FEASIBILITY OF FISSILE MASS ASSAY OF SPENT NUCLEAR FUEL USING \(^{252}\)Cf-SOURCE-DRIVEN FREQUENCY-ANALYSIS

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Feasibility of Fissile Mass Assay of Spent Nuclear Fuel Using 
$^{252}\text{Cf}$-Source-Driven Frequency-Analysis

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Abstract

Any measurement capable of directly estimating the fissile material content of a nuclear assembly has obvious utility in safeguards, control and accountability, and criticality safety applications. Such a measurement applied to the fissile mass assay of spent nuclear fuel is clearly useful as a method of obtaining burnup credit for storage pool and shipping cask loading. The feasibility of applying $^{252}\text{Cf}$-source-driven frequency-analysis to perform fissile mass assay of spent nuclear fuel assemblies has been evaluated using MCNP-DSP, an analog Monte Carlo transport code to simulate source-driven measurements. This feasibility study employed models of an isolated Westinghouse 17x17 pressurized water reactor fuel assembly in a 1500-ppm borated water storage pool. In the models, the fuel burnup profile was represented using seven axial burnup zones, each with isotopics estimated by the PDQ code. Four different fuel assemblies with average burnups from fresh to 32 GWd/MTU were modeled and analyzed. Analysis of the fuel assemblies was simulated by inducing fission in the fuel using a $^{252}\text{Cf}$ source adjacent to the assembly and correlating source fissions with the response of a bank of $^3\text{He}$ detectors adjacent to the assembly opposite the source. This analysis was performed at seven different axial positions on each of the four assemblies, and the source-detector cross-spectrum signature was calculated for each of these 28 simulated measurements. The magnitude of the cross-spectrum signature follows a smooth upward trend with increasing fissile material ($^{235}\text{U}$ and $^{239}\text{Pu}$) content, and the signature is independent of the concentration of spontaneously fissioning isotopes (e.g., $^{144}\text{Ce}$) and $(\alpha,\text{n})$ sources. Furthermore, the cross-spectrum signature is highly sensitive to changes in fissile material content. This feasibility study indicated that the signature would increase ~100% in response to an increase of only 0.1 g/cm$^3$ of fissile material.

Introduction

Measurements employing $^{252}\text{Cf}$-source-driven frequency-analysis have been performed since 1974 at eight Department of Energy sites to estimate properties of over 30 different fissile systems for applications in nuclear weapons identification, nondestructive analysis for nuclear material control and accountability, initial loading of reactors, fuel preparation and processing, and reactor fuel element quality assurance. Measurements of moderated fresh fuel arrays performed in 1984 at the former Babcock & Wilcox critical facility in Lynchburg, Virginia constitute initial attempts to apply this measurement to the analysis of nuclear fuel. The results of these earlier experiments, in particular the experiment performed on a single 17x17 fuel array in borated water, prompted further evaluations of the feasibility of applying $^{252}\text{Cf}$-driven frequency-analysis to perform fissile mass assay of spent nuclear fuel.

$^{252}\text{Cf}$-Source-Driven Frequency-Analysis

Figure 1 illustrates the configuration of a typical $^{252}\text{Cf}$-source-driven frequency-analysis measurement. Fission is induced in the subcritical system by the $^{252}\text{Cf}$ source, and the response of the system is measured by one or more detectors appropriate to the measurement (e.g., $^3\text{He}$, fission chamber, Li-glass, plastic detectors, etc.). For application to the fissile mass assay of spent nuclear fuel, only the neutron response of the system is of interest, so either $^3\text{He}$ or fission detectors will be employed. The $^{252}\text{Cf}$ source is contained in an ionization chamber, and for typical source sizes...
and ionization chamber configurations, the source chamber counting efficiency exceeds 99.9%. Consequently, the timing of source fission events is observable and constitutes a time-dependent signal hereafter indexed as signal \( \#1 \). Similarly, the response of the external detectors is measured to acquire time-dependent signals \( \#2, \#3 \), etc.

Consequently, the detector cross-spectra \( (G_{ij}) \) correlate events in one detector with events in another detector. So their magnitude indicates the amount of both source-induced and inherent fission occurring in the system analyzed. The source-detector cross-spectra have the following properties.

The signatures obtained from the described measurement configuration involve all two-channel combinations (i.e., \( 1\&2, 1\&3, 2\&3, \) etc.) including signatures that correlate an individual signal with itself (i.e., \( 1\&1, 2\&2, 3\&3, \) etc.). The signatures between different signals are termed cross-spectra, while those involving the same signal twice are termed autospectra. The measured autospectra and cross-spectra have the following properties.

The source autospectrum \( G_{11} \) is simply a measure of the fission source strength. The source-detector cross-spectra \( (G_{12}, G_{13}) \) measure counting events in the detectors correlated with fission events in the source. Consequently, the source-detector cross-spectra indicate the amount of source-induced fission occurring in the system analyzed. The detector cross-spectra \( (G_{ij}) \) correlate events in one detector with events in another detector, so their magnitudes indicate the amount of both source-induced and inherent fission occurring in the system analyzed. Finally, the detector autospectra \( (G_{11}, G_{22}, G_{33}) \) are a measure of the source-induced and inherent fission rate of the system and the background rate.

Consequently, these spectral signatures characterize the fission system by separately measuring the system's response to induced fission and its response to inherent fission. The system's response to source-induced fission indicates the concentration of fissile materials like \( ^{234}\text{U} \) and \( ^{239}\text{Pu} \). The system's response to inherent fission indicates the concentration of spontaneously fissioning isotopes like \( ^{244}\text{Cm} \).

So, of particular interest in the application of this measurement to fissile mass assay of spent nuclear fuel is the source-detector cross-spectrum \( G_{12} \). Since the source-detector signature \( G_{12} \) correlates events in the detector with events in the source, its magnitude changes only in response to changes in the induced fission rate. Changes in the inherent fission rate only affect the uncertainty in the magnitude of the signature, and this uncertainty may be reduced by increasing the measurement time. Because the induced fission rate is indicative of the concentration of fissile materials, the magnitude of the source-detector cross-spectrum \( G_{12} \) directly indicates the fissile material content of the system. Consequently, this signature is ideal for fissile mass assay.

In implementation, a measurement system for fissile mass assay of spent fuel would reduce to include the source ionization chamber and instrumentation (signal \#1), a bank of neutron detectors (e.g., \( ^{3}\text{He} \) or fission chambers) and instrumentation (signal \#2), and a Fourier analyzer to acquire signals \#1 and \#2 and to estimate the source-detector cross-spectrum signature \( G_{12} \).

To assess the feasibility and to characterize the sensitivity of such a measurement, calculational models of the measurement were constructed for analysis by MCNP-DSP.

Calculation Tool: MCNP-DSP

MCNP-DSP\(^2\) is an ORNL modification of the code MCNP-4A\(^{10}\). The modification is an analog Monte Carlo code to simulate source-driven measurements like the one previously described. The DSP extension indicates that the modification implements digital signal processing algorithms to calculate the signatures that are acquired from the simulated measurement.

An MCNP-DSP calculation of a source-driven measurement essentially follows the natural chain of events. Source particle histories are initiated by \( ^{235}\text{Cf} \) fission, and these particles and their progeny are tracked to their extinction by

![Figure 1. \( ^{235}\text{Cf} \)-driven frequency-analysis measurement configuration](image-url)
absorption or escape. During the particles' histories, appropriate interactions with detectors initiate detection events, and the timing of events is tracked to generate time-dependent detector responses. The spectral signatures are then estimated from the time-dependent detector responses using standard Fourier DSP algorithms.11

MCNP-DSP has been validated against numerous measurements. For example, the code has been validated against measurements of uranium metal cylinders12 and storage vault arrays of uranium metal annular castings.13 Furthermore, the code has been validated against measurements of a single 17×17 pressurized water reactor (PWR) fuel assembly in 1510-ppm borated water performed in 1984 at the former Babcock & Wilcox critical facility in Lynchburg, Virginia.14 This last validation is directly relevant to the examination of the feasibility of applying the measurement to fissile mass assay of single moderated fuel assemblies.

**Calculational Model: 17×17 PWR Assembly**

Figure 2 illustrates the geometry of the MCNP-DSP models employed in this feasibility study. Central to the model is an isolated Westinghouse 17×17 PWR spent fuel assembly in a 1500-ppm borated water storage pool. The axial burnup profile of the spent fuel was represented using seven axial zones, each with isotopics estimated by the PDQ code.15 In the model, the measurement system consists of a 235U source adjacent to the fuel assembly exterior and a bank of five 1-in-diam x 1-ft-long 10-atm 3He detectors adjacent to the fuel assembly opposite the source. The source and detectors were surrounded by a 1-in-thick polyethylene moderator to improve the measurement efficiency. Four different fuel assemblies with average burnups from fresh to 32 GWD/MTU were modeled; the axial burnup profile of each spent assembly is plotted in Fig. 3.

For each of the four average burnup assemblies, measurements at each of the seven axial positions indicated in Fig. 2 were simulated by MCNP-DSP. The source-detector cross-spectrum signature for each of these 28 simulated measurements was estimated by MCNP-DSP.

**Results**

Figure 4 plots the average magnitude |G1| of the source-detector cross-spectrum signature versus the local concentration of fissile material (235U + 239Pu) in g/cm³. First note that the signature follows one trend at the middle positions (2 through 6) while it follows a different trend at the end positions (1 and 7). This difference occurs because at the end positions the detectors extend beyond the fuel so that they in effect "look at" transmission through the borated water as well as fission induced in the fuel. A more careful selection of measurement positions to ensure that the detectors do not extend beyond the fuel would eliminate the difference between the trends observed at the middle and end positions. Also note that the spread of the signatures acquired at the end positions is significantly larger than the spread of the signatures acquired at the middle positions, which follow the fitted trend very closely. The spread observed in the end position signatures occurs because at the top position (#7), the detectors also "view" the fuel expansion plenum, whereas at the bottom position (#1), the detectors view only the fuel and borated water. Again, this effect can be eliminated by a more careful selection of detector positions to ensure that the top position does not "see" a significant amount of the expansion plenum.

With these caveats in mind, note the high sensitivity of the signature to small changes in fissile material content. Over the range of fissile material concentrations shown, which spans an increase of roughly 0.25 g/cm³, the signature |G1| changes by roughly 250%. Consequently, since the observed trend is nearly linear over the range of concentrations investigated, the signature can be expected to double for each increase of ~0.1 g/cm³ in fissile material.

Furthermore, because the signature correlates detection events with source fission events, the signature is independent of inherent sources like 238Cm. However, the uncertainty in the measured signature is affected by the presence of inherent fission sources, and this uncertainty is reduced by increasing the measurement time. Table I summarizes measurement times to achieve 5% uncertainty in the estimated signature at a single position using 3He detectors. These measurement times were estimated assuming that the assembly analyzed was discharged from the core after three years' irradiation (~36 GWD/MTU burnup). For lower burnups the measurement times decrease significantly. Consequently, the time required to acquire the signatures plotted in Fig. 4 would be less than one minute using 3He detectors if the fuel assemblies were allowed to cool 10 years and a 20-µg 235U source were employed.

**Conclusions**
Figure 2. 17×17 PWR fuel assembly model

Figure 3. Axial burnup profile of spent fuel assemblies
Figure 4. Source-detector cross-spectrum magnitude $|G_{12}|$ versus local fissile material content

Table 1. Estimated measurement time in minutes to achieve 5% uncertainty in $|G_{12}|$ at one measurement position

<table>
<thead>
<tr>
<th>$^{252}$Cf source ($\mu$g)</th>
<th>Discharge</th>
<th>5 yr</th>
<th>10 yr</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>10.0</td>
<td>4.4</td>
<td>4.0</td>
</tr>
<tr>
<td>10</td>
<td>5.0</td>
<td>2.2</td>
<td>2.0</td>
</tr>
<tr>
<td>20</td>
<td>2.5</td>
<td>1.1</td>
<td>1.0</td>
</tr>
</tbody>
</table>

- Fuel irradiation: three years (36 GWh/MTU).
- Accounts for spontaneous fission and (α,n) sources.
- Assumes $^1$He detectors used.
This preliminary investigation indicates that $^{252}$Cf-driven frequency-analysis may be applied to perform fissile mass assay of single spent nuclear fuel assemblies in storage pools. The calculational results obtained during this feasibility study predict that the signature acquired in the proposed measurement will possess high sensitivity to small changes in fissile material content. Results indicate that the signature will increase by ~100% in response to an increase in fissile material of only 0.1 g/cm$^2$. Furthermore, this study indicated that the measured signatures may be acquired in less than one minute if 10-year-cooled fuel is analyzed using $^3$He detectors. A more extensive study is required to determine the applicability of $^3$He detectors in the strong gamma fields surrounding spent fuel assemblies. Earlier experiments with modified $^3$He detectors$^{16}$ have succeeded in operating these modified detectors in gamma fields as high as 200 R/h. These same experiments have shown that the modified detectors may be operated in gamma fields as high as 60,000 R/hr when the detectors are shielded by two inches of lead. The implementation of lead-shielded modified $^3$He detectors for the proposed spent fuel measurements should be investigated. Furthermore, further studies should also investigate measurements on other fuel assembly configurations in addition to the 17x17 PWR assemblies modeled in this study. However, in general, the results of this study indicate high potential for the practical implementation of $^{252}$Cf-driven frequency-analysis to perform fissile mass assay of spent nuclear fuel.

References


