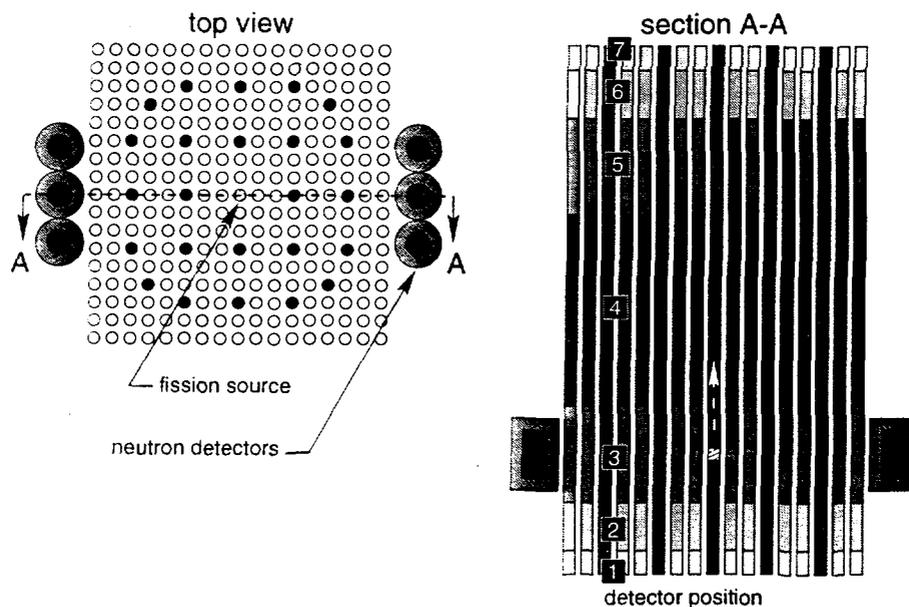


Fig. 3. Source-detector-fuel pin configuration for a 17 x 17 fuel pin PWR fuel assembly.

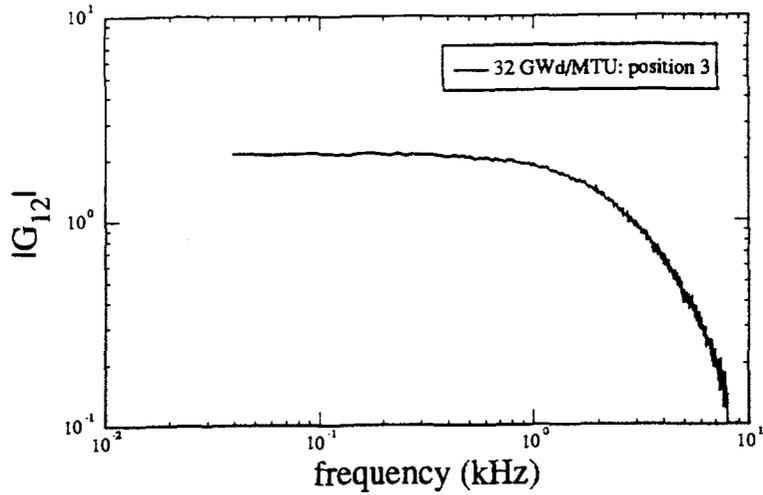


Fig. 4. Magnitude of cross power spectral density G_{12} as a function of frequency for source and detectors located at position 3 for 32-GWd/MTU burnup.

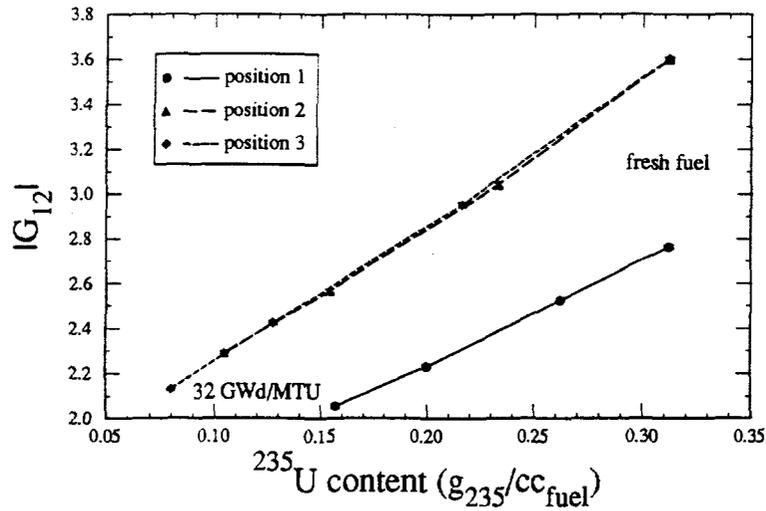


Fig. 5. Magnitude of G_{12} at low frequency vs ^{235}U content of fuel for various burnups up to 32 GWd/MTU.

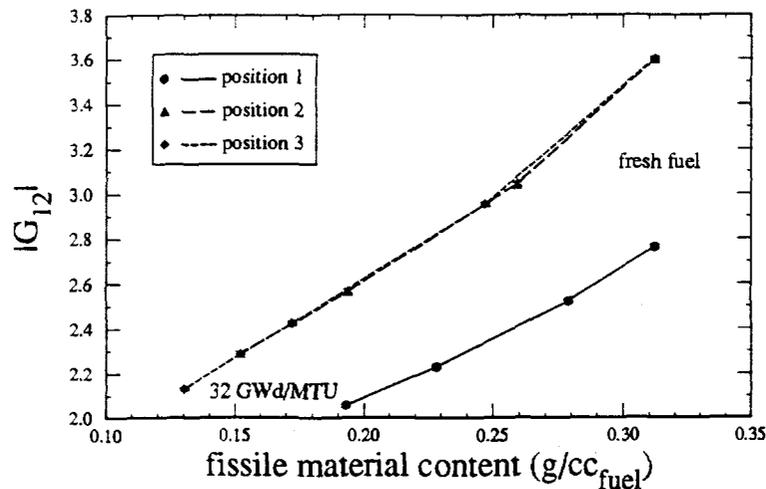


Fig. 6. Magnitude of G_{12} at low frequency vs fissile content ($^{235}\text{U} + ^{239}\text{Pu}$) of fuel for various burnups up to 32 GWd/MTU.

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